Public Comments on Draft NPDES General Permit #WAG130000

Compiled February 6, 2023

This document includes all comments received during the public comment period for EPA's draft *NPDES General Permit for Federal Aquaculture Facilities and Aquaculture Facilities Located in Indian Country in Washington* (Permit Number: WAG130000) from September 7 through December 22, 2022.

Comments were received from:

- Columbia River Inter-Tribal Fish Commission
- Northwest Indian Fisheries Commission
- Tulalip Tribes
- U.S. Army Corps of Engineers
- Wild Fish Conservancy
- Inland Empire Paper Company

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COLUMBIA RIVER INTER-TRIBAL FISH COMMISSION

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October 8, 2022

Submitted via email

Sally Goodman, Environmental Engineer
U.S. Environmental Protection Agency
1200 6th Avenue, Suite 155
Seattle, WA 98101
epar10wd-npdes@epa.gov

RE: Columbia River Inter-Tribal Fish Commission Public Comments on EPA proposed NPDES Permit WAG130000

Dear Ms. Goodman:

The Columbia River Inter-Tribal Fish Commission (CRITFC) supports the proposed NPDES General Permit #WAG130000 with the inclusion of Fish Sampling Programs at Dam Fish Passage Facilities. The permit will allow discharge of water that has been treated with fish anesthetic (Aqui-S20E). This anesthetic is an essential tool in valuable research programs that CRITFC and other fishery co-managers conduct at the facilities.

CRITFC serves the four Columbia Basin treaty fishing tribes – the Confederated Tribes and Bands of the Yakama Nation, the Confederated Tribes of the Warm Springs Reservation of Oregon, the Confederated Tribes of the Umatilla Indian Reservation, and the Nez Perce Tribe. CRITFC works to protect the treaty-reserved fishing rights of its member tribes and conducts research that informs fish restoration and management. These dual goals collectively ensure the preservation of fisheries resources that are critical to the culture and identity of the member tribes. CRITFC runs an essential salmon and steelhead sampling program at Bonneville Dam that is necessary for tribal co-management of the salmon and steelhead resources, the implementation of the U.S. Canada Pacific Salmon Treaty, *U.S. v. Oregon* Harvest Management Agreement for the Columbia River Basin, the 2019 Columbia River System BiOp (formerly FCRPS BiOp), as well as other research needs.

CRITFC's program at the Bonneville Dam Adult Fish Facility samples salmon and steelhead. Fish are anesthetized in Aqui-S20E, examined, measured, tissue and scale sampled, tagged, and released. The information collected is used in spawning escapement monitoring, assessing indicator stocks, population estimates, spawner recruit relationships, developing harvest management approaches, and ESA status trends monitoring and reporting.

Aqui-S20E, an anesthetic covered under the U.S. Fish and Wildlife Service's Investigational New Animal Drug program, is the only conditionally approved immediate-release fish anesthetic available. Immediately releasing anesthetized fish is crucial for this program since the sampling occurs during active tribal and non-tribal fisheries. The updated General Permit #WAG130000 would allow NPDES coverage for the discharge of Aqui-S20E treated water.

Please contact John Whiteaker, fisheries scientist, at whij@critfc.org with follow-up questions or comments.

Sincerely,

Aja K. DeCoteau
Executive Director

Northwest Indian Fisheries Commission
6730 Martin Way E., Olympia, Washington 98516-5540
Phone (360) 438-1180 www.nwifc.org FAX # 753-8659

December 15, 2022

Dan Opalski, Director
Office of Water and Watersheds
U.S. Environmental Protection Agency, Region 10
1200 Sixth Avenue, Suite 155
Seattle, WA 98101

Re: Public Comments for WAG130000. Comments on Draft NPDES General Permit for Federal Aquaculture Facilities and Aquaculture Facilities Located in Indian Country

Dear Mr. Opalski:

Thank you for the opportunity to provide comments on the National Pollution Discharge Elimination System (NPDES) General Permit for Federal Aquaculture Facilities and Aquaculture Facilities located in Indian Country within the bounds of Washington State (Permit WAG130000). The following comments represent the concerns of the Northwest Indian Fisheries Commission and its member tribes, which are affected by the issuance of the permit.

We appreciate that EPA incorporated many of the comments we provided regarding the preliminary draft permit. We wish to draw attention to several issues we raised but were not addressed or incorporated in the draft permit.

General Comments

NWIFC member tribes operate hatcheries to support tribal fisheries and maintain the treaty right to harvest fish, which have been severely curtailed due to habitat loss and degradation. These facilities are valued by the tribes for the cultural, ecological, subsistence and commercial benefits they provide. Tribal hatcheries are important to all Washingtonians as they support the rearing and releasing of salmon and steelhead for harvest by Indian and non-Indian fisheries in the U.S. and Canada. The majority of salmon harvested in western Washington come from hatchery production, which is a necessary tool due to the lack of natural production.

Tribal hatchery facilities are regulated by EPA under the NPDES permits, as well as by state water quality standards where hatcheries discharge to state waters. This dual state and federal regulation of tribal facilities underscores the importance of EPA's trust responsibility as it relates to the issuance and enforcement of the permit. It is also worth noting that tribes with water quality standards are the appropriate regulating entities for discharges into tribal waters covered by tribal water quality standards. This fact should be expressly stated in the permit.

Tribal hatchery programs operate under tight budget constraints with limited staff. Compliance with the permit includes expenses for such activities as transporting samples and laboratory analysis. These expenses can be burdensome for the smaller tribal facilities, which are often located a great distance from accredited labs. EPA should address these circumstances when crafting the final permit language.

Specific Comments

In our response to the preliminary draft, we requested that EPA remove the phrase "as determined by the Department of Ecology" within the definition of "Toxic Substances." EPA responded by replacing "As determined by Ecology" to specifically reference WAC173-201A-240. This change does not address scenarios where tribal hatcheries discharge into tribal waters where tribal water quality standards exist. As you are aware, several of our member tribes have promulgated water quality standards for water bodies on their reservations. Therefore, we reiterate our previous request to make specific reference to tribal water quality standards as the applicable regulatory authority where it exists.

A final point in our response to the preliminary draft, we identified inconsistencies between the penalty structure in the Washington State Department of Ecology General Permit, which covers state facilities (oftentimes located within the same watershed as tribal hatcheries) and this permit. The penalty structure under the state permit is strikingly different, with lower penalties and different language than described in the EPA draft permit. We again question why this higher standard is being applied to tribal and federal hatcheries, and how EPA plans to address this disparity.

Thank you for the opportunity to comment on this draft permit. For additional information, please contact Marissa Paulling at mpaulling@nwifc.org. We look forward to our continued cooperation in the operation of tribal hatchery facilities.

Sincerely,

Justin R. Parker
Executive Director

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Tulalip, WA 98271-9694
360-716-4500
Fax 360-716-0628

November 22, 2022

Via Electronic Mail

Daniel Opalski, Director
Office of Water and Watersheds
U.S. Environmental Protection Agency, Region 10
1200 Sixth Avenue, Suite 155
Seattle, WA 98101

Dear Mr. Opalski,

The Tulalip Tribes ("Tulalip") writes this letter regarding the *Draft NPDES General Permit for Federal Aquaculture Facilities Located in Indian Country, within the Boundaries of Washington State, NPDES Permit No. WAG13000* ("*Draft Permit*") drafted by the United States Environmental Protection Agency, Region 10 ("EPA") for Tulalip's aquaculture programs. After reviewing the *Draft Permit*, Tulalip has concerns with two definitions and propose edits to address these concerns. To the extent government-to-government consultation is necessary to resolve these issues, then Tulalip hereby requests such consultation.

First, the definition of "toxic pollutants" on page 58 (pdf) of the draft permit is the definition from the Clean Water Act ("CWA") § 502(13). This definition alone reads quite broadly because it does not recognize that the CWA and accompanying regulations only apply to "the list of toxic pollutants or combination of pollutions to [the CWA] shall consist of those toxic pollutants listed in the 40 C.F.R. 401.15. CWA § 307(a)(1). Accordingly, Tulalip requests that the following sentence immediately follows the current definition in the *Draft Permit*:

> The list of applicable toxic pollutants or combination of pollutants shall consist of those toxic pollutants listed in 40 C.F.R. 401.15.

Second, based on feedback from several tribes regarding the definition of "toxic substances", the EPA proposed to replace reference to the Washington State Department of Ecology ("Ecology") with a reference to Ecology's water quality standards for toxic substances as described in WAC 173-201A-240. For the same reason that Tulalip and other tribes took issue with the reference to Ecology, Tulalip seeks to avoid citing to WAC 173-201A-240. To that end, Tulalip proposes that the substance of WAC 173-201A-240 be included as an appendix, which is referenced in the definition of toxic substances.

Further, Tulalip has Treatment as a State ("TAS") authority under the CWA and Tulalip's water quality standards are currently being evaluated by the EPA. It is Tulalip's understanding that its water quality standards are likely to be fully promulgated before the expiration of Tulalip's next NPDES permit. Accordingly, Tulalip proposes the following definition for toxic substances in the *Draft Permit*:

> Toxic substances - Substances that when discharged above natural background levels in waters of the state have the potential either singularly or cumulatively to adversely affect characteristic water uses, cause acute or chronic toxicity to the most sensitive biota dependent upon those waters, or adversely affect public health as described in Appendix __ or in the Tulalip Tribes' water quality standards that are hereafter approved by the EPA.

Tulalip is hopeful that the EPA will agree to make these narrow changes to the *Draft Permit*. Please reach out to Mike Crewson at 360-222-2690 or 425-754-0955 if you have any questions or concerns.

Sincerely,

Jason Gobin, Natural & Cultural Resources Director, Tulalip Tribes

cc: Mike Crewson, Salmon Enhancement Scientist, Tulalip Tribes

DEPARTMENT OF THE ARMY
U.S. ARMY CORPS OF ENGINEERS, NORTHWESTERN DIVISION
PO BOX 2870
PORTLAND, OR 97208-2870

22 December 2022

SUBJECT: USACE Comments to EPA's DRAFT Proposed NPDES Permits for Federal Aquaculture Facilities and Aquaculture Facilities Located in Indian Country (#WAG130000).

Sally Goodman
Environmental Engineer, NPDES Permitting Section
U.S. EPA, Region 10
1200 6th Ave, Suite 155 (19-CO4)
Seattle, WA 98101

Dear Ms. Goodman:

On behalf of the U.S. Army Corps of Engineers ("Corps") Portland District, I submit the following comments on the U.S. Environmental Protection Agency's ("EPA") draft National Pollutant Discharge Elimination System ("NPDES") permits for the Corps' Federal Aquaculture Facility and Aquaculture Facilities Located in Indian Country. An example of the permit is as follows:

- Bonneville Lock and Dam, Adult Fish Facility (#WAG130000)

On 6 September 2022, EPA issued proposed draft NPDES permits for public comment to authorize the discharges from federal aquaculture facilities. The Corps requests EPA's consideration of the following comments by submittal of this letter.

Comment 1
Part II.B.2.c (Washington Hatchery General Permit)

"Dam fish passage activities" should be referred to as such consistently throughout the document. In some sections the dam fish passage activities are referred to as "facilities" and in others they are described such as "fish passage facilities" that "contain grow, or hold aquatic animals in tanks, or similar structures, which discharge water treated with Aqui-S20E . . . ." this could lead to confusion as the permit and notice of intent (NOI) are implemented. Additionally, the permit conflicts with the NOI as the NOI states that '"fish sampling programs at federal hydroelectric dams that result in discharges of water treated with Aqui-S20E" [be] considered research facilities'; however, Part II.B.2 establishes a separate "Dam Fish Passage" activity that specifically includes sampling programs where discharges of Aqui-S20E are occurring. EPA appears to be confusing or conflating physical facilities with permitted activities that result in a discharge. The Corps owns and operates fish passage "facilities" where Corps personnel and third parties carry out permitted activities that result in point source discharges. The Corps requests EPA clearly treat fish passage facilities separate and apart from those activities (occurring at those facilities) that result in discharges. In short, the NPDES permit should appropriately distinguish owners from operators, and facilities from activities.

Comment 2

Part II.B.2.c (Washington Hatchery General Permit)

Under permit requirements for fish sampling as part of dam fish passage activity at an aquaculture facility, the document seems to restrict discharges to only water treated with "Aqui-S20E" and exclude other fish anesthetics. The Corps requests that EPA consider extending the permit to apply to discharges of Aqui-S20E and other fish anesthetics.

Comment 3
Part II.E.1.j (Washington Hatchery General Permit)

Limitations to permit coverage explains that discharges of water treated with Aqui-S20E are not covered if other non-discharge disposal options are "feasible." The Corps requests EPA either define "feasible" or incorporate a definition of "feasible" from other regulatory context.

Thank you for the continued and positive coordination. The Corps appreciates the opportunity to submit these comments for consideration. We look forward to continuing to work closely with EPA on the draft NPDES permits for Federal Aquaculture Facilities and Aquaculture Facilities Located in Indian Country. If you have any questions regarding the comments above, please contact Mr. Carson Freels at 503-808-4323.

Sincerely,

for
Kymberly C. Anderson
Chief, Operations Division
Portland District, USACE

Wild Fish Conservancy
NORTHWEST
SCIENCE EDUCATION ADVOCACY

Wild Fish Conservancy offers the following comments on the draft NPDES General Permit for Federal Aquaculture Facilities and Aquaculture Facilities Located in Indian Country in Washington (WAG130000) proposed by the US Environmental Protection Agency (EPA).

Summary:

Overall, Wild Fish Conservancy believes that EPA should open a new public comment period after completing ESA Section 7 consultation, because EPA identifies a number of topics that they plan to re-evaluate. We believe that chemical use, especially the use of formalin, has greatly increased in some hatcheries and that EPA's assumptions in its 2015 Biological Evaluation may no longer be valid.

Nitrogen: The nitrogen loading of some of these facilities may be similar to municipal wastewater treatment plants, and those discharging to impaired waterways should have permit conditions similar to facilities subject to the Puget Sound Nutrient General Permit. Effluent limitations should be consistent with the developed Total Maximum Daily Load (TMDL) that was prepared for the receiving water.

Biological agents: Through effluent discharges and release of diseased fish, hatcheries (and perhaps some research facilities) are discharging microbes (e.g, fungi, bacteria, protozoans, viruses) that can adversely affect wild fish and the surrounding aquatic environment. Unnaturally high densities of confined animals are known to result in amplification of such microbes. EPA should place conditions into this permit requiring monitoring and subsequent reporting as a first step, then effluent limitations on microbial agents if necessary. In addition, the requirement for a facility operator to develop "procedures" to "minimize" the release of diseased animals runs contrary to a common hatchery practice to release fish early in the event of a disease outbreak. The permit should prohibit the transfer or release of diseased fish.

Chemicals/drugs: In part due to warming waters and elevated fish densities, disease outbreaks at hatcheries have gotten worse and hatchery operators have countered with an increase in chemical and drug use. The assumptions on chemical and drug use that EPA made in its 2015 risk assessment (included in the ESA biological evaluation) are no longer valid, and EPA needs to evaluate current hatchery practices. EPA should again solicit public comments on this permit after a re-evaluation and completion of ESA consultation.

Facility effects: EPA should work with Ecology and the operators of the aquaculture facilities to ensure that all activities of the facilities protect aquatic life and comply with Washington's water quality standards. We offer more detail below.

Nitrogen and other Pollutants

EPA has extended considerable support to the Washington Department of Ecology (Ecology) in modeling dissolved oxygen variations in Puget Sound. Modeling results have shown that anthropogenic inputs of nitrogen exacerbate violations of the applicable water quality criterion for dissolved oxygen, and that the effects of nitrogen discharges are "far-field"; that is, the nitrogen discharged may cause violations distant from the discharge point.

Ecology has recently issued a "Puget Sound Nutrient General Permit" (PSNGP) to control nitrogen inputs into Puget Sound. The targets of the permit are the fifty-eight publicly owned domestic wastewater treatment plants (WWTP) discharging into Washington waters of Puget Sound (WWTPs on federal or tribal lands are not covered, but some EPA-permitted facilities have received Clean Water Act Section 401 certifications from Ecology that include conditions identical to provisions of the PSNGP).

We appreciate and support that EPA, in this permit, recognizes that aquaculture facilities can be responsible for significant nutrient loads to waterbodies by requiring monitoring for nutrients when the facility discharges into impaired waters. Table 2 in Part IV of the Fact Sheet, however, does not include the areas of Puget Sound which are in Category 5 (waters requiring a TMDL) for dissolved oxygen.

Facilities covered under this general permit that discharge to Puget Sound should monitor and report the concentrations of nitrogenous compounds and $CBOD_5$ in their effluent, consistent with the requirements found in Ecology's Puget Sound Nutrient General Permit.

Similarly, other chemicals, such as polychlorinated biphenyls (PCBs) are known to be present in fish food and discharged through hatchery facilities. The State of Washington has been working on draft PCB reduction plans within their hatchery facilities, but we know of no limits that have been placed on discharges. As facilities are increasing these types of discharges associated with increased hatchery production, or changes in food sourcing, it is appropriate to analyze that information and present the findings to the public before re-issuing this general permit.

Biological agents

Fish hatcheries propagate and grow fish in conditions and at densities far more extreme than anything found in nature, and factors such as temperature, other water quality parameters, feeding regimes (to name only a few) are controlled as much as practical by hatchery operators. Despite this, the very high densities of confined fish that hatcheries routinely operate at are perfect incubators for disease-causing organisms. Wild Fish Conservancy staff have been researching how aquaculture facilities "amplify" disease-causing organisms, and through the discharge into public waters, have facilitated their spread. And these are not just disease-causing organisms native to the Pacific Northwest, but exotic ones as well brought in through the aquaculture industry. Fish disease-causing organisms can be released, and distributed to infect biota in the natural environment, in the effluent discharged by a facility as well as through the release of diseased hatchery fish.

There are no numeric criteria for fish disease-causing organisms, but narrative criteria found in Washington's water quality standards (WAC 173-201A-260(2)(a)), prohibit the discharge of "deleterious material," and apply to all fresh and marine waters of the state.

Facilities discharging these disease-causing organisms should, at a minimum, be required to periodically monitor their effluent. A panel of fish-health and water-quality experts should be assembled to develop appropriate limits for these facilities.

Control of biological agents through water quality standards is not unheard of, and in fact Washington's water quality standards include a numeric criterion for *E. coli* to protect human health in waters designated for the primary contact recreation use. There is no reason why an effort cannot be made in this case to control the discharge of biological agents deleterious to wild fish, many of which are also protected under the Endangered Species Act. EPA needs to include an analysis of the discharges of disease-producing organisms in its ESA Section 7 Biological Evaluation.

Section VI.B.4.(a)(v)(h) states: "[p]rocedures must be implemented to minimize the release of diseased aquatic animals from the facility." Wild Fish Conservancy believes that this provision needs to include a *prohibition on the release of diseased fish*. It is common hatchery practice to prematurely release fish (e.g., salmon smolts) earlier than originally intended if a disease outbreak occurs. For the most part, hatcheries are unconcerned with receiving waters or wild native fish, except insofar as they affect hatchery operations (e.g., the common hatchery practice of restricting migration upstream of the hatchery in disregard of the needs of wild fish or the ecosystem). A threshold of disease incidence in a raceway or tank, when exceeded, should require hatchery operators to euthanize and properly dispose of all infected fish in the affected raceway or tank.

Chemicals/drugs

The other side of the coin of amplification of disease-causing organisms is an increase in the use of chemicals and drugs. They are not used to eliminate the discharge of these organisms to receiving waters but instead to keep infection rates inside the hatchery to an acceptable level. This draft General Permit, like its predecessor, has provisions for reporting usage of chemicals and drugs, through the annual report submitted by the facility's operator, but neither this permit nor its predecessor provides any constraints. Chemicals and drugs must be used in accordance with their labels, unless "extra-label" use is approved by a veterinarian, or it is considered an Investigational New Animal Drug (INAD). Amounts have to be reported, but there are no considerations regarding ambient receiving water conditions such as flow or temperature.

EPA conducted a ESA Section 7 Biological Evaluation (BE) in 2015 that included a risk assessment for some of the chemical quantities commonly used in hatcheries. EPA and the Department of Ecology also studied formalin discharges at selected hatcheries in 2016 (the study was published in 2017). Our evaluation of state hatcheries indicates a sharp increase in the use of these chemicals, especially formalin, and the assumptions on chemical use employed in the risk assessment found in EPA's 2015 Biological Evaluation may not reflect our more recent observations of production facilities in Washington and Oregon.

In the Fact Sheet (p. 72) EPA states

> An additional focus of this re-initiation will be confirming that data collected during the previous permit cycle regarding fish drug and chemical discharge concentrations does not exceed the conservative assumptions used in the risk assessments conducted in the development of the 2016 BE.
>
> EPA will reinitiate a focused consultation in coordination with the Services and intends to complete ESA consultation with the Services prior to issuance of this General Permit.

This implies that EPA has not yet examined the drug and chemical use of the past few years before releasing this draft permit for public comment. WFC has reason to believe, however, that the assumptions in both the BE and the 2016 study are no longer valid, as chemical use has changed, along with the changing climate.

WFC submitted a Freedom of Information Act request for drug and chemical use by holders of the current permit, but an expedited processing request was denied and we did not receive adequate information in time to inform these comments. Once we receive these records, we will be sure to make them available for the purposes of this permitting process.

The FDA label for at least one brand name of commonly used formalin, "Parasite-S" has changed since the BE and the 2017 study. The original language in the section entitled "Environmental Precautions" stated

> Do not discharge the contents of fish treatment tanks into natural streams or ponds without thorough dilution (greater than or equal to 10X). Do not discharge the contents of egg treatment tanks without a 100X dilution. This will avoid damage to PARASITE-S sensitive phytoplankton. zooplankton, and fish populations and avoid depletion of dissolved oxygen.

The current label[1] states

> Do not discharge effluent containing this product into lakes, streams, ponds, estuaries, oceans, or other waters unless in accordance with the requirements of a National Pollutant Discharge Elimination System (NPDES) permit and the permitting authorities are notified in writing prior to discharge. Notify the NPDES authority that water quality benchmarks for the protection of freshwater aquatic life have been derived for formaldehyde by Hohreiter and Rigg, 2001 (Chemosphere, 45:471-486) following EPA guidelines. The acute benchmark value for formaldehyde is 4.58 mg/L (12.4 mg formalin/L). The chronic benchmark value is 1.61 mg/L (4.35 mg formalin/L). Water quality benchmark concentrations are not discharge limits, but may be used by the NPDES authority to derive such limits for the permit.

[1] https://syndel.com/wp-content/uploads/2019/01/P-S-Package-Insert-ver.-030519-1-1.pdf Accessed November 29, 2022. Concerned individuals found identical language on a drum of Parasite-S being used for treatment at a WDFW Hatchery, and documented it with a photo on August 26, 2022.

It is understandable that the manufacturer changed the label, as the older label implied that discharge was allowed provided the water was sufficiently diluted but legally, an FDA label cannot serve as a substitute for an NPDES permit. Unfortunately, the draft NPDES permit requires nothing except annually reporting formalin use. There is nothing constraining unlimited use and discharge of formalin if the "extra-label" use is sanctioned by a veterinarian. Unfortunately, the concern of the veterinarian is the health of the hatchery fish, and not the condition of the receiving water.

Formalin use has changed significantly over the life of the current permit. We compared formalin use as documented in the 2017 EPA sampling study to more recent annual reports in some select hatcheries tested in the previous biological evaluation:

Facility	Formalin use (gallons/yr) from 2017	Formalin use (gallons/yr) from more recent annual reports (yr)
Kalama Falls Hatchery	1554 (2017)	4370 (2020) 5105 (2021)
Wallace River Hatchery	843 (2017)	2214 (2021) 3072 (2020)
Priest Rapids Hatchery	3493 (2017)	4722 (2018) 4435 (2019) 3044 (2020)
*Many of the National Fish Hatcheries have no data in Washington Department of Ecology's PARIS database.		

It appears that EPA considered recent formalin use to be sufficiently similar to past use because the Fact Sheet refers to the BE and the 2017 study and implies that the conclusions still hold as far as the chemicals that were assessed. EPA and the Services are going to reinitiate consultation, however, because, in part, other chemicals are being considered. We urge EPA to re-evaluate formalin because WFC believes that formalin has the potential to be used in much greater amounts than was the case when the BE and 2017 study were developed. We also urge EPA to again solicit public comments after this re-evaluation and completion of ESA consultation on this draft permit.

In addition to increased use of formalin, hatcheries in Washington have increased their use of other drugs and chemicals. For example, in 2018 the Kalama Falls Hatchery (WDFW) used 992.5 lbs of feed containing 4.0 g terramycin/lb, but increased its use to 1125 lbs in 2020.

Again, we believe EPA needs to re-evaluate all chemical and drug use, complete ESA Section 7 consultation with the Services, and then re-solicit public comment afterwards.

Facility effects

We provided comments to Ecology on their Clean Water Act Section 401 certification of this permit, urging them to condition the permit to include evaluations of the "facility effects" of these aquaculture facilities (sent to EPA under separate cover). Examples of facility effects include but are not limited to disruption of native fish migration, dewatering of stream reaches, and competition between native fish and hatchery fish (after release). We urge EPA to support our request and work with Ecology and the operators of the aquaculture facilities to ensure that the facilities protect aquatic life and comply with Washington's water quality standards.

I appreciate the opportunity to comment on behalf of Wild Fish Conservancy. Please contact me at the provided email address if you have any questions.

Sincerely,

Emma Helverson
Executive Director
emma@wildfishconservancy.org

Appendix 1 – Commonly used drugs and chemicals in salmonid hatcheries from the Mitchell Act Biological Opinion (NMFS Consultation Number: NWR-2014-697, 2017)

16 **TABLE 3-35. PROPERTIES OF CHEMICALS COMMONLY USED AT HATCHERY FACILITIES.**

CHEMICAL	HAZARD RANK[1]	LD_{50} (MG/KG)[2]	SKIN OR LUNG IRRITANT	CARCINOGENIC RATING[3]
DISINFECTANTS				
Chloramine T	1	935_{rat}	Corrosive to skin and respiratory irritant	N/A -- N/A[4]
Formalin	2	100_{rat}	Skin and respiratory irritant	1 -- B1
Hydrogen Peroxide	1	$>2{,}000_{mouse}$	Mildly irritating to skin or lungs	3 -- N/A
Iodophor	0	$10{,}000_{rabbit}$	Skin irritant	N/A -- N/A
Quaternary Ammonia (Hyamine)	2	350_{rat}	Skin and respiratory irritant	N/A -- N/A
Chlorine (Sodium Hypochlorite)	0	$5{,}800_{mouse}$	Skin and respiratory irritant	3 -- N/A
THERAPAUTICS				
Amoxicillin	N/A	N/A	Skin irritant	N/A -- N/A
Erythromycin	0	$9{,}272_{rat}$	Mild skin, eye, and respiratory irritant	N/A -- N/A
Florfenicol	1	800_{rat}	Mild skin, eye, and respiratory irritant	N/A -- N/A
Oxytetracycline (terramycin)	0	$7{,}200_{mouse}$	Mild skin, eye, and respiratory irritant	N/A -- N/A
Penicillin	N/A	N/A	Skin irritant	N/A -- N/A

TABLE 3-35. PROPERTIES OF CHEMICALS COMMONLY USED AT HATCHERY FACILITIES (CONTINUED).

CHEMICAL	HAZARD RANK[1]	LD_{50} (MG/KG)[2]	SKIN OR LUNG IRRITANT	CARCINOGENIC RATING[3]
Potassium Permanganate	1	750_{rat}	Skin, eye, and respiratory irritant	N/A -- N/A
Romet®	1	665_{rat}	Skin, eye, and respiratory irritant	N/A -- N/A
Sulfamethoxazole Trimethoprim	0	$5{,}513_{mouse}$	Skin irritant	N/A -- N/A
ANESTHETICS				
Benzocaine	N/A	N/A	NA	N/A -- N/A
Tricaine Methanesulfonate (MS-222)	N/A	N/A	Skin, eye, and respiratory irritant	N/A -- N/A
PESTICIDES/HERBICIDES				
2,4-Dichlorophenoxyacetic Acid	2	275_{rat}	Skin, eye, and respiratory irritant	2B -- N/A
2-Butoxyethyl 2,4-Dichlorophenoxy Acetate	1	831_{rat}	Skin, eye, and respiratory irritant	2B -- N/A
Chelated Copper	N/A	N/A	Skin, eye, and respiratory irritant	N/A -- N/A
Dichlobenil	1	$2{,}126_{rat}$	Mild skin and respiratory irritant	N/A -- N/A
Diquat	2	130_{rat}	Skin, eye, and respiratory irritant	N/A -- N/A
Endothall	3	$>38_{rat}$	Skin, eye, and respiratory irritant	N/A -- N/A
Fluridone	0	$>10{,}000_{rat}$	Mild skin and respiratory irritant	N/A -- N/A
Glyphosate	1	$1{,}568_{mouse}$	Skin, eye, and respiratory irritant	N/A -- D
Rotenone	2	60_{rat}	Skin, eye, and respiratory irritant	N/A -- N/A
MISCELLANEOUS				
Alcohol Anhydrous (ethyl alcohol)	1	$3{,}450_{mouse}$	Skin, eye, and respiratory irritant	N/A -- N/A
Lime (calcium hypochlorite)	1	850_{rat}	Skin, eye, and respiratory irritant	3 -- N/A
Salt (NaCl)	1	$3{,}000_{rat}$	Mild eye, irritant	N/A -- N/A
Sodium Thiosulfate	N/A	N/A	Skin, eye, and respiratory irritant	N/A -- N/A

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December 21, 2022

Via E-mail (epar10wd-npdes@epa.gov)

Ms. Sally Goodman
Region 10
U.S. Environmental Protection Agency

Re: Public Comments on WAG13000: Draft NPDES General Permit for Federal Aquaculture Facilities and Aquaculture Facilities Located in Indian Country within the Boundaries of the State of Washington

Dear Ms. Goodman:

Inland Empire Paper Company (IEP) appreciates the opportunity to provide these comments to the draft federal hatchery permit for the state of Washington. The conditions of the draft permit as applied to Washington State Aquaculture Facilities, WAG13000, are important to the work of IEP and others to reduce toxic loading to the Spokane River. With respect to toxic loading to the Spokane River, these comments extend to any and all hatcheries covered under the permit that are located within the Spokane River watershed and to those that may be the source of fish stocked in the Spokane River watershed.

It is unacceptable that EPA is proposing to delete all PCB monitoring requirements in the permit based on one limited study of PCB impacts from a state trout hatchery that is not regulated under the permit. Draft Fact Sheet, 54-55. This approach ignores extensive scientific research substantiating that Tribal and federal hatcheries in Washington are significant sources of fish tissue concentrations of PCBs. It is hypocritical of EPA to encourage extensive sampling and monitoring using an unpromulgated test method for other dischargers to the Spokane River watershed (included in the EPA comments on the draft 2022 NPDES permits on the Spokane River) and then disregard similar requirements for Tribal hatcheries, which are known and significant sources of PCBs. Finally, the approach in this permit is directly contrary to the position taken by the Spokane Tribe of Indians regarding the need for robust monitoring and control of PCBs discharged to the Spokane River.

IEP has been addressing Polychlorinated Biphenyls (PCBs) through its NPDES permit and, as required under the permit, as a member of the Spokane River Regional Toxics Task Force (Task Force). IEP was also a party to an appeal of the Washington Department of Fish and

Wildlife (WDFW) Permit for its Spokane Hatchery under permit number WAG137007. That appeal resulted in the issuance of Administrative Order No. 13422, which required WDFW to engage in more specific efforts to address PCBs.

By way of background, IEP operates a pulp and paper mill discharging to the Spokane River. IEP does not produce or generate PCBs, but receives them as a consequence of inks and pigments contained on the recycled paper coming into its facility. These inks and pigments contain inadvertent PCBs (iPCBs) allowable under the Toxics Substance Control Act (TSCA) at levels billions of times higher than the water quality standards for WA State. IEP has the most advanced wastewater treatment system in the pulp and paper industry, including being the first to use membrane technology to treat its effluent. This state-of-the-art treatment system removes and destroys over 99% of iPCBs entering the facility, thus eliminating them as a continued source to the ecosystem. Even with all this investment into state-of-the-art technology and efficient removal and destruction of iPCBs, the facility falls short of attaining the water quality standards, jeopardizing the future of paper recycling. Despite the best efforts of IEP and others, EPA has refused to review its TSCA authorizations for iPCBs that continue to be a source to the environment and an impediment to attaining the water quality standards.

EPA has placed an unfair burden on the Spokane River watershed to address a problem that is of EPA's creation. The draft hatchery permit further exacerbates this effort by discounting the substantial loading of PCBs associated with hatcheries. The great irony is that IEP removes over 99% of PCBs through its treatment system in a manner that also eliminates these PCBs from the environment. The remaining PCBs in IEP's effluent are typically lower molecular weight congeners that are soluble and cannot be removed by advanced filtration systems. However, these lower molecular weight congener PCBs are not bio-accumulating in fish tissue as shown from Spokane River fish tissue studies. In contrast, the draft federal permit for Aquaculture Facilities Located in Indian Country gives hatcheries a pass for releasing fish with PCB concentrations that violate both the state and proposed federal PCB criteria, and allows effluent discharges that will likely violate WA State's water quality standards and most certainly the Spokane Tribe's own water quality standards.

1. Substantial scientific research documents the impact of hatcheries on PCBs in Washington waters.

It is well documented that tribal and federal fish hatcheries discharge a significant percentage of the annual PCB loading to Washington waters. Ecology has identified hatcheries as a significant source of PCB loading to waters of the state and has estimated that as much as ten percent of annual PCB loading to Puget Sound is attributable to returning salmon.[1]. In 2011,

[1] Ecology, Control of Toxic Chemicals in Puget Sound: Assessment of Selected Toxic Chemicals in the Puget Sound Basin, 2007-2011, at 93 (2011)(Ecology Pub. 11-03-055)(04297-4593).

Ecology calculated that returning salmon contribute up to 0.3 kg/yr based on PCB residues per whole-body fish ranging from 7 µg for pink salmon to 336 µg for Chinook salmon.[2]

Ecology has also acknowledged, in addition to the PCB loading from returning salmon, that PCB contaminated hatchery fish play a significant role in CWA Section 303(d) listings for PCBs.[3] Ecology concluded that hatchery fish "may contribute to impairment and, in some cases, may cause the bulk of impairment."[4]

A 2006 Ecology report on hatchery fish included an analysis of skin-on fillets of pre-release rainbow trout from 11 hatcheries with PCB concentrations ranging from <2.3 to 67 ng/g (wet weight) with an average of 13.0 ng/g (wet weight) PCBs.[5] Other researchers have found between 39 and 59 ng/g total PCBs in whole-body juvenile Chinook salmon from six west coast hatcheries.[6] The authors concluded, "contaminated salmon may be a significant source of toxicants in the environment and in the food chain."[7] A study of British Columbia hatcheries found on average 25.5 and 48.5 ng/g (wet weight) PCBs in Chinook smolts from two hatcheries and 34.9 ng/g (wet weight) in Coho smolts from a third (BC) hatchery.[8] An analysis of pre-release juvenile Chinook from eight hatcheries feeding on the Columbia River found whole body concentrations of PCBs ranging from 6.9 to 61 ng/g (wet weight), corresponding to 22 to 323 ng/g per fish (individual hatchery-specific average weights from 3.2 to 6.2 g).[9] An analysis of pre-release juvenile Chinook salmon from the Soos Creek hatchery on Puget Sound over a three year period found total PCB concentrations ranging from 10 to 50 ng/g (wet weight), corresponding to 90 to 125 ng PCBs per fish (fish weight ranged from 2.5-9.4 g).[10] NOAA

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[2] *Id.*

[3] Ecology, Persistent Organic Pollutants in Feed and Rainbow Trout from Selected Trout Hatcheries (April 2006)(Ecology Pub. No. 06-03-017)(04681-4732).

[4] *Id.* at 30.

[5] *Id.* at 29.

[6] L. Johnson *et al*, Contaminant Exposure in Outmigrant Juvenile Salmon from Pacific Northwest Estuaries of the United States, 124 ENVIRON. MONIT. ASSESS., 167-194 (2007)(04955-4982).

[7] *Id.*

[8] B. Kelly *et al*, Persistent Organic Pollutants in Aquafeed and Pacific Salmon Smolts from Hatcheries in British Columbia, Canada, 285 AQUACULTURE, 224-233 (2008).

[9] L. Johnson *et al*, Contaminant Concentrations in Juvenile Fall Chinook Salmon from Columbia River Hatcheries, 72 N. AMERICAN J. AQUACULTURE, 73-92 (2010).

[10] J. Meador *et al*., Bioaccumulation of Polychlorinated Biphenyls in Juvenile Chinook Salmon (*Oncorhynchus tshawytscha*) Outmigrating through a Contaminated Urban Estuary: Dynamics and Application, 19 ECOTOXICOLOGY, 141-152 (2010).

Fisheries has also documented the significant PCB concentrations in hatchery fish feed and in hatchery origin fish.[11]

Tribal and federal hatcheries are undoubtedly an increasing source of PCB loading to Washington waters. In 2010, the combined hatchery release in Washington was 229.5 million fish including 117.4 million Chinook salmon.[12] In 2015, the Northwest Indian Fisheries Commission reported that tribal hatcheries alone released 40 million salmon and steelhead.[13]

In light of this information, IEP requests that EPA respond to the following comments:

Comment 1: Why has EPA ignored the extensive scientific information regarding the loading of PCBs from hatcheries in developing the subject permit?

Comment 2: Does EPA agree that information in the studies referenced in this letter is relevant regarding PCB loading from hatcheries?

Comment 3: Does EPA contend that it is appropriate to rely on one weak study conducted by Ecology at a state hatchery rather than the extensive weight of evidence that hatcheries in fact have a substantial impact on PCB concentrations in fish tissue within Washington waters?

Comment 4: The data of PCB concentrations in Pacific Northwest hatcheries indicate levels of PCBs that exceed the Department of Ecology Fish Tissue Equivalent Concentrations for PCBs that would be a basis for section 303(d) impairment listings. How does EPA justify not requiring rigorous monitoring and Best Management Practice requirements to eliminate this loading of PCBs?

Comment 5: Can EPA confirm that releases from hatcheries governed by this permit will be in compliance with applicable Federal, State and Tribal water quality standards?

Comment 6: Can EPA explain why hatcheries governed by this permit that are known to be significant sources of PCBs do not have similar permit requirements and limits for PCBs similar to those being required by all other NPDES permitted discharges to the Spokane River?

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[11] NOAA Fisheries, Draft Environmental Impact Statement on Two Joint State and Tribal Resource Management Plans for Puget Sound Salmon and Steelhead Hatchery Programs, Appendix K: Chemicals Used in Hatchery Operations (July 2014)(04257-4273).

[12] C. Roheim *et al,* The Role of Hatcheries in North American Wild Salmon Production, The Great Salmon Run: Competition Between Wild and Farmed Salmon, TRAFFIC North America, World Wildlife Fund (January 2007), Table IV-1: Salmon Fry Releases by Species, Region, and Area, 2000, at 44 (06739-6752).

[13] Northwest Indian Fisheries Commission, Tribal Natural Resources Management, A Report from the Treaty Indian Tribes in Western Washington (2015), at 4 (06530-6545).

2. The Little Spokane River Fish Hatchery study does not support the permit decisions on the general permit.

EPA relies on the Little Spokane Fish Hatchery study to conclude that the hatchery contributes only a small percentage of PCB loading to the Spokane River. The study provides a limited snapshot in time of PCB loading, and there is no basis in the study for assessing the relative contribution of PCB loading based on the study results. Even that question is misleading as the real issue in the Spokane River is PCB concentrations in fish tissue. With advanced filtration, IEP is not discharging PCBs that are bio-accumulating PCBs. In contrast the Tribal hatcheries on the Spokane River and its tributaries are contributing PCBs in fish tissue. Even if the contribution is a fraction of the total PCB loading to the river, it may be the largest source of PCBs found in fish tissue.

Comment 5: Why has EPA cherry picked one weak and general study to make a permit decision on PCB limits and monitoring in the permit?

Comment 6: What has EPA done to assess the relative contribution of hatchery releases to PCB concentrations in fish tissue?

Comment 7: How can EPA assess the relative contribution of fish hatchery releases to fish tissue concentrations without rigorous tissue monitoring and characterizations by hatcheries?

Comment 8: How will EPA determine if the releases from Tribal hatcheries are not causing or contributing to violation of PCB water quality standards without monitoring?

3. EPA should consider and apply the Spokane Tribe of Indian comments on the IEP NPDES permit to the coverage of the Tribes facilities under the general hatchery permit.

In 2022 the Spokane Tribe of Indians made the following comments on the IEP draft NPDES permit that should be considered and included in the general hatchery permit:

Commenter: Chad McCrea - Comment T-1-1

> On behalf of the Spokane Tribe of Indians' Department of Natural Resources (Tribe), please accept these comments on the Washington State Department of Ecology's (Ecology) draft NPDES Permit for IEP Empire Paper Company. These comments do not, nor are they intended in any way to impact the Tribe's August 18, 1877 federally reserved water rights within the Spokane River ("River") which includes quantities necessary to carry out the purposes of the Spokane Indian Reservation ("Reservation") which include but are not limited to guaranteeing the Tribe access to fish for food within Tshimakain Creek and the Columbia and Spokane Rivers. This fishing purpose of the Reservation also includes water of a quality necessary to carry out that purpose. Unfortunately,

pollution that originates upstream of the Tribe's waters impacts the Tribe's citizens ability to safely exercise their fishing rights.

As you are aware, the Spokane Tribe received "treatment in the same manner as a state" (TAS) status under the Clean Water Act in 2002. The Tribe's first water quality standards were approved by the EPA in 2003. Since that time, attainment of the Tribe's water quality standards within its jurisdictional waters has been difficult, particularly in regard to toxins that bioaccumulate such as PCBs. Accordingly, the Tribe in 2013 intervened in a lawsuit1 over the lack of a PCB TMDL for the 303d listed sections of the Spokane River. On February 12, 2022 the Court entered a consent decree that binds EPA to develop a PCB TMDL for the Spokane River by no later than December 29, 2024. (Dkt. 253). The Tribe has a significant interest in ensuring that the PCB TMDL is developed and implemented to lead to the attainment of the Tribe's downstream water quality standards. Accordingly, these comments will primarily focus on what it views as shortcomings in these permits as they relate to PCBs.

For background, the Tribe's first EPA approved fish consumption rate ("FCR") in 2003 was 86.3 grams/per day. This resulted in a surface water quality standard of 3.37 pg/L for PCBs. The Tribe adopted new standards in 2010 based on a fish consumption rate of 865 grams of fish per day to recognize a subsistence quantity of fish consumption, along with 4 liters of water intake all calculated utilizing a 1/1,000,000 cancer risk rate. This resulted in a PCB surface water quality standard of 1.3 pg/L for total PCBs. These new standards were approved by EPA in December of 2013. [Washington State's current water column PCB standard is based on a 1/100,000 cancer risk rate and is 170 pg/L, which is currently subject to two lawsuits in the Western District of Washington]. Discharges from upstream NPDES permittees contribute to violations of the Tribe's water quality standards for PCBs and other parameters.

The Tribe recognizes the current loophole in the enforcement of PCB water quality standards eloquently described by Justice Gonzalez dissenting in Puget Sound Keeper v. Dep't of Ecology, et al., 191 Wn.2d 631, 646-653 (2018). This as interpreted by Ecology requires that enforcement monitoring for PCBs only be conducted with a method that cannot detect down to the water quality standards for PCBs, method 608. PCBs are currently an unenforceable limit in Ecology's view.

With that said, it is critically important that Ecology revise these draft permits to include appropriate monitoring for PCBs utilizing Method 1668 or an equal and similar method for all monitoring purposes. This is an appropriate use of Method 1668. Nw. Pulp & Paper Ass'n v. Dep't of Ecology, No. 55164-1-II, 2021 Wash. App. LEXIS 2970, at *7–8 (Ct. App. Dec. 14, 2021).

It is important that all discharges into the Spokane River be monitored for PCBs appropriately for three important reasons.

First, the PCB TMDL will be completed by the EPA and will include appropriate Waste Load Allocations (WLA) for PCBs. The data EPA uses to develop the WLAs should be the best quality possible to increase the PCB TMDL's effectiveness. Requiring the entities that discharge toxic pollution into the Spokane River to monitor their effluent at all discharge points will help gather the most relevant and current data and will in turn make the PCB TMDL more accurate.

Second, apart from the numeric limits for PCBs, Ecology has narrative limits that must be monitored which Method 1668 can assist with...Here, the Tribe is a downstream state (with a PCB water column standard of 1.3 pg/L) and Method 1668 monitoring of effluent can help provide data on whether this standard can be attained and maintained under the permit conditions.

Third, 40 C.F.R. Section 122.4(d) requires that: "No permit may be issued: (d) When the imposition of conditions cannot ensure compliance with the applicable water quality requirements of all affected States." Again, the Tribe is an "affected" State for purposes of the Clean Water Act and requiring Method 1668 for effluent monitoring will allow the Tribe and the EPA to better monitor the effectiveness of the permit conditions.

The Clean Water Act does not exempt Tribal and federal hatcheries from compliance with applicable water quality standards. As the Spokane Tribe of Indians comments suggest, it is not possible for EPA to assess the relative source of PCBs to fish tissue or assess the ability and means to reduce PCB fish tissue concentrations in the Spokane River without rigorous monitoring.

Comment 9: Does EPA agree with the Spokane Tribe of Indians that toxic pollution discharged from hatcheries covered under this permit in the form of effluent and fish releases should be monitored to gather the most relevant and current data for the PCB TMDL that EPA is developing?

4. IEP requests that EPA equitably include PCB limits, sampling, monitoring, Source ID Studies, and Best Management Practices for the federal hatchery permits similar to the requirements included for the WA NPDES permitted facilities on the Spokane River.

A. <u>The permit should require a procurement preference for low or non-PCB containing products including feed.</u>

WA State hatcheries are required to eliminate the use of PCB containing products including fish feed unless it is “not cost effective or technically feasible to do so.” RCW 39.26.280(2). This statute applies to all state agencies and prohibits the knowing purchase of products containing PCBs above the practical quantification limit absent such documentation.

Numerous studies have documented high concentrations of PCBs in fish feed. Regardless of whether the Food and Drug Administration (FDA) authorizes these concentrations, the use of such feed should be prohibited in the permit unless they can document the basis for not doing so under the statute. The permit should require for all of its hatcheries including the Spokane Tribal Hatchery, to prepare an assessment of alternative feed sources that do not contain PCBs or have reduced PCB levels.

B. The permit should require a PCB Source Identification Study.

Hatcheries covered under this permit that discharge to the Spokane River and its tributaries as well as hatcheries covered under the permit that release fish to the Spokane River should be required to develop a scope of work for a PCB Source Identification Study within two to three years of permit issuance that includes a list of raw materials used at the facility that may contain PCBs, a review of the facility identifying where PCB containing equipment was or may have been used, a sampling plan with proposed sampling locations, quality control protocols, sampling protocols, and PCB test methods. The PCB test method for its source identification study should have a target detection limit of 50 pg/L, similar to that required for all NPDES permitted discharges to the Spokane River. The permit should provide a deadline within the term of the permit for submission of the study once the scope of work is approved by EPA.

This condition is roughly the same as the PCB source identification study provisions in Washington and EPA NPDES permits on the Spokane River. All NPDES permitted facilities that have the potential to impact Lake Spokane and the Spokane River for PCBs should be subject to parallel conditions. If EPA deems this to be important work for individual NPDES permits, it is as equally important for hatcheries that discharge to the Spokane River as well as hatcheries that are a source of fish stocked within the watershed.

C. The permit should require a PCB BMP Plan.

The hatcheries covered under the permit that discharge to the Spokane River, its tributaries, or are a source of fish released in the river should be required to prepare and submit a PCB Best Management Practices Plan within the term of the permit that includes the following elements:

a. A list of members of a cross-functional team responsible for developing the BMP plan including the name of the designated leader.

b. A description of current and past source identification, source control, pollution prevention, and pollutant loading reduction efforts.

c. Preparation of a technical/economical evaluation of new BMPs. BMPs should include, but are not limited to: modification of equipment, facilities, technology, processes, and procedures; source control; remediation of any contaminated areas.

d. A schedule for implementing economically feasible BMPs.

e. Methods used for measuring progress towards the BMP goals and updating the BMP plan.

f. Results from testing of any waste streams, including all effluent from the hatchery, taken in support of the BMP plan and PCB Source Identification Plan.

g. Annual reports to EPA after submission of the BMP plan.

D. Spokane River Regional Toxics Task Force or future organization.

The permit should require all hatcheries covered under the permit that discharge to the Spokane River, its tributaries, or are the source of fish released in river to participate in the Spokane River Regional Toxics Task Force or future organizational structure on the same terms of every individual NPDES permitted facility on the Spokane River in Washington and Idaho. It is essential that the subject hatcheries work closely with everyone on the Task Force or its substitute to monitor, document, and reduce PCB loadings.

E. PCB monitoring.

The permit should require intake and effluent monitoring for all hatcheries covered under the permit that discharge to the Spokane River, its tributaries, or are the source of fish released in the Spokane River on the same terms as all other NPDES permits on the Spokane River. The permit should also require PCB monitoring of the effluent using EPA test method 1668C.

Hatcheries discharging or stocking fish in the Spokane River watershed should have an additional obligation to document the PCB levels in the fish feed used to rear the fish, PCB levels in the fish being stocked and the locations where the fish are being stocked. This information will be important to the work of the SRRTTF and future organizational structure in determining the loadings and sources of PCBs to the river.

I appreciate your consideration of these comments and invite EPA staff to contact me for further information and clarification.

Sincerely,

Douglas P. Krapas
Environmental Manager

Control of Toxic Chemicals in Puget Sound

Assessment of Selected Toxic Chemicals in the Puget Sound Basin, 2007-2011

Publication No. 11-03-055

DEPARTMENT OF ECOLOGY
State of Washington

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Control of Toxic Chemicals in Puget Sound

Assessment of Selected Toxic Chemicals in the Puget Sound Basin, 2007-2011

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November 2011

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Abstract

The Washington State Department of Ecology and other agencies initiated a multi-phase project in 2006, the Puget Sound Toxics Loading Analysis (PSTLA), to evaluate sources of toxic chemicals entering Puget Sound. The analysis focused on an abbreviated list of chemicals that were known to, or threaten to, harm the Puget Sound Ecosystem. The study included an assessment of major delivery pathways such as surface water runoff, groundwater, publicly-owned treatment works (POTWs), and direct air deposition. An assessment of the relative hazards posed by target chemicals was also performed.

The overall goal of PSTLA is to provide technical information to help develop toxic chemical control strategies for the Puget Sound basin. This report is a synthesis of information generated on (1) chemical releases from human-caused sources, (2) the rates of chemical loading through various pathways, and (3) a basin-wide hazard evaluation for chemicals of concern.

For most of the chemicals addressed, the rate of loading to Puget Sound was estimated to be one to three orders of magnitude lower than the rate of release from human-caused sources. In most cases, surface runoff was found to contribute the largest loads to Puget Sound, typically accounting for more than one-half of the total loads from all environmental pathways combined. Loads delivered through POTWs were generally the smallest among the pathways assessed, typically accounting for less than 10% of the total loading for each of the chemicals addressed.

Results of the hazard evaluation suggest that the following chemicals are most likely to be found at concentrations where effects are documented or at levels above criteria used to protect aquatic organisms and consumers of aquatic organisms:

- copper
- mercury
- polychlorinated biphenyls (PCBs)
- polychlorinated dioxins and furans (PCDD/Fs)
- the pesticide DDT (and its metabolites DDD and DDE)
- polycyclic aromatic hydrocarbons (PAHs)
- bis(2-ethylhexyl) phthalate (DEHP)

Results of the hazard evaluation were coupled with information on chemical sources and loading to suggest priorities for source control among the chemicals assessed. Recommendations are provided for source control strategies, and data needs are identified.

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Executive Summary

Background and Approach

The Washington State Department of Ecology (Ecology) and other agencies initiated the Puget Sound Toxics Loading Analysis (PSTLA) in late 2006 to provide scientific information that could be used to guide decisions about how best to direct and prioritize resources and strategies for controlling toxic chemicals in the Puget Sound basin. The primary focus of PSTLA was to estimate toxic chemical loading to Puget Sound through major pathways such as surface water runoff, publicly-owned treatment works (POTWs), and direct air deposition.

PSTLA used a phased approach to develop technical information on toxics chemicals in the Puget Sound basin.

- Phases 1 and 2 relied on existing data to estimate chemical loadings and identify the most important delivery pathways.
- Phase 3 studies included collection of new monitoring data to fill data gaps identified during earlier phases.

Other important components of the project included:

- Assessing pharmaceuticals and personal care products in wastewater treatment plants.
- Assessing persistent organic pollutants in three guilds of marine species.
- Developing numerical models for polychlorinated biphenyls (PCBs) in Puget Sound.
- Developing a framework for a toxicant-based biological monitoring system.
- Preparing a report on the estimated release of chemicals from human-caused (anthropogenic) sources.

The present *Assessment Report* aims to synthesize information from all phases of the PSTLA, focusing primarily on the Phase 3 loading studies. Since information on loading and delivery pathways, primary chemical sources, and other PSTLA studies may not by itself be sufficient to meet the overall goal of PSTLA, a screening-level hazard evaluation of selected chemicals was conducted and included in this report. The hazard evaluation provides information about the relative risk of toxic effects posed by selected chemicals at observed concentrations in the Puget Sound basin. The information provides a scientific basis to develop a source control strategy for toxic chemicals in the Puget Sound Basin, and to prioritize actions.

The report is organized in a chemical-by-chemical fashion. For each of the selected chemicals addressed in the report, the major ongoing anthropogenic sources are discussed and release rates are estimated, an assessment of loading to Puget Sound and major pathways is presented, and the results of the hazard evaluation for the specific chemical are discussed. These elements are expressed by asking the following questions about toxic chemicals in the Puget Sound basin:

- Where do they come from?
- How much is being delivered?
- What delivery pathways contribute to the loading?
- What is the relative toxic hazard posed by these chemicals at observed concentrations?

In order to focus source control actions, a lines-of-evidence approach was developed to identify (1) chemicals with large ongoing anthropogenic releases from primary sources (does not include natural sources or legacy pollution, *and* (2) a relatively high potential to elicit effects based on the results of the hazard evaluation and regionally important biological-effects data. Combining information on chemical releases and loadings with the relative potential for effects provides for a more robust prioritization of possible future source control efforts. Recommendations for filling data gaps are also included.

The PSTLA focused on an abbreviated list of chemicals of concern (COCs). This list was developed during Phase 1 of the project based on observed harm or the threat of harm to the Puget Sound ecosystem. There is a wide range of chemicals in the Puget Sound basin for which we lack environmental information, and yet the chemicals may have the potential to cause biological or ecological harm. In addition, there is a large degree of uncertainty about the sources, pathways, and hazards for the chemicals that have been addressed. Therefore, this assessment should be viewed as the starting point for developing a larger toxic chemical control strategy in which a much broader spectrum of chemicals is considered.

Summary of Major Findings

Sources, Loads, and Pathways

COCs (listed in Table ES-1) were selected as the core group of chemicals analyzed in PSTLA studies (1) based on a documented history of their presence in Puget Sound and their capacity to harm or threaten the Puget Sound ecosystem and (2) to ensure that a broad variety of delivery pathways would be represented. While there is general consensus that a much larger number of potentially harmful chemicals are released to Puget Sound, the identification and evaluation of all of these chemicals were beyond the scope of the PSTLA projects.

To remain consistent with other PSTLA projects, the geographical scope of this *Assessment Report* includes Puget Sound, the U.S. portions of the Straits of Georgia and Juan de Fuca, and the entire U.S. watershed for Puget Sound and the Straits.

Chemical loads were calculated for most of the major pathways identified during the initial phase of the PSTLA effort. These include surface water runoff, POTWs, atmospheric deposition directly to marine waters, and direct groundwater discharge. Table ES-1 summarizes the estimated quantities of COCs released in the Puget Sound basin and the loads delivered to Puget Sound. Although these estimates are based on the best available information, releases and loads for some COCs remain incomplete or reflect high levels of uncertainty. In particular, air deposition and groundwater loading data are unavailable for many of the organic COCs.

In general, the load of metals to Puget Sound is approximately an order of magnitude lower than the total release from ongoing anthropogenic sources. Arsenic is an exception, apparently due to a high level of enrichment from natural sources.

For organic chemicals, loads are generally one to three orders of magnitude lower than releases from ongoing anthropogenic sources. The comparatively large differences between release and loading rates for organics may simply reflect the fewer number of pathways assessed for some

organics. Closer agreement between anthropogenic releases and loads for metals may also be due in part to natural enrichment which contributes to the loads. Historic releases may also be a factor in loading of both metals and organic compounds.

Table ES-1. Toxic Chemical Releases and Loading in the Puget Sound Basin (metric tons/year).

COC	Total Release in the Puget Sound Basin[a]	Major Sources	Total Load to Puget Sound[b,c]	Major Pathway(s)
Arsenic	0.8	Industrial air emissions. CCA-treated wood leaching. Roofing material leaching.	14 – 25	Surface Runoff
Cadmium	1.0	Roofing material leaching.	0.05 – 0.53	Groundwater Atm. Deposition
Copper	180 - 250	Pesticides use on urban lawns and gardens.[d] Residential plumbing component leaching. Brake pad abrasion. Roofing material leaching. Vessel anti-fouling paint leaching.	33 – 80[e]	Surface Runoff
Lead	520	Ammunition and hunting shot use. Loss of fishing sinkers and wheel weights. Roofing material leaching. Aviation fuel combustion.	3.6 – 12	Surface Runoff
Mercury	0.5	Consumer product improper disposal. Crematoria and industrial air emissions.	0.11 – 0.37	Surface Runoff
Zinc	1,500	Roofing material leaching. Vehicle tire abrasion.	140 - 200	Surface Runoff
Total PCBs	2.2	Electrical equipment spills and leakage.[d] Residential trash burning. Building sealant (caulk) volatilization and abrasion.	0.003 – 0.02	Surface Runoff
Total PBDEs	0.7	Furniture, computer monitors, and other components of residential and commercial indoor environments.	0.028 – 0.054	Atm. Deposition POTWs
PCDD/Fs	0.000009[f]	Backyard burn barrels.	NA	NA
Total DDT	NA	NA	0.0025 – 0.032	Surface Runoff
Total PAHs	310	Woodstoves and fireplace combustion emissions. Vehicle combustion emissions. Creosote-treated piling, railroad ties, and utility poles.	0.19 – 1.0[e]	Groundwater Surface Runoff
DEHP	17	Polymer (primarily PVC) off-gassing. Industrial, commercial, and institutional air emissions. Roofing material leaching.	2.0 – 3.2	Surface Runoff
Triclopyr	150	Herbicide use on crops and golf courses.	0.64 – 0.69	Surface Runoff
Nonylphenol[g]	0.18	Industrial, commercial, and institutional air emissions.	0.023 – 0.024	Surface Runoff
Petroleum	9,300	Motor oil drips and leaks. Used oil improper disposal. Gasoline spillage during fueling.	330 – 500	Surface Runoff
Oil & Grease[h]	NA	NA	8,500 – 11,000	Surface Runoff

NA=Not analyzed

[a] Includes the Puget Sound, the U.S. portions of the Straits of Georgia and Juan de Fuca, and the entire U.S. watershed for Puget Sound and the Straits.

[b] Includes the Puget Sound and the U.S. portions of the Straits of Georgia and Juan de Fuca.

[c] Range of all pathways combined expressed as the sum of the 25th percentile values for each pathway – sum of the 75th percentile values for each pathway.

[d] Estimate is highly uncertain.

[e] Does not include estimated direct releases to marine waters (54 metric tons/yr for PAHs and 26 metric ton/yr for copper).

[f] Expressed as Toxic Equivalents (TEQs).

[g] Sources were not fully assessed.

[h] Category includes all hexane extractable material

The relationships between rates of initial release, rates of loading to Puget Sound, and the major delivery pathways are discussed for each COC. For most COCs, it is clear that a simple assessment of overall release from primary sources will not translate to levels measured in the environment. The type, mechanism, and setting of a chemical release are important factors governing their presence in environmental pathways. For instance, large quantities of polycyclic aromatic hydrocarbons (PAHs) are potentially released from creosote-treated railroad ties and utility poles throughout the basin, but the mechanisms and settings of release (volatilization and near-field leaching primarily to soil) do not translate to elevated concentrations of PAHs in surface runoff. In contrast, estimated zinc releases from major sources such as roof materials and vehicle tires occur in manners and settings (leaching from precipitation, abrasion to roadway surfaces) that result in substantial entrainment of zinc to surface waters.

For the majority of COCs, surface runoff contributed the largest loads to Puget Sound, typically accounting for more than one-half of the total loads from all pathways combined. Surface runoff from commercial/industrial land covers typically had the highest concentrations. However, agricultural areas produced the highest concentrations for several metals. Loading calculations were strongly influenced by the areas occupied by different land cover types. As a result, the largest loads were typically from forested areas (occupying 83% of all land cover), even though COC concentrations in forest areas were often the lowest among land covers or below reporting limits.

Loads delivered directly to the Puget Sound marine environment through groundwater were estimated using literature values for COC concentrations and discharge estimates. Groundwater loads were estimated to be an order of magnitude lower than surface runoff for most COCs. Exceptions were PAH loads, which were similar to surface water, as well as cadmium, gasoline, and diesel fuel, which had low to non-detectable surface water loads. Like surface runoff, load calculations for groundwater were strongly influenced by methods used to estimate concentrations from non-detected values.

Atmospheric deposition directly to marine waters was an important loading pathway for polybrominated diphenyl ethers (PBDEs) and high molecular weight PAHs. PBDEs was the only COC for which direct deposition from air appeared to be the largest delivery pathway to Puget Sound. COC flux from the atmosphere was comparatively high at a monitoring station located in a high-density urban area with nearby commercial, industrial, and major roadway contaminant sources.

POTWs generally accounted for less than one-tenth of the delivery to Puget Sound for each of the COCs assessed. Exceptions were diethylhexyl phthalate (DEHP, a.k.a. bis(2-ethylhexyl) phthalate) and PBDEs, the latter of which had POTW loads larger than those in surface runoff. POTW loads for other chemicals not specifically addressed in this report, such as pharmaceuticals, are expected to be much higher in POTWs than in other delivery pathways.

Hazard Evaluation

To assess the relative hazards, observed environmental concentrations of COCs in various environmental media (surface water, sediment) were compared to available data on biological effects or to established criteria to protect aquatic life and consumers of aquatic organisms.

Evaluations were conducted for the following categories:

- Direct hazards to aquatic life through surface water exposure
- Direct hazards to benthic organisms through sediment exposure
- Direct hazards to aquatic life based on tissue residue levels
- Hazards to wildlife based on ingestion of prey, water, and sediment
- Hazards to human health through fish/seafood consumption

The results of these comparisons were grouped into three broad "level of concern" categories: Priority 1 level of concern, Priority 2 level of concern, or unknown (U) level of concern. Results were classified as Priority 1 when the upper end of a set of observed concentrations (e.g. 90^{th} percentile values) exceeded the lower end of a set of effects concentrations (e.g. 10^{th} percentile values), or exceeded selected threshold values such as water quality criteria. A Priority 2 level of concern was assigned in cases where the upper end of a set of observed concentrations was below the lower end of a set of effects concentrations or other threshold values. In cases where there were not sufficient data to make a meaningful comparison, results were assigned a U.

The hazard evaluation has several limitations that should be considered prior to acting on the results. In particular, the hazard evaluation is not a risk assessment but is instead designed to assess the *relative* level of concern of COCs *across the entire Puget Sound basin*. Although a COC may be assigned Priority 2 or U for a particular sub-category, this should not be interpreted to mean there are no hazards associated with that COC. All of the COCs evaluated pose some level of concern for Puget Sound. Locally, concentration hot spots may exist near major sources and may cause localized toxicity to aquatic organisms or lead to violations of standards that would not necessarily be emphasized in this broad regional assessment. To address this shortcoming in the hazard assessment, a limited review was conducted of regionally important biological effects information.

Results of the hazard evaluation showed that all of the COCs except lead are a Priority 1, where sufficient data were available for evaluation, for at least one of the categories evaluated. There were not sufficient data to conduct evaluations for PBDEs, triclopyr, nonylphenol, or petroleum. PCBs is the only COC assigned a Priority 1 for all five categories evaluated.

COC concentrations in surface waters and sediments – particularly freshwater – resulted in the most COCs assigned Priority 1. Tissue residue effects, wildlife, and human health evaluations generally resulted in fewer COCs receiving a Priority 1 assignment. However, only bioaccumulative chemicals were evaluated for these latter categories.

In addition to the hazard evaluation, reviews of regionally important biological-effects data showed that levels of the following chemicals found in the Puget Sound basin result in documented or potentially adverse effects to a variety of aquatic organisms:

- copper
- mercury
- PCBs
- PBDEs
- polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs)
- dichlorodiphenyltrichloroethane (DDT) and metabolites DDD and DDE
- PAHs

Priorities for Source Control Actions

A lines-of-evidence approach was used to move further toward a goal of deciding how best to prioritize actions and resources for controlling toxic chemicals in the Puget Sound basin. This lines-of-evidence approach considers information on the four major components of the assessment (sources, loading, pathways, and the relative hazards), but the approach mostly relies on COC sources and the relative hazards posed by COCs as determined by the hazard evaluation and review of other regional studies.

This approach adopts the rationale that chemicals with the greatest potential to elicit toxic effects at existing concentrations should be an important factor in determining the priority for source control efforts. Priority was also given to COCs for which we have large opportunities for source control. This reflects the extent to which there are existing regulatory actions to control releases, such as bans, management of materials, or other permanent actions which reduce releases to the environment. Given the uncertainty associated with individual estimates of releases or loadings, this lines-of-evidence approach provides a supportable rationale for establishing relative priorities for control actions.

Opportunities for source control are considered large where the major sources of a COC have not been addressed by control actions; where some of the major sources have been addressed, opportunities may be considered medium. In cases where actions have been implemented to control and reduce all or most of the major sources and this appears to have resulted in low rates of loading to Puget Sound, the opportunities for controlling a COC are considered small. This assessment relies principally on the *Sources Report* (Ecology, 2011) with limited input by Ecology staff and management to gauge the opportunities for source control; the assessment was not intended to be a detailed review of management initiatives. Table ES-2 summarizes major sources for each COC and possible opportunities for reducing those sources.

Based on the lines-of-evidence approach, copper, PAHs, DEHP, and petroleum sources were rated as have the highest priority for early actions. The reasoning for this determination is as follows:

- A substantial portion of the fresh and marine water copper data observed basin-wide falls within concentrations where effects have been documented (including reduced olfactory function in salmonids). Copper is released in large quantities from a variety of sources which appear to translate to substantial loads to the Puget Sound ecosystem. The use of copper in pesticide applications and the release of copper from roofing materials are sources which warrant further investigation. In addition, the effectiveness of recent legislation to limit copper in brake pads and vessel anti-fouling paint should be evaluated.
- A number of individual PAHs surpass (do not meet) freshwater sediment guidelines and human health criteria. In addition, a variety of studies have demonstrated links between PAH exposure and adverse effects to regionally relevant aquatic species. There appear to be numerous opportunities for control actions, primarily for combustion sources and for creosote-treated wood.

- Observed DEHP concentrations in both freshwater and marine environments exceed (do not meet) criteria for protection of benthic species and human health. Substantial amounts of DEHP are released in the Puget Sound basin, much of which occurs initially through releases to air from off-gassing of plasticized polymers and point-source air emissions. Several non-polymer uses of DEHP may also provide opportunities for source reduction.
- The relative hazard posed by petroleum in the Puget Sound basin was not able to be evaluated due primarily to the lack of biological-effects data and the absence of criteria to protect aquatic organisms, wildlife, or human health. However, some of the COCs addressed in this assessment are components of petroleum and may be released in substantial quantities along with the release of petroleum. In particular, substantial releases of PAHs are estimated to be released from petroleum. The major sources of petroleum are diffuse, such as motor oil drips and leaks and minor gasoline spillage during vehicle fueling, and therefore offer ample opportunities for reduction efforts.

Several COCs were found to be a Priority 1 level of concern based on the hazard evaluation but were not determined to be among the highest priorities for reduction actions since the major sources have been addressed through regulatory programs or other efforts. For instance, mercury poses a relatively high hazard to freshwater and marine aquatic organisms and wildlife based on doses calculated from observed data. However, many of the historical regional sources of mercury to the Puget Sound basin have been eliminated or are being addressed by the Mercury Chemical Action Plan (Ecology and WDOH, 2003). Similarly, PCBs are a Priority 1 level of concern for all hazard evaluation categories, but PCBs have been banned for decades, the major sources (use in electrical equipment) are highly regulated, and current loads to Puget Sound appear to be small.

Although the systematic prioritization approach identified four COCs for early actions, other factors should be considered to determine the need and feasibility for developing control and reduction strategies for other COCs. For instance, PBDEs are ubiquitous environmental contaminants, and although voluntary actions and bans have removed major PBDE formulations from new consumer products, much of the PBDEs produced historically may remain in consumer products and commercial office products and these potentially represent substantial diffuse ongoing sources. The hazard evaluation was not able to adequately assess the relative hazards associated with PBDEs due to a lack of environmental standards, although there is evidence in the available literature to suggest this COC may pose a hazard at observed concentrations.

Additional research is needed to assess the relative hazards posed by PBDEs and other COCs for which there are only limited environmental data. By the same token, COCs with limited source information should be further evaluated to assess additional opportunities for source control. Of the COCs addressed in this report, PBDEs and nonylphenol were the COCs that should receive top attention for further research on potential hazard as well as possible opportunities for source control.

Table ES-2. Summary of Possible Actions to Reduce COCs in the Environment.

COC	Opportunities for Source Control	Major Ongoing Anthropogenic Sources	Possible Actions for Reductions
Arsenic	Medium	Industrial air emissions	Maintain existing permit controls.
		CCA-treated wood leaching	Continue ban for most non-structural uses.
		Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
Cadmium	Medium	Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
Copper	Large	Pesticides use on urban lawns and gardens	More data needed on actual pesticide use.
		Residential plumbing component leaching	Continue to implement Lead and Copper Rule.
		Brake pad abrasion	Continue to implement legislation enacted to reduce source.
		Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
		Vessel anti-fouling paint leaching	Continue to implement legislation enacted to reduce source.
Lead	Small	Ammunition and hunting shot use	Implement CAP and enforce existing regulations.
		Loss of fishing sinkers and wheel weights	Implement CAP and enforce existing regulations.
		Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
		Aviation fuel combustion	Implement CAP and enforce existing regulations.
Mercury	Medium	Consumer product improper disposal	Continue to implement CAP and enforce existing regulations.
		Crematoria and industrial air emissions	Continue existing permit limits.
Zinc	Large	Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
		Vehicle tire abrasion	Investigate source where it poses local concern.
Total PCBs	Small	Electrical equipment spills and leakage	Continue programs for management and disposal.
		Residential trash burning	Continue enforcing existing ban.
		Building sealant (caulk) volatilization and abrasion	Investigate source where it poses local concern.
Total PBDEs	Medium	Furniture, computer monitors, and other components of residential and commercial indoor environments	Enforce ban on new products but consider control actions to reduce the release from existing products.
PCDD/Fs	Small	Backyard burn barrels	Continue enforcing existing ban.
Total DDT	Small	None apparent	Investigate source where it poses local concern.
Total PAHs	Large	Woodstoves and fireplace combustion emissions	Continue change out programs, investigate catalysts/capture devices, promote alternatives to wood heat.
		Vehicle combustion emissions	Anti-idling programs, continue/expand engine retrofits for private section engines, enforce existing vehicle controls.
		Creosote-treated piling, railroad ties, and utility poles	Control actions needed, gather information to identify highest priority areas.
DEHP	Large	Polymer (primarily PVC) off-gassing	Gather additional information on extent of releases.
		Industrial, commercial, and institutional air emissions	Maintain existing permit controls.
		Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
Triclopyr	Medium	Herbicide use on crops and golf courses	More data needed on pesticide use.
Nonylphenol	Unknown	Industrial, commercial, and institutional air emissions	More information needed on emissions from these sources and unidentified releases.
Petroleum	Large	Motor oil drips and leaks Used motor oil improper disposal	Expand existing education/workshop programs.
		Gasoline spillage (minor) during fueling	Possible opportunity for source control, but more data needed on extent of releases.

Bold=Recommended as priority for near-term actions based on lines-of-evidence approach.

Recommendations

Broad recommendations are provided below. These are intended to guide development of a long-term strategy to reduce toxic threats to Puget Sound. The reader is directed to the main body of the report for more detailed results of this assessment that should provide further direction towards specific chemical control actions and further source/pathway investigations.

While this report identifies sources of toxic chemicals entering Puget Sound and recommends ways to reduce this contamination, these recommendations should be prioritized and balanced alongside current efforts and regulatory programs that already keep millions of pounds of business-generated COCs safely managed.

In 2010 the U.S. Environmental Protection Agency selected Ecology to lead the development and implementation of a long-term toxic chemical control strategy for Puget Sound. Results from the PSTLA will be a key piece of information to help design and implement actions to reduce threats from the most important sources of toxic chemicals to the Puget Sound ecosystem. Ecology will use funding from a National Estuary Program grant to implement priority actions under this long-term toxics control strategy.

Major recommendations from this assessment can be summarized as follows:

- A variety of diffuse (nonpoint) sources appear to account for the majority of contaminant releases in the Puget Sound basin. In addition, surface water runoff during storms was identified as the major delivery pathway for most contaminants. High priority should be given to (1) implementing control strategies to prevent the initial release of contaminants and (2) reducing or treating stormwater inputs.

- Vehicles and vehicle-related activities represent an important source of a number of contaminants. Examples include: copper and zinc from brake and tire wear, PAHs from fuel combustion, and petroleum from motor oil drips and leaks as well as refueling operations. Source control strategies should be developed around reducing contaminant inputs from vehicles.

- Runoff and leaching from roofing materials were estimated to be a major source of several metals, particularly cadmium, copper, and zinc. Roof runoff may also be a substantial source of DEHP. Field investigations should be conducted to gauge the accuracy of this information, and if warranted, alternative assessments should be considered for this source category.

- Developed lands (commercial/industrial, agricultural, and residential) had higher concentrations of COCs compared to undeveloped forest land. Source control strategies should focus on identifying and controlling contaminant releases from existing and new developments.

- This assessment focused on a short list of contaminants that were known to, or threaten to, harm the Puget Sound ecosystem. Data are needed on the spatial distribution and impacts from a much wider range of potential contaminants (e.g. pharmaceuticals and personal care products, brominated flame retardants, nanomaterials) in the basin.

- Businesses in Washington that routinely handle large amounts of COCs should be inspected on a routine basis; once every three years appears to be a reasonable schedule.

Examples of other recommendations are provided below:

- One of the largest potential releases of copper is due to the urban lawn and garden use of products containing copper. Due to the lack of good pesticide-use information, there is a high degree of uncertainty surrounding this conclusion. Additional information is needed to determine release rates for this potentially important source of copper.

- Wood-burning stoves and fireplaces along with vehicle emissions were identified as some of the largest sources of PAHs in the Puget Sound watershed. Regional air programs should continue to pursue abatement programs to reduce wood smoke emissions and vehicle emissions.

- Creosote-treated wood represented approximately one-third of the PAHs released to the Puget Sound basin. In particular, direct release to Puget Sound occurs from treated pilings in marine (salt) water. Programs such as the Department of Natural Resources Marine Piling Removal program should be supported to reduce the release of PAHs from marine pilings and bulkheads.

- More information is needed to help distinguish natural and legacy sources of contaminants in environmental pathways such as surface water runoff. This will help gauge the feasibility and effectiveness of actions taken to reduce releases of chemicals from contemporary anthropogenic (human-caused) releases.

- Resources should be provided for local source control programs that identify and prevent the release of contaminants on a local scale.

Introduction

Background

The Puget Sound Basin covers more than 43,400 square kilometers (16,800 square miles) of land and water (Hart Crowser et al., 2007) and is home to 4.5 million people (U.S. Census Bureau, 2010) including large urban metropolitan centers such as Seattle and Tacoma (Figure 1). Although large urban and industrial areas have developed along the shores of Puget Sound and near the mouths of major rivers – particularly on the east side of the central Sound – much of the shoreline and the watershed remains undeveloped and the bulk of the upland basin is forest.

During the past 150 years, humans and their activities have released a wide variety of chemicals into Puget Sound and its surrounding watershed, many of which are toxic to humans and aquatic organisms. Due to its fjord-like structure and shallow sills, the entry of deep oceanic water into Puget Sound is restricted, which reduces flushing of the inland marine and estuarine waters (PSAT, 2007). As a result, toxic chemicals (toxicants) and other pollutants may accumulate in some inlets and embayments of Puget Sound, increasing their exposure to aquatic organisms.

While the marine waters of Puget Sound may be the ultimate sink for many of the toxic chemicals released in the basin, it is universally recognized that freshwater streams, rivers, and lakes in the basin may be at risk from contamination as toxic chemicals travel from their points of initial release to the Puget Sound. The health of Puget Sound ultimately depends on the health of its upland watershed, particularly since one of its most ecologically important and iconic organisms – pacific salmon – rely on the upland watersheds for some of their most vulnerable stages of life.

For several decades the loading of toxicants to Puget Sound has been recognized as a serious problem and has been documented in a number of reviews (e.g. Dexter et al., 1981; Romberg et al., 1984; PSWQA, 1986; PTI, 1991; PSAT, 2003; Redman et al., 2006). These reviews have primarily focused on identifying chemicals of concern (COCs), concentrations in marine sediments, and effects to aquatic organisms, but generally provided only conjecture about delivery pathways.

Puget Sound Toxics Loading Analysis

In December 2005, Governor Christine Gregoire and the Washington Legislature launched the Puget Sound Initiative, a comprehensive effort by local, state, federal, and tribal governments; business, agriculture and environmental communities; scientists; and the public to restore, protect, and preserve the Sound by 2020. Among the top recommendations put forth by the original Puget Sound Partnership (Partnership) in 2006 was to make the reduction of toxic chemicals entering Puget Sound waters a primary objective for the long-term agenda.

The Washington State Department of Ecology (Ecology) and other agencies responded to the original Partnership's "reducing toxics" recommendation by initiating the Puget Sound Toxics Loading Analysis (PSTLA). Phase 1 of PSTLA was an initial estimate of toxicant loading to Puget Sound through various pathways such as surface runoff and direct air deposition. The

analysis relied on readily accessible data to estimate chemical loading to the marine basin, and the authors of the report acknowledged that there remained a number of significant gaps in determining an accurate toxics budget for the Sound (Hart Crowser et al., 2007). However, the Phase 1 study satisfied its primary goal of identifying data gaps and needs for additional studies, and informed the Puget Sound Action Agenda (PSP, 2008), the plan for restoring Puget Sound.

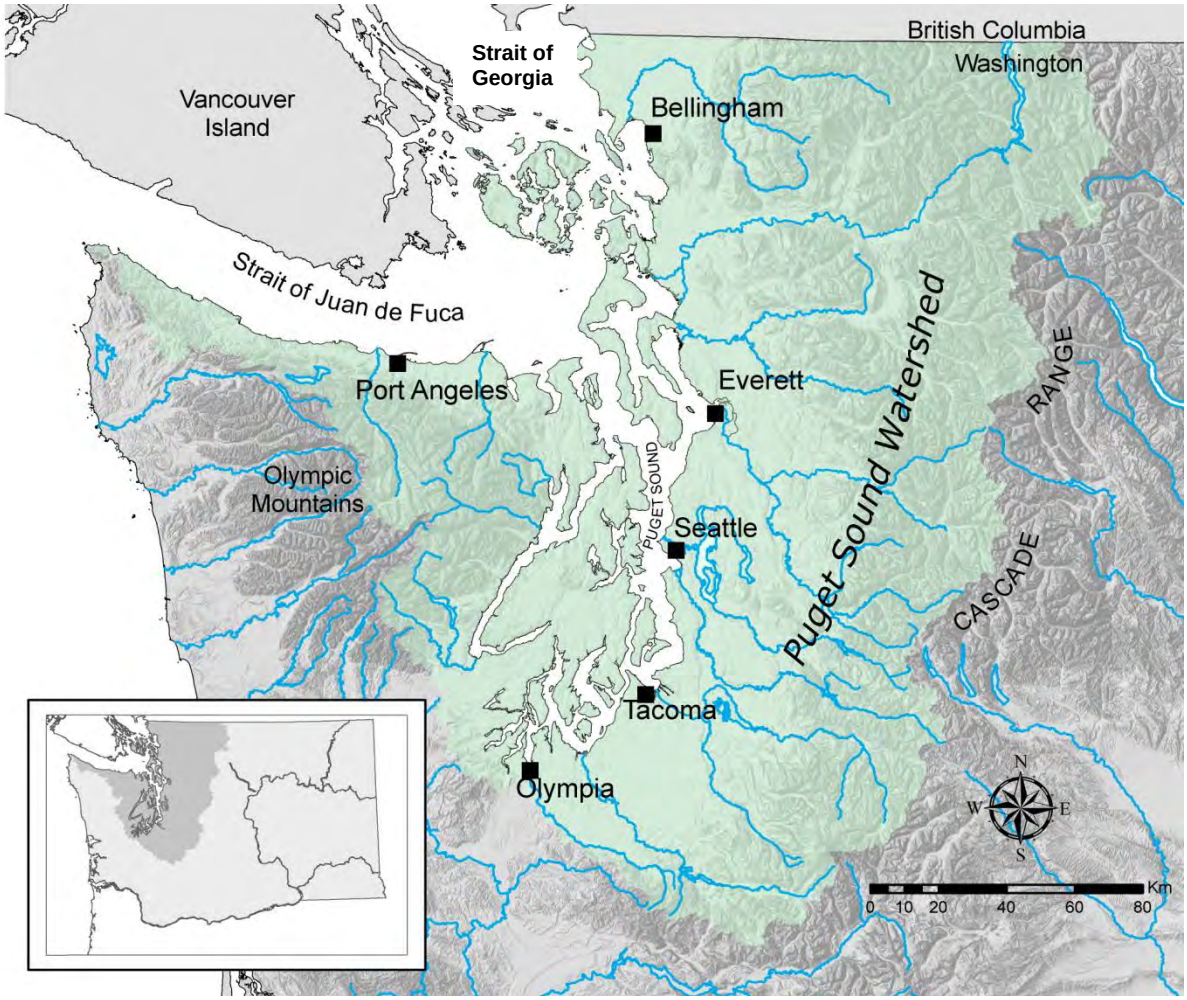


Figure 1. Watershed area for Puget Sound and the U.S. Portion of the Strait of Georgia and the Strait of Juan de Fuca.

The Partnership's Action Agenda re-iterated the conclusion that toxic chemical contamination is one of the largest problems facing Puget Sound, and a number of recommendations were developed to tackle various aspects of this complex problem. Although many of the recommendations were centered on implementation (e.g. programmatic, education, policy), the Partnership also recognized the need to gain a more thorough understanding of the problem as it currently exists. This need was expressed in the Action Agenda as Priority C.1.1.10: *Continue scientific work to better understand the sources of toxics, as well as transport and fate in the Puget Sound ecosystem, to better refine reduction strategies. This includes the toxic loadings assessments.*

In keeping with recommendations from the Action Agenda, two additional phases of PSTLA projects were mapped out:

- Phase 2 PSTLA studies were generally designed to refine loading estimates obtained during the Phase 1 effort and relied on readily available information much like the Phase 1 study.
- Phase 3 represented a departure from the earlier approaches in that nearly all of the Phase 3 projects included a sampling component so that refinements would include up-to-date and region-specific data.

In general, PSTLA projects were designed to assess the loading, sources, and to some degree, the impacts of toxic chemicals in Puget Sound. But the bulk of the PSTLA effort has been devoted to studying the delivery of toxicants through various pathways, such as direct atmospheric deposition, surface water runoff, and water exchange at the ocean boundary.

Purpose

The overall goal of PSTLA is to provide scientific information that will help guide decisions about how best to direct and prioritize resources and strategies for controlling toxic chemicals in the Puget Sound basin.

The purpose of this *Assessment Report* is to (1) distill the data and information generated by the PSTLA projects into a single document and (2) provide conclusions about toxic chemicals and their sources that can be used by those developing strategies to control toxic chemicals in Puget Sound.

The *Assessment Report* does not address all threats to Puget Sound. The issues regarding cleanup and restoration of Puget Sound are complicated and multi-faceted. For every pound of toxic chemicals released to Puget Sound by people, cars, households, etc., Washington businesses produce and handle hundreds to thousands times the amount of these chemicals, both in the form of chemical products and hazardous waste. While the *Assessment Report* identifies sources of pollution entering Puget Sound and recommends priorities to address this contamination, these recommendations should be balanced alongside current efforts that already keep millions of pounds of business-generated chemicals safely managed.

In 2010 the U.S. Environmental Protection Agency (EPA) selected Ecology to lead development and implementation of a long-term toxic chemical control strategy for Puget Sound. Results from the PSTLA will be a key component to (1) identify the most significant sources and delivery pathways of toxic chemicals and (2) prioritize reduction efforts to reduce threats from toxic chemical to the Puget Sound ecosystem. Ecology will use funding from a National Estuary Program grant to implement priority actions under the toxics control strategy.

Scope, Approach, and Organization of the Report

Many projects are included under the umbrella of the PSTLA, as shown in Figure 2. Appendix A provides additional information on PSTLA project subject matter, authorship, and completion

status. All completed PSTLA reports are posted on the PSTLA internet homepage: www.ecy.wa.gov/programs/wq/pstoxics/index.html.

This *Assessment Report* focuses primarily on the PSTLA Phase 3 loading studies and draws on information provided in other PSTLA studies where appropriate. Since information on loading and delivery pathways, primary chemical sources, and other PSTLA-derived data may not by itself be sufficient to meet the overall goal of PSTLA, a screening-level hazard evaluation of selected chemicals was conducted and included in the *Assessment Report*. The hazard evaluation provides information about the relative risk of toxic effects posed by selected chemicals at observed concentrations in the Puget Sound basin.

This *Assessment Report* is organized in a chemical-by-chemical fashion. For each of the selected chemicals addressed in the report, the major ongoing anthropogenic sources are discussed, an assessment of loading to Puget Sound and major pathways is presented, and the results of the hazard evaluation for the specific chemical is discussed. These elements may be re-phrased as the following questions for specific toxic chemicals in the Puget Sound basin:

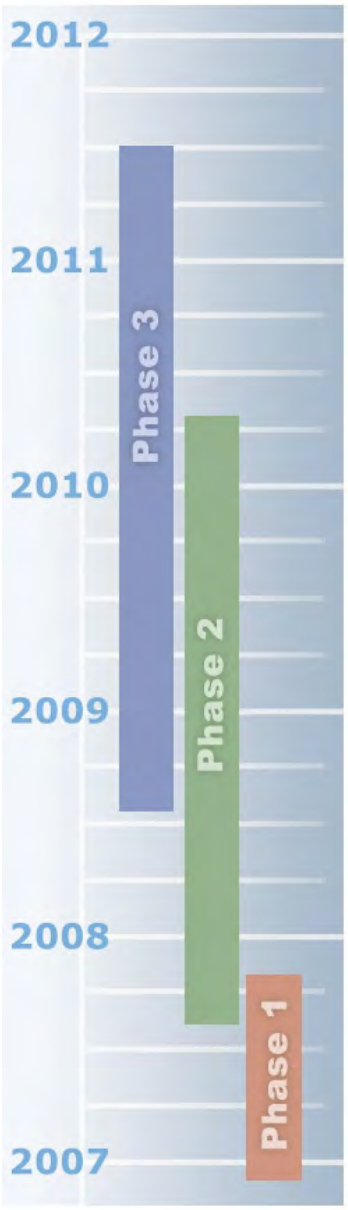
- Where do they come from?
- How much is being delivered to Puget Sound?
- What delivery pathways contribute to the loading?
- What is the relative toxic hazard posed by these chemicals at observed concentrations?

Figure 3 shows the major elements considered for this *Assessment Report*. As mentioned previously, loading estimates from different pathways come primarily from the PSTLA Phase 3 loading studies. Loading information from PSTLA Phases 1 and 2, as well as additional information on chemical transport and attenuation, may be used to supplement the PSTLA Phase 3 loading studies.

The PSTLA Phase 3 report on primary sources provides estimates of ongoing anthropogenic releases of selected chemicals. For each chemical addressed in this *Assessment Report*, the information on loading rates and pathways is discussed in context of their ongoing releases from primary sources (i.e. how these chemicals get in the environment in the first place).

As mentioned previously, the hazard evaluation provides information about the relative risk of toxic effects posed by selected chemicals at observed concentrations in the Puget Sound basin. The hazard evaluation was not produced as a separate project; it was conducted specifically for this Assessment Repot to enhance and supply additional context to the information on loading, pathways, and sources of toxic chemicals.

The major components used for this *Assessment Report* are described in more detail in subsequent sections. The reader should be aware that the *Assessment Report* does not consider these components exclusively. For instance, additional information on bioaccumulative chemicals in three guilds of marine organisms (West et al., 2011a and b; Noel et al., 2011) are used to provide additional context to the hazard evaluation for specific chemicals.



Phase 3: Targeting Priority Toxic Sources

Phase 3 Projects:

3A: Toxic Chemical Loadings via Surface Runoff
3B: Modeling Surface Runoff in Two Pilot Watersheds
3C: Evaluate Air Deposition
3D: Toxic Chemicals in Marine Waters and from Ocean Exchange
3E: Numerical Models and Scenarios
3F: Priority Pollutant Scans for POTWs
3G: Primary Sources of Toxic Chemicals
3H: Pharmaceuticals and Personal Care Products (PPCPs)
3J: Persistent Organic Pollutants in Three Guilds of Pelagic Marine Species from the Puget Sound
3K: Assessment Report
3L: Groundwater Discharge Directly to Puget Sound

Phase 2: Improved Loading Estimates

Phase 2 Projects:

2A: Loadings from Surface Runoff and Roadways
2B: Loadings from Dischargers of Municipal and Industrial Wastewater
2C: Bioaccumulation Model Estimates of Toxics from Sediments
2D: Water Column Data for Puget Sound and its Ocean Boundary
2E: Support for a Human Health Risk Assessment
2F: Numerical Models for Polychlorinated Biphenyls in Puget Sound
2G: Biological Observing System (TBiOS) for Toxics in Puget Sound

Phase 1: Initial Estimate of Toxic Chemical Loadings to Puget Sound

Phase 1 Project:

1A: Initial Toxics Loading Estimates

Figure 2. Timeline and Complete List of All PSTLA Projects.

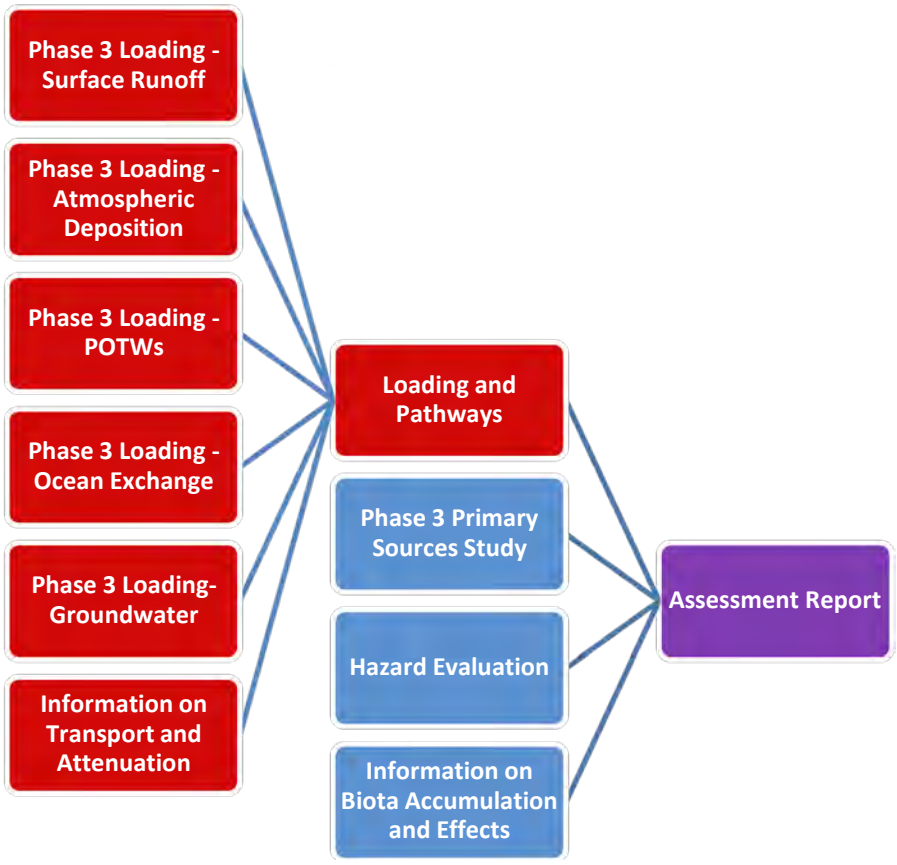


Figure 3. Elements Incorporated into the Assessment Report.

Selection of Chemicals

While there is general consensus that a large number of potentially harmful chemicals may be present in Puget Sound, the identification and evaluation of all chemicals was beyond the scope of the PSTLA projects. In order to focus on a group of chemicals that are known to be important and likely represent at least some of the most important ones in Puget Sound, the chemicals addressed in this *Assessment Report* are the chemicals of concern (COCs) first selected during the initial phase of the PSTLA.

The COCs were selected by a Chemicals of Concern Workgroup that had been convened to recommend a list of chemicals based on previous work and using best professional judgment. The workgroup sought to choose chemicals that had a documented history of presence in Puget Sound and "… that harm or threaten to harm the Puget Sound ecosystem and those that represent, or serve as an indicator for, a particular class of chemicals." The COCs list was developed to ensure that a broad variety of delivery pathways would be represented.

The workgroup settled on a list of chemicals that largely mirrored those identified by the then-lead agency for Puget Sound (Puget Sound Action Team; Redman et al., 2006). These chemicals became the core group of chemicals analyzed for the subsequent loading studies, although a few chemicals were excluded for particular studies while other studies included chemicals beyond the COCs.

The COCs are as follows:

- Arsenic
- Cadmium
- Copper
- Lead
- Mercury
- Zinc
- Polychlorinated biphenyls (PCBs)
- Polybrominated diphenyl ethers (PBDEs)
- Polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs)
- Dichlorodiphenyltrichloroethane (DDT) and metabolites DDD and DDE
- Low molecular weight polycyclic aromatic hydrocarbons (LPAHs)
- High molecular weight polycyclic aromatic hydrocarbons (HPAHs)
- Carcinogenic polycyclic aromatic hydrocarbons (cPAHs)
- Diethylhexyl phthalate (DEHP, a.k.a. bis(2-ethylhexyl) phthalate)
- Triclopyr
- Nonylphenol
- Oil and petroleum hydrocarbons

It is possible that this list of COCs represents only a small subset of those chemicals that may be impacting Puget Sound. Any conclusions drawn from the assessment of these COCs should not signify that other chemicals may not be of equal or greater concern.

Geographical Study Area

The geographical study area addressed in this *Assessment Report* is Puget Sound, the U.S. portions of the Straits of Georgia and Juan de Fuca, and the entire U.S. watershed for Puget Sound and the Straits (Figure 1). This is consistent with all of the land-based PSTLA loading projects (e.g. Hart Crowser et al., 2007; Envirovision et al., 2008a), except the groundwater loading analysis which excludes loads from the western Strait of Juan de Fuca.

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Puget Sound Toxics Loading Analysis (PSTLA) Projects

The bulk of COC data analysis in this report is contained in the Chemical-Specific Assessments section. The COC data analyzed in the Chemical-Specific Assessments section are primarily from the PSTLA studies. The following section summarizes the PSTLA studies used for these analyses in order to familiarize the reader with the subject and scope of the studies conducted under PSTLA. For more detailed information, the reader is advised to review the Chemical-Specific Assessments section or the original PSTLA studies which are referenced in the following section and are available online at www.ecy.wa.gov/programs/wq/pstoxics/index.html.

Loading Projects

PSTLA Phase 3 loading studies are described in the following sub-sections. PSTLA Phase 1 and 2 projects that were conducted to estimate COC loading are not included because their goal was generally to focus information for the development of the more refined Phase 3 loading studies. Aside from a few possible exceptions, the Phase 3 loading studies contain the best PSTLA loading data available.

Surface Water Runoff

Description

The Phase 3 study on COCs and other chemicals in surface water runoff (Herrera, 2011) was the primary off-shoot of the Phase 1 and 2 loading studies which identified surface runoff as the principal delivery pathway for most COCs. Like the other loading studies conducted under PSTLA, the surface runoff study was essentially a modeling effort which included the collection of sampling data to provide input data for the model used to estimate surface runoff loads of COCs.

The primary objective of the surface runoff study was to quantify the annual loading of COCs to Puget Sound through surface runoff, defined as the water flowing over the surface of the land at some point, including stream baseflow, stormwater, and groundwater discharging to surface waters. In addition to calculating COC mass loading to Puget Sound, the surface runoff study also provided data on chemical concentrations in surface runoff among different land cover types (commercial/industrial, residential, agricultural, and forests) and during different streamflow regimes (baseflow and storm flow).

Four sub-basins from the each of the representative land covers (commercial/industrial, residential, agricultural, and forests) were sampled twice during baseflow and six times during storm flow conditions between October 2009 and July 2010. Sampling was limited to stream sub-basins in the Snohomish River and Puyallup River watersheds for logistical reasons. Samples were analyzed for the entire list of COCs (except PCDD/Fs) as well as additional chemicals such as phthalates, semi-volatile organic compounds, pesticides, herbicides, and

nutrients (Appendix B). Continuous stream discharges (flows) were recorded during the study period. The complete results of the chemistry and discharge data are included in the surface runoff report (Herrera, 2011).

The COC concentration data obtained from sampling were coupled with stream gauging data and converted to annual unit-area loads (e.g. kg COC/km^2) specific for a land cover and flow regime (baseflows or storm flows). The land area of the entire Puget Sound basin was assigned one of the four land covers, and the unit-area COC loads computed from study results were assigned to the corresponding land cover. Using this "scale-up" methodology, COC loads for the Puget Sound basin were calculated for both storm flows and baseflows.

Summary of Major Findings

The surface runoff project team found that PAHs, phthalates, semi-volatile organic compounds, pesticides, herbicides, and petroleum hydrocarbons were rarely detected or not detected at all. PCBs and PBDEs were detected in a majority of samples; however, only a few individual chemicals from each of these classes were commonly present. Most nutrients and six of the 15 metals evaluated in this study were detected in nearly all the samples. The frequency of detection and concentrations for most chemicals was generally higher for samples collected during storm flows than baseflow samples, a pattern generally consistent among all land cover types.

Since COC concentrations were typically higher during storm events, the increased discharge during storm events amplified the COC loads compared with baseflow loads. As a result, storm flow unit-area loads were much larger than baseflow unit-area loads for most chemicals. This suggests that environmental chemicals are mobilized during storm events; otherwise increased storm flows would simply dilute chemical concentrations and loads would remain the same regardless of discharge.

Although the surface runoff study demonstrated that chemicals are mobilized during storm events, it is unclear to what extent chemicals are transported to the stream corridors versus re-mobilization of chemicals residing in the stream corridors (e.g. sediments). It is likely that both circumstances occur to varying degrees, and may depend largely on the chemical in question.

COCs were generally detected more frequently and at higher concentrations in the commercial/industrial sub-basins compared to the other land cover types with a few exceptions; metals were occasionally detected more frequently and at higher concentrations in the agricultural areas. Metals aside, agricultural and residential areas had roughly the same level of chemical detection frequencies and concentration. Streams sampled in forested areas had the lowest overall concentrations as well as frequency of COC detection.

In general, COC unit-area loading rates for the four land cover types generally showed the same pattern as concentrations (commercial/industrial > agricultural ≥ residential > forest). However, since approximately 83.4% of the land base in the Puget Sound basin is forested and only 0.8% is commercial/industrial land cover, absolute loads from forested areas dominate the overall loading from surface runoff.

As with any study designed to extrapolate large values from a limited sample set, there are a number of uncertainties associated with the results. The low frequencies of detection for many organic COCs were particularly troubling since summary statistics for concentrations were required for calculation of loads. This required substitution methods for non-detected values within a concentration dataset, with one-half of the reporting limit typically used to substitute for non-detected results. Appendix B shows the method used to establish representative COC values for datasets with non-detected values. The result is that COC concentrations used to derive loads were determined largely by reporting limit values rather than measured concentrations.

The use of unit-area loads to compute basin-wide loading has the potential to introduce bias to the final load estimates. The unit-area load method assumes that all of a single land cover type (e.g. agricultural) in the Puget Sound basin delivers the same COC load regardless of the runoff volume (the runoff volume method was applied in the Phase 2 surface runoff estimates). This essentially has the effect of dampening the load signal from forest areas where the percentage of total runoff volume is even greater than the area. At the same time, loads from commercial/ industrial areas (and agricultural areas for some COCs) may be higher using the unit-area load method compared with loading calculations that use precipitation-driven runoff volumes. However, the overall effect of the unit-area load methodology results in lower absolute loads for the Puget Sound basin due to the diminished contribution of the forest loads.

Atmospheric Deposition

Description

The study of COCs atmospherically deposited directly to the surface of the Puget Sound marine waters (Brandenberger et al., 2010) was conducted based on recommendations from the Phase 1 report which concluded that PBDE and PAH loads delivered through this pathway may be comparable or greater than from surface runoff. The Phase 1 report also noted that there were few data to assess atmospheric deposition loads, and that what little data that existed was two decades old. There were no efforts among the Phase 2 projects to improve air depositions loads.

In order to obtain estimates of air deposition loads to Puget Sound, the project team designed a sampling network of seven locations geographically dispersed around Puget Sound that represented a variety of possible air pollution influences and precipitation patterns. One station was located in a high-density urban area of Tacoma with numerous potential industrial and roadway influences; a companion station was located in a nearby undeveloped area to assess the influence of highly localized air deposition.

Sampling was conducted at two-week intervals over the course of 14 months during 2008 – 2009. The funnel-type sampling devices used for the study collected bulk samples (dry + wet deposition) and were not designed to distinguish between dry and wet deposition. However, the large number (19) of discrete sampling events permitted a partial evaluation of differences between dry and wet deposition since sampling spanned wet and dry seasons.

Samples collected from the bulk deposition collectors were analyzed for the COC metals, PCBs, PBDEs, HPAHs, and cPAHs (Appendix B). Additional analyses were conducted for PAH

markers and anhydrosugars in order to conduct analyses of PAH sources. Once COC concentrations from a sample were obtained, fluxes (e.g. ng COC/m^2/day) were calculated based on the sample volumes, funnel area, and elapsed collection time.

Using the COC fluxes, three scenarios were used to estimate total annual loads to Puget Sound:

1. The first scenario applied summary statistics derived from all of the pooled results to the entire marine surface.
2. The second scenario used location-specific deposition results and applied them to geographically associated marine compartments based on the Puget Sound Box Model (Pelletier and Mohamedali, 2009).
3. The third scenario used the same method as for the second scenario, but further divided the loading into dry and wet seasons.

The three scenarios for calculating annual loads from atmospheric deposition produced similar results, and the authors of the air deposition study did not explicitly state a preference for any particular scenario. Loading estimates from the third scenario are used for the present report because (1) values typically fell between estimated loads derived from the other scenarios, and (2) loads calculated for both dry and wet deposition could provide useful information for further analysis, much the same way baseflow and storm flow data from the surface runoff study are able to be evaluated independently.

Summary of Major Findings

All of the COCs analyzed were detected at all stations, and fluxes and loads were calculated based on summary statistics computed for each COC at each location. The high-density urban station in Tacoma had COC fluxes much higher than most other stations – an order of magnitude higher in most cases – including the nearby companion station. This finding supports the view that strong air deposition signals may be highly localized, particularly in high-density urban areas. Seasonal differences suggest a higher rate of wet deposition for metals, while the opposite appears to be the case for PBDEs. There does not appear to be any effect on seasonality for PAHs, although biomarker fluxes suggest a larger proportion of PAHs deposited during the winter originates from biofuel (e.g. firewood) combustion.

Users attempting to interpret results of the air deposition study should be aware of several limitations. The difficulties of attempting to extrapolate atmospheric deposition of chemicals across a large area using a limited sampling coverage are evident. As noted by the authors of the study (Brandenberger et al., 2010), the sampling coverage was able to capture only one location with elevated deposition rates due to highly localized conditions. Perhaps more important, mercury aside, there are few data on which to compare the results of the study. Most comparable data were collected 20 years prior to this study, and advances in analytical methods, increases in population, and emissions reduction efforts have all occurred during the intervening years. The current COC fluxes are much lower than those reported during the early 1990s.

Users of the data should also be aware that the method for handling non-detected PBDE congeners (14 were analyzed) was different from methods used in other loading studies. The method detection limit was used to replace each non-detect congener result during the

summation of individual congeners to derive a total PBDE sum. The authors of the study acknowledged this resulted in a conservative (upward) bias of the results. This bias appears to result in fluxes (and resulting loads) on the order of 50% higher than those derived from using summing methods which substitute a zero for non-detected congeners when calculating a total PBDE sum.

In contrast to the non-detect substitution method described for PBDEs, non-detected PCB congeners were substituted with a zero, unless no congeners were detected for a sample, in which case one-half the highest method detection limit among congeners was used as the result. This likely resulted in a downward bias of the results. Appendix B shows the method used to establish representative COC values for datasets with non-detected values.

Publicly-Owned Treatment Works (POTWs)

Description

The Phase 3 project to assess COC loading through POTWs (Ecology and Herrera, 2010) was conducted based on recommendations from the Phase 1 report and results of a Phase 2 study of discharge data from POTWs and industrial wastewater discharges (Envirovision et al., 2008b). Both the Phase 1 and 2 studies found a limited amount of data which could be used to calculate reliable load estimates and recommended collection of additional data in order to refine an assessment of loading.

In order to estimate loads from POTWs, ten facilities were sampled during two events each. Sampling was conducted during February and July 2009 to represent wet and dry seasons, respectively. The POTWs were selected to represent varying types of treatment process, size, and source of wastewater, and were geographically distributed around the Puget Sound region. To obtain load estimates, the project team computed summary statistics for representative COC concentrations then multiplied concentrations by the average annual volume of treated wastewater discharged from all of the 96 POTWs in the Puget Sound basin.

Samples collected from POTWs were analyzed for the COCs (except arsenic, cadmium, mercury, and PCDD/Fs) as well as additional chemicals such as phthalates, semi-volatile organic compounds, pesticides, and herbicides (Appendix B). Few of these chemicals are routinely monitored by POTW operators.

Summary of Major Findings

Most classes of chemicals were detected in POTW effluent, and loads were calculated for all of the COCs analyzed except DDT, cPAHs, nonylphenol, triclopyr, and oil/petroleum. To obtain load estimates, the project team computed summary statistics for representative COC concentrations then multiplied concentrations by the average annual volume of treated wastewater discharged from all of the 96 POTWs in the Puget Sound basin. Due to the limited number of sampling events and atypical weather during the sampling period, the project team was not able to assess any seasonal variations in loadings.

Like the other loading studies, methods for handling non-detected results were required in order to obtain representative COC concentrations for datasets that contained a combination of detected and non-detected results. The POTW project team used the regression-on-order statistical analysis to calculate summary COC concentration statistics rather than simple substitution with one-half of the reporting limit. Perhaps more importantly, no loads were calculated for chemicals that had less than 50% overall frequency of detection. This appears to have introduced a downward bias in basin-wide loading estimates when compared with loading estimates that would have been obtained using the same non-detect substitution methodology that was applied in the surface runoff study. Appendix B shows the method used to establish representative COC values for datasets with non-detected values.

The absolute Puget Sound COC loads were calculated based on discharge volumes from 96 POTWs in the Puget Sound basin, yet not all of these POTWs discharge directly to the marine waters of Puget Sound. Efforts to distinguish POTW loads from surface runoff loads potentially run the risk of double-counting surface runoff loads if surface water sampling is conducted downstream of a POTW outfall. However, none of the surface runoff sampling stations were located downstream of POTW outfalls, and therefore distinct COC loads can be attributable to POTWs and surface runoff based on the methodologies used in the respective projects.

Ocean Exchange and Major Tributaries

Description

Ecology's study of chemical load exchange at the ocean boundary (Gries and Osterberg, 2011) was originally conceived to provide chemical input data for the Puget Sound Box Model (Pelletier and Mohamedali, 2009) after a review of existing data (Serdar, 2008) found the available data were inadequate for the model. The ocean exchange project was designed with a number of marine water column stations to provide calibration data for the box model. The study design was later expanded to include measurements of chemical concentrations in the five rivers having the greatest annual discharges to Puget Sound in order to assess the relationships between river COC concentrations and those in corresponding inland marine waters.

The final sample design for the project included shallow and deep water sampling at three ocean boundary stations (eastern Strait of Juan de Fuca and Haro Strait), marine water at four locations representing the Main, Whidbey, South Sound, and Hood Canal basins, and freshwater and suspended particle sampling near the mouths of the Skagit, Snohomish, Nooksack, Stillaguamish, and Puyallup Rivers. Samples collected from freshwater and marine waters were analyzed for the COCs (except mercury, PCDD/Fs, and triclopyr; oil and petroleum was analyzed in freshwater only) as well as additional chemicals such as phthalates, semi-volatile organic compounds, and chlorinated pesticides (Appendix B).

The exchange of chemical loads at the ocean boundary was calculated by using concentrations in the deep water boundary stations to represent inflow to Puget Sound, and chemical concentrations from the Main, Whidbey, and Hood Canal basin sites to represent water flowing out of Puget Sound. The concentrations representing the inflow and outflow were then multiplied by the known volumes of water flowing in and out of Puget Sound, respectively.

Summary of Major Findings

The study found metals, PCBs, and PBDEs at detectable concentrations in the marine water column, but other COCs were either not detected or detected at such low frequencies that load estimates were not calculated.

Metals, PCBs, and PBDEs were found at detectable concentrations in the water column of rivers, but other COCs were either not detected or detected at such low frequencies that load estimates were not calculated. However, PAHs and DEHP were detected at high frequencies in suspended particles (sampled once in each river versus three water sampling events for each river).

Due to the inherent variability of chemical loads carried by rivers due to constantly shifting stage and conditions, the authors (project team) did not feel that annual loading estimates were supportable based on three sampling events. However, instantaneous daily loads were calculated and reported based on the river discharges recorded during the sampling events.

Uncertainties and limitations of the ocean exchange and major tributaries project were due more to assumptions about hydrology than difficulties associated with handling non-detected laboratory results. Perhaps the largest assumption used by the project team is that COC concentrations at deep water locations in the eastern Strait of Juan de Fuca and Haro Strait represent those flowing into Puget Sound, and COC concentrations from various Puget Sound locations represent concentrations in the outflow. Recommendations were made to improve the assessment of chemical exchange by sampling at more representative boundary locations, namely Admiralty Inlet (sill) and Deception Pass.

Aside from PBDEs, there was little need for the project team to adopt a method for calculating summary statistics for datasets containing non-detects. For PBDEs, the method of substituting a non-detect result with one-half of the estimated quantitation limit was adopted (Appendix B).

PBDE results also showed a very high degree of variability during the study, particularly for marine waters. The project team was unable to provide an explanation for the high degree of variability, but they did note that there was no evidence of sample contamination.

Groundwater

Description

The 2007 Phase 1 report included a specific recommendation for the development of loading estimates for *direct* groundwater discharge to Puget Sound. Although the authors of the Phase 1 report (Hart Crowser et al., 2007) acknowledged the potential for the groundwater pathway to contribute significant toxic chemical loads at the local scale, this pathway was not addressed at | a basin-wide scale. In light of the low mobility characteristics of many of the COCs and the assumed contaminant attenuation capacity of subsurface sediments, the transport of toxic chemicals to Puget Sound via direct groundwater discharge was generally considered to represent a comparatively minor component of the overall loading to Puget Sound marine waters, but this assumption had not been confirmed by a formal technical analysis.

The Phase 1 report recommendation to conduct a sampling-based groundwater loading assessment was not carried forward to a Phase 3 project. Instead, a loading study was conducted using readily available data to address the absence of data on COC loading through a direct groundwater pathway (Pitz, 2011). The study produced estimates of upper-bound (worst-case) mass loads of COCs delivered annually to Puget Sound through the groundwater pathway. The reader should note that i*ndirect* groundwater contributions of toxic chemicals to freshwater streams and rivers draining to Puget Sound are assumed to be represented in the load estimates developed for the surface runoff pathway (Herrera, 2011).

A substantial majority of the chemistry data used for the groundwater loading analysis was from industrial or commercial sites or from facilities that are known or suspected to have point-source-related toxic contamination, typically sampled in response to the Model Toxics Control Act (MTCA) or state Dangerous Waste regulations. Additional sources of data were also found to represent un-impacted (i.e. ambient) groundwater, but these datasets were small compared to the amount of data from impacted areas since most of the COCs are not typically analyzed during routine groundwater monitoring. Only data collected within 500 meters of the marine shoreline were used to derive representative COC concentrations for load calculations.

In an attempt to minimize potential bias from the high proportion of data from contaminated sites, groundwater data were divided into three categories: impacted areas, urban ambient areas, and non-urban ambient areas. Data from each category were pooled separately, and representative concentrations from each category were derived for loading estimates. Fluxes were then calculated by multiplying by the discharge (flow) for each shoreline segment by the COC concentration representing the groundwater data category for the associated 500-m buffer. This approach reduced the groundwater discharge associated with impacted areas to approximately 1% of the overall volume, while non-urban ambient areas discharged approximately 75% of the groundwater.

Sufficient groundwater data were generally available for metals but comparatively scarce for organic compounds. In particular, data on PCBs, PBDEs, PCDD/Fs, triclopyr, and nonylphenol were not sufficient to derive usable representative concentrations for groundwater.

Summary of Major Findings

The groundwater loading analysis found that COC loads from ambient areas generally exceed loads from impacted areas as a result of the comparatively high flow volume in ambient areas. This is similar to the dominant effect that forest land has on absolute COC loads as reported in the surface runoff study. For many of the PAHs, however, the highly elevated concentrations in groundwater from impacted areas more than compensated for lower flows, resulting in comparatively high PAH loads from impacted areas.

Users of the groundwater loading data should be aware of the many limitations and assumptions used in the report. It is an initial effort to calculate direct groundwater loads and has not benefited from the refinement process used for some of the other loading projects. The author (Pitz, 2011) found a low frequency of detection for nearly all of the COCs, and therefore the concentrations used for loading are largely driven by non-detected values (and the substitution methods used to handle non-detects). Appendix B shows the method used to establish

representative COC values for datasets with non-detected values. Complex decision processes were developed to derive COC concentration for use in loading computations. Even the discharge volume estimates used to calculate loads, typically a consistent factor in loading studies for other pathways, ranged by an order of magnitude.

In addition to the limitations and assumptions discussed above, there are also uncertainties about the mobility and attenuation of COCs in groundwater. The groundwater loading study does not attempt to account for attenuation – an additional reason the results represent upper-bound estimates – but instead assumes that the COCs will migrate to the marine boundary in the same concentrations measured in upland groundwater. These and other uncertainties and limitations are thoroughly documented in the groundwater loading report (Pitz, 2011).

Other Projects

Inventory of COC Releases from Primary Sources

Description

In recognition that loading analyses by themselves may not provide adequate information to help Ecology, the Puget Sound Partnership, and others to develop and implement a toxics reduction and control strategy, an inventory of COC releases was undertaken by Ecology as part of the PSTLA Phase 3 effort. This sources inventory, referred hereto after as the *Sources Report* (Ecology, 2011), was conducted with the objectives of (1) identifying major ongoing anthropogenic sources of COCs in the Puget Sound basin and (2) estimating the quantities in which they are released into the environment.

The *Sources Report* focused specifically on the release of COCs from their primary sources. The term *source* was strictly defined as *the object or activity from which a COC is initially released to environmental media or released in a form which can be mobilized and transported in an environmental pathway* (Ecology, 2011). The term *primary source* was used to distinguish the initial release of a COC from a secondary release, such as mobilization of a chemical from a toxic cleanup site.

Examples of releases from primary sources include copper and zinc released from tire and brake pad wear, PAHs formed and released from combustion sources, and motor oil released from vehicle drips and leaks. The *Sources Report* did not estimate releases from secondary or natural sources.

All COCs except DDT were addressed in the *Sources Report*. The quantity of COCs released to the environment from approximately 110 primary sources was estimated from available information; no sampling was conducted for the project.

Summary of Major Findings

The study found that petroleum is released in the largest quantity among COCs, followed by zinc which is the only additional COC released at a rate greater than 1,000 metric tons (t) per year.

Lead, PAHs, and copper are released at rates greater than 200 t/yr, and triclopyr is released at a rate over 100 t/yr. Approximately 30 t/yr of phthalates are released, but the organic chemicals PCBs, PBDEs, PCDD/Fs, and nonylphenol, as well as the metals mercury, arsenic, and cadmium, are generally released at rates near 1 t/yr or less. A summary of release estimates for all COCs is included as Appendix C.

While the *Sources Report* provides only rough estimates for many of the COC release rates, useful information about the relative magnitude of releases from each source is contained in the report. The *Sources Report* also discusses how COCs are released from their sources, providing insight into their transport and fate in the environment following release.

The study of COC releases from primary sources was limited in its ability to provide refined estimates of COC releases, and the authors (project team) were not able to consider all sources of COCs to the Puget Sound environment. The release estimates were based on many assumptions and limited data in many cases. However, these assumptions and associated uncertainties are documented in the report.

Evaluation of Fate and Transport Mechanisms

Description

Understanding the behavior, transport, and fate of chemicals following their initial release from primary sources is a key element in developing strategies for controlling chemicals at their source as well as along their transport pathways following release. The transport and fate of chemicals in the environment is complex, particularly in the upland environment where various media and management practices can affect chemical concentrations, loads, sequestration, and removal. In order to better understand the transport and fate of chemicals in the environment, staff at the U.S. Geological Survey (USGS; Paulson et al., 2011-Draft) conducted an evaluation of transport and fate for copper released from several of the major sources identified in the *Sources Report* (Ecology, 2011). The evaluation of fate and transport mechanisms was not conducted as a discrete PSTLA project but instead was done specifically to enhance this *Assessment Report*. Upon its completion, it will be included as an amendment to this report.

The authors (Paulson et al., 2011-Draft) of the transport and fate evaluation used a mass-balance case study approach to examine factors affecting copper attenuation following release from roof runoff, brake pads, and vehicle tires. Using information from the *Sources Report*, the authors calculated the mass of copper released annually from these sources in two small urban watersheds in King County, Washington. The annual mass of copper discharged from these watersheds was estimated from historical sampling data for the streams comprising the respective watershed outlets. Finally, the authors incorporated information on the types of copper release, the likely locations of the releases, watershed characteristics including best management practices, and stream and water quality characteristics to provide possible explanations for copper attenuation.

The initial scope of this project included a proposal to conduct a similar evaluation for PCB and PBDE transport and fate. However, the lack of data on these chemicals in urban streams and

stormwater precluded the authors' ability to conduct an evaluation for these chemicals using a case study mass-balance approach.

Summary of Findings

Findings of the copper transport and fate evaluation suggested that less than 10% of the copper mass estimated to be released in the watersheds was discharged at the respective watershed outlets. It appeared that the retention of copper in the watersheds was largely governed by the limited amount of water volume discharged from the watershed at the surface outlet; only about 15% of the estimated storm precipitation for each watershed was discharged at the surface outlets. The authors speculated that the retention of copper was due to infiltration into groundwater, trapping of particles by lawns, grassy road-side ditches, road shoulders and road right-of-ways, retention in the many structures installed in the watershed, and settling of particles in vegetated channels in low-gradient portions of the watersheds. Some of the copper released in the watersheds may have been removed by street sweeping, but this portion was likely minor.

The movement of copper by a variety of transport mechanisms through a variety of pathways was discussed by the authors of the fate and transport report (Paulson et al., 2011-Draft), although most of the information provided was speculative due to a lack of data for the multiple environmental media components required for an exhaustive evaluation. In cases where there were available data, the information did not always yield clear conclusions regarding copper transport and fate. For instance, data on copper analyzed from road surfaces resulted in divergent lines of evidence as to whether copper released from brake pads remains on the road surfaces or alternatively is suspended in air and blown away from the roads. This example underscores the complexity and difficulties in understanding the behavior, transport, and fate of chemicals at a small scale.

Hazard Evaluation for COCs in the Puget Sound Basin

The hazards posed by different COCs are not simply associated with the quantities released to the environment or loaded to Puget Sound, but are rather more appropriately evaluated by assessing their concentrations in various media. To assess the relative hazards posed by COCs, Ecology consulted with the ecological toxicology assessment team at King County Natural Resources and Parks to design and conduct a hazard screening of COCs in the Puget Sound basin. The hazard evaluation was not conducted as a discrete PSTLA project but instead was done specifically to enhance this *Assessment Report*.

To evaluate hazards potentially posed by COCs in the Puget Sound basin, King County used a methodology in which readily available observed environmental data for each COC were compiled then compared to concentrations where effects are documented, or to criteria established to protect aquatic life or consumers of aquatic organisms. These comparisons do not attempt to estimate absolute hazards but instead provide a rough discriminator of relative hazards among COCs using a specified methodology. A similar approach was used in a survey conducted several years earlier to evaluate endocrine disrupting chemicals in King County surface waters (King County, 2007).

The hazard evaluation was conducted for the following categories:

- Direct hazard to aquatic life through surface water exposure
- Direct hazard to benthic organisms through sediment exposure
- Direct hazard to aquatic life based on tissue residue levels
- Hazard to wildlife based on ingestion of prey, water, and sediment
- Hazard to human health through fish/seafood consumption

The results of these comparisons were grouped into three broad "level of concern" categories: a Priority 1 level of concern, a Priority 2 level of concern, or unknown (U) level of concern due to lack of sufficient data for an assessment. Results were classified as a Priority 1 when high observed concentrations (e.g. 90^{th} percentile values) exceeded low effects concentrations (e.g. 10^{th} percentile values), selected criteria, or other threshold values. A Priority 2 level of concern was assigned in cases where high observed concentrations were below threshold values. In cases where there were not sufficient data to make a meaningful comparison, results were assigned a U. Appendix D-1 details the thresholds used for comparisons and the minimum data required for the comparisons.

Environmental COC concentrations used for the hazard evaluation were obtained from a number of data sources including Ecology's Environmental Information Management (EIM) system, King County Laboratory Information Management System, USGS data obtained from their online database, the National Oceanic and Atmospheric Administration (NOAA) mussel watch program, and ENVVEST data from the U.S. Department of Defense. Data from the PSTLA Phase 3 Ocean Exchange, Pelagic Fish, and Surface Runoff studies were included among the observed data compilations. Due to the complexities associated with handling non-detects, only detected values were used to represent observed environmental COC concentrations for each of the media assessed.

Effects concentrations were obtained from a variety of sources. For surface waters, effects data were obtained from EPA's ECOTOX database for surface water. Freshwater and marine sediment data were compared to Washington State's Sediment Quality Standards (2003 Floating Percentile values for freshwater). Effects resulting from fish and invertebrate tissue burdens were evaluated using data from the Lower Duwamish Waterway Group and the Lower Willamette Group Remedial Investigation Baseline Ecological Risk Assessments. Wildlife hazards were evaluated by comparing daily COC doses through various exposure routes with daily doses where effects have been demonstrated. National Toxics Rule criteria were used as a basis for evaluating human health hazards.

The hazard evaluation has several limitations that should be considered prior to acting on the results. In particular, the hazard evaluation is not a risk assessment but is instead designed to assess the *relative* level of concern of COCs *across the entire Puget Sound basin*. Although a COC may be assigned Priority 2 or U, this should not be interpreted to mean there are no hazards associated with that COC. Locally, concentration hot spots may exist near major sources, and may cause localized toxicity to aquatic organisms or lead to violations of standards. Finally, no attempt was made to evaluate hazards due to multiple COC exposures.

Details of the methodology and assumptions used for the hazard evaluation are in Appendix D-1. This includes rules for assigning COCs to Priority Levels for each media or receptor evaluated. Additional sections of Appendix D include box plots of the observed and effects concentrations and tabular summaries of the data and the results.

Puget Sound Box Model

Description

The Puget Sound Box Model study was developed as a tool to predict concentrations of PCBs in water, sediment, and biota of Puget Sound (Pelletier and Mohamedali, 2009). Three separate existing models were linked to achieve this task:

- A model to predict the circulation and transport of between regions of Puget Sound and between surface and deep layers of the water column.
- A contaminant fate and transport model to predict water and sediment concentrations of PCBs in response to external loading and internal processes.
- A food web bioaccumulation model to predict PCBs in Puget Sound biota in response to water and sediment concentrations.

Summary of Findings

Based on model outputs, concentrations of PCBs in sediments and biota were found to be very sensitive to external loading. However, the authors found that conclusions about increasing or decreasing trends in Puget Sound PCB mass could not be made due to the wide range in uncertainties regarding current external loading rates.

The median estimates of PCB loading used for the model showed slight increases in the total PCB mass for Puget Sound. However, the model was conducted using available information at the time and did not benefit from data collected during Phase 3 studies that may have aided in model calibration.

Persistent Organic Pollutants in Three Guilds of Marine Species

Description

Investigations of persistent organic pollutants (POPs) in three guilds of marine species were comprised of three separate studies carried out by Washington Department of Fish and Wildlife (WDFW), NOAA, and Fisheries and Oceans Canada. POPs were analyzed in plankton (West et al., 2011a), pelagic fish (West et al., 2011b), and harbor seals (Noël et al., 2010).

Previous studies have suggested that some pelagic species may accumulate higher levels of POPs from the water column and other pelagic components of the food web than from contaminated bottom sediment. In addition, pelagic fish are considered to be the primary source of POPs to southern resident killer whales. The overall goal of the studies on POPs in marine species was to assess where geographically the POPs enter the pelagic food web from stormwater and the

atmosphere, the pathways of POPs within the pelagic food web, and the sources of POPs to species occupying the highest trophic levels (marine mammals, seabirds, and humans). The data from these studies were also intended to be used to refine the Puget Sound Box Model (Pelletier and Mohamedali, 2009).

For the plankton study, investigators analyzed phytoplankton (and other organisms and particles retained in a 20-micron net) and three species of krill which graze on phytoplankton (primarily *Euphausia pacifica*, but also *Thysanoessa spinifera* and *T. raschii).* Samples were obtained from numerous locations around Puget Sound during 2009. Phytoplankton and krill were analyzed for PCBs, PBDEs, PAHs, chlorinated pesticides, and ancillary parameters to assist with interpretation of the results.

In the study of pelagic fishes, researchers analyzed Pacific hake (*Merluccius productus*), which are an important prey item for harbor seals, and walleye pollock (*Theragra chalcogramma*). These species consume a variety of zooplankton such as the krill analyzed in the phytoplankton study, as well as small pelagic forage fishes. Hake or pollock were collected from a number of locations representing six hydrologically distinct waterbodies and one urbanized embayment during 2009. All fish were analyzed whole for PCBs, PBDEs, chlorinated pesticides, and ancillary parameters to assist with interpretation of the results.

The harbor seal (*Phoca vitulina*) study consisted of sampling 24 pups from four widely dispersed locations in Puget Sound. Investigators collected blood, fur, and skin/blubber biopsy samples from the pups. Skin/blubber samples were analyzed for PCBs, PCDD/Fs, PBDEs, chlorinated pesticides, and mercury. Hair and blood samples were analyzed for a variety of parameters to assess the feeding ecology, contaminant trends over space, and effects on their health.

Summary of Findings

For the plankton study, PCBs, PBDEs, DDTs, and PAHs in both particulate organic matter and krill exhibited a correlation with urban waters, and for PCBs and PBDEs in particular, concentrations were lower in less developed, more ocean-influenced basins. This suggests that urban waters represent areas where POPs enter the pelagic food chain.

Although PAHs are known to be metabolized and therefore do not accumulate in tissues of aquatic vertebrates (they were not analyzed for the companion fish and harbor seal studies), the authors of the plankton study found high levels of PAH accumulation in both phytoplankton and krill compared to other POPs. They also noted that a potentially significant implication of this finding was that pacific herring, a primary predator of krill in Puget Sound, exhibited significant exposure to PAHs possibly pointing to krill as a major contaminant transfer pathway. Another finding regarding PAHs was the relatively high concentrations in phytoplankton from non-urbanized basins, and in particular from samples collected near marinas, ferry terminals, or shoreline roadways. This suggests that shoreline development may play an important role in PAH transfer to the pelagic food web.

Patterns of PCB, PBDE, and chlorinated pesticide accumulation similar to plankton were found by authors of the pelagic fish study. Greater size- and lipid-specific accumulations of these chemicals were observed in Pacific hake from more developed basins compared to those with

less development. As pointed out by the authors, the lipid-weighted PCB concentrations in Pacific hake were similar to Pacific herring and higher than in Chinook salmon, all species consumed by harbor seals. However, harbor seals may selectively prey on larger hake, which are typically females and have lower lipid and PCB concentrations, potentially making Pacific herring a greater overall contaminant transfer pathway than hake.

Results of the harbor seal study indicated that Hood Canal (south) seal pups were the least contaminated overall. Pups from the Main Basin of Puget Sound had the highest PCB and mercury levels. PBDE levels from all locations were similar, except Hood Canal which had the lowest concentrations. There was no discernible geographical trend in chlorinated pesticide levels, and PCDD/Fs were detected so infrequently that it was difficult to distinguish trends, although concentrations from the South Sound (east) basin were highest.

The authors of the harbor seal study note that several indicators suggest that the health of harbor seals may be impaired due to the contaminant exposure, particularly PCBs. These indicators include both threshold values based on effects and developed to protect marine mammals, as well as the health indicators measured during the study. They also noted that, while PCB exposure and accumulation may pose health risks to harbor seals, concentrations in seals have decreased appreciably during the 2000s.

There appear to be similarities in contaminant accumulation among the three studies conducted on POPs in marine organisms, and some inferences can be drawn from the results. For instance, there is a consistent geographical pattern seen in PCB concentrations across all species, which indicates the greatest exposure occurs in the Main Basin or embayments therein (e.g. Elliott Bay). However, at the time of this writing there has been no assessment of the results considered as a whole to: (1) Evaluate the consistencies in geographical patterns and (2) Assess the trophic transfer of contaminants between plankton and hake, and between hake and harbor seals. This may be best accomplished through updates and refinements to the Puget Sound Box Model.

A Toxics-Focused Biological Observing System for Puget Sound

Description

The concept of a toxics-focused biological observing system (TBiOS) was developed by authors from NOAA Fisheries, WDFW, and University of California (UC) Davis as a framework for assessing and monitoring toxic chemicals in Puget Sound through biological components (e.g. accumulation, responses, effects) rather than simply monitoring toxic chemicals through the more conventional analysis of water and sediments (Johnson et al., 2010a). Examples of recent research showing adverse effects to aquatic organisms from contaminant exposure are provided. The authors point out that biological monitoring would allow us to evaluate the impacts of toxic chemicals, the effectiveness of efforts to reduce toxic chemicals, effects to the ecosystem and the food web, and the effects of toxic chemical exposure coupled with other stressors.

As proposed, TBiOS would have three major components:

- Region-wide monitoring of toxic chemicals to assess large-scale geographical or temporal trends.
- Localized effectiveness monitoring to assess the impact of local source control or cleanup efforts.
- Diagnostic studies that would help uncover biological effects caused by toxic chemicals and develop monitoring tools to measure these effects.

The authors propose general ideas for the type and scale of monitoring and assessment programs that might be conducted under TBiOS. These programs would be a combination of new initiatives and building upon existing programs, such as the Puget Sound Assessment and Monitoring Program (PSAMP).

Benefits of TBiOS would include:

- Identifying toxic chemical-associated injury to the Puget Sound ecosystem, including the geographic extent and severity of the problem.
- Increasing our understanding of how toxic chemicals move through the Puget Sound ecosystem and accumulate in shellfish, fish, wildlife, and consumers of these organisms.
- Guiding our toxics reduction strategy efforts by helping to identify those watersheds where contaminants are the greatest problem and help us focus where detailed evaluations are most needed.
- Helping us evaluate the effectiveness of regional and localized toxics reductions strategies and actions.
- Establishing cause-and-effect linkages between toxicant exposure and biological impacts.
- Helping develop and establish more protective water quality and sediment guidelines.

Bioaccumulation from Sediments

Description

The study on bioaccumulation modeling was conducted to predict the concentrations of toxic chemicals in organisms resulting from specific concentrations in Puget Sound sediments (Ecology and Environment, 2009). In particular, the model was applied to the organic chemical criteria of Washington's Sediment Quality Standards (SQS). Since the model can also predict water column concentrations, surface water concentrations were predicted to provide comparisons with surface water criteria.

The model used for this effort was based on the Condon bioaccumulation model (Condon, 2007) which was developed to predict PCBs in biota from the Strait of Georgia, and therefore was deemed (with some modifications) adaptable for Puget Sound. This was the same model used as the food-web bioaccumulation component of the Puget Sound Box Model (Pelletier and Mohamedali, 2009; described above) to predict PCB concentrations in biota. The report documents modifications made to the Condon model to accommodate chemicals other than

PCBs, and in some cases, specimens in addition to those used by Condon (e.g. herring and salmon). PCBs, PAHs, and DEHP were the only COCs analyzed for the sediment bioaccumulation study.

Summary of Findings

Results of the modeling exercise showed that, at SQS levels in sediment, predicted water concentrations of PCBs and several PAHs may not be protective of human health, marine organisms, and piscivorous wildlife (PCBs only). Several non-COC organic chemicals were also predicted to be found at non-protective concentrations in water.

The authors of the report conclude that the model provides insights into the behavior and transfer of contaminants in the food web. However, they note the vast complexity of food-web modeling and caution users to consider the numerous assumptions and uncertainty before applying this or other generalized models, particularly if the model may be used for regulatory and management decisions.

Pharmaceuticals and Personal Care Products in POTWs

Description

The study of pharmaceuticals and personal care products (collectively referred to as PPCPs) was a screening-level effort carried out by Ecology and EPA Region 10 staff during 2008 (Lubliner et al., 2010). The study consisted of analyzing one-day composite samples of influent, effluent, and sludge from four POTWs in the Puget Sound region and one POTW in Hayden, Idaho. All of the plants had different processes for treatment of wastewater, with two plants employing secondary treatment and three plants employing tertiary treatment for nitrogen and phosphorus removal.

The screening-level of PPCPs was conducted due to concerns emerging from recent studies (cited in Lubliner et al., 2010) documenting PPCP presence in the aquatic environment and the possibility PPCPs may lead to effects which are not widely understood or have not been investigated. Since POTWs are a major step along the pathway from consumer use to release in the environment, the study sought to generate information about the effectiveness of POTWs in removing these chemicals. The study analyzed 72 PPCPs, 27 hormones and steroids, and 73 semi-volatile organic chemicals. PAHs, DEHP, and nonylphenol were the only COCs among the analytes selected for the PPCP study.

Summary of Findings

The authors of the study found that PPCPs are detected routinely in municipal wastewater and that their removal in POTWs varies by chemical and treatment process. Approximately one-fifth to one-half of the analytes were reduced to levels below reporting limits in the effluent. Overall, the combination of enhanced biological nutrient removal and filtration processes was found to provide the greatest PPCP removal effectiveness, although the authors note that this treatment process is employed by relatively few POTWs in the Puget Sound basin.

As for the COCs analyzed, DEHP was found in all influent samples, and concentrations were greatly reduced in all but one of the POTW effluents. Nonylphenol (4-nonylphenol) was rarely detected in POTW influent or effluent. None of the 16 PAHs analyzed were detected in either influent or effluent samples, although indeno(1,2,3-cd) pyrene was selected as one of the few target analytes for biosolids and was detected in three-quarters of the samples analyzed.

Chemical-Specific Assessments

Arsenic

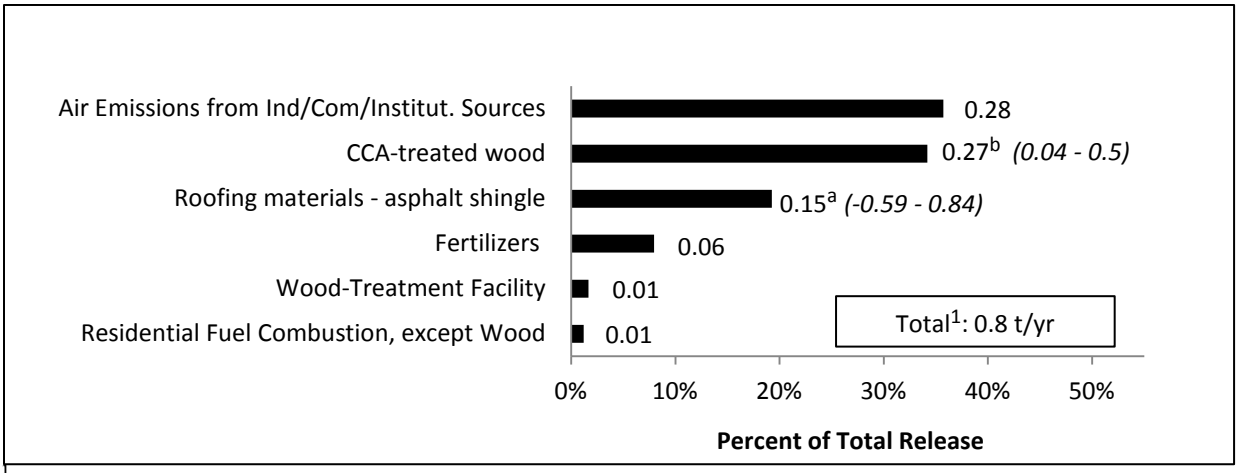
Arsenic is a naturally occurring element in the earth's crust and is nearly always detectable at concentrations >0.1 ug/l in water and >100 ug/kg (dw) in sediments from freshwater and marine environments (PTI, 1991; Serdar, 2008; Hallock, 2010; Appendix D). Toxicity in water varies due to pH and redox potential (Eisler, 1988a).

Historically, approximately 70% of the global arsenic emissions are from anthropogenic sources, with the remaining 30% due to weathering of soils and rock (Moore and Ramamoorthy, 1984). In the Puget Sound region, historical releases may have occurred due to uses of arsenic compounds as pesticides as well as releases from large industrial sources. The Asarco Smelter in Tacoma emitted arsenic for decades and may have resulted in large swaths of the Puget Sound area with elevated arsenic (PTI, 1991; San Juan, 1994).

Major Releases from Primary Sources

An inventory of primary arsenic sources suggests that approximately 0.8 metric tons (t) is released annually from anthropogenic sources in the Puget Sound basin (Ecology, 2011). The largest current source of arsenic to the environment is point-source air emissions, accounting for approximately one-third of the total release (Figure 4).

There is a large amount of uncertainty around estimates of arsenic released from roof runoff and CCA-treated wood, sources which combined account for about one-half of anthropogenic arsenic released in the Puget Sound basin. For other sources – fertilizer application, a wood treatment facility that releases arsenic primarily to surface water, and residential fuel use (excluding wood) – arsenic releases are relatively minor.



[1] Sum of best estimates. Best estimates are either the mean, mid-point, median, or most reasonable estimate for each source.

[a] Mean

[b] Mid-point of range

Figure 4. Total Arsenic Release in the Puget Sound Basin (values shown are t/yr).

Major Delivery Pathways and Loading

Data on major delivery pathways for arsenic loading were obtained from PSTLA loading studies and are included in Table 1. As mentioned previously, loading through direct groundwater discharge was estimated from literature values, whereas loadings through other pathways were estimated from field studies specifically designed to estimate loads.

Table 1. Total Arsenic Loads (t/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater (a)	0.06	0.43	0.79
Air Deposition	0.25	0.35	0.54
Surface Runoff	13.5	16.9	23.4
POTWs	NA	NA	NA
Ocean Exchange (b)	-28	-23	-24

(a) Lowest, highest, and mid-point of estimated loads
(b) Negative values indicate a net outflow at the ocean boundary
NA=not analyzed

Estimated groundwater loads of arsenic range by an order of magnitude (0.06 – 0.8 t/yr). The range in estimates is due primarily to differences in flows used to establish the possible range of loads; groundwater discharges used in the groundwater loading study ranged by an order of magnitude.

Arsenic was not measured in POTWs for the Phase 3 loading study. Phase 1 estimates suggest that arsenic loads from industrial wastewater are potentially substantial (0.2 – 14.6 t/yr; Hart Crowser et al., 2007), but this is based on a limited dataset and is far in excess of industrial discharge of arsenic reported in the Toxics Release Inventory (TRI) (0.01 t/yr total) (Ecology, 2011). In contrast, Phase 1 estimates for arsenic loading via municipal wastewater were based on a small set of non-detected sample results, and the load estimates appear to be unrealistically small (0 – 1 kg/yr). Due to the unreliability of the arsenic loads discharged through wastewater treatment, estimates from this pathway were not included in Table 1.

The deposition of atmospheric arsenic directly to the marine waters of Puget Sound is approximately 0.25 – 0.54 t/yr, based on fluxes ranging from 0.06 – 0.65 ug/m^2/d. Median arsenic fluxes are generally ≤0.2 ug/m^2/d, except for one location within a high-density urban (Tacoma) area including a nearby pulp mill, metal refiners, other industrial activities, and in close proximity to major roadways including interstate highways. Fluxes at this location are consistently several times higher than at other locations.

Surface runoff loads for arsenic are estimated to be approximately 13.5 – 23.4 t/yr for the entire Puget Sound basin based on the surface runoff study. Nearly all (97%) of the total arsenic was in the dissolved form during baseflows; the fraction of dissolved arsenic decreased during storm flows to 74% of the total. Arsenic concentrations observed across base and storm flows were found to significantly correlate with total suspended solids (TSS) in residential and forest areas, partly due to a seasonal first-flush episode in the forested sub-basins.

Forested areas had much lower arsenic concentrations than other land covers. Commercial/ industrial and agricultural land covers had the highest arsenic concentrations, approximately double those in residential areas and four-fold higher than forests during baseflows. For all of the land covers, arsenic concentrations decreased during storm events, particularly in commercial basins.

Overall, there appears to be a net export of arsenic out of the Puget Sound due to exchange of marine waters, although total arsenic concentrations in the incoming marine waters (1.44 – 1.52 ug/l; 25^{th} -75^{th} percentile) are slightly higher than concentrations in the outgoing marine waters (1.36 – 1.49 ug/l). Total net export was 24 – 28 t/yr based on an inflow of 842 – 889 t/yr and an outflow of 870 – 913 t/yr.

The net sum of arsenic loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25^{th}, median (50^{th}), and 75^{th} percentile values is -14, -5.3, and 0.7 t/yr, respectively. Under estimates at the 25^{th} and median levels, there is a net export of arsenic out of Puget Sound due to the large mass exchanged at the ocean boundary. Estimates at the 75^{th} percentile suggest that a net outflow at the ocean boundary is balanced by loads from the watershed and from air deposition.

Arsenic loading from pathways that may represent *partially controllable* sources of COCs (i.e. all of the major pathways except ocean exchange) is shown as ranges (25^{th} -75th percentiles) and median for each pathway in Figure 5. Each pathway represented as a contribution to the total load is displayed in Figure 6.

Total arsenic loading from the major pathways assessed is 14 – 25 t/yr. Surface runoff accounts for the largest pathway (95% – 98%), followed by air deposition (2%). Groundwater potentially accounts for up to 3% at the upper end of the estimated range, but this value should be viewed with caution since it is based on literature values of arsenic in wells and variable estimates of groundwater flow. At the lower end of the load range, arsenic in groundwater is estimated to account for <1% of the load to Puget Sound. The loading studies failed to provide estimates of arsenic loading through POTWs, and earlier estimates (Envirovision et al., 2008b) provide little information on which to base reasonable load estimates.

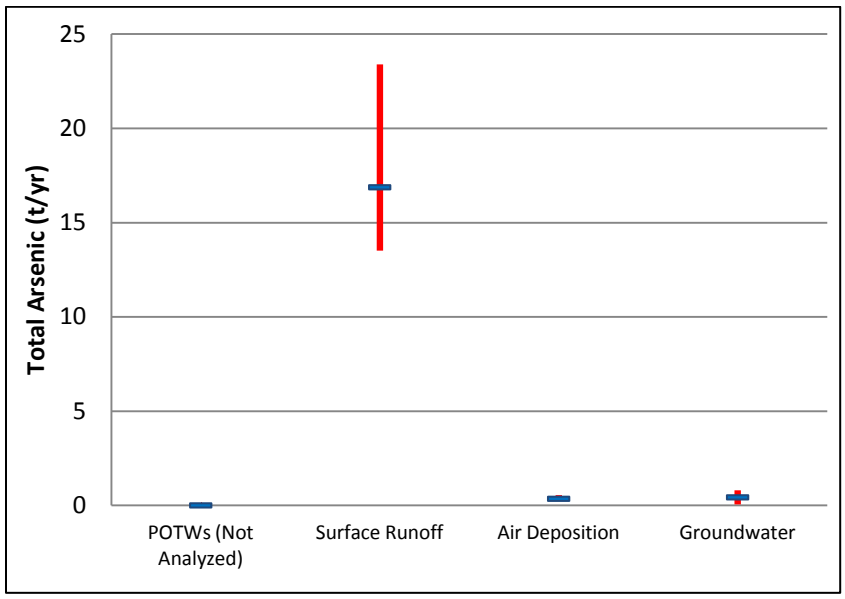


Figure 5. Range (25th - 75th percentiles) and Median Total Arsenic Loads to Puget Sound from Each Major Delivery Pathway.

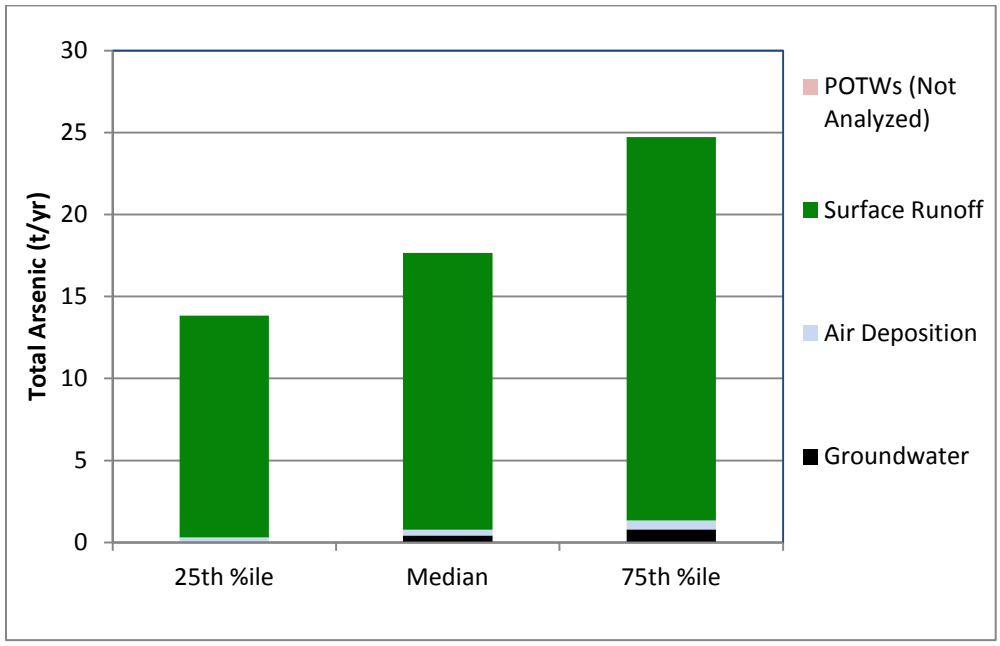


Figure 6. Total Arsenic Loads to Puget Sound as the Sum of Major Delivery Pathways.

Relationship Between Sources and Pathways/Loading

The conceptual model of arsenic transport and fate following release suggests that much of the arsenic will be transported to Puget Sound through surface runoff. Direct deposition to marine waters from atmospheric deposition and loading through direct groundwater discharges appear to be only a small portion of overall loading.

Although not all anthropogenic sources of arsenic were assessed in estimates of primary releases, the total release of arsenic was only a small fraction (3 - 6%) of estimated loads delivered from major controllable pathways. Total arsenic initially emitted to the air (0.3 t/yr) is similar to air deposited on the marine waters (0.25 – 0.54 t/yr), although estimates for releases are for the entire Puget Sound basin and it is not known what portion of the air releases in the basin are transported out of the airshed and what portion of deposited arsenic is imported from outside airsheds.

The inventory of anthropogenic releases suggests that little arsenic is released directly to road surfaces or released through leaching by precipitation. Leaching of arsenic from CCA-treated wood, asphalt shingle roofs, and leaching or mobilization of arsenic-containing agricultural fertilizers only accounts for approximately 0.5 t/yr. In contrast, surface runoff loads range from 13.5 – 23.4 t/yr.

Arsenic loads in surface runoff during baseflow and storm flow conditions are roughly equal, and arsenic concentrations in streams from all land covers decline during storm events. This suggests little enrichment of streams by arsenic mobilized during storms. Instead, stormwater delivered to streams appears to simply dilute the baseflow levels of arsenic. This may indicate that arsenic originates primarily from groundwater.

Ongoing releases of anthropogenic sources of arsenic appear to be minimal, suggesting that a high proportion of the load is due to natural sources or historical releases which reside in the aquatic freshwater environment. Large historical sources of arsenic in the region include the Asarco Smelter in Tacoma which emitted arsenic for decades and may have resulted in large swaths of the Puget Sound area with elevated arsenic (PTI, 1991; San Juan, 1994).

Hazard Evaluation

The hazard evaluation indicates that arsenic is a Priority 2 or unknown level of concern for the categories assessed except for freshwater sediment (Table 2). There were few or no effects data available to adequately compare observed concentrations to surface water effects or human health criteria. Hazard due to tissue residue effects and effects to wildlife were not evaluated.

The limited set of freshwater effects for dissolved arsenic is three orders of magnitude above the 90^{th} percentile value for dissolved arsenic in freshwater (approx. 2 ug/l) and two orders of magnitude below the chronic and acute water quality criteria.

Table 2. Summary of Hazard Evaluation for Arsenic.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.* or *Acute WQC* or *Chronic WQC***	**Level of Concern**
	Freshwater	4,528	85%	No/INS	U
	Nearshore Marine	43	93%	INS	U
	Offshore Marine	58	91%	No/INS	U
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. > *SQS***	**Level of Concern**
	Freshwater	623	82%	Yes	Priority 1
	Nearshore Marine	399	70%	No	Priority 2
	Offshore Marine	372	84%	No	Priority 2
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.***	**Level of Concern**
	Not Analyzed				
Wildlife		**Total N**	**FOD**	**Daily Dose > *10% of Lowest Effects Dose***	**Level of Concern**
	Not Analyzed				
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. > *NTR Criterion***	**Level of Concern**
	Freshwater	a	>99%	INS	U
	Nearshore Marine	a	100%	INS	U
	Offshore Marine	a	100%	INS	U

Yellow highlight indicates Priority 1 Level of Concern
FOD=Frequency of detection
INS=Insufficient observed data available for comparison to effects data
U=Unknown level of concern due to lack of sufficient data for adequate comparison
[a] N = 8 for freshwater fish, 11 for nearshore marine bivalves, 2 for nearshore marine invertebrates, 2 for offshore marine bivalves, and 0 for all other categories

Median arsenic concentrations in both freshwater and marine sediments are below the lowest guidelines or standards. However, in freshwater sediments the 90th percentile (approx. 40 mg/kg dw) of observed arsenic concentrations exceed the floating percentile SQS concentration. More than 25% of observed concentrations exceed the Canadian threshold effect level (TEL) and the consensus-based threshold effects concentration (TEC), more than 10% of values exceed the Canadian probable effects level (PEL) and the consensus-based probable effects concentration (PEC), and 5% of observed freshwater arsenic concentrations are at the floating percentile cleanup screening level (CSL).

In marine sediments, arsenic concentrations are nearly all (>95%) well below established guidelines and standards (including the SQS) except for the Canadian TEL which is exceeded by arsenic more than 25% of the observed concentrations in both nearshore and offshore sediments.

Arsenic hazards to human health were difficult to assess due to the paucity of data on inorganic arsenic, the form of arsenic for which the NTR has established criteria. No data were available for inorganic arsenic in edible freshwater fish or invertebrates. In marine waters, the few available data on inorganic arsenic in edible tissues (2 samples each for nearshore invertebrates and offshore fish, 11 samples for nearshore bivalves) all exceed (did not exceed) the NTR criteria. However, due to the small sample size available for assessment and lack of any data for one or more seafood categories, the overall human health level of concern for marine seafood was assigned an unknown level of concern.

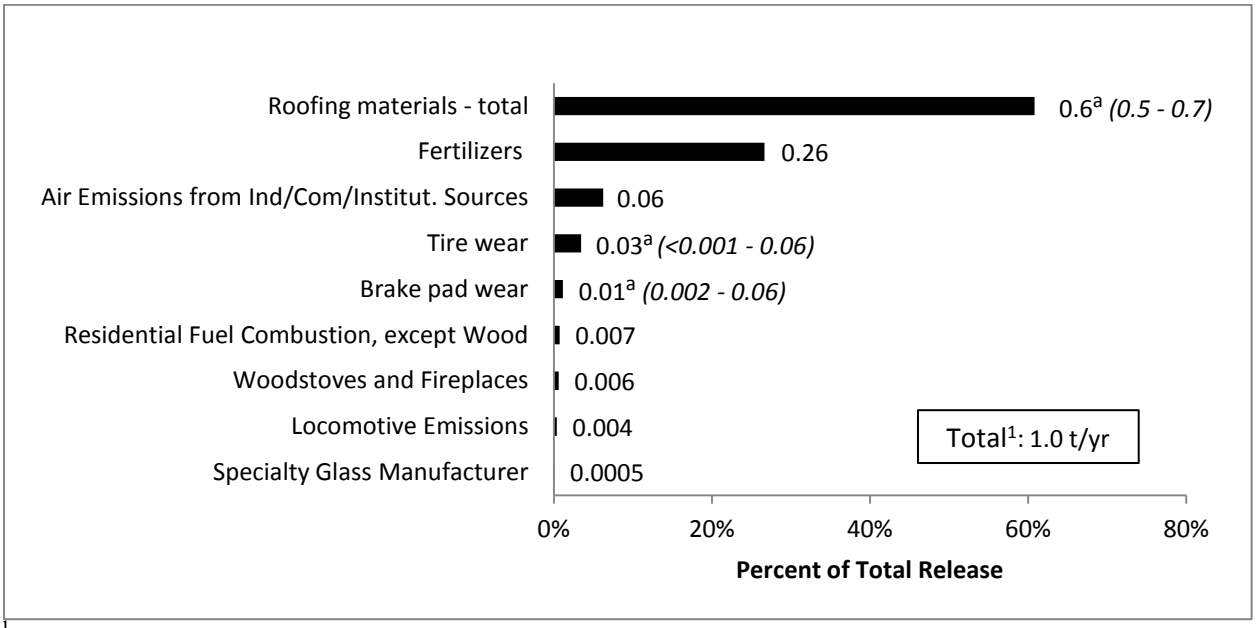
Cadmium

Cadmium is a heavy metal naturally occurring in the earth's crust and is nearly always detectable at concentrations >0.01 ug/l in water and >50 ug/kg (dw) in sediments from freshwater and marine environments (PTI, 1991; Serdar, 2008; Hallock, 2010; Appendix D). Environmental levels of cadmium have been increased above natural levels due largely to manufacturing-related releases, combustion of fossil fuels, and the use of phosphorus fertilizers. Historically, the largest source of cadmium contamination was associated with waste from the now-defunct Asarco smelter in Tacoma (PTI, 1991).

Major Releases from Primary Sources

An inventory of primary cadmium sources suggests that approximately 1.0 metric tons (t) is released annually from anthropogenic sources in the Puget Sound basin. The largest current source of cadmium to the environment appears to be leaching of cadmium from roofing material (Figure 7). Approximately 0.6 t/yr is released from rooftops, with 0.4 t/yr from asphalt composite shingles, 0.14 from built-up roofs, and a small fraction (0.03 t) from metal roofs.

Release from fertilizers accounts for most of the remainder of cadmium release (0.26 t/yr, 27% of total). Road-related sources such as tire and brake pad wear account for approximately 4% of the total release (0.04 t/yr). Comparatively little cadmium is released from industrial sources, with only a single facility reporting fugitive air releases during the previous ten years. Other air releases include locomotives and residential fuel use, but combined, these sources account for only about 6% of the total cadmium release.



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

[a] Mean

Figure 7. Total Cadmium Release in the Puget Sound Basin (values shown are t/yr).

Major Delivery Pathways and Loading

Data on major delivery pathways for cadmium loading were obtained from PSTLA loading studies and are included in Table 3. As mentioned previously, loading through direct groundwater discharge was estimated from literature values, whereas loading through other pathways was estimated from field studies specifically designed to estimate loads.

Table 3. Total Cadmium Loads (t/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater (a)	0.012	0.22	0.43
Air Deposition	0.031	0.052	0.074
Surface Runoff (b)	0.01	0.01	0.02
POTWs	NA	NA	NA
Ocean Exchange	3.2	2.9	3.9

(a) Lowest, highest, and mid-point of estimated loads

(b) Surface runoff loads based on storm flows only; not detected in any land covers during baseflows and in commercial/industrial areas only during storm flows

NA=not analyzed

The estimated range of groundwater cadmium loads is large (0.012 – 0.43 t/yr). The range in estimates is due primarily to differences in flows used to establish the possible range of loads; groundwater discharges used in the groundwater loading study ranged by an order of magnitude. Cadmium was not measured in POTWs for the Phase 3 loading study. Phase 1 estimates suggest that cadmium loads through industrial wastewater may range from 0.02 to 0.9 t/yr (Hart Crowser et al., 2007), but this is based on a limited dataset and exceeds the industrial discharge of

cadmium reported in the TRI by one to three orders of magnitude (Ecology, 2011). Phase 1 estimates for cadmium loading via municipal wastewater were based on a small set of non-detected sample results, and the load estimates appear to be small (1 – 4 kg/yr).

The deposition of atmospheric cadmium directly to the marine waters of Puget Sound is approximately 0.031 – 0.074 t/yr, based on fluxes ranging from 0.007 – 0.11 ug/m^2/d. Median cadmium fluxes were generally <0.02 ug/m^2/d, except for one location within a high-density urban (Tacoma) area including a nearby pulp mill, metal refiners, other industrial activities, and close to major roadways including interstate highways. Cadmium fluxes at this location were elevated three- to four-fold above fluxes at other locations.

Surface runoff loads for cadmium are estimated to be approximately 0.01 – 0.02 t/yr for the entire Puget Sound basin based on the surface runoff study. Total cadmium was not detected in baseflow samples, although dissolved cadmium was detected in 13% of baseflow samples. Dissolved cadmium was also detected at a higher frequency than total cadmium in storm flow samples (34% and 8%, respectively). In either case, cadmium was detected at such low rates that estimates to calculate loads in surface waters were driven by the analytical reporting limits, and no estimate was derived for baseflow loads.

No cadmium was detected in surface runoff from residential or forested areas. Total cadmium was only detected in commercial/industrial areas – largely as a result of a seasonal first-flush episode – but the overall low frequency of detection was low (27%). However, dissolved cadmium was detected at a high rate in commercial/industrial areas (87%) and a more moderate rate in agricultural land covers (34%).

Overall, there appears to be a large net import of cadmium through exchange at the ocean boundary compared to other load pathways, although this estimate is based on very limited data. Total cadmium concentrations (25^{th} -75^{th} percentile) in the incoming marine waters are 0.091 – 0.097 ug/l, and total cadmium concentrations in the outgoing marine waters are 0.072 – 0.077 ug/l. Total net import was 3.2 – 3.9 t/yr based on an inflow of 53 – 57 t/yr and an outflow of 50 – 53 t/yr.

The net sum of cadmium loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25^{th}, median (50^{th}), and 75^{th} percentile values is 3.3, 3.0, and 4.4 t/yr, respectively. Under all of these estimates, there is a net cadmium load to Puget Sound.

Cadmium loading from pathways that may represent *partially controllable* sources of COCs (i.e. all of the major pathways except ocean exchange) are shown as ranges (25^{th} -75th percentiles) and median for each pathway in Figure 8. Each pathway represented as a contribution to the total load is displayed in Figure 9.

Total cadmium loading from the major pathways assessed is estimated to be 0.05 – 0.53 t/yr. Groundwater potentially accounts for the largest loading pathway, at 24 – 82% of the total, but the groundwater numbers should be viewed with caution since they are driven largely by analytical reporting limits rather than measurable sample concentrations. The comparatively

small contribution from surface runoff (4% – 15% of total) is also derived by non-detected values. Since the groundwater loading estimates are highly variable and load estimates at the low end of the range are small (0.012 t/yr), atmospheric deposition potentially represents the largest loading pathway (up to 62% of total load) if all estimates are assumed to be at the low end of the range.

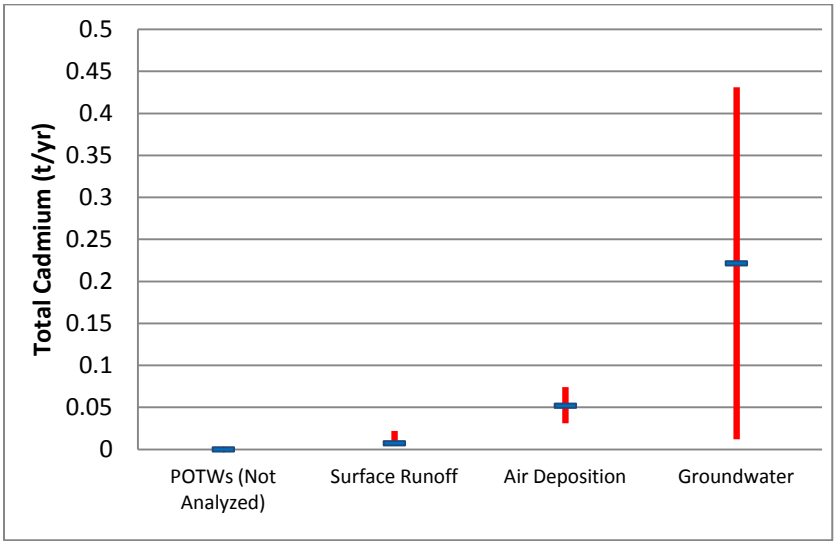


Figure 8. Range (25th - 75th percentiles) and Median Total Cadmium Loads to Puget Sound from Each Major Delivery Pathway.

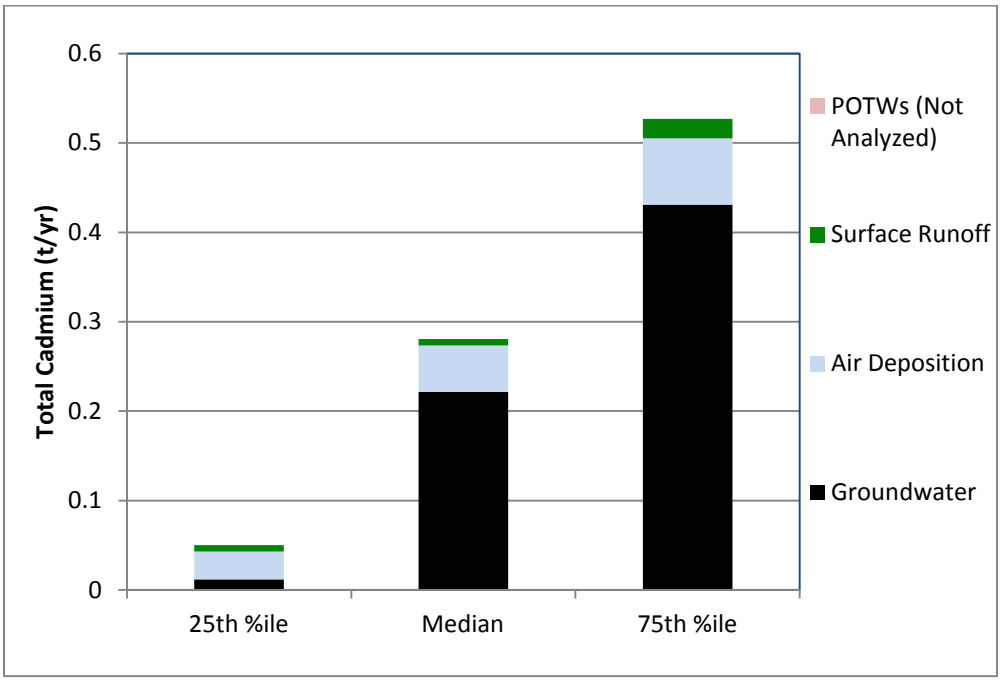


Figure 9. Total Cadmium Loads to Puget Sound as the Sum of Major Delivery Pathways.

Relationship Between Sources and Pathways/Loading

The conceptual model of cadmium transport and fate following release is limited by a lack of data on cadmium in major loading pathways. The available information suggests that most of the anthropogenic cadmium released from primary sources – leaching from roofing material and fertilizers – would be initially mobilized in surface runoff. This may be reflected in the presence of cadmium in storm samples and a lack of cadmium in baseflow samples, but the data are too limited to establish any patterns with even moderate confidence.

The combined loading of cadmium from major controllable pathways (0.050 – 0.53 t/yr) is approximately 20 times lower than releases from all primary sources combined. Air releases of cadmium to the watershed (0.06 t/yr) are not substantially different than atmospheric deposition in marine waters (0.03 – 0.07 t/yr), although marine areas represent only about one-sixth of the watershed. It is not known what portion of the air releases in the basin is transported out of the airshed and what portion of deposited cadmium is imported from outside airsheds.

Hazard Evaluation

The hazard evaluation indicates that cadmium is a Priority 2 or unknown level of concern for the categories assessed except for freshwater sediment (Table 4). Hazards due to tissue residue effects, effects to wildlife, and effects to human health were not evaluated.

The 90th percentile values for dissolved cadmium in freshwater (approx. 0.3 ug/l) are one-half of the concentrations in freshwater where 10% of effects have been documented, and are below the chronic and acute water quality criteria. However, more than 5% of the observed cadmium concentrations in freshwater are above the chronic water quality criterion, and at least 10% of the observed values are above concentrations where 5% or more of the effects have been documented.

Despite enrichment from oceanic waters, the gap between observed cadmium concentrations and effects levels or criteria is much larger for marine waters than for freshwater. The 90th percentiles of dissolved cadmium concentrations in both nearshore and offshore marine waters (approx. 0.09 ug/l and 0.06 ug/l, respectively) are two orders of magnitude below the lowest 5-10% of effects as well as the acute and chronic water quality criteria. However, the limited dataset ($n<50$) for dissolved cadmium nearshore and offshore marine waters does not meet the criteria established for an adequate comparison, and this evaluation was assigned an unknown level of concern.

Median cadmium concentrations in both freshwater and marine sediments are below the lowest guidelines or standards. However, in freshwater sediments the 75th percentile (approx. 0.9 mg/kg dw) of observed cadmium concentrations exceed the floating percentile SQS concentration, and more than 10% of the observed values exceed the floating percentile CSL.

Cadmium concentrations in marine nearshore and offshore sediment have 90th percentile levels (approx. 1.0 mg/kg dw) one-fifth the SQS. However, the observed concentrations for both nearshore and offshore sediments exceed the Canadian TEL at the 75th percentile levels, and 5% of the nearshore concentrations are at the Lowest Apparent Effects Threshold (LAET).

Table 4. Summary of Hazard Evaluation for Cadmium.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.* or *Acute WQC* or *Chronic WQC***	**Level of Concern**
	Freshwater	4,166	7%	No	Priority 2
	Nearshore Marine	32	100%	INS	U
	Offshore Marine	42	100%	INS	U
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. > *SQS***	**Level of Concern**
	Freshwater	764	67%	Yes	Priority 1
	Nearshore Marine	462	62%	No	Priority 2
	Offshore Marine	471	70%	No	Priority 2
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.***	**Level of Concern**
	Not Analyzed				
Wildlife		**Total N**	**FOD**	**Daily Dose > *10% of Lowest Effects Dose***	**Level of Concern**
	Not Analyzed				
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. > *NTR Criterion***	**Level of Concern**
	Not Analyzed				

Yellow highlight indicates Priority 1 Level of Concern
FOD=Frequency of detection
INS=Insufficient observed data available for comparison to effects data
U=Unknown level of concern due to lack of sufficient data for adequate comparison

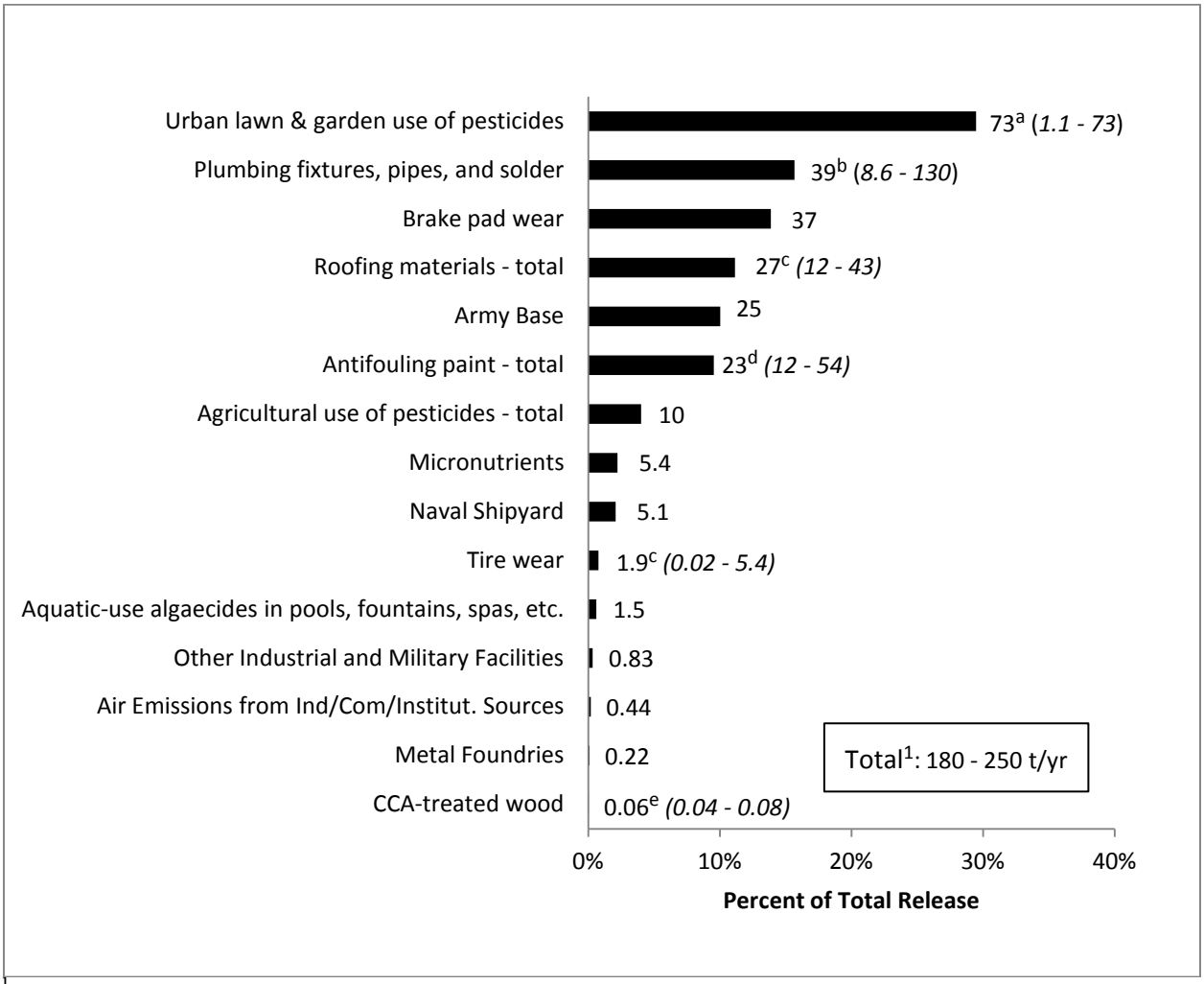
Copper

Copper is a heavy metal naturally occurring in the earth's crust and is nearly always detectable at concentrations >0.5 ug/l in water and >3,000 ug/kg (dw) in sediments from freshwater and marine environments (PTI, 1991; Serdar, 2008; Hallock, 2010; Appendix D). Copper is an essential element for all living organisms and is generally not toxic to humans and terrestrial wildlife at typical environmental concentrations. However, copper can be highly toxic to aquatic organisms at low concentrations.

Major Releases from Primary Sources

An inventory of primary copper sources suggests that approximately 180 – 250 metric tons (t) is released from anthropogenic sources annually in the Puget Sound basin. The largest current

source of anthropogenic copper to the environment could potentially be due to urban lawn and garden use (Figure 10). Copper use in urban landscaping as an herbicide/fungicide or possibly as a micronutrient accounts for approximately 73 t/yr by some estimates, but may be as little as 1 t/yr based on other estimates. The authors of the *Sources Report* note the difficulty in obtaining accurate information on pesticide use and note that these data should be viewed with caution since they were not derived from market data, and use rates are difficult to estimate without this information (Ecology, 2011). Estimates of copper used as an agricultural pesticide and micronutrient in the Puget Sound basin are probably more accurate than estimates for urban use, but agricultural use of copper only represents 4% – 6% of the total annual release.



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

[a] High end of range

[b] Median

[c] Average

[d] Sum of means for recreational and commercial and mid-point for naval vessels

[e] Mid-point of range

Figure 10. Total Copper Release in the Puget Sound Basin (values shown are t/yr).

Other major releases of copper are leaching from plumbing components (39 t/yr), vehicle brake pad and tire wear (37 t/yr and 2 t/yr, respectively), and leaching from vessel anti-fouling paint (26 t/yr). Industrial, commercial, and institutional facilities are estimated to release a total of 31 t/yr of copper, with approximately four-fifths reported from activities at the Fort Lewis Army Base, possibly as solid copper from munitions use.

Smaller releases of copper also occur following the use of copper **c**ompounds in fountains and spas as an algaecide (1.3 t/yr), and from leaching of copper from CCA-treated wood (0.05 t/yr). There are no currently permitted uses of copper as an aquatic herbicide/algaecide in surface waters of the Puget Sound basin.

Major Delivery Pathways and Loading

Data on major delivery pathways for copper loading were obtained from PSTLA loading studies and are included in Table 5. As mentioned previously, loading through direct groundwater discharge was estimated from literature values, whereas loadings through other pathways were estimated from field studies specifically designed to estimate loads.

Table 5. Total Copper Loads (t/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater (a)	0.099	2.19	4.27
Air Deposition	1.9	2.7	4.1
Surface Runoff	28.4	35.7	66.1
POTWs	2.5	4.33	5.5
Ocean Exchange (b)	-110	-100	-30

(a) Lowest, highest, and mid-point of estimated loads
(b) Negative values indicate a net outflow at the ocean boundary

Estimated groundwater loads of copper range by an order of magnitude (0.1 – 4.3 t/yr). The range in estimates is due primarily to differences in flows used to establish the possible range of loads; groundwater discharges used in the groundwater loading study ranged by an order of magnitude.

Copper discharged from POTWs in the Puget Sound basin accounts for loads estimated to be 2.5 – 5.5 t/yr. Phase 1 estimates suggest that copper loads from industrial wastewater are potentially substantial (6 t/yr; Hart Crowser et al., 2007), but this is based on a limited dataset and is far in excess of industrial discharge of copper to surface waters or transferred to POTWs as reported in the TRI (<0.7 t/yr total) (Ecology, 2011).

The deposition of atmospheric copper directly to the marine waters of Puget Sound is approximately 1.9 – 4.1 t/yr, based on fluxes ranging from 0.5 - 19 ug/m^2/d. Copper fluxes were generally <2 ug/m^2/d, except for one location within a high-density urban area (Tacoma) including a nearby pulp mill, metal refiners, other industrial activities, and close to major roadways including interstate highways. Copper fluxes at this location were consistently an order of magnitude higher than other locations around the Puget Sound region, and were elevated

five-fold above a nearby station that did not have the same air pollution influences in such close proximity.

Surface runoff loads for copper are estimated to be approximately 28.4 – 66.1 t/yr for the entire Puget Sound basin based on the surface runoff study. Overall, dissolved copper accounts for 63 – 76% of total copper concentrations in surface water, with the higher proportion of dissolved copper occurring during baseflows.

Total copper concentrations were highest in agricultural areas, whereas dissolved copper concentrations were similar in agricultural and commercial/industrial area. In agricultural, commercial/industrial, as well as forested areas, elevated copper concentrations appeared to correspond with elevations in TSS. The apparent links with TSS in forested sub-basins may be related to a substantial seasonal first-flush episode in forests. However, there appeared to be little overall increase in forest copper concentrations during storm events, whereas the median total copper concentrations increased by two- to three-fold during storms for all other land covers.

Based on sampling marine waters, there appears to be a large net export of copper at the ocean boundary, although this estimate is based on very limited data. Total copper concentrations (25^{th} -75^{th} percentile) in the incoming marine waters are 0.24 – 0.41 ug/l, and total copper concentrations in the outgoing marine waters are 0.38 – 0.46 ug/l. Total net export was 30 – 110 t/yr based on an inflow of 140 – 240 t/yr and an outflow of 250 – 270 t/yr.

The net sum of copper loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25^{th}, median (50^{th}), and 75^{th} percentile values is -77, -55, and 50 t/yr, respectively. Under the 25^{th} percentile and median estimates, there is a net export of copper out of Puget Sound due to the large mass exchanged at the ocean boundary. However, when the 75^{th} percentile values are summed, the large surface water load outweighs the smaller export at the ocean boundary, leading to a net load of copper load to Puget Sound.

Copper loading from pathways that may represent *partially controllable* sources of COCs (i.e. all of the major pathways except ocean exchange) are shown as ranges (25^{th} -75^{th} percentiles) and median for each pathway in Figure 11. Each pathway represented as a contribution to the total load is displayed in Figure 12.

Total copper loading from the major pathways assessed is 33 – 80 t/yr. Surface water runoff accounts for the largest pathway (83 – 86%), followed by POTWs (7 – 8%) and air deposition (5 – 6%). Groundwater potentially accounts for up to 5% at the upper end of the estimated range, but this value should be viewed with caution since it is based on literature values of copper in wells and rough estimates of groundwater flow. At the lower end of the load range, copper in groundwater is estimated to account for <1% of the load to Puget Sound.

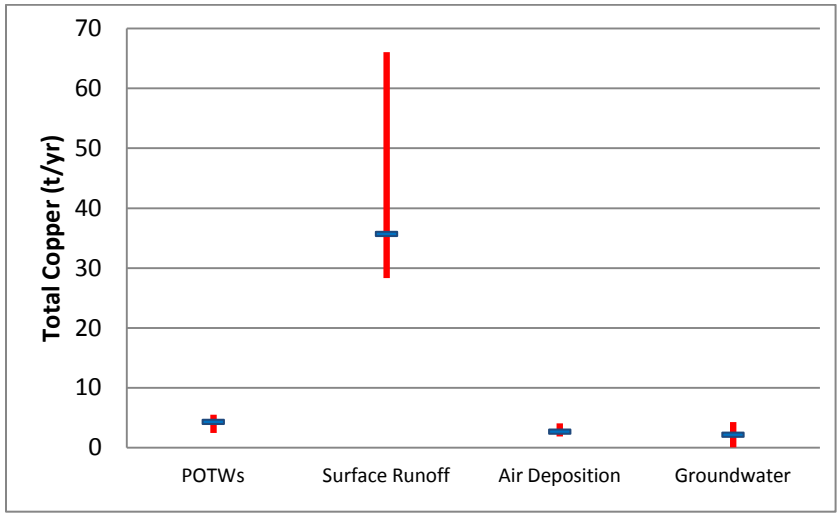


Figure 11. Range (25th - 75th percentiles) and Median Total Copper Loads to Puget Sound from Each Major Delivery Pathway.

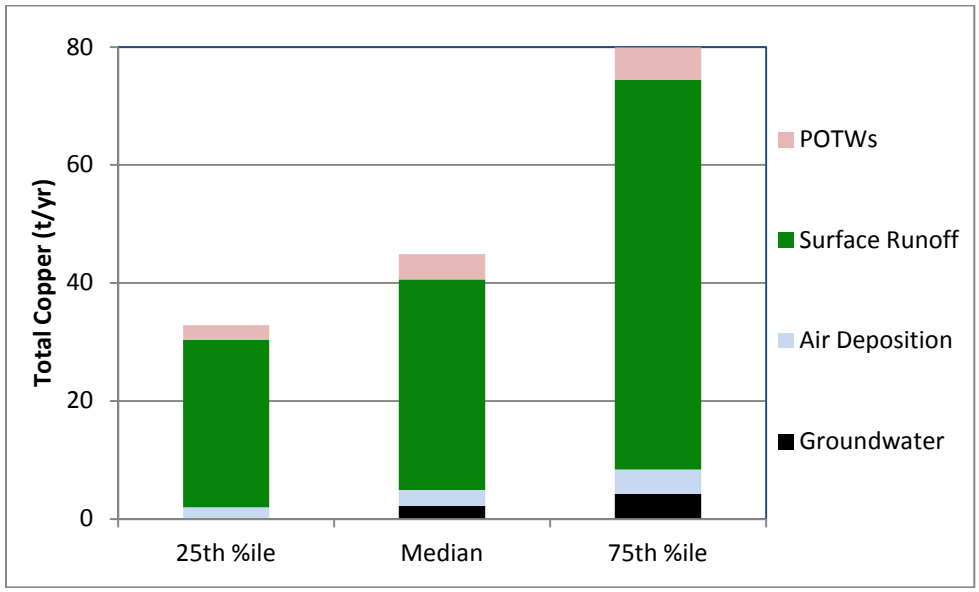


Figure 12. Total Copper Loads to Puget Sound as the Sum of Major Delivery Pathways

Relationship Between Sources and Pathways/Loading

The conceptual model of copper transport and fate following release suggests that much of the copper transported to Puget Sound will occur through surface runoff during storm events. Direct deposition to marine waters from atmospheric deposition and loading through direct groundwater discharges appear to be only a small portion (<15%) of overall loading. The high proportion of copper loading contributed by surface runoff is consistent with the types of sources accounting for the major copper releases.

Major unconstrained releases of copper are potentially from its use in urban landscaping and pesticides, leaching from rooftops, and from vehicle component wear. Together, these mostly urban sources account for as much as 140 t/yr of copper released to the environment in the Puget Sound basin.

It is difficult to draw conclusions regarding the fate of copper released from urban landscaping use and pesticide use. For instance, copper may be highly soluble depending on the formulation applied, and it may be released in dissolved form during storms or irrigation. However, since copper is likely to be used on recently disturbed soil in many instances, release through mobilization of particle-bound copper may also occur. This latter form of release is less likely to be infiltrated into groundwater, a seemingly distinct possible fate for copper solubilized in permeable soil. At the lower estimate of copper pesticide use in urban areas (1 t/yr), the comparatively low rate of use may be much more important locally than on a basin-wide scale.

Copper released from rooftops is likely to be in dissolved form since leaching is the likely release mechanism, but anticipating its pathway following initial release is difficult since it may run along roads and other impervious areas to surface waters, while some may be infiltrated into the soil or delivered to storm sewers and thereafter POTWs.

Brake pad and tire wear may result in a relatively high proportion of copper finding its way to surface runoff since approximately 50% is transported off-roadway as fugitive dust (Sinclair-Rosselot, 2006), although the fate of brake pad dust may vary greatly due to local conditions (Paulson et al., 2011-Draft). This and the possible attenuating factors discussed previously for copper initially released in water, mobilized by water, or released to an impervious surface such as a roadway likely account for much of the difference between amounts released from the unconstrained urban sources mentioned above (140 t/yr) and the amounts loaded to Puget Sound via surface runoff (28 – 66 t/yr).

The differences in copper releases from the unconstrained urban sources and the surface runoff on the regional scale are much smaller than the differences between releases and loads calculated during the mass-balance analysis of two small urban King County watersheds conducted by Paulson et al. (2011-Draft). They calculated that the annual copper mass discharged from these watersheds were less than 10% of the copper estimated to be released just from vehicle component (brake pad and tire) wear and roof runoff. At the much larger scale (entire Puget Sound basin), the high end (75^{th} percentile) estimate of copper discharged to Puget Sound through surface water is equal to the mass of copper released just from vehicle wear and rooftops (66 t/yr). This suggests that while the conceptual relationships between copper sources and loading/pathways may be valid, these relationships may be much more complex and uncertain at finer scales.

Copper deposited atmospherically is difficult to link with its primary source(s). Copper released from stack air emissions is small compared to overall loads (<1%), and copper reported as fugitive air releases in the TRI accounts for only about 2% of the total annual release, for a total maximum release to air of approximately 6 t/yr to the Puget Sound basin. This is higher than the range of copper deposited directly to marine water from the atmosphere (1.9 – 4.1 t/yr), but the latter only accounts for one-sixth of the basin area. However, copper released as fugitive dust

from brake pad wear is potentially substantial (>17 t/yr). This may account for the high levels of copper in atmospheric deposition samples at the high-density urban location, which is located near Interstates 5 and 705, associated interchanges, and other major roadways. Copper fluxes at this location were an order of magnitude higher than at most other locations, and the inventory of primary releases indicates that industrial sources may not be large contributors to airborne emissions. Copper fluxes at a location nearby but removed from close-proximity industrial and roadway influences were one-fifth those in the high-density urban area. This indicates that urban locations have strongly localized high copper flux signals, which are not reflected to the same degree in regional elevations of copper.

Disposal of copper at the Ft. Lewis Army Base (25 t/yr) presumably consists of solid copper material placed in waste piles or scattered throughout firing ranges. Solid copper disposal in the terrestrial environment will be retained in a soil reservoir and will only be transported to surface waters through gradual erosion or storm events with high energy flows.

Copper leached from vessel anti-fouling paint (26 t/yr) is presumed to be released entirely to marine waters with the possible exception of the Lake Washington/Lake Union system and freshwater marina areas near the mouths of the Snohomish and Duwamish Rivers.

Of the primary sources inventoried, copper released from residential plumbing components represents the only constrained source assessed. The annual load of copper discharged to POTWs from this source is estimated to be 28 t; discharges to septic systems were estimated to be an additional 11 t/yr (Ecology, 2011). The difference between the amount released and loading from POTWs (2.5 – 5.5 t/yr) is presumably due to the removal of solid material during the treatment process, an assumption that might be easily checked by conducting sampling of representative sludge material. Of course, copper from other constrained sources, naturally occurring copper in water, and copper in stormwater represent additional releases of copper to POTWs, but the exact extent of these contributions are not known.

Although patterns in copper loading pathways appear to be consistent with the types of primary sources, one confounding result is the high concentrations of copper in agricultural areas compared to other land covers. Overall copper releases as agricultural pesticides and micro-nutrients appear to be small (15.3 t/yr combined) compared to the other unconstrained releases previously mentioned, most of which are expected to occur in commercial/industrial and residential areas.

The high concentrations cannot simply be explained by excessive soil erosion in agricultural areas during storms, a scenario that would be expected to cause waters to become enriched with particle-bound copper. While copper in agricultural streams experienced a three-fold increase in concentration during storms, increases in TSS loads were lowest among all land covers (Herrera, 2011). One possible explanation for relatively high copper levels in agricultural streams may simply be that the form(s) of copper used and methods of application in agricultural settings are particularly prone to result in copper migration to surface waters.

Hazard Evaluation

The hazard evaluation indicates that copper is a Priority 1 level of concern for freshwaters, nearshore marine surface waters, and freshwater sediments (Table 6). Copper is a Priority 2 level of concern for offshore marine surface waters and marine sediments. Hazards due to tissue residue effects, effects to wildlife, and effects to human health were not evaluated.

Table 6. Summary of Hazard Evaluation for Copper.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.* *or* *Acute WQC* *or* *Chronic WQC*	**Level of Concern**
	Freshwater	5,378	92%	Yes	Priority 1
	Nearshore Marine	107	100%	Yes	Priority 1
	Offshore Marine	71	100%	No	Priority 2
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. >** *SQS*	**Level of Concern**
	Freshwater	826	>99%	Yes	Priority 1
	Nearshore Marine	519	>99%	No	Priority 2
	Offshore Marine	560	98%	No	Priority 2
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.*	**Level of Concern**
	Not Analyzed				
Wildlife		**Total N**	**FOD**	**Daily Dose >** *10% of Lowest Effects Dose*	**Level of Concern**
	Not Analyzed				
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. >** *NTR Criterion*	**Level of Concern**
	Not Analyzed				

Yellow highlight indicates Priority 1 Level of Concern

FOD=Frequency of detection

Dissolved copper concentrations in freshwater have a 90th percentile value of approximately 3.5 ug/l, higher than the level where more than 10% of effects documented for aquatic organisms occur and above the chronic water quality criterion (calculated at 25 mg/l calcium carbonate). At least 5% of the concentrations are above the acute water quality criterion (also calculated at 25 mg/l calcium carbonate).

In marine waters, nearshore concentrations of dissolved copper are distinctly higher than those observed offshore. At the 90th percentile level, the nearshore concentration (approx. 5 ug/l) is more than double the offshore concentration (approx. 2 ug/l) and exceeds both the chronic and acute water quality criteria. The upper levels (90th – 95th percentile values) of both the observed nearshore and offshore dissolved copper levels approach or exceed the 10th percentile values) of the ECOTOX dataset used for these comparisons, but only the observed nearshore data have 90th percentile values that exceed this threshold for assigning a Priority 1 level of concern.

Median copper concentrations in both freshwater and marine sediments are below all guidelines and standards except the marine Canadian TEL. However, in freshwater sediments the 90th percentile (approx. 90 mg/kg dw) of observed copper concentrations exceed the floating percentile SQS as well as the Canadian TEL and the consensus-based TEC. More than 5% of the observed freshwater sediment values exceed the Canadian PEL and the consensus-based PEC for copper.

In marine sediments, the 90th percentiles of the observed copper concentrations (approx. 90 mg/kg dw for nearshore and 70 mg/kg dw for offshore) are similar to those for freshwater sediments, but the marine SQS is much higher than the freshwater floating percentile SQS. As a result, all but possible outlier concentrations exceed the SQS in marine sediments.

Regionally Important Biological-Effects data

Copper is one of the most far-reaching potential priority toxicants in the Puget Sound region due largely to its ability to alter the sensory capacity and behavior of a wide variety of aquatic organisms. A number of local researchers have documented these effects in different organisms and in different ways.

Tierney et al. (2010) reviewed over 150 papers and found that avoidance behaviors were common in a variety of fresh and salt water fishes at less than 1 ug/l to concentrations ranging up to 20-30 ug/l. Tested species included coho and Chinook salmon as well as rainbow trout and golden shiner. Hecht et al. (2007) compiled a similar body of evidence for the disruptive effects of copper on juvenile salmonids. They used EPA methodologies to calculate benchmark concentrations predicted to represent 10% and 50% reductions in chemosensory response at 0.18 ug/l and 2.1 ug/l respectively. These values bracket a variety of other regional primary literature sources which confirm that the environmentally relevant range of <1.0 to 5.0 ug/l copper adversely impacts a variety of Puget Sound basin fish, particularly salmonids. Similar neurologic impacts were found by Linbo et al. (2006) on the mechanosensory lateral line of fish.

Sandahl et al. (2004) found copper concentrations of 4.4 ug/l produced sublethal neurotoxicity in coho salmon. In this laboratory study, copper reduced the ability of coho salmon to detect the natural odorants taurocholic acid and L-serine. Further study by Sandahl et al. (2007) confirmed that concentrations as low as 2 ug/l copper not only affect the neurologic systems of fish but also alter their behavioral responses to alarm pheromones. Other studies such as Baldwin et al. (2003) have also found olfactory inhibition at a comparable environmentally relevant concentration of 2.3 ug/l.

Hansen et al. (1999) produced a seminal work which was used by several of the subsequent reviews discussed above. In it they documented Chinook salmon avoidance behaviors at concentrations as low as 0.7 ug/l dissolved copper. However, Chinook also failed to avoid concentrations >44 ug/l due to the extensive neural saturation. This window of effect potentially contributes to mortality from prolonged copper exposure or impairment of olfactory dependent behaviors such as homing. Additional studies by McIntyre et al. (2008) found that water hardness had very little effect on copper's ability to alter olfactory function in coho salmon despite water hardness being a variable influencing the Washington State water quality criteria. All of these reviews and studies on regionally relevant species provide an additional line of evidence suggesting that copper is a very important toxicant at concentrations well within the range found it the Puget Sound regional environment.

Lead

Lead is a naturally occurring heavy metal that is a major constituent of more than 200 minerals and is nearly always detectable at concentrations > 0.01 ug/l in water and >100 ug/kg (dw) in sediments from freshwater and marine environments (PTI, 1991; Serdar, 2008; Meredith and Furl, 2009). Lead also accumulates in fish, particularly in bony material, but does not biomagnify to any meaningful extent (Eisler, 1988b).

Although lead occurs naturally in the earth's crust, human activity has increased environmental levels by one-thousand-fold during the past three centuries (ATSDR, 2007). Most recently, the use of tetra-ethyl lead in gasoline ("leaded gasoline") accounted for hundreds of millions of pounds of lead per year in the U.S. alone before being phased out from 1973 – 1996; by 1995, lead emissions from gasoline were estimated to be less than 1% of the level prior to initiation of the phase out (EPA, 1996).

Historically, concerns surrounding the effects of lead have been focused on toxicity to humans, particularly children. Ecology and WDOH (2009) have reviewed the history, exposure pathways, and effects of lead on humans in Washington. Environmental effects, particularly in the aquatic environment, are less apparent.

Major Releases from Primary Sources

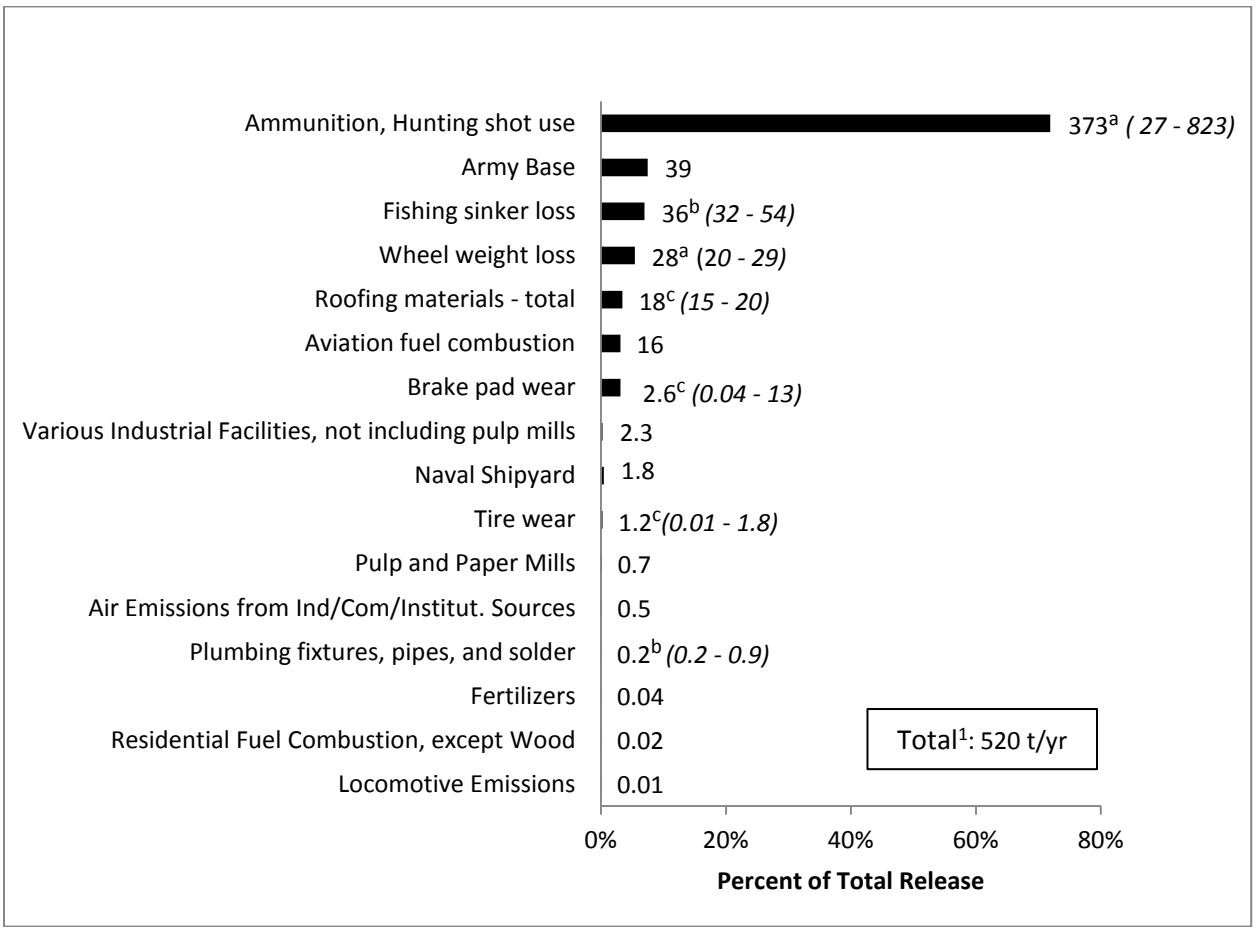
An inventory of primary lead sources suggests that approximately 520 metric tons (t) is released annually from anthropogenic sources in the Puget Sound basin. The largest current source of lead to the environment is the use of ammunition and lead shot (Figure 13). Together with lost wheel weights, fishing sinkers, and a self-reported release at Fort Lewis presumed to be from ammunition use, approximately 90% (480 t) of the anthropogenic releases of lead in the basin may be as solid metallic lead in bulk form.

Other sources of lead include leaching from materials such as roof runoff (18 t/yr) and abrasion of vehicle brake pads and tires (4 t/yr combined). Although these releases are small compared to ammunition use, they represent the most likely sources of lead to be mobilized in stormwater following release. To be more precise, lead in roof runoff requires rain storms for its release, and approximately 50% of brake pad particles are released to the road surface (Garg et al., 2000;

Sinclair- Rosselot, 2006) and may be subsequently entrained in stormwater (depending on season and conditions). Tire particles are presumably released mainly to road surfaces, although some portion becomes transported away from the release point as fugitive dust.

Additional releases of lead include emissions from combustion of aviation fuel (16 t/yr) and approximately 0.5 t/yr released to the air from point sources; pulp mill emissions appear to make up the bulk of this latter category.

Other smaller releases include approximately 1 t/yr released from residential plumbing components, and 0.040 t/yr (40 kg/yr) released through fertilizer application.



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

[a] Most reasonable estimate

[b] Median

[c] Mean

Figure 13. Total Lead Release in the Puget Sound Basin (values shown are t/yr).

Major Delivery Pathways and Loading

Data on major delivery pathways for lead loading were obtained from PSTLA loading studies and are included in Table 7. As mentioned previously, loading through direct groundwater discharge was estimated from literature values, whereas loadings through other pathways were estimated from field studies specifically designed to estimate loads.

Table 7. Total Lead Loads (t/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater (a)	0.044	1.07	2.10
Air Deposition	0.63	1.1	1.5
Surface Runoff	2.80	4.67	7.64
POTWs	0.14	0.18	0.25
Ocean Exchange	21	21	18

(a) Lowest, highest, and mid-point of estimated loads

Estimated groundwater loads of lead range by two orders of magnitude (0.044 – 2.1 t/yr). The range in estimates is due to differences in flows used to establish the possible range of loads as well as differences in methods used to estimate representative lead concentrations; most of the data were non-detected values.

Lead discharged from POTWs in the Puget Sound basin accounts for loads estimated to be 0.14 – 0.25 t/yr. Phase 1 estimates suggest that lead loads from industrial wastewater are potentially substantial (0.3 – 9 t/yr; Hart Crowser et al., 2007). This is based on a limited dataset yet brackets the estimate of the industrial discharge of lead to surface waters or transferred to POTWs as reported in the TRI (1.4 t/yr total) (Ecology, 2011).

The deposition of atmospheric lead directly to the marine waters of Puget Sound is approximately 0.63 – 1.5 t/yr, based on fluxes ranging from 0.15 - 8.6 ug/m^2/d. Lead fluxes were generally <1 ug/m^2/d, except for one location within a high-density urban area (Tacoma) including a nearby pulp mill, metal refiners, other industrial activities, and close to major roadways including interstate highways. Lead fluxes at this location were consistently an order of magnitude higher than other locations around the Puget Sound region, and were elevated five-fold above a nearby station that did not have the same air pollution influences in such close proximity

Surface runoff loads for lead are estimated to be approximately 2.8 - 7.6 t/yr for the entire Puget Sound basin based on the surface runoff study. Dissolved lead accounted for only a small portion of the total lead concentrations, and elevated concentrations of total lead appeared to correspond with elevated TSS (Herrera, 2011). Lead has a high affinity for particulate matter and is generally found at low proportions in the dissolved phase (Meredith and Furl, 2009; Hallock, 2010). Lead is therefore much more likely to be transported as particle-bound lead rather than in the dissolved phase.

The ratio of lead concentrations in storm flows-to-baseflows was consistently higher for lead than for other COCs analyzed. Lead in storm flows was higher compared to baseflows by factors of 3 to 6 depending on land cover (commercial/industrial was highest). This is consistent with particle-bound constituents which require storm flows to become mobilized.

Total lead was present at the highest concentrations in commercial/industrial land covers, generally by factors of 2 to 5. Overall median total lead concentrations in commercial/industrial areas increased by six-fold during storm events, while only increasing by factors of 2 to 3 in other land cover types. Although elevations in lead during storm flows were most pronounced in commercial/industrial areas, seasonal first-flush signals were only evident in residential (dissolved lead) and forest sub-basins (total lead).

Like cadmium, there appears to be a large import of lead through exchange at the ocean boundary compared to other load pathways, although this estimate is based on very limited data. Total lead concentrations (25^{th}-75^{th} percentile) in the incoming marine waters are 0.087 – 0.125 ug/l, and total lead concentrations in the outgoing marine waters are 0.047 – 0.087 ug/l. Total net import was 18 – 21 t/yr based on an inflow of 51 – 73 t/yr and an outflow of 30 – 55 t/yr.

The net sum of lead loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25^{th}, median (50^{th}), and 75^{th} percentile values is 25, 28, and 30 t/yr, respectively. Under all of these estimates, there is a net lead load to Puget Sound.

Lead loadings from pathways that may represent *partially controllable* sources of COCs (i.e. all of the major pathways except ocean exchange) are shown as ranges (25^{th} - 75th percentiles) and median for each pathway in Figure 14. Each pathway represented as a contribution to the total load is displayed in Figure 15.

Total lead loading from the major pathways assessed is 3.6 – 12 t/yr. Surface water runoff accounts for the largest pathway (66 – 77%), followed by air deposition (13 – 17%). Ground-water potentially accounts for up to 18% at the upper end of the estimated range, but this value should be viewed with caution since it is based on literature values of lead in wells and rough estimates of groundwater flow. At the lower end of the load range, lead in groundwater is estimated to account for 1% of the load to Puget Sound. Loading through POTWs represents only a small portion of total lead loads to Puget Sound (2 – 4%).

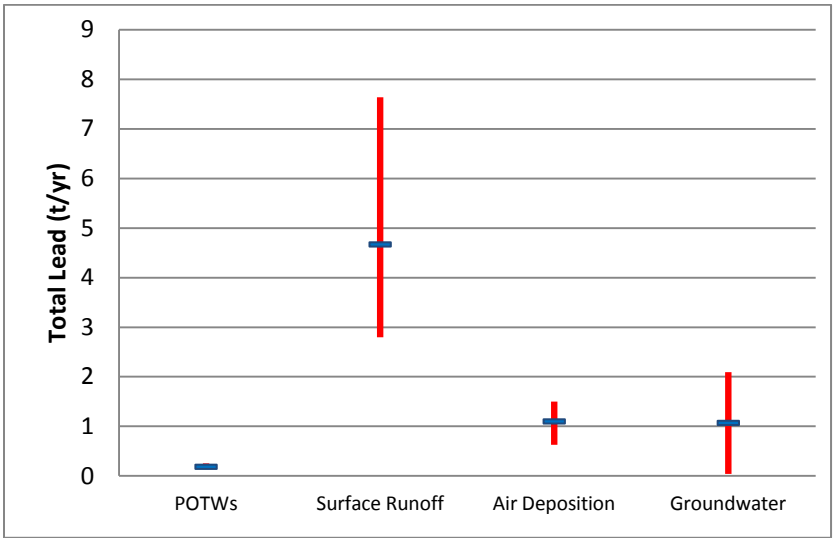


Figure 14. Range (25th - 75th percentiles) and Median Total Lead Loads to Puget Sound from Each Major Delivery Pathway.

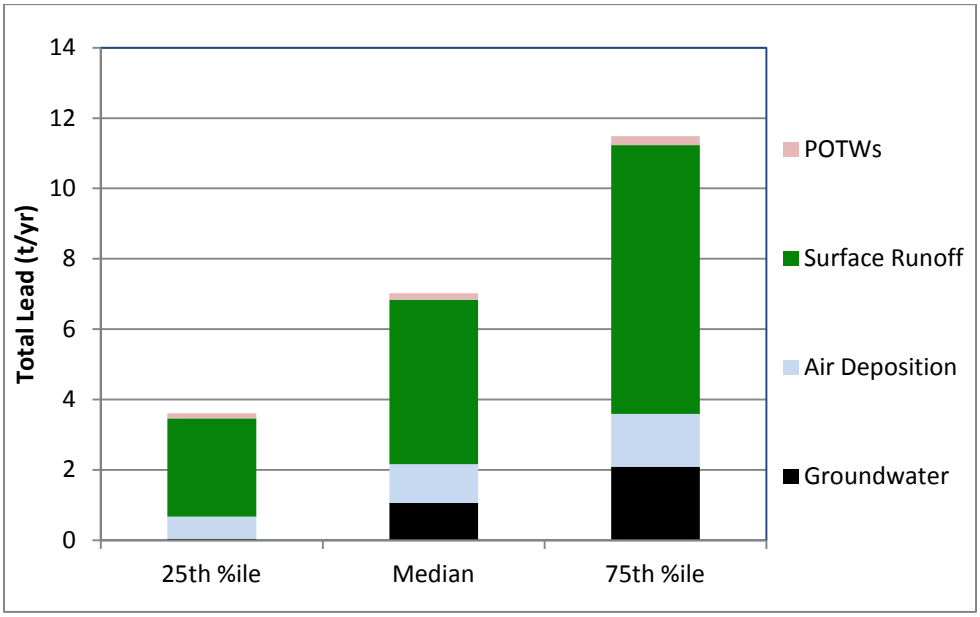


Figure 15. Total Lead Loads to Puget Sound as the Sum of Major Delivery Pathways.

Relationship Between Sources and Pathways/Loading

The conceptual model of lead transport and fate following release suggests that much of the lead transported to Puget Sound will do so through surface runoff during storm events. Lead released to road surfaces through roof runoff, lost wheel weights, and tire and brake pad wear appears to represent the largest release to road surfaces. Combined, these account for an estimated 51 t/yr of lead released, or about 7-18 times the estimated lead load in surface runoff. These differences can be explained largely by the source for lead released to road surfaces. For instance, not all

rooftop runoff will be delivered to roadways; some may be infiltrated or delivered to storm sewers and thereafter to POTWs. Lost wheel weights will largely remain in bulk metallic form and only gradually become pulverized and leached. Brake pad and tire wear may represent the lead source with the highest proportion of lead to find its way to surface runoff, even though a substantial fraction may be transported off-roadway as fugitive dust.

Lost fishing sinkers and lead shot and other ammunition landing in surface waters will gradual leach lead to the aquatic environment, although this likely results in only small increases in water column concentrations. Lead ammunition landing in the terrestrial environment will likely be retained in a soil reservoir and will only be transported to surface waters through gradual erosion or storm events with high energy flows. In general, lead from ammunition will be retained in the soil and not pose a problem to surface waters, with the exception of shooting ranges which may accumulate large enough masses of lead to impact nearby waterbodies. Two instances of high lead concentrations in streams and soils resulting from spent lead at shooting ranges have been documented recently in western Washington (Era-Miller, 2009; Ecology and WDOH, 2009).

The ultimate fate of lead emitted to air is less certain; only a small fraction of the estimated releases to air are deposited directly to the waters of Puget Sound, and it is not known what portion of the air releases in the basin are transported out of the airshed and the portion of deposited lead that is imported from outside airsheds. Approximately 19 t/yr are released to the air within the Puget Sound basin (not counting fugitive brake pad and tire dust) compared to 0.6 – 1.5 t/yr directly deposited to marine waters. Considering the area of the entire Puget Sound basin (six times the marine area) and assuming the same rate of deposition across the basin (an assumption that has not been verified), the ranges of air deposition fall within a factor of 2 compared with lead emissions to air. Atmospherically deposited lead would presumably be washed off surfaces in runoff – most likely in particulate form – and ultimately be transported to the Puget Sound through surface runoff, although infiltration to groundwater and paths to POTWs are other possible scenarios for secondary and tertiary pathways.

Although comparatively small amounts of lead are released in constrained sources, release of lead from residential plumbing fixtures (0.2 t/yr) almost certainly is received at POTWs. Annual loads of lead released to POTWs from this source appear to be nearly identical to the amount discharged from POTWs. Presumably, removal of lead would occur during the removal of solid material during the treatment process. Lead from other constrained sources, naturally occurring lead in water, and lead in stormwater represent additional releases of lead to POTWs, but the extent of these contributions are not known.

Hazard Evaluation

The hazard evaluation indicates that lead is a Priority 2 or unknown level of concern for all categories assessed (Table 8). Hazards due to tissue residue effects, effects to wildlife, and effects to human health were not evaluated.

The 90th percentile values for dissolved lead in freshwater (approx. 0.5 ug/l) are an order of magnitude below the 10th percentile of effects data and the acute water quality criterion, although the values are only slightly less than the chronic water quality criterion. In marine waters, the

gap between observed concentrations and those where criteria are set and where effects occur is similar to that for freshwater. However, the dataset for observed nearshore marine water was too small (n<50) to assign a level of concern.

Table 8. Summary of Hazard Evaluation for Lead.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.* or *Acute WQC* or *Chronic WQC***	**Level of Concern**
	Freshwater	4,427	33%	No	Priority 2
	Nearshore Marine	44	68%	INS	U
	Offshore Marine	77	88%	No	Priority 2
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. > *SQS***	**Level of Concern**
	Freshwater	838	96%	No	Priority 2
	Nearshore Marine	472	95%	No	Priority 2
	Offshore Marine	478	99%	No	Priority 2
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.***	**Level of Concern**
	Not Analyzed				
Wildlife		**Total N**	**FOD**	**Daily Dose > *10% of Lowest Effects Dose***	**Level of Concern**
	Not Analyzed				
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. > *NTR Criterion***	**Level of Concern**
	Not Analyzed				

FOD=Frequency of detection
INS=Insufficient observed data available for comparison to effects data
U=Unknown level of concern due to lack of sufficient data for adequate comparison

Unlike most other metals, lead appears to have a fairly distinct pattern of higher concentrations in offshore marine waters compared with concentrations observed nearshore. This may reflect lead enrichment of marine waters through ocean exchange. For cadmium, the only other metal assessed which is comparatively high in oceanic waters, the higher concentrations in offshore water may also be a reflection of this ocean enrichment process.

At least 10% of the observed lead concentrations in freshwater sediments exceed the Canadian TEL and PEL as well as the consensus-based TEC and PEC. However, the 90th percentile value (approx. 200 mg/kg dw) did not exceed the floating percentile SQS, making lead one of the few COCs that did not exceed this threshold.

Lead concentrations in marine nearshore and offshore sediment have 90th percentile values of approximately 100 mg/kg dw and 70 mg/kg dw, respectively. These concentrations are not above the SQS and exceed only the consensus-based TEC among the guidelines and standards used for comparison.

As mentioned previously, lead hazards to fish, wildlife, and human health were not assessed due to the lack of effects data. However, although lead shot has been prohibited in Washington for all waterfowl, coot, and snipe hunting since a nationwide phase-in of non-toxic shot was implemented during 1986-1991, lead poisoning of birds due to ingestion of lead shot remains a concern (Ecology and WDOH, 2009).

Mercury

Mercury is a naturally occurring heavy metal in the earth's crust and is nearly always detectable at concentrations >0.005 ug/l in water and >5 ug/kg (dw) in sediments from freshwater and marine environments (PTI, 1991; Serdar, 2008; Hallock, 2010; Appendix D). Unlike other metals, mercury is liquid at typical ambient temperatures, is volatile, and bioaccumulates to a high degree in edible tissues of fish (i.e. fillet), particularly those occupying high trophic positions.

Mercury naturally degasses from soils, the rate of which may be increased dramatically by disturbances such as logging and land development, and inundation caused by dam construction. Historically, a large source of mercury to Puget Sound was a now-defunct chloralkali plant in Bellingham (PTI, 1991), although mercury is released through numerous industrial and combustion sources as well.

Major Releases from Primary Sources

An inventory of primary mercury sources suggests that approximately 0.54 metric tons (t) is released annually from anthropogenic sources in the Puget Sound basin. The largest current source of anthropogenic mercury to the environment may be due to thermostat and fluorescent lamp disposals (24% and 18%, respectively) (Figure 16).

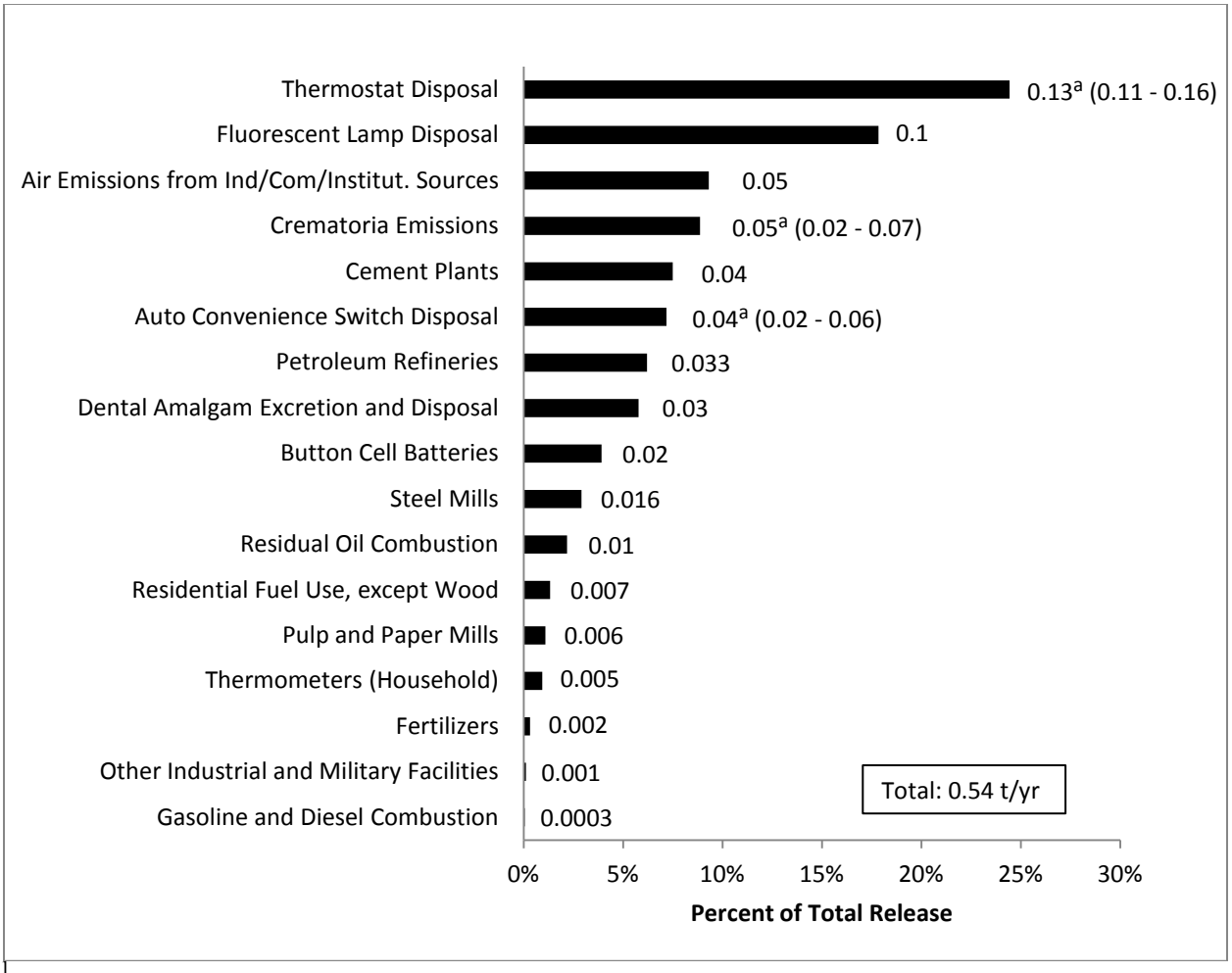
Mercury releases from improper disposal of other materials may account for an additional 11% of the total release in the Puget Sound basin. These products contain mercury as liquid, or as in the case of fluorescent lamps, in vapor form. Since mercury has the potential to volatilize, there is likely some portion of liquid mercury that is released from landfills due to disposal of these products, even though collection and recycling efforts are underway to prevent these circumstances. Nearly all of the anthropogenic mercury sources assessed in the *Sources Report* have been addressed in the Mercury Chemical Action Plan (Ecology and WDOH, 2003), and efforts are underway to reduce or eliminate mercury releases from these sources.

Air emissions from major industrial facilities reporting under TRI represent approximately 18% of the total mercury release, and four-fifths of these are stack air emissions. As much as of one-quarter of the mercury release to air in the Puget Sound basin may be through combustions

emissions. Much of this may be due to fossil fuel combustion, although mercury emissions from crematoria and cement plants may originate from the source material rather than the fuel.

Mercury is also released from the TransAlta Centralia Generating Plant at an annual rate of approximately 0.15 t (Ecology, 2011). Although the TransAlta plant was not included in the source inventory since its location falls outside of the Puget Sound basin, it is upwind of the basin and at least two nearby lakes in the Puget Sound basin have mercury levels in sediment that appear to reflect deposition from the plant (Furl and Meredith, 2010).

Other mercury releases which do not fall under air emission or landfill disposal categories include disposal and excretion of dental amalgam (0.03 t/yr, 6% of total) and mercury contained in fertilizer, particularly nitrogen and potassium material (0.002 t/yr, <1% of total).



[1] Sum of best estimates. Best estimates are either the mean, mid-point, median, or most reasonable estimate for each source.

[a] Mid-point of range

Figure 16. Total Mercury Release in the Puget Sound Basin (values shown are t/yr).

Major Delivery Pathways and Loading

Data on major delivery pathways for mercury loading were obtained from PSTLA loading studies and are included in Table 9. As mentioned previously, loading through direct groundwater discharge was estimated from literature values, whereas loadings through other pathways were estimated from field studies specifically designed to estimate loads. Mercury was not measured in marine waters to assess exchange at the ocean boundary.

Table 9. Total Mercury Loads (t/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater (a)	0.0047	0.049	0.094
Air Deposition	0.0094	0.02	0.033
Surface Runoff	0.091	0.136	0.238
POTWs (b)	NR	0.0024	NR
Ocean Exchange	NA	NA	NA

(a) Lowest, highest, and mid-point of estimated loads
(b) Estimate from Phase 2 (Envirovision et al., 2008b)
NR=not reported
NA=not analyzed

The estimated groundwater mercury load ranges by an order of magnitude (0.005 – 0.09 t/yr). The range in estimates is due primarily to the range in flows used to calculate loads; groundwater discharges used in the groundwater loading study ranged by an order of magnitude.

Mercury was not measured in POTWs for the Phase 3 loading study. Phase 2 estimates for mercury loading through wastewater calculated an estimated release of 0.002 t/yr from municipal wastewater treatment plants and an additional 0.007 t/yr from industrial discharges, the latter of which is considered an underestimate due to incomplete sampling (Envirovision et al., 2008b).

The deposition of atmospheric mercury directly to the marine waters of Puget Sound is approximately 0.009 – 003 t/yr, based on fluxes ranging from 0.003 – 0.020 ug/m^2/d. Median mercury fluxes were generally ≤0.010 ug/m^2/d, except for one location within a high-density urban (Tacoma) area including a nearby pulp mill, metal refiners, other industrial activities, and close to major roadways including interstate highways. Fluxes at this location were two- to three-fold higher than at other locations.

Surface water runoff loads for mercury are estimated to be approximately 0.09 – 0.24 t/yr for the Puget Sound basin based on the surface runoff study. Elevations in total mercury concentrations appeared to correspond with elevated TSS in all areas except agriculture land covers, although dissolved mercury accounted for one-half to two-thirds of the total mercury concentration.

During storm events, mercury concentrations in surface water increased by factors of 2 to 3 over baseflow concentrations. Agricultural areas have the highest mercury concentrations in surface water during both baseflow and storm flow conditions, followed by residential areas, commercial/industrial sub-basins, and forests. A substantial seasonal first-flush episode was evident for total mercury concentrations in forests but was not seen in other land covers.

The net sum of mercury loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25th, median (50th), and 75th percentile values is 0.1, 0.2, and 0.3 t/yr, respectively. Under all of these estimates, there is a net mercury load to Puget Sound, although the possibility of a net export out of Puget Sound could not be explored since ocean boundary water was not sampled during the loading studies.

Mercury loading from pathways that may represent *partially controllable* sources of COCs (i.e. all of the major pathways except ocean exchange) are shown as ranges (25th - 75th percentiles) and median for each pathway in Figure 17. Each pathway represented as a contribution to the total load is displayed in Figure 18.

Total mercury loading from the major pathways assessed is 0.11 – 0.36 t/yr. Surface runoff accounts for the largest pathway (65 – 87%), followed by air deposition (9%). Groundwater potentially accounts for up to 26% at the upper end of the estimated range, but this value should be viewed with caution since it is based on literature values of mercury in wells and rough estimates of groundwater flow. At the lower end of the load range, mercury in groundwater is estimated to account for 4% of the load to Puget Sound. The Phase 2 estimate of loading through POTWs – data obtained from the literature – is roughly 2% of the total mercury load to Puget Sound.

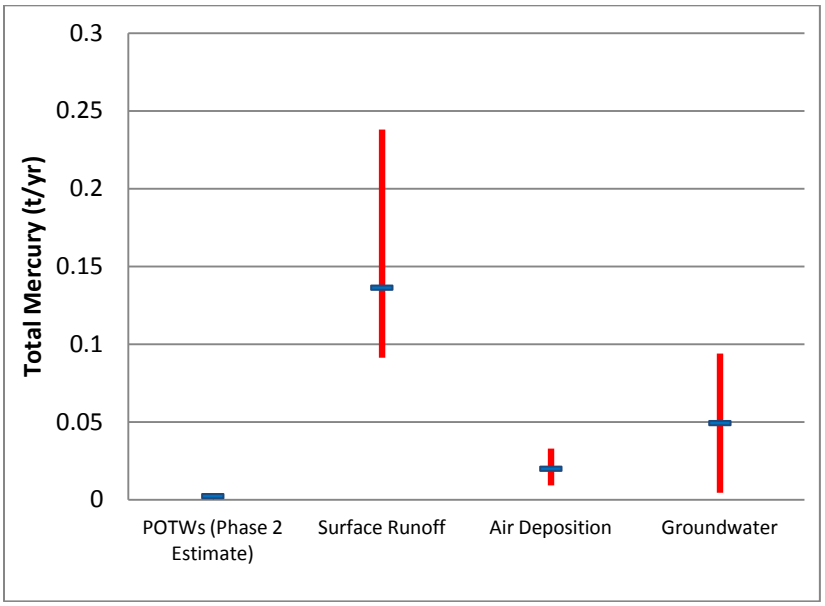


Figure 17. Range (25th - 75th percentiles) and Median Total Mercury Loads to Puget Sound from Each Major Delivery Pathway.

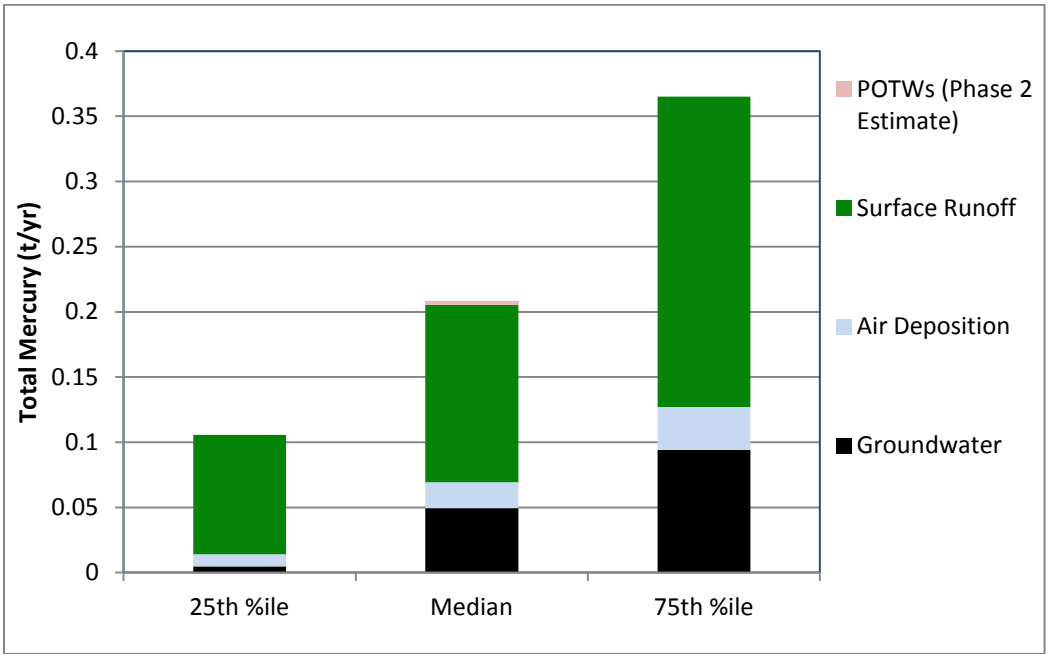


Figure 18. Total Mercury Loads to Puget Sound as the Sum of Major Delivery Pathways.

Relationship Between Sources and Pathways/Loading

The conceptual model of mercury transport and fate following release suggests that much of the mercury transported to Puget Sound will occur through surface runoff, mostly during storm events, even as a high proportion of initial releases are emissions to air. Since most of the anthropogenic mercury releases occur as air emissions, the surface water pathway may be assumed to represent a secondary or tertiary pathway. Other possible explanations for the differences between the high proportion of air emission as a mercury source and the high proportion of surface runoff as a delivery pathway are natural sources of mercury or continuous mobilization of historic releases of mercury remaining in aquatic systems.

Total inventoried anthropogenic mercury releases to air in the Puget Sound basin are approximately 0.5 t/yr; closer to 0.7 t/yr if mercury released from the TransAlta plant is included. Since releases to air are at least double the loads from all major pathways combined, it seems reasonable that some of the mercury atmospherically deposited on land is entrained in surface runoff and transported to Puget Sound. Applying the known median atmospheric mercury deposition rates to marine waters to the entire Puget Sound watershed would result in an additional 0.1 t/yr of deposition. If all of the mercury assumed to be deposited to land surfaces under this scenario were mobilized in surface runoff, it would closely match the median load delivered to Puget Sound through the surface runoff pathway.

The air deposition study found that, unlike other trace elements measured, mercury did not vary significantly among stations, and much of the loading occurred as a result of washout during rain events. These findings appear to support widespread "dosing" of the watershed with aerially-transported mercury. However, the authors of the study (Brandenberger et al., 2010) note that

while mercury deposition in the Puget Sound appears to be a result of regional transport, the bulk deposition methods used to estimate fluxes may underestimate the dry deposition of mercury. As a result, strong localized signals from industrial areas may be diluted out.

A consistent level of aerial mercury "dosing" across a watershed would likely result in similar mercury concentrations among land uses, particularly since much of the load is due to washout. The surface runoff results do not reflect a widespread dosing and washout scenario since forested areas have lower mercury concentrations than commercial/industrial and residential areas where combustion sources are likely to be located. However, higher mercury loads from commercial/ industrial and residential areas may simply be a result of enhanced mobilization from surfaces (due to the comparatively high proportion of impervious surfaces) rather than localized mercury sources.

The comparatively high concentration of mercury in agricultural area runoff samples remains a puzzle. Loss of mercury-enriched soil during storms may be one possible explanation, although baseflow mercury concentrations in agricultural areas are also higher than in other land covers, and TSS increases during storm flows are smaller in agricultural areas compared to other land types. It appears unlikely that major mercury sources would typically be located in close proximity to agricultural lands, and the one inventoried mercury release that is specific to agricultural practices is comparatively small (releases from fertilizer application, 0.002 t/yr).

The single inventoried source of mercury released in a constrained pathway is loss of dental amalgam through disposal (approximately 95% of the mercury used in dental offices is currently recovered) and excretion. Approximately 0.03 t/yr of mercury release in the Puget Sound basin is attributed to this source. This is approximately tenfold the estimated load from POTWs during the Phase 2 Study (0.002 t/yr) (Envirovision et al., 2008b). The differences are presumably due to the removal of solid material during the treatment process. Mercury from other constrained sources, naturally occurring mercury in water, and mercury in stormwater represent additional releases of mercury to POTWs, but the extent of these contributions are not known.

Hazard Evaluation

The hazard evaluation indicates that mercury is a Priority 1 level of concern for a range of media and receptors in both the freshwater and marine aquatic environments (Table 10).

Most (>95%) of the observed mercury concentrations in surface waters are one to two orders of magnitude below concentrations where most (95%) of the effects have been documented. The 90^{th} percentile of observed total mercury concentrations are slightly above the chronic water quality criterion (0.012 ug/l), but observed dissolved concentrations are well below the acute criterion (2.1 ug/l). Interestingly, the chronic water quality criterion is based on a value designed to avoid exceedance of the Food and Drug Administration Action Level for mercury in seafood (1.0 mg/kg; EPA, 1985).

Table 10. Summary of Hazard Evaluation for Mercury.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.* or *Acute WQC* or *Chronic WQC***	**Level of Concern**
	Freshwater	4,313	63%	Yes	Priority 1
	Nearshore Marine	7	100%	INS[a]	U
	Offshore Marine	14	93%	INS[a]	U
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. > *SQS***	**Level of Concern**
	Freshwater	803	66%	Yes	Priority 1
	Nearshore Marine	459	70%	Yes	Priority 1
	Offshore Marine	367	79%	Yes	Priority 1
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.***	**Level of Concern**
	Freshwater	b	100%	INS[a]	U
	Nearshore Marine	c	≥95%	INS[d]	U
	Offshore Marine	e	100%	INS[d]	U
Wildlife		**Total N**	**FOD**	**Daily Dose > *10% of Lowest Effects Dose***	**Level of Concern**
	Great Blue Heron (FW)	--	--	Yes	Priority 1
	Osprey (SW)	--	--	Yes	Priority 1
	River Otter (FW)	--	--	Yes	Priority 1
	Harbor Seal (SW)	--	--	Yes	Priority 1
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. > *NTR Criterion***	**Level of Concern**
	Freshwater	f	>99%	No	Priority 2
	Nearshore Marine	g	>92%	No	Priority 2
	Offshore Marine	h	100%	No	Priority 2

Yellow highlight indicates Priority 1 Level of Concern

FOD=Frequency of detection

INS[a] =Insufficient observed data available for comparison to effects data

U=Unknown level of concern due to lack of sufficient data for adequate comparison

[b] N range is 11 – 16 and varies for organism type and tissue type

[c] N range is 42 – 169 and varies for organism type and tissue type

INS[d] =Insufficient effects data available for comparison to observed data

[e] N range is 5 – 190 and varies for organism type and tissue type

FW=Freshwater

SW=Saltwater

[f] N range is 34 – 776 and varies for organism type and tissue type

[g] N range is 107 – 197 and varies for organism type and tissue type

[h] N range is 37 – 346 and varies for organism type and tissue type

There are few observed data to compare mercury concentrations to effects data or criteria in marine waters. For instance, only seven usable values were found for nearshore marine waters and only 14 values for offshore waters. Ninety percent of the effects data were at least two orders of magnitude above the 90th percentile values for both the nearshore and offshore datasets. Although more than 25% of the offshore values exceed the chronic water quality criterion, the paucity of observed values precluded an adequate evaluation for any of these comparisons.

Mercury concentrations in freshwater sediments exceed the floating percentile at the 90th percentile concentration (approx. 0.6 mg/kg dw) of the observed dataset. Marine sediments also have observed mercury concentrations that exceed the SQS at 90th percentile levels for both the nearshore and offshore datasets (both approx. 0.5 mg/kg dw); these concentrations are also at or near the LAET.

Observed mercury concentrations in fish tissues are nearly identical to effects levels, but there were insufficient observed data for an adequate evaluation. Fewer effects values are available for marine waters. No marine effects concentrations were available for non-decapod invertebrates and fish, and only one marine decapod effect concentration was available. There are observed concentrations for all tissue types although only five for offshore decapods.

Evaluation of daily mercury doses based on fish and incidental sediment ingestion for the four species evaluated – great blue heron, osprey, river otter, and harbor seal – indicate that all species would be exposed to doses equal to or greater than the lowest effects dose. For the bird species, the lowest effects doses are based on reproductive effects, although heron also exceed a dose for growth effects. For the mammal species evaluated, both seal and otter exceed the lowest dose calculated for growth effects, while the otter also exceeds the lowest dose for mortality.

Edible tissues evaluated for comparison to the NTR criterion (based on the default consumption rate of 6.5 g/d) reveal that fish tissue generally has higher observed mercury concentrations than bivalves or other invertebrates, but for all organisms more than 95% of the observed values fell below the criterion (approx. 800 ug/kg wet weight).

Comparisons were also made using exposure assumptions outlined in the NTR but with varying daily consumption rates. More than one-half of the freshwater and marine fish tissue samples exceed the acceptable risk level using the EPA recommended subsistence rate of 142.4 g/d. In the nearshore marine areas, more than one-half of the mercury concentrations found in invertebrates other than bivalves exceed the acceptable risk at the 142.4 g/d rate, and more than one-half of the bivalves exceed the acceptable risk based on the Suquamish tribal rate (769 g/d). In the offshore marine areas, more than one-half of the mercury concentrations found in invertebrates other than bivalves exceed the acceptable risk at the 142.4 g/d rate, and more than one-half of the bivalves exceed the acceptable risk based on the Tulalip tribal/King Co. American Petroleum Institute (API) rate (242.5 g/d).

Regionally Important Biological-Effects Data

Grove and Henny (2008) analyzed mercury in river otter carcass livers obtained from trappers working in western Oregon and western Washington, including Puget Sound. Mercury liver concentrations were higher in Puget Sound adult river otters (mean of 7.89 mg/kg dw) than those from the northwest Washington area (mean of 5.85 mg/kg dw). Concentrations in Willamette River and coastal Oregon otter livers were slightly higher (mean of 9.2-9.3 mg/kg dw) but similar to Puget Sound levels. This study demonstrates that river otters living in the Puget Sound area bioaccumulate mercury.

In addition to the bioaccumulation of mercury by otters, accumulation in fish tissue has led to advisories for human consumption of fish in Puget Sound. The advisories, issued by Washington State Department of Health (WDOH), are based on data and consumption of particular species, and vary by region (Hardy and Palcisko, 2006). For instance, WDOH advises no consumption of yelloweye rockfish (*Sebastes ruberrimus*) caught anywhere in Puget Sound, and advises limited consumption of English sole (*Parophrys vetulus*) and other flatfish based on the marine area in which they are caught. In addition, there is a statewide mercury advisory for smallmouth bass (*Micropterus dolomieui*), largemouth bass (*M. salmoides*), and northern pikeminnow (*Ptychocheilus oregonensis*) with specific advice based on risks to population segments (e.g. children, nursing mothers) (McBride, 2003; WDOH, 2011).

Zinc

Zinc is a naturally occurring heavy metal abundant in the earth's crust and is nearly always detectable at concentrations >0.5 ug/l in water and >10,000 ug/kg (dw) in sediments from freshwater and marine environments (PTI, 1991; Serdar, 2008; Hallock, 2010; Appendix D). It occurs at comparatively high concentrations in natural waters, but zinc's wide use as a protective coating and alloy with other metals to reduce corrosion in outdoor environments may increase levels in the aquatic environment.

Major Releases from Primary Sources

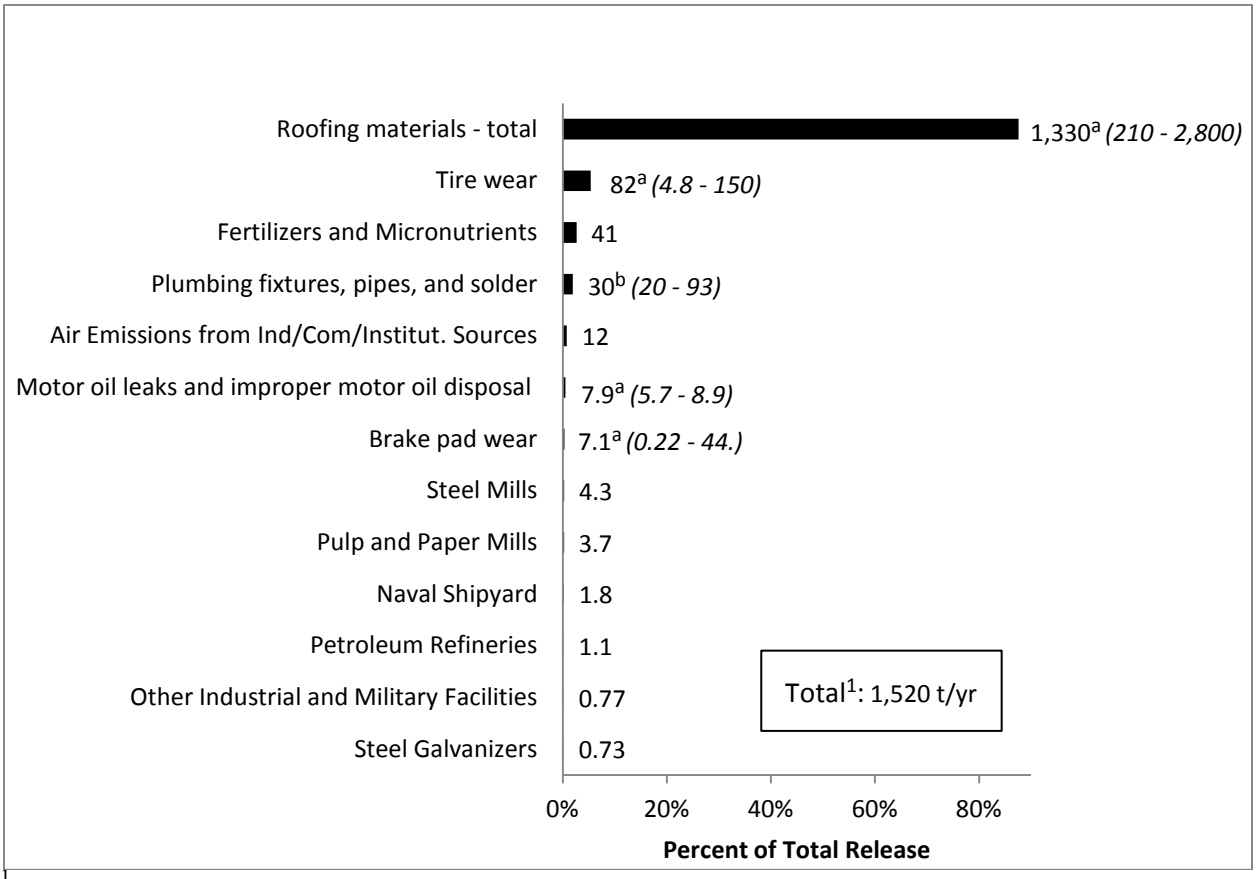
An inventory of zinc released from primary sources suggests that approximately 1,500 metric tons (t) is released annually from anthropogenic sources in the Puget Sound basin. The largest current source of zinc to the environment is the leaching of zinc from rooftops, particularly those with galvanized components, accounting for approximately 1,300 t/yr of zinc released (Figure 19). The authors of the *Sources Report* note that total inventoried zinc releases to the Puget Sound basin probably underestimate the true extent of release since leaching from other galvanized items (e.g. culverts, light standards, guardrails) was not assessed (Ecology, 2011).

Although small in comparison to rooftop releases, zinc released from tire wear is substantial (82 t/yr), accounting for approximately 6% of the total. Brake pad wear accounts for approximately 5 t/yr of zinc released in the Puget Sound basin.

Zinc contained in fertilizers and micronutrients used in agricultural applications accounted for large zinc releases (41 t/yr). Similar to most other COC metals, concentrations were highest in

phosphate fertilizers. However, zinc use in fertilizer applications (4 t/yr) is small compared to its use as an agricultural micronutrient (37 t/yr).

Other inventoried sources of anthropogenic release of zinc in the Puget Sound basin included leaching from residential plumbing components (21 t/yr) and industrial, commercial, and institutional emissions (approximately 24 t/yr). Of this latter category, approximately one-third of the zinc releases were from steel mills and pulp and paper mills.



[1] Sum of best estimates. Best estimates are either the mean, mid-point, median, or most reasonable estimate for each source.

[a] Mean

[b] Median

Figure 19. Total Zinc Release in the Puget Sound Basin (values shown are t/yr).

Major Delivery Pathways and Loading

Data on major delivery pathways for zinc loading were obtained from PSTLA loading studies and are included in Table 11. As mentioned previously, loading through direct groundwater discharge was estimated from literature values, whereas loadings through other pathways were estimated from field studies specifically designed to estimate loads.

Table 11. Total Zinc Loads (t/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater (a)	1.97	10.8	19.7
Air Deposition	11	18	26
Surface Runoff	113	122	134
POTWs	16	19	24
Ocean Exchange (b)	-150	-80	10

(a) Lowest, highest, and mid-point of estimated loads
(b) Negative values indicate a net outflow at the ocean boundary

The estimated range of groundwater zinc loads is large (2.0 – 20 t/yr). The range in estimates is due almost entirely to differences in flows used to establish the possible range of loads; groundwater discharges used in the groundwater loading study ranged by an order of magnitude.

Zinc discharged from POTWs in the Puget Sound basin has a much narrower range of loading, accounting for loads of 16 – 24 t/yr. Phase 1 estimates suggest that zinc loads from industrial wastewater are potentially substantial (16 t/yr; Hart Crowser et al., 2007), but this is based on a limited dataset and is far in excess of industrial discharge of zinc to surface waters or transferred to POTWs as reported in the TRI (<4 t/yr total) (Ecology, 2011).

The deposition of atmospheric zinc directly to the marine waters of Puget Sound is approximately 11 – 26 t/yr, based on fluxes ranging from 2.2 - 52 ug/m^2/d. Median zinc fluxes were generally <10 ug/m^2/d, except for one location within a high-density urban (Tacoma) area including a nearby pulp mill, metal refiners, other industrial activities, and close to major roadways including interstate highways. Zinc fluxes at this location were consistently an order of magnitude higher than other locations around the Puget Sound region, and were elevated five-fold above a nearby station that did not have the same air pollution influences in such close proximity. This pattern closely mirrored the deposition patterns of copper and lead among the air sampling locations.

Surface runoff loads for zinc are estimated to be approximately 113 – 134 t/yr for the entire Puget Sound basin based on the surface runoff study. Overall, dissolved zinc accounted for approximately one-half to three-quarters of the total zinc concentrations in surface water.

Absolute zinc loads were highest in forests, with little change in loading during storms compared to baseflows, although a seasonal first-flush episode was in forest, as well as agricultural areas. Zinc loads in other land covers increased dramatically during storms, particularly commercial/ industrial areas, where zinc loads increased by an order of magnitude. This leads to a disproportionately high loading of zinc in commercial/industrial areas relative to other land areas.

Based on sampling marine waters, there appears to be a large net export of zinc at the ocean boundary using estimates derived from the 25th and 50th percentiles of the data, although the dataset is very limited. However at the high end of the reported data range (75th percentiles), there is virtually no net flux across the ocean boundary. Total zinc concentrations (25th -75th percentile) in the incoming marine waters are 0.53 – 0.88 ug/l, and total zinc concentrations in

the outgoing marine waters are 0.70 – 0.80 ug/l. Zinc inflow is estimated to be 310 – 520 t/yr, and zinc outflow is estimated to be 460 – 510 t/yr.

The net sum of zinc loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25th, median (50th), and 75th percentile values is -8, 90, and 210 t/yr, respectively. Under the 25th percentile estimate, there is a comparatively small net export of zinc out of Puget Sound due to the large mass exchanged at the ocean boundary. However, when the median values are summed, the large surface water load outweighs the smaller export at the ocean boundary, leading to a net load of zinc load to Puget Sound, and all loads estimated at the 75th percentile indicate net loads to Puget Sound.

Zinc loading from pathways that may represent *partially controllable* sources of COCs (i.e. all of the major pathways except ocean exchange) are shown as ranges (25th - 75th percentiles) and median for each pathway in Figure 20. Each pathway represented as a contribution to the total load is displayed in Figure 21.

Total zinc loading from the major pathways assessed is 140 – 200 t/yr. Surface water runoff accounts for the largest pathway (66 – 83%), followed by POTWs (11 – 12%) and air deposition (8 – 13%). Groundwater potentially accounts for up to 10% at the upper end of the estimated range, but this value should be viewed with caution since it is based on literature values of zinc in wells and rough estimates of groundwater flow. At the lower end of the load range, zinc in groundwater is estimated to account for 1% of the load to Puget Sound.

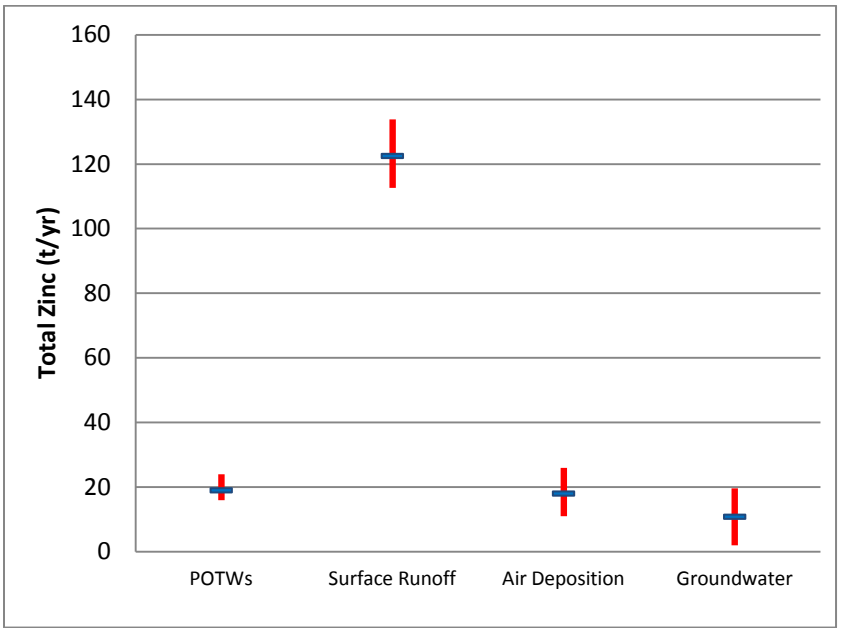


Figure 20. Range (25th - 75th percentiles) and Median Total Zinc Loads to Puget Sound from Each Major Delivery Pathway.

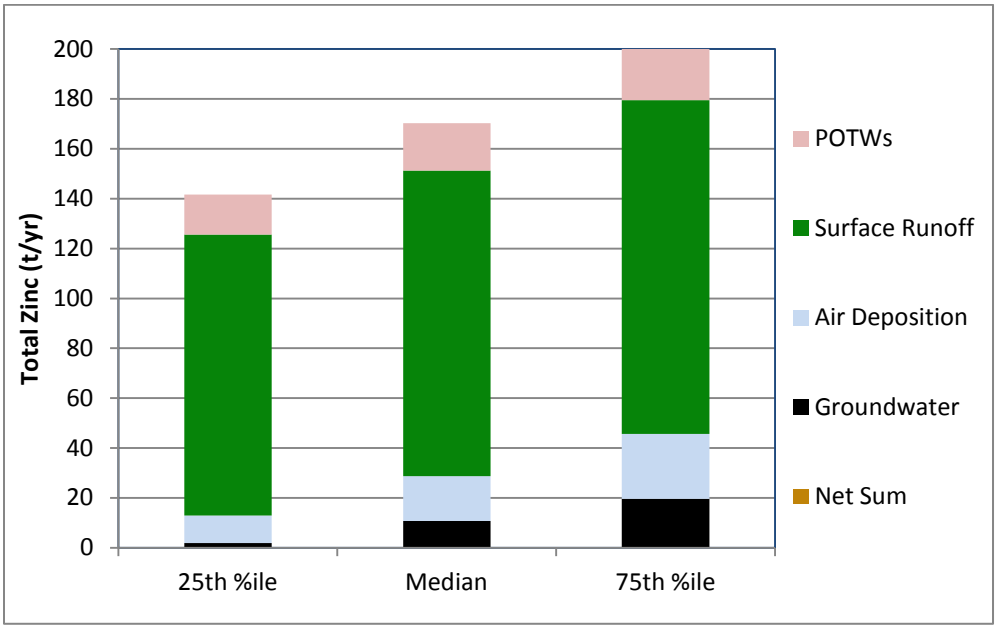


Figure 21. Total Zinc Loads to Puget Sound as the Sum of Major Delivery Pathways.

Relationship Between Sources and Pathways/Loading

The conceptual model of zinc transport and fate following release suggests that much of the zinc transported to Puget Sound will occur through surface runoff during both baseflow conditions and storm events. The high proportion of zinc loading contributed by surface runoff is consistent with the types of sources accounting for the major zinc releases.

Major unconstrained releases of zinc are leaching from rooftops and from vehicle component wear. Together, these sources account for approximately 1,400 t/yr of zinc released to the environment in the Puget Sound basin.

Once zinc is released from rooftops it may be transported in runoff to any number of pathways. Zinc may continue to remain in runoff on impervious surfaces until it reaches surface waters or is diverted to wastewater treatment plants, or zinc may be initially or secondarily allowed to infiltrate into soils where it can migrate to groundwater or become retained in a soil reservoir. Once zinc is in soil, it can be slowly leached out in dissolved form, migrate to groundwater, or become released as soil particles during high-energy storms where it settles as aquatic sediments, including those found in catch basins.

The surface runoff results appear to be consistent with major zinc releases from rooftops. During storms, zinc is released through leaching, and in commercial/industrial areas, the high proportion of rooftop area and relative dearth of attenuating components would allow for the enriched runoff to reach surface waters. Most of the zinc released from rooftop and galvanized materials would presumably be in the dissolved form, consistent with elevations of dissolved zinc during storms.

Although this conceptual model appears to fit with commercial/industrial areas during storms, it does not explain the comparatively low concentrations of dissolved zinc from residential areas, nor does it explain the high zinc concentrations in commercial/industrial areas during baseflows. While there are few clues to explain the finding of low zinc in residential areas, the high levels in commercial/industrial area baseflows may be due to a high density of galvanized culverts or a reservoir of zinc-enriched sediments residing in aquatic systems.

Aside from zinc released from rooftops, zinc released from vehicle component wear would most likely occur in residential and commercial/industrial areas. Abraded particles released to roadways and mobilized in storm runoff would presumably result in increases in total zinc in excess of comparable increases in dissolved zinc. The increase in concentrations of total zinc in commercial/industrial and residential areas appears to be consistent with this supposition, rising two- to three-fold, with concentrations significantly correlated with TSS. There are virtually no elevations in total zinc in agricultural and forested land covers during storm events.

Overall zinc releases to air across the entire basin (not including fugitive dust from tire and brake pad wear) are estimated to be approximately 18 t/yr. Zinc released as fugitive dust from tire and brake pad wear is potentially substantial (48 t/yr), assuming 50% is emitted as airborne particles or becomes suspended in air following release to the road surface. This may account for the high levels of zinc in atmospheric deposition samples at the high-density urban location, which is located near Interstates 5 and 705, associated interchanges, and other major roadways. Zinc fluxes at this location were an order of magnitude higher than at other locations, and the inventory of primary releases indicates that industrial sources are not large contributors to airborne emissions. The total zinc air emissions across the basin (up to 66 t/yr) appears to match the deposition on marine waters (11 – 26 t/yr) when one considers that marine waters only constitute one-sixth of the basin area.

Of the primary sources inventoried, zinc released from residential plumbing components represents the only constrained source assessed. The annual load of zinc discharged to POTWs is estimated to be 21 t; an additional 8 t/yr is discharged to septic systems from this source. The estimate of zinc released from plumbing components is nearly identical to loads released from POTWs (16 – 24 t/yr), but it is unlikely that a mass balance has been achieved simply from these loading terms. It is much more likely that zinc from other constrained sources is delivered to POTWs, and naturally occurring zinc in water, as well as zinc in stormwater, represents additional releases of zinc to POTWs, but the exact extent is not known. At the treatment end, some quantity of zinc is presumably lost via solids removal prior to discharge.

Hazard Evaluation

The hazard evaluation indicates that zinc is a Priority 1 level of concern for offshore marine surface waters and for freshwater sediments (Table 12). A Priority 2 level of concern was assigned to fresh surface waters and marine sediments. The number of observed data in nearshore marine waters was insufficient for an adequate comparison with effects data or criteria. Hazards due to tissue residue effects, effects to wildlife, and effects to human health were not evaluated.

The 90th percentile values for dissolved zinc in freshwater (approx. 15 ug/l) are one-half the 10th percentile of effects levels and the chronic and acute water quality criteria. For marine waters, the differences between the observed and effects/criteria is even larger, with the difference between the 90th percentile of observed data (approx 2.1 ug/l for nearshore and 1.2 ug/l for offshore) are one to two orders of magnitude below the 10th percentile of the effects data and the chronic and acute water quality criteria.

Median zinc concentrations in both freshwater and marine sediments are about one-half of the lowest guidelines or standards. However, in freshwater sediments the 90th percentile (approx. 300 mg/kg dw) of observed zinc concentrations exceeds the floating percentile SQS concentration and floating percentile CSL.

Zinc concentrations in marine nearshore and offshore sediment have 90th percentile levels (approx. 180 mg/kg dw and 120 mg/kg dw, respectively) less than one-half the SQS concentration, although they exceed Canadian TEL.

Table 12. Summary of Hazard Evaluation for Zinc.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.* *or* *Acute WQC* *or* *Chronic WQC*	**Level of Concern**
	Freshwater	4,844	88%	No	Priority 2
	Nearshore Marine	33	100%	INS	U
	Offshore Marine	57	95%	Yes	Priority 1
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. >** *SQS*	**Level of Concern**
	Freshwater	822	>99%	Yes	Priority 1
	Nearshore Marine	513	100%	No	Priority 2
	Offshore Marine	513	100%	No	Priority 2
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.*	**Level of Concern**
	Not Analyzed				
Wildlife		**Total N**	**FOD**	**Daily Dose >** *10% of Lowest Effects Dose*	**Level of Concern**
	Not Analyzed				
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. >** *NTR Criterion*	**Level of Concern**
	Not Analyzed				

Yellow highlight indicates Priority 1 Level of Concern
FOD=Frequency of detection
INS=Insufficient observed data available for comparison to effects data
U=Unknown level of concern due to lack of sufficient data for adequate comparison

PCBs

Polychlorinated biphenyls (PCBs) were manufactured commercially in the U.S. from 1929 until their ban in 1979 after the negative health and environmental impacts associated with PCBs became apparent (Sittig, 1980; EPA, 1999). EPA (1997) estimated that as of 1977, U.S. manufacturers had produced a total of 635,000 metric tons (t) of PCBs. Prior to federally imposed use restrictions, the PCB market spanned a wide range of end products. While electrical equipment represents the majority of PCB use – 77% from 1929-1975 according to EPA (1997) – their chemical stability and plasticizing properties made them useful in a variety of applications. PCBs in open system applications such as plasticizers, hydraulics fluids and lubricants, and carbonless copy paper accounted for >20% of their historic use.

Although banned more than three decades ago, PCBs continue to be found in environmental media. Many of the same properties that made PCBs commercially desirable – their stability and resistance to degradation – make them extremely persistent in the environment, and they have become one of the most ubiquitous of all environmental contaminants.

There are 209 individual forms of PCBs, known as congeners, based on the degree of chlorine substitution and arrangement on the biphenyl molecule. The persistence of PCBs increases with the degree of chlorination. Mono-, di-, and tri-chlorinated biphenyls biodegrade relatively rapidly, tetrachlorinated biphenyls biodegrade slowly, and higher chlorinated biphenyls are resistant to biodegradation. PCBs accumulate in the lipids (fats) of fish and other animals, with lipid solubility typically increasing with the degree of chlorination (Mabey et al., 1982).

PCBs are typically present at very low concentrations in ambient waters, with water column concentrations typically in the 10 – 1,00 pg/l range for total PCBs (Dangerfield et al., 2007; Appendix D), although few data are available for marine waters (Serdar, 2008). In sediments, total dry weight PCB concentrations are typically found in the 1 – 100 ug/kg (dw) range (Appendix D).

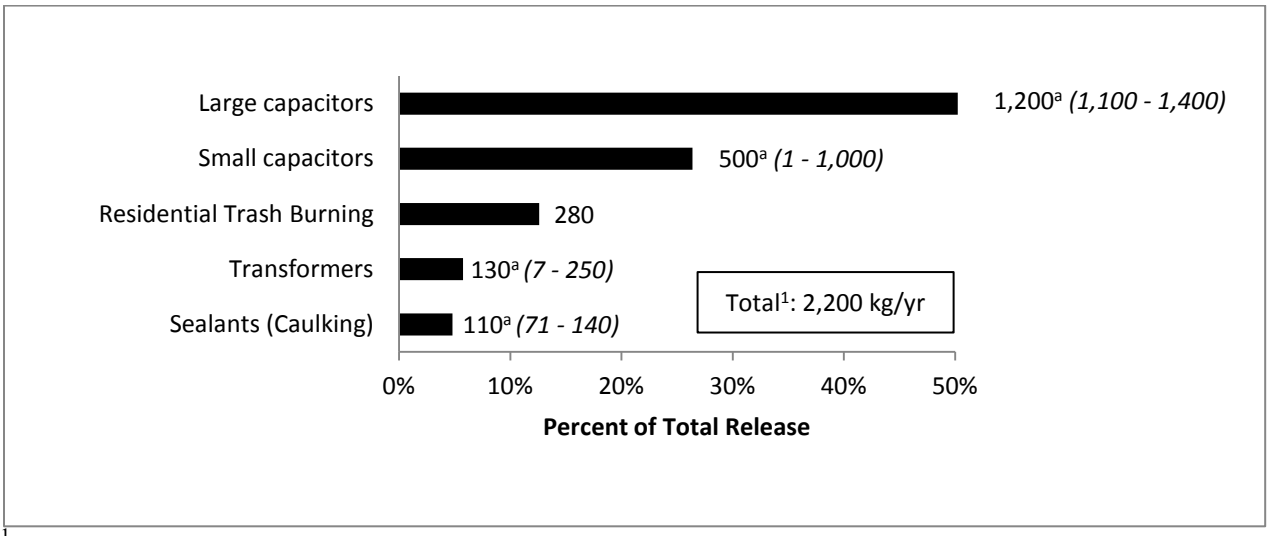
Major Releases from Primary Sources

An inventory of PCBs released from primary sources suggests that approximately 2,200 kg is released annually in the Puget Sound basin (Figure 22). PCB use can be placed in two categories: closed systems and opens systems. Closed systems include PCBs used in electrical transformers and capacitors, including those used in light ballasts. The release of PCBs due to leakage of closed systems was estimated at 1,800 kg/yr in the Puget Sound basin.

PCBs historically were used in a number of open-system products such as adhesives, carbonless copy paper, flame retardant coatings, pesticide extenders, lubricants, and caulking sealants. Of the products that incorporated PCBs, caulking sealants are among the most durable. Caulking used in commercial, industrial, and institutional buildings constructed during the 1940s through the 1970s may contain PCBs which may continue to be released through volatilization, leaching, or abrasion of the material. Based on the volume of commercial, industrial, and institutional buildings constructed in the Puget Sound basin during the era of PCB caulk use, an estimated 110 kg of PCBs are released from this source annually.

An additional source of PCB release is from residential trash burning. Based on modeling information generated by Ecology's Air Quality Program, PCBs are emitted from this source at a rate of 281 kg/yr in the Puget Sound basin.

The annual rate of PCB release, estimated to be 2,200 kg/yr, should be viewed with caution and likely overestimates actual releases. No regional sampling efforts to inventory PCB releases from primary sources have been conducted, and therefore PCB release estimates are based on literature values. There are also no regional field studies to estimate leakage rates, relative amounts released indoors and outdoors, and the proportion cleaned up and contained following spillage. The estimates displayed in Figure 22 assumed all leakage was unconfined.



[1] Sum of best estimates. Best estimates are either the mean, mid-point, median, or most reasonable estimate for each source.
[a] Mid-point of range

Figure 22. Total PCB Release in the Puget Sound Basin (values shown are kg/yr).

Major Delivery Pathways and Loading

Data on major delivery pathways for PCB loading were obtained from PSTLA loading studies and are included in Table 13. No PCB loading data are available for groundwater.

The estimated discharge of PCB from POTWs is 0.13 – 1.75 kg/yr. The POTW load estimates were based on limited sampling conducted only during the wet season.

The deposition of atmospheric PCBs directly to the marine waters of Puget Sound is approximately 0.7 – 3.7 kg/yr, based on fluxes ranging from 0.16 – 2.8 ng/m^2/d. Median PCB fluxes were generally <0.7 ng/m^2/d, except for one location within a high-density urban (Tacoma) area including a nearby pulp mill, metal refiners, other industrial activities, and close to major roadways including interstate highways. PCB fluxes at this location were elevated five-fold above most other locations around the Puget Sound region, including a nearby station that did not have the same air pollution influences in such close proximity.

Table 13. PCB Loads (kg/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater	NA	NA	NA
Air Deposition	0.68	1.32	3.76
Surface Runoff	2.55	5.29	15.77
POTWs	0.126	0.342	1.75
Ocean Exchange (a)	-1.4	0.8	0.6
Returning Salmon (b)	NC	0.265	NC

NA=not analyzed
NC=not calculated
(a) Negative values indicate a net outflow at the ocean boundary
(b) Best estimate using available data

Surface runoff loads for PCBs are estimated to be approximately 2.6 – 15.8 kg/yr for the entire Puget Sound basin based on the surface runoff study. PCBs were detected in all land use types, and median concentrations among land covers were within a factor of 3 during baseflows. However, during storm flows, concentrations in commercial/industrial areas increased by an order of magnitude while concentrations in other areas remained virtually unchanged. PCB concentrations were particularly elevated during seasonal first-flush episodes in all land types except agricultural areas.

Overall PCB loads were generally proportional to land area when evaluated by land cover type, except commercial/industrial loads during storm events (percentage of the total PCB load was 15-fold higher than the percentage of commercial/industrial land cover in the Puget Sound basin). However, due to the large area and flows of forested areas, overall loads from forests accounted for 83% of the total PCB load.

Based on the sampling of marine waters, there appears to be an annual net export of 1.4 kg total PCBs at the ocean boundary using estimates derived from the 25th percentile of the data, although the dataset is very limited. At the median and higher range (75th percentile) of the estimates, there is less than 1 kg annual flux into Puget Sound at the ocean boundary. Total PCB concentrations (25th-75th percentile) in the incoming marine waters are 15 – 35 pg/l, and total PCB concentrations in the outgoing marine waters are 14 – 47 pg/l. PCB inflow is estimated to be 8.6 – 21 kg/yr, and PCB outflow is estimated to be 10 – 20 kg/yr.

In addition to major loading pathways, estimates of PCB influx to Puget Sound through returning adult salmon were based on typical whole-body PCB concentrations measured during 2004-2005 and escapement estimates from 2001. Rough estimates for five species of pacific salmon (*Oncorhynchus spp.*) suggest that the total influx of PCBs from this pathway is approximately 0.265 kg/yr based on whole-body PCB concentrations ranging from 4 ug/kg to 51 ug/kg (Sandie O'Neill, National Oceanic and Atmospheric Administration, written communication).

The net sum of PCB loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25th, median (50th), and 75th percentile values is 2.2, 8.0, and 22 kg/yr, respectively. Under all of these estimates, there is a net PCB load to Puget Sound.

PCB loading from pathways that may represent *partially controllable* sources of COCs is shown as ranges (25th - 75th percentiles) and median for each pathway in Figure 23. Each pathway represented as a contribution to the total load is displayed in Figure 24.

Total PCB loading from the major pathways assessed is 3.3 – 21 kg/yr. Surface runoff accounts for the largest pathway (74 – 76%), followed by atmospheric deposition (18 – 20%) and POTWs (4 – 8%).

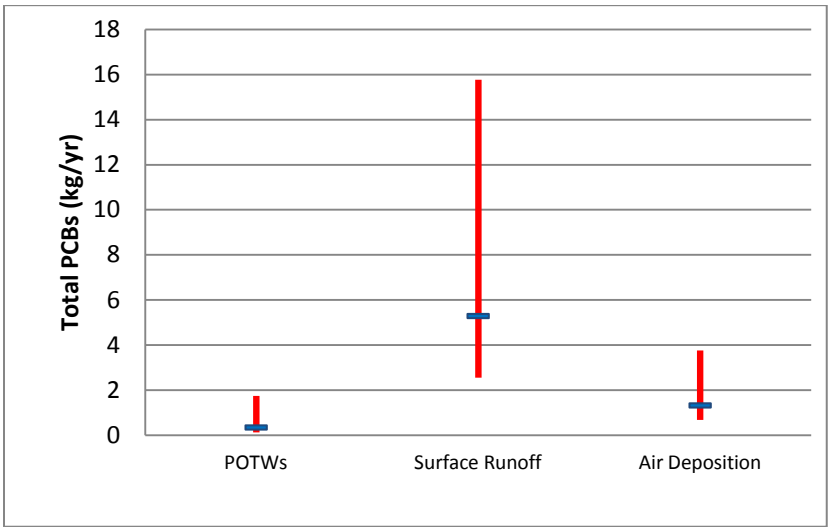


Figure 23. Range (25th - 75th percentiles) and Median Total PCB Loads to Puget Sound from Each Major Delivery Pathway.

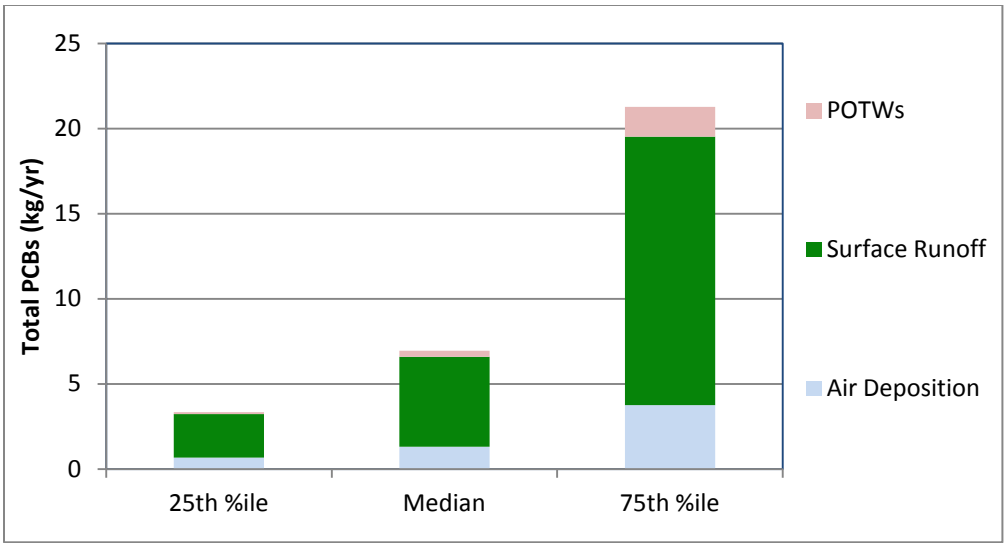


Figure 24. Total PCB Loads to Puget Sound as the Sum of Major Delivery Pathways.

Relationship Between Sources and Pathways/Loading

The conceptual model of PCB transport and fate following release suggests that much of the PCBs transported to Puget Sound will do so through surface runoff during both baseflow and storm flow conditions. Surface runoff data suggest that in residential, agricultural, and forested areas, a continuous and low level of PCB dosing from instream or upland sources occurs during baseflow. PCB concentrations remain the same or decrease slightly during storms, indicating that stormwater contains similar or lower PCB concentrations than the stream baseflows, and any mobilization of instream PCB reservoirs do not effectively increase concentrations.

In commercial/industrial areas, the dynamics of PCBs in surface runoff appear to be much different. PCB concentrations during baseflow conditions are 40% - 180% higher than in other land covers. During storm events, PCB concentrations in commercial/industrial areas increase six-fold. Based on the available information, it is impossible to ascertain whether the increase is due to mobilization of land surface PCBs, re-suspension of instream PCB reservoirs, or a combination of the two circumstances.

The major PCB sources identified in the *Sources Report* (Ecology, 2011) do not appear to have a direct link with the surface runoff pathway. Most of the PCBs releases are likely to occur in and around buildings and become bound to soil following release, volatilize and become transported off-site, or occur indoors. Residential trash burning presumably occurs in residential areas. However, commercial/industrial areas are the most likely to deliver PCBs to surface waters based on the primary sources since PCBs released from buildings and from transformers/ capacitors are more likely to occur in commercial/industrial areas than in the other land covers assessed. In addition, PCBs atmospherically deposited on land surfaces are more likely to become mobilized during storms if they are deposited on impervious surfaces which are more prevalent in commercial/industrial areas.

One other possible source of PCB enrichment of streams is marine-derived PCBs delivered upstream by salmon returning to spawn. PCB residues per whole-body fish range from approximately 7 ug for pink salmon (*O. gorbuscha*) to 336 ug for Chinook (*O. tshawytscha*) (Sandie O'Neill, National Oceanic and Atmospheric Administration, written communication). While the total annual PCB load entering Puget Sound from salmon is estimated to be approximately 0.3 kg/yr, the impact to streams is likely to be somewhat less. The load estimate does not include losses from commercial and recreational takes, live fish and carcasses removed from streams by wildlife, and maternal transfer to eggs (and subsequent flux from out-migrating smolts). In addition, approximately one-third of the PCB burden is carried by two species – pink salmon and chum salmon (*O. keta)* – which spawn much lower in the watershed than other species, thus diminishing upstream PCB transport.

PCB releases to air from inventoried sources are several orders of magnitude greater than air deposition to marine water (approx. 300 – 400 kg/yr versus 0.7 – 3.7 t/yr), and this discrepancy is large even when the deposition rates to marine water are scaled to the entire watershed area. The primary releases to air are combustion emissions originating from residential trash burning (280 kg/yr) and volatilization from PCB-containing building sealants (maximum of 140 kg/yr). There is a large degree of uncertainty surrounding release estimates from both of these sources;

neither has been sufficiently analyzed to determine if reported release rates are representative of the Puget Sound region.

PCB deposition patterns indicate similar fluxes occur throughout Puget Sound, suggesting a widespread atmospheric deposition pattern compounded with additional deposition from near-field sources in high-density urban areas. PCB emissions from building sealants are expected to be concentrated in cities, particularly older industrial cities, although it is not clear if they are likely to be deposited near their point of release.

Based on the inventory of sources, none of the PCB releases were exclusive to constrained systems such as sanitary sewers. Therefore it is difficult to quantitatively assess the relationship between releases and POTW loads (0.1 – 1.8 kg/yr). Some of the PCBs released from building sealants are likely to occur indoors and find their way to sanitary sewers after attaching to dust particles, although the quantity has not been determined. It is likely that some portion of the PCBs delivered to POTWs occurs through stormwater, but the extent of this contribution is not known. Since PCBs were historically used in a variety of commercial, industrial, and consumer applications (see *Sources Report* [Ecology, 2011] for a discussion of PCB uses), releases to constrained and unconstrained sources are likely to continue from a variety of sources.

Hazard Evaluation

Comparisons to effects levels, criteria, and guidelines are done separately for both PCB Aroclor and congener data. PCBs were historically marketed in the U.S. as Aroclors, mixtures of individual PCB compounds (a.k.a. congeners) based on average chlorine content. Environmental analysis of PCBs historically has focused on these Aroclor mixtures, although once in the environment Aroclors quickly alter their original composition due to unequal degradation, fugacity, and bioaccumulation rates of their individual components.

Beginning in the mid-1990s, the analysis of PCB congeners gained wider acceptance as the laboratory capacity for this method became more widely available and the utility of congener analysis became more evident. Although much more expensive than Aroclor analysis, congener analysis provides detection limits several orders of magnitude lower than Aroclors (e.g. mid parts per quadrillion levels versus mid parts per trillion levels in water). As a result, environmental sample datasets are generally a mix of Aroclor and congener data. Due to the lower congener detection limits, results of measurable PCBs are typically lower for congeners (as mentioned previously, the hazard evaluation results are shown for detectable concentrations only).

The hazard evaluation indicates that PCBs are a Priority 1 level of concern for a range of media and receptors in both the freshwater and marine aquatic environments (Table 14).

Table 14. Summary of Hazard Evaluation for PCBs.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.* or *Acute WQC* or *Chronic WQC***	**Level of Concern**
	Freshwater	1,248[a]	4%[a]	Yes	Priority 1
	Nearshore Marine	11[a]	0%[a]	INS[b]	U
	Offshore Marine	84[c]	100%[c]	No	Priority 2
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. > *SQS***	**Level of Concern**
	Freshwater	506[a]	43%[a]	Yes	Priority 1
	Nearshore Marine	612[a]	63%[a]	No	Priority 2
	Offshore Marine	387[a]	47%[a]	Yes	Priority 1
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.***	**Level of Concern**
	Freshwater	d,e	88%	Yes/INS[b]	Priority 1/U
	Nearshore Marine	d,f	100%	No/INS[g]	Priority2/U
	Offshore Marine	d,h	≥99%	No/INS[i]	Priority 2/U
Wildlife		**Total N**	**FOD**	**Daily Dose > *10% of Lowest Effects Dose***	**Level of Concern**
	Great Blue Heron (FW)	--	--	Yes	Priority 1
	Osprey (SW)	--	--	Yes	Priority 1
	River Otter (FW)	--	--	Yes	Priority 1
	Harbor Seal (SW)	--	--	Yes	Priority 1
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. > *NTR Criterion***	**Level of Concern**
	Freshwater	d,j	≥77%	Yes	Priority 1
	Nearshore Marine	d,k	>33%	Yes	Priority 1
	Offshore Marine	d,l	>66%	Yes	Priority 1

Yellow highlight indicates Priority 1 Level of Concern
FOD=Frequency of detection
[a] Based on Aroclor data
INS[b]=Insufficient observed data available for comparison to effects data
U=Unknown level of concern due to lack of sufficient data for adequate comparison
[c] Based on congener data
[d] Based on Aroclor and congener data
[e] N = 123 – 142 for non-decapod invertebrates. Observed data insufficient to evaluate fish tissue.
[f] N = 27 – 28 for decapods, N = 57 – 99 for non-decapod invertebrates, and N = 70 – 96 for fish tissue. Effects data insufficient to evaluate fish tissue.

(continued on next page)

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INS[g]=Insufficient effects data available for comparison to observed data

[h] N = 11 – 32 for non-decapod invertebrates and N = 26 – 324 for fish tissue. Observed data insufficient to evaluate decapods. Effects data insufficient to evaluate fish tissue.

INS[i]=Insufficient observed or effects data available for comparison

FW=Freshwater

SW=Saltwater

[j] N range is 51 – 918 and varies for organism type, tissue type, and PCB type.

[k] N range is 68 – 344 and varies for organism type, tissue type, and PCB type.

[l] N range is 10 – 477 and varies for organism type, tissue type, and PCB type.

PCB concentrations in surface waters vary depending on whether Aroclor or congener data are considered. The 90th percentile of observed freshwater Aroclor concentrations (approx. 0.05 ug/l) is several times higher than the chronic water quality criterion, but the 90th percentile of observed freshwater congener concentrations (approx. 0.002 ug/l) is several times lower than the criterion. Although there are more Aroclor data available, the detection frequency is much higher for congener data. For both sets of observed data, most (>95%) of the observed concentrations are one to two orders of magnitude below concentrations where most (95%) of the effects have been documented. It is notable that the numerical value for the chronic water quality criterion (0.014 ug/l) is driven by adverse reproductive effects to mink through consumption of fish, demonstrating the importance of PCBs' high bioconcentration potential and manifestation of effects on higher trophic organisms.

There are no detectable PCB concentrations observed in nearshore marine waters to compare to effects data or criteria. For offshore marine waters, the 90th percentile of congener concentrations (approx. 0.00005 ug/l) is five orders of magnitude less than the 10th percentile of effects data and three orders of magnitude below the chronic water quality criterion.

In freshwater sediments, at least 25% of the observed PCB concentrations analyzed as congeners or Aroclors exceed the floating percentile SQS, although the number of congener samples is comparatively small. At least 5% of the Aroclor data exceed all of the guidelines and standards used for comparison.

PCB concentrations in marine sediments are difficult to characterize due to the vast differences between observed congener and Aroclor concentrations (Aroclors are two to four orders of magnitude higher), as well as large differences between concentrations in nearshore and offshore sediments (offshore Aroclors concentrations are two orders of magnitude higher than nearshore Aroclor concentrations). The congener concentration, assessed on either dry weight or organic-carbon normalized bases, are well below any guideline, whereas median nearshore Aroclor concentrations fall in the midst of guidelines, with the 75th percentile of the concentrations (approx. 20,000 ug/kg organic carbon) above the SQS.

Interestingly, although the nearshore Aroclor concentrations are two orders of magnitude higher than congeners, the concentrations are nearly identical on an organic-carbon normalized basis, suggesting that the differences can be partly attributed to high levels of organic carbon in the samples analyzed for Aroclor. The magnitude of difference between nearshore and offshore Aroclor concentrations remain after accounting for organic carbon. All level-of-concern

assignments for sediments were based on Aroclor data since the congener datasets were not deemed sufficient for adequate comparisons (n ≥100; Appendix D-1).

Comparisons of observed PCB concentrations in tissues of fish, decapods, and other invertebrates to effects due to PCB burdens were difficult to evaluate due to a paucity of either observed or effects data. In freshwater where sufficient observed and effects data were available for non-decapod invertebrate, most of the observed concentrations measured as both Aroclor and congeners exceeded all of the effects concentrations.

For nearshore marine waters, both decapods and non-decapod invertebrates had sufficient (observed and effects) data to conduct an evaluation. For both organism types, the 90th percentile of observed data was at least five-fold lower than the 10th percentile of effects data. In offshore marine water, non-decapods invertebrate data were available to conduct comparisons; the 90th percentile value of observed concentrations was an order of magnitude below the 10th percentile of effects concentrations. There were not sufficient effects data for fish tissue to conduct an adequate evaluation in marine surface waters.

Evaluation of daily PCB doses based on fish and incidental sediment ingestion for the four species evaluated – great blue heron, osprey, river otter, and harbor seal – indicate that all species would be exposed to doses equal or greater than the lowest effects dose (assuming a 4% sediment ingestion rate for heron). For the bird species, the lowest effects doses are based on reproductive effects; osprey are exposed to doses three times the lowest of the effects level. For the mammal species evaluated, both seal and otter exceed the lowest dose calculated for reproductive effects by an order of magnitude.

Edible tissues evaluated for comparison to the NTR criterion (based on the default consumption rate of 6.5 g/d) indicate that PCB concentrations in nearly all of the fish and non-bivalves are higher than the NTR criterion. For freshwater, nearly all of the bivalve tissue had concentrations above the criterion as well. Bivalve PCB concentrations in nearshore areas are higher than those from offshore areas.

Regionally Important Biological-Effects data

PCBs have been detected in outmigrant juvenile salmon (Johnson et al., 2007) from multiple northwest estuaries and hatcheries, including three in the Puget Sound. Whole-body juvenile Chinook salmon from the Duwamish River contained the highest PCB concentration [103 ng/g wet weight (ww) or 3,100 ng/g lipid] of any of the locations tested. Johnson et al. (2007) note that this concentration is higher than the 2,400 ng/g lipid developed by Meador et al. (2002) as a tissue threshold for adverse health effects including reduced growth, altered enzyme and hormone activity, and increased mortality.

Separately, juvenile salmonid PCB exposures were documented as occurring via food source by an analysis of stomach content of outmigrants at three locations in Puget Sound (Stein et al., 1995). Meador et al. (2010) found that PCB tissue concentrations in outmigrant juvenile Chinook salmon from the Duwamish estuary varied by time and location within the estuary, suggesting that localized heterogeneity of sediment concentrations may substantially impact accumulation in fishes.

PCB concentrations in adult Puget Sound Chinook salmon tissues were found to be three to five times higher than those measured in six other populations of Chinook salmon on the West Coast of North America (O'Neill and West, 2009). Approximately 22% of maturing and sub-adult Puget Sound Chinook salmon had concentrations above the 2,400 ng/g lipid threshold mentioned previously. O'Neill and West (2009) note that these elevated tissue concentrations have resulted in consumption advisories, and have implications for the viability of these fish and southern resident killer whales. Cullon et al. (2009) found elevated PCBs in adult Chinook returning to the Duwamish River, as well as in Puget Sound Chinook smolts.

PCB concentrations in Puget Sound herring and Puget Sound flatfish have also been evaluated. Puget Sound herring were found to contain three to nine times higher concentrations of PCBs than herring from the Strait of Georgia, with Puget Sound whole-body concentrations ranging from about 120 to 160 ng/g wet weight (ww) (West et al., 2008).

The Puget Sound Partnership has adopted a target for PCB concentrations in fish as one of the first "dashboard indicators" for toxic chemicals in fish (PSP, 2011a). The dashboard indicators were developed to provide a broad range of measurements to assess the health of Puget Sound. The PCB target is based on the documented accumulation of PCBs in a variety of Puget Sound fish species and the availability of a PCB tissue threshold concentration (i.e., 2,400 ng/g lipid; Meador et al., 2002). Specifically, the target is to reduce PCB levels in Puget Sound so that 95% of the sampled species have tissue concentrations below the threshold. Currently, at least 15% and up too 100% of the Chinook salmon, Pacific herring, and English sole analyzed have PCB concentrations exceeding the threshold.

In addition to measurements of PCB accumulation in tissues, analyses of various biomarkers of pollution exposures in benthic flatfish were shown to successfully differentiate between sites with differing degrees of sediment contamination (Stein et al., 1992). Cullon et al. (2005) also found about seven times higher levels of PCBs in a mixture of fishes designed to represent the diet of Puget Sound harbor seals than in a similar mixture of fish designed to represent the diet of harbor seals from the Strait of Georgia. Sol et al. (2008) found a statistically significant correlation between PCB concentrations in English sole livers and two biological effects parameters.

A number of studies have been conducted in the Puget Sound region investigating exposure and/or effects of PCBs and other persistent and bioaccumulative contaminants on wildlife, particularly marine mammals. Johnson et al. (2009) measured PCB concentrations in osprey eggs from the Lower Duwamish River and compared them to those sampled from the upper Willamette River. Total PCB residues were significantly higher in Lower Duwamish River osprey eggs (geometric mean = 897 ug/kg ww) compared to those from the Willamette River (geometric mean = 182 ug/kg ww). These results demonstrate that adult osprey bioaccumulation and maternal transfer of PCBs is occurring in osprey nesting in PCB contaminated areas of Puget Sound. This study also compared egg residues over time and determined that PCB concentrations in osprey eggs from the Lower Duwamish River had decreased 53% between 2003 and 2007.

Grove and Henny (2008) also demonstrated the bioaccumulation of PCBs in river otter livers from Puget Sound. The Puget Sound area river otters accumulated more PCBs (as total PCBs)

than otters from other areas in western Washington. PCBs and other organochlorines have been shown to cause immunosuppression, thyroid disruption, and possibly cancer in harbor seals (Tabuchi et al., 2006; Ylitalo et al., 2005; Simms et al., 2000; Ross et al., 1996; Ross et al., 1995; de Swart et al., 1996; de Swart et al., 1995; Van Loveren et al., 1994). Vitamin A disruption has also been observed in harbor seal pups found on the Washington State coast whose mothers contained high PCB residues in their blubber (Simms et al., 2000). This effect on seal pups is suspected to result from exposure to contaminated milk.

There is substantial evidence that Puget Sound harbor seals and killer whales are bioaccumulating PCBs at very high concentrations in their blubber. The prey items of Puget Sound harbor seals were measured to have seven times higher concentrations of PCBs than prey from Strait of Georgia on a lipid basis (Cullon et al., 2005), which corresponds to PCB concentrations measured in harbor seal blubber.

Tissue concentrations of PCBs have often been reported as dioxin toxicity equivalents (i.e. TEQs) which are toxicities of dioxin-like PCBs, dioxins, and furans relative to the most toxic dioxin (2,3,7,8-TCDD). Puget Sound harbor seals have significantly higher TEQs in blubber (158 ng/kg lipid weight) compared to seals from the Strait of Georgia (33 ng/kg lipid weight) (Ross et al., 2004), and the TEQ contribution was greater from PCBs than dioxins and furans. Levin et al. (2005) also found that the majority of TEQs in harbor seal pups (from southern B.C.) were from PCBs, not dioxins and furans.

Ross et al. (2000) reported measured mean total PCBs in transient and Southern resident male killer whales were 251 and 146 mg/kg lipid, respectively; the authors concluded these marine mammals are among the most contaminated in the world.

Further research on the northern, southern, and transient killer whale communities have discovered that males bioaccumulate more PCBs than females due to maternal transfer (Krahn et al., 2007; Krahn et al., 2009). PCB concentrations in the blubber of mothers decrease temporarily during nursing and can reach concentrations below those in their calves. Mothers initiate bioaccumulation again after calves are weaned. Total PCB concentrations in southern resident killer whales were measured to range from about 5,000 to 180,000 ug/kg lipid. For all but three recent mothers, the measured concentrations exceed a marine mammal threshold for blubber concentrations (17,000 ug/kg lipid). Although environmental concentrations of PCBs are gradually declining, one modeled estimate of southern resident killer whale recovery projects that blubber concentrations will not reach the marine mammal threshold until 2063 (Hickie et al., 2007).

Accumulation of PCBs in fish tissue has led to advisories for human consumption of fish from Puget Sound marine waters, as well as limited freshwaters in the basin. The advisories, issued by WDOH, are based on data and consumption of particular species, and vary by region (Hardy and Palcisko, 2006). For instance, WDOH advises limited consumption of rockfish and flatfish based on the marine area in which they are caught. Consumption limits are also recommended for Chinook salmon due to elevated PCB levels, and vary depending on whether fish are migratory or the resident (blackmouth) species. WDOH also provides consumption advice for several Puget Sound region freshwater lakes and rivers due to PCBs in fish, including Lake Washington, Green Lake, and the Lower Duwamish River (WDOH, 2011).

PBDEs

Polybrominated diphenyl ethers (PBDEs) have been incorporated as flame retardants in numerous consumer products for decades and as a result, have gradually been released to the environment where they persist and accumulate in biota. Unlike most other persistent organic pollutants addressed in this report, environmental concentrations of PBDEs appear to be escalating in some cases, although available data on which to assess trends are also much more limited than for other chemicals.

Manufacturers of many different materials and products have used PBDEs as flame retardant additives in their products since the 1960s. These products include fabrics, television sets, computers, ABS resins, high impact polystyrene, textile coatings, carpet, polyurethane foams, cushions, mattresses, and insulation for wire and cables.

PBDEs are not chemically bonded to the matrices of those materials and products, and therefore they potentially escape from their matrix through volatilization to the air. Products and materials partially composed of or treated with PBDEs off-gas PBDEs to the environment during the useful lifetime of the product or material (i.e., while the product or material is still in use). Volatilization is one of the primary mechanisms of the release of PBDEs to the environment (Lorber and Cleverly, 2010).

Since PBDEs are a complex mixture of 209 congeners (varying by the number of bromine atoms and location on the molecule), their use and behavior in manufactured materials, as well as their behavior once released from materials, varies substantially. Major homolog groups (groups classified by the number of bromine atoms per molecule) include penta-, octa-, and deca-brominated diphenylethers (commonly referred to as Penta, Octa, and Deca, respectively). Penta was used widely in polyurethane foam and textiles, while the heavier homologs (Octa and Deca) were used primarily in polymers and electronics. The heavier homologs tend to be less volatile than the lighter BDEs, although once in the environment they may degrade to the lighter homologs. Heavier congeners such as Deca may also bind to dust more strongly than the more volatile congeners.

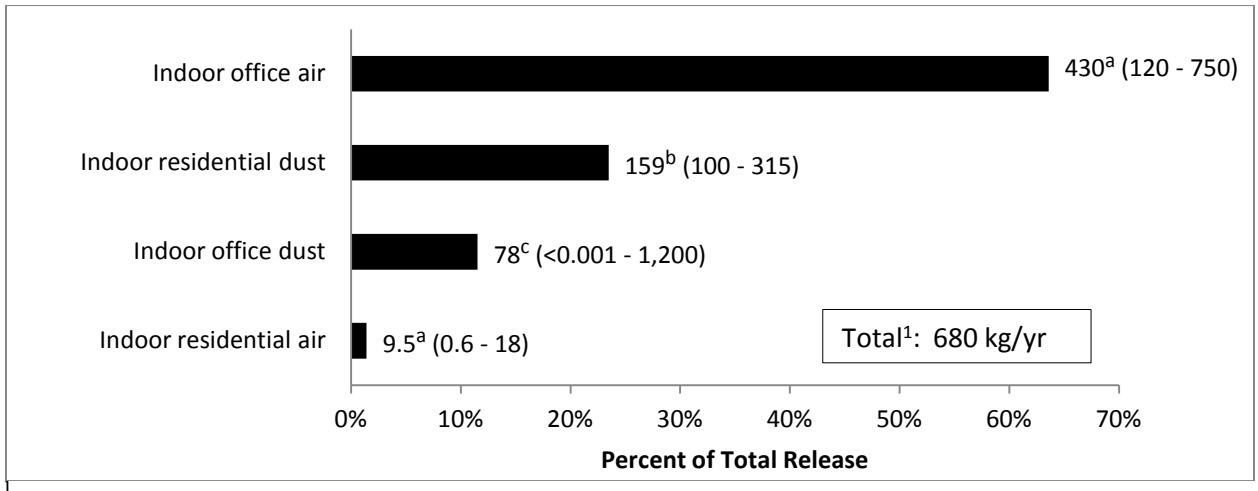
Beginning in the late-1990s, concerns began to emerge over the accumulation of PBDEs in animal tissues and the potential toxicity of PBDEs. Pressure to limit or ban PBDEs continued to mount until manufacturers of Penta and Octa voluntarily ceased production beginning in 2004. Deca manufacturers have agreed to discontinue the manufacture, import, and sales of Deca at the end of 2012, but in Washington State, Deca has been banned from mattresses since 2008 and was banned from televisions, computers, and residential upholstered furniture beginning January 1, 2011.

Major Releases from Primary Sources

Unlike most of the other COCs addressed in the present report, PBDEs are nearly all released by consumer products in constrained systems (i.e. indoors). For the inventory of primary sources (Ecology, 2011), no attempt was made to quantify environmental releases based on emissions from categories of PBDE-containing products such as computer monitors and mattress pads since most releases occur indoors and the attenuation between initial emissions and release to an environmental medium or pathway is uncertain. Therefore, release estimates were based on air exchange and dust generation in residential and commercial office spaces. This approach was taken to integrate individual component emissions and quantify PBDE releases in a simplified manner.

Total PBDE release from the four sources assessed totaled approximately 680 kg/yr (Figure 25). Indoor office space air accounted for 64% of the total release, while indoor residential air accounted for only 1%; indoor residential dust was the other major contributor at 23% while the indoor office dust contributed 12% to the overall releases. These release estimates should be viewed with caution since there is a high degree of uncertainty around all of the variables used to derive the values.

The PBDE Chemical Action Plan (Ecology and WDOH, 2006) largely addresses these sources and is consistent with the bans and restriction on PBDEs mentioned previously. However, many PBDE-containing consumer and office products are still in use and may represent diffuse sources of PBDEs to the environment during the remainder of their life cycle.



[1] Sum of best estimates. Best estimates are either the mean, mid-point, median, or most reasonable estimate for each source.

[a] Mid-point of range

[b] Median

[c] Geometric mean

Figure 25. Total PBDE Release in the Puget Sound Basin (values shown are kg/yr).

Major Delivery Pathways and Loading

Data on major delivery pathways for PBDE loading were obtained from PSTLA loading studies and are included in Table 15. No loading data are available for groundwater.

Table 15. PBDE Loads (kg/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater	NA	NA	NA
Air Deposition	15.7	20.3	23.8
Surface Runoff	5.14	5.67	9.95
POTWs	7.01	10.6	20.7
Ocean Exchange (a)	41	-11	240

NA=not analyzed
(a) Negative values indicate a net outflow at the ocean boundary

Loads from atmospheric deposition are estimated to be 16 – 24 kg/yr based on fluxes ranging from 4.5 to 27.3 ng/m^2/d (25th – 75th percentiles). Samples collected from the high-density urban station had higher PBDE fluxes than at other sites by factors of 3 to 4. PBDE flux patterns reflect some increased localized input, but regional sources may also play an important role in overall loading. The authors of the air deposition loading study point out that the estimates are likely to be conservative (high) due to the use of conservative assumptions to handle non-detected results.

PBDEs were frequently detected in the POTW loading study, with annual loads estimated at 7.0 – 21 kg. Although the authors of the POTW study caution against drawing conclusions about seasonal differences, they point out that PBDE concentrations are generally higher during the dry season.

Surface runoff loads for PBDEs are estimated to be approximately 5.1 – 10 kg/yr for the entire Puget Sound basin based on the surface runoff study. PBDEs were detected more frequently in storm runoff samples compared to baseflow. All samples from commercial/industrial areas contained detectable PBDE concentrations, but detection frequencies were close to 50% in other land covers (38% in forests).

Concentrations of PBDEs in commercial/industrial areas were higher than in other areas during both baseflows and storm flows. Like PCBs, concentrations in residential, agricultural, and forested areas were nearly identical among land types and were similar between baseflows and storm flows. PBDE concentrations in commercial/industrial areas increased by an order of magnitude during storms, although a seasonal first-flush episode was not evident in commercial/ industrial areas whereas it was seen in other land covers. Normalized to land cover area, commercial/industrial area loads were 10- to 20-fold above other areas. However, absolute loads from forests were larger overall due to the large area of forested land cover.

Based on marine water sampling and subsequent calculation of loads at the ocean boundary, there appears to be an annual net export of 11 kg total PBDEs at the ocean boundary using estimates derived from the median concentrations, while loads calculated from the 25th percentile and the 75th percentile concentrations appear to show net PBDE imports of 41 and 240 kg/yr, respectively. Total PBDE concentrations (25th-75th percentile) in the incoming marine waters are 760 – 1,600 pg/l, and total PBDE concentrations in the outgoing marine waters are 603 – 1,071 pg/l. PBDE inflow is estimated to be 440 – 940 kg/yr, and PBDE outflow is estimated to be 400 – 700 kg/yr.

The net sum of PBDE loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25th, median (50th), and 75th percentile values is 69, 26, and 290 kg/yr, respectively. Under all of these estimates, there is a net PBDE load to Puget Sound.

PBDE loading from pathways that may represent *partially controllable* sources of COCs is shown as ranges (25th - 75th percentiles) and median for each pathway in Figure 26. Each pathway represented as a contribution to the total load is displayed in Figure 27.

Total PBDE loading from the major pathways assessed is 28 – 54 kg/yr. Atmospheric deposition accounts for the largest pathway (44 – 56%), followed by POTWs (25 – 38%) and surface runoff (18%). This pattern of source contribution is notably different than other COCs which typically have the largest, and often the majority, load input from surface runoff.

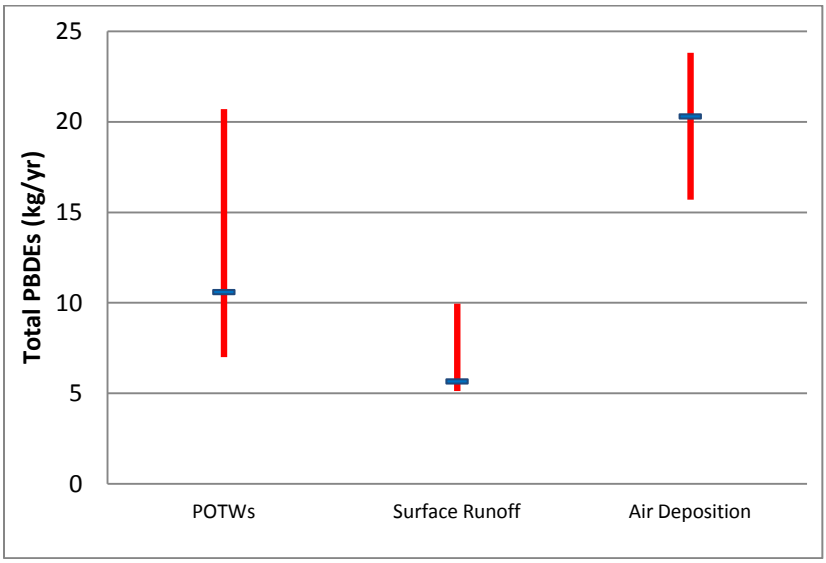


Figure 26. Range (25th - 75th percentiles) and Median Total PBDE Loads to Puget Sound from Each Major Delivery Pathway.

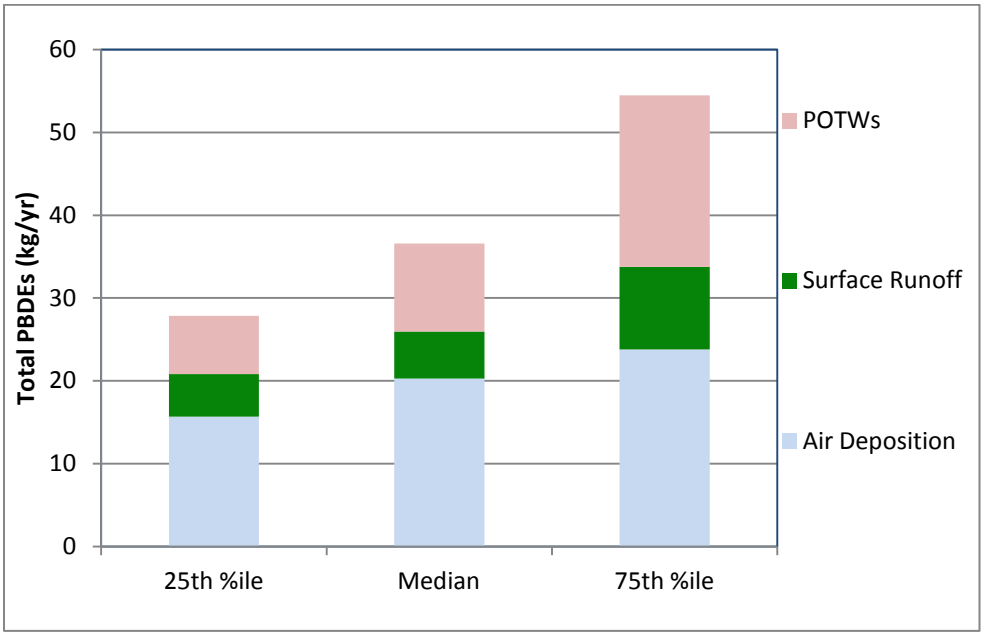


Figure 27. Total PBDE Loads to Puget Sound as the Sum of Major Delivery Pathways.

Relationship Between Sources and Pathways/Loading

The conceptual model of PBDE transport and fate following release suggests that much of the PBDEs will be initially released to air and atmospheric transport will deliver comparatively high loads directly to Puget Sound. PBDEs deposited on land will also be mobilized during storm events and delivered to surface waters, but in quantities lower than for direct atmospheric deposition. Some of the PBDEs deposited to land are also likely to be transported in storm sewers and delivered directly to Puget Sound or indirectly by way of POTWs.

The inventory of primary sources (Ecology, 2011) suggests that PBDEs are released in the highest quantities in commercial areas compared to other land covers; this notion is supported by results of the loading studies. The air deposition study found higher PBDE fluxes in the high-density urban (Tacoma) location relative to other sites. The surface runoff study found much higher PBDE concentrations in commercial/industrial areas compared to other land covers. The potential for large releases from indoor air is also supported by seasonal PBDE fluxes in atmospheric deposition; the authors found higher flux rates during the warm season and surmised that they may have been due to increased indoor air release (i.e. windows left open and increased ventilation).

PBDEs are released from indoor consumer and office products, become attached to dust particles, and are subsequently delivered to the sanitary sewer through washing machine rinse water during the washing of fabrics with the attached PBDE-enriched dust, and rinsing other materials with attached dust particles. This appears to be a reasonable pathway for PBDE release and transport, although the estimated quantity delivered to POTWs via this route is highly uncertain. However, the high frequency of detection and relative large loads from POTWs lends support to this concept. Furthermore, due to the nature of these PBDE sources, washing machine rinse water concentrations would be expected to remain steady throughout the year. This appears to be consistent with PBDE loads from POTWs which did not vary appreciably between wet and dry seasons.

Hazard Evaluation

PBDE data are available for surface waters and sediment, yet the paucity of documented effects, standards, or guidelines for PBDEs consistent with those used for other COCs preclude the assignment of a Priority 1 or Priority 2 level of concern (Table 16).

Table 16. Summary of Hazard Evaluation for PBDEs.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.* or *Acute WQC* or *Chronic WQC***	**Level of Concern**
	Freshwater	255	59%	INS	U
	Nearshore Marine	0	--	INS	U
	Offshore Marine	126	20%	INS	U
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. > *SQS***	**Level of Concern**
	Freshwater	77	97%	INS	U
	Nearshore Marine	1	100%	INS	U
	Offshore Marine	45	98%	INS	U
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.***	**Level of Concern**
	Not Analyzed				
Wildlife		**Total N**	**FOD**	**Daily Dose > *10% of Lowest Effects Dose***	**Level of Concern**
	Not Analyzed				
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. > *NTR Criterion***	**Level of Concern**
	Not Analyzed				

FOD=Frequency of detection
INS= Insufficient effects data available for comparison to observed data
U=Unknown level of concern due to lack of sufficient data for adequate comparison

Regionally Important Biological-Effects Data

PBDEs were detected in outmigrant Chinook salmon tissue and their stomach contents from four sites in Puget Sound (Sloan et al., 2010). Levels in wild outmigrant juveniles were higher than in hatchery fish, ranging from 67 to 13,000 ug/kg lipid, generally comparable to those measured in the Lower Columbia River and Estuary. Sloan et al. (2010) conclude that PBDEs may be contributing to reduced health and fitness in outmigrant juvenile Chinook salmon. PBDEs were detected in adult Chinook salmon returning to the Duwamish River and were not detected in adult Chinook returning to the Johnstone Strait, Lower Fraser River, or Deschutes River (Cullon et al., 2009).

Lema et al. (2008) demonstrated that dietary exposures to certain PBDEs by adult fathead minnows can alter thyroid status and thyroid hormone-regulated gene transcription. Arkoosh et al. (2010) found that juvenile Chinook salmon exposed to moderate doses of PBDEs in their diet may be at increased risk of disease relative to those exposed to higher or lower doses of

PBDEs in their diet. PBDE levels were found to be about four to five times higher in a mixture of fishes designed to represent the diet of Puget Sound harbor seals than in a similar mixture of fish designed to represent the diet of harbor seals from the Strait of Georgia (Cullon et al., 2005).

Very few studies have been conducted examining effects of PBDEs on birds. The studies reviewed indicate that PBDEs impact the reproduction and endocrine system similarly to PCBs. Exposure to BDE-71 for 75 days adversely impacted courtship and mating behavior of American kestrels (*Falco sparverius*)(Fernie et al., 2008). These birds also displayed significant delays in clutch initiation and smaller eggs (Fernie et al., 2009). Eggshell thinning and reduced hatching success also resulted. A study of species sensitivity to PBDEs (PBDE-71) observed that pentabrominated diphenyl ether (Penta BDE) exposure to eggs at 0.01 to 20 mg/kg caused decreased pipping and hatching success in American kestrels but not chickens (*Gallus gallus*) or Mallard ducks (*Anas platyrhynchus*)(McKernan et al., 2009). Species sensitivity was concluded to be Mallard ducks <chickens <American kestrels.

Total PBDE concentrations in osprey eggs and nestling plasma are significantly lower in the Lower Duwamish River (eggs: 321 ug/kg ww; plasma: 6 ug/kg ww) compared to those from the upper Willamette River (eggs: 897 pb ww; plasma: 22 ppb ww) (Johnson et al., 2009). Total PBDE concentrations in the osprey eggs did not change significantly between 2003 and 2007. Reproductive failure was observed in four of nine nests in the Lower Duwamish area. A small dataset from this study suggests that some nestlings may have experienced immunosuppression. However, the results were inconclusive due to the small sample size.

Compared to birds, a larger but still limited number of publications exist on the effects of PBDEs in mammals. Rodent exposure studies have demonstrated thyroid hormone disruption (Hallgren et al., 2001; Zhou et al., 2002) and developmental neurotoxic and behavioral effects (Ericksson et al., 2001; Viberg et al., 2003a; Viberg et al., 2003b). A study of grey seal pups and juveniles observed a relationship between circulating thyroid hormones, transport proteins, and PBDE uptake (Hall et al., 2003).

Similar to PCBs, there is evidence of bioaccumulation of PBDEs in marine mammals at high concentrations in blubber. However, absolute concentrations of total PBDEs appear to be lower than total PCBs. Cullon et al. (2005) measured PBDE concentrations five times higher in harbor seal prey from Puget Sound than the Strait of Georgia, but the mean PBDE concentration was five times lower than that measured for PCBs. Krahn et al. (2009) and Rayne et al. (2004) found the same pattern of killer whale blubber concentrations as found for PCBs in males, mothers, and calves. Krahn et al. (2009) measured total PBDE concentrations ranged from 680 to 15,000 ug/kg lipid. Mean PBDE concentrations in northern male killer whale blubber have been found to be significantly lower (203 ug/kg lipid) than those of southern resident (942 ug/kg lipid) and transient males (1,015 ug/kg lipid).

Although a quantitative effects assessment was not conducted for PBDE exposure to marine mammals, published research demonstrates that PBDEs are bioaccumulating to high concentrations in Puget Sound killer whales. This coupled with the growing evidence that PBDE exposure can cause thyroid and developmental effects in mammals strongly suggest that PBDEs are an important contaminant to monitor.

PCDD/Fs

Polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs; a.k.a. dioxins) are a class of persistent bioaccumulative compounds ubiquitous in the environment at low concentrations. There are 210 individual PCDD/F congeners (75 PCDDs and 135 PCDFs), but only the 17 congeners (seven PCDD and ten PCDF) with chlorines occupying the 2,3,7, and 8 positions on the molecules are considered toxic.

PCDD/Fs are generally found in mixtures, with the toxicity of the mixture translated to that of TCDD (and more recently, PeCDD) which is the most toxic congener. Each of the 17 PCDD/Fs are assigned a toxicity factor relative to that of TCDD and PeCDD, and the toxicity factor multiplied by the congener concentration is termed the toxic equivalent (TEQ) when the congeners are summed. The TEQ of an environmental sample is generally useful shorthand for assessing comparisons to regulatory thresholds and for assessing risks. Sampling and source data are commonly expressed as TEQs, and this convention has been adopted in the present document.

Unlike many other environmental contaminants, PCDD/Fs are not produced intentionally, but instead are formed and released as by-products of industrial production and combustion of certain chlorinated materials. As a result, industrial and combustion sources are responsible for the majority of PCDD/F releases. In the Puget Sound region, the major historical sources of PCDD/Fs – use of elemental chlorine in pulp bleaching, pentachlorophenol wood treatment operations, and combustion of saltwater-infused hog fuel – are all but gone (Yake et al., 1998). However, PCDD/Fs are extremely persistent in the environment, particularly at sites where these activities historically occurred.

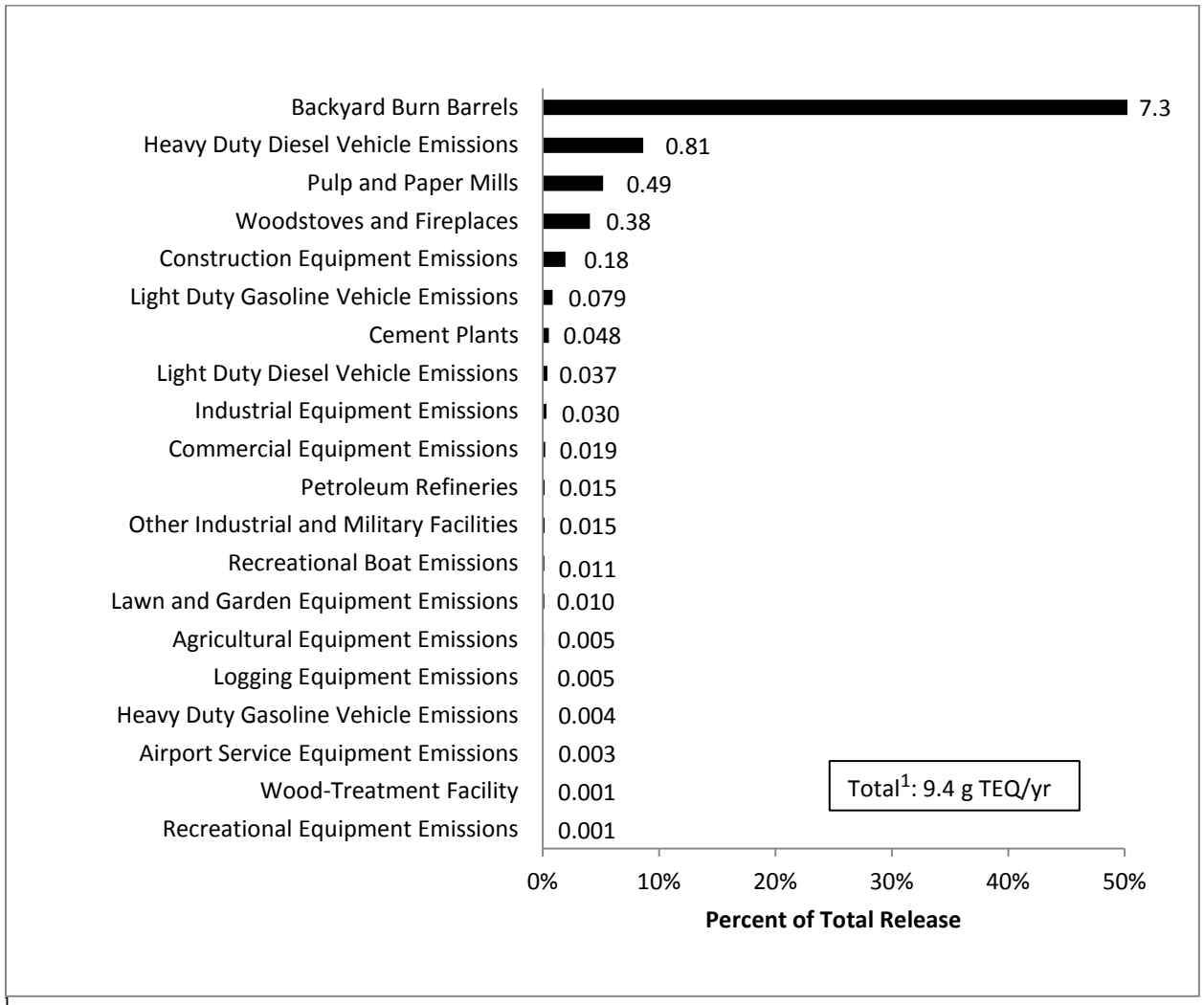
One other characteristic also distinguishes PCDD/Fs from most other environmental toxicants: their analysis requires expensive analytical techniques and is performed by only a limited number of commercial laboratories. As a result, they are commonly excluded from screening level investigations and are generally analyzed only if they are the focus of an environmental investigation. Since environmental PCDD/F data are not as prevalent as for other chemicals, there are fewer data on sources of their release to the environment.

PCDD/Fs are not typically detectable in ambient waters using conventional sampling and analytical techniques, even at detection limits in the low parts per quadrillion (pg/l) range. PCDD/Fs in sediments and biota are more typically found at low parts per trillion (ng/kg) levels, depending on the specific compound, organic carbon content of sediment, and lipid content in tissue.

Major Releases from Primary Sources

Annual PCDD/F release from the 20 sources assessed totaled approximately 9 g TEQ/yr (Figure 28). The largest single source is backyard burn barrels, accounting for nearly three-quarters of the PCDD/F release to the Puget Sound basin.

Combustion emissions to air account for 97% of the PCDD/F release. The remaining 3% of the release is to water, primarily from pulp and paper mills. This represents a substantial change from two to three decades previous when PCDD/F discharges to water from pulp mills represented the bulk of all releases to Washington State (EPA, 1991).



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

Figure 28. Total PCDD/F Release in the Puget Sound Basin (values shown are g TEQ/yr).

Major Delivery Pathways and Loading

None of the Phase 3 sampling studies included analysis of PCDD/Fs. The groundwater loading analysis included an estimate of PCDD/Fs from values reported in the literature. However, the groundwater data only included PCDD/Fs reported as total TCDD or total TCDF, with no indication of whether these are 2,3,7,8-substituted congeners.

Surface runoff load estimates were estimated during PSTLA Phase 2 based on a variety of U.S. and European runoff data, most of which were from urban areas (Envirovision et al., 2008a). The best estimates of the medians from these concentrations were 10 pg/l for commercial/ industrial areas, 5 pg/l for residential and agricultural areas, and 0.1 pg/l for forested area. When applied to the hydrologic model used in Phase 2, loads were estimated to be 6.1 – 103 g TEQ/yr (25^{th} – 75^{th} percentiles), with a median estimate of 25 g TEQ/yr. Most of the PCDD/F load (59%) was from residential areas, with the smallest percentage (6%) from commercial/industrial areas.

Estimates of PCDD/F loads to marine waters from atmospheric deposition were derived in Phase 1 from fluxes reported in Europe and adjusted to the Puget Sound region based on relative differences in air concentration (Hart Crowser at al., 2007). Fluxes of 0.1 – 10 pg/m^2/day (low to high end of range, 1 pg/m^2/day as medium value) were used to estimate the aerially deposited loads (0.31 – 31 g TEQ/yr, medium estimate of 3.1 g TEQ/yr).

Both the atmospheric deposition (Phase 1) and surface runoff (Phase 2) load estimates for PCDD/Fs have a large degree of associated uncertainty. The paucity of PCDD/F stream and air deposition data from the Puget Sound region makes gauging the representativeness of other data difficult. Therefore, these estimates should not be treated with the same level of confidence as those derived from Phase 3 sampling efforts.

Relationship Between Sources and Pathways/Loading

The conceptual model of PCDD/F transport and fate following release is complicated by the lack of sampling data. Based on the inventoried sources (Ecology, 2011), release to air is likely to be the primary initial pathway for PCDD/Fs. The dominance of combustion as a formation mechanism suggests that much of the PCDD/F release may be associated with airborne particles.

There are no reliable data to assess the degree of deposition for particle-bound PCDD/Fs in the Puget Sound basin, but near-field deposition may be expected for much of the emitted PCDD/Fs since the bulk of emission is from numerous non-point sources as opposed to high stacks designed to disperse emissions. Based on the primary sources in the basin, the release of PCDD/Fs is likely to occur in a mix of urban and rural locations, and may occur at a distance from the Puget Sound marine waters. Delivery to Puget Sound is therefore likely to occur through secondary pathways (e.g. surface runoff, POTWs) as well as direct deposition and would not be expected to be dominated by a single delivery mechanism.

Hazard Evaluation

The hazard evaluation indicates that PCDD/Fs are a Priority 1 level of concern for a wildlife mammalian species (river otter) and human health due to concentrations found in both the freshwater and marine aquatic environments (Table 17). However, there are insufficient observed or effects data to adequately evaluate PCDD/Fs in surface waters, sediments, or for tissue residue effects.

In fresh surface waters only five observed results are available to compare dioxin concentrations with effects data. Observed concentrations of 1,2,3,7,8-PeCDD and 1,2,3,4,7,8-HxCDD are one to two orders of magnitude below the lowest effects concentrations, and 2,3,7,8-TCDD is three orders of magnitude below levels where effects are documented. There are no observed environmental data available for PCDD/Fs in the marine water column.

In freshwater sediments, the median PCDD/F concentration (expressed as TEQ) is approximately equal to the lowest available guideline, the Canadian TEL. However, the interquartile range of concentrations spans four orders of magnitude, and at least 25% of the observed TEQs are well above the Canadian PEL where adverse effects are expected to frequently occur.

Median PCDD/F TEQ concentrations in marine sediments are similar to freshwater sediments, but the distribution of concentrations occupies a much narrower range. For both nearshore and offshore sediments, the median values exceed the Canadian TEL, but only 5 – 10% of the observed concentrations exceed the Canadian PEL.

There are few effects data to assess the potential concern of PCDD/Fs associated with tissue residue. For freshwater, there were sufficient effects data available for decapods, but no observed data. No tissue residue effects data were available for marine waters.

For the two bird species evaluated – great blue heron and osprey – only two daily effects doses of PCDD/Fs (as TEQ) were calculated: one for reproductive effects and one for mortality. Neither of the species are exposed to these doses, but heron are exposed to doses that are one-fifth to one-half of the lowest (reproductive) dose. Osprey are exposed to doses three orders of magnitude below the lowest effects dose. However, the low number of dose effects was not deemed sufficient for an adequate comparison with calculated doses (Appendix D-1).

Based on environmental concentrations of PCDD/Fs, river otter receive approximately five to ten times the daily doses (as TEQs) where reproductive effects have been documented, and within 10% of the lowest dose associated with mortality. Harbor seals are exposed to much lower daily PCDD/F doses; less than 10% of the lowest effect dose.

Concentrations of 2,3,7,8-TCDD in freshwater fish, bivalves, and other invertebrates were compared to the NTR criterion. Median concentrations for fish and tissues from other invertebrates were above the NTR criterion in both fresh and marine waters, although no detectable concentrations are available for 2,3,7,8-TCDD in marine nearshore fish. At least 90% of the 2,3,7,8-TCDD concentrations in freshwater bivalves are above the criterion, but in marine nearshore areas, less than 25% of the concentrations are above the criterion.

Table 17. Summary of Hazard Evaluation for PCDD/Fs.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.* or *Acute WQC* or *Chronic WQC***	**Level of Concern**
	Freshwater	7	>14%	INS[a]	U
	Nearshore Marine	0	--	INS[a]	U
	Offshore Marine	0	--	INS[a]	U
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. > *SQS***	**Level of Concern**
	Freshwater	36	89%	INS[b]	U
	Nearshore Marine	219	>99%	INS[b]	U
	Offshore Marine	106	>99%	INS[b]	U
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.***	**Level of Concern**
	Freshwater	c	57%	INS[b]	U
	Nearshore Marine	d	≥97%[e]	INS[b]	U
	Offshore Marine	f	≥86%	INS[b]	U
Wildlife		**Total N**	**FOD**	**Daily Dose > *10% of Lowest Effects Dose***	**Level of Concern**
	Great Blue Heron (FW)	--	--	INS[b]	U
	Osprey (SW)	--	--	INS[b]	U
	River Otter (FW)	--	--	Yes	Priority 1
	Harbor Seal (SW)	--	--	No	Priority 2
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. > *NTR Criterion***	**Level of Concern**
	Freshwater	g	≥25%	Yes	Priority 1
	Nearshore Marine	h	>18%[e]	Yes	Priority 1
	Offshore Marine	i	≥5%	Yes	Priority 1

Yellow highlight indicates Priority 1 Level of Concern
FOD=Frequency of detection
INS[a]=Insufficient observed data available for comparison to effects data
U=Unknown level of concern due to lack of sufficient data for adequate comparison
INS[b]=Insufficient effects data available for comparison to observed data
[c] N = 175 for non-decapod invertebrates. No observed data available for other organism types
[d] N = 76 for non-decapod invertebrates, N = 13 for decapods, and N = 1 for fish tissue.
[e] FOD=0% for fish tissue
[f] N = 28 for non-decapod invertebrates, N = 9 for decapods, and N = 10 for fish tissue.
[g] N range is 35 – 72 and varies for organism type and tissue type
[h] N range is 8 – 129 and varies for organism type and tissue type
[i] N range is 32 – 53 and varies for organism type and tissue type

Regionally Important Biological-Effects Data

Studies in Puget Sound of harbor seal and southern resident killer whale prey items have higher PCDD/Fs compared to the same prey items from the Strait of Georgia and British Columbia coast (Cullon et al., 2005, Cullon et al., 2009). Harbor seal prey were three to four times higher on a lipid basis than prey from the Strait of Georgia (Cullon et al., 2005). However, Ross et al. (2000) found that PCDD/F concentrations in killer whale blubber were much lower than PCBs, and there were no differences between whales from the northern and southern resident and transient communities. This was suspected to be due to metabolic removal of dioxins and furans.

DDT

The chlorinated insecticide dichlorodiphenyltrichloroethane (DDT) was banned in 1972 from general agricultural uses in the U.S. following concerns over its effects on wildlife and human health. However, potentially harmful levels are still found in the environment, together with its major breakdown and metabolic products dichlorodiphenyldichloroethylene (DDE) and dichlorodiphenyldichloroethane (DDD) due to their persistence and tendency to accumulate in fish and wildlife. In western Washington, DDT is only rarely detected in water, even at a typical analytical detection limit of about 1 ng/l (parts per quadrillion), but is nearly always detected in fish due to its extremely high bioaccumulation potential.

Major Releases from Primary Sources

DDT sources were not inventoried in the analysis of primary sources since it was banned nearly four decades ago and DDT had no uses other than as an agricultural insecticide and limited use as an urban pesticide. Due to its persistence, however, it continues to be found in environmental media, particularly soils and sediments, and is delivered to Puget Sound when these soil and sediment particles become entrained in surface water runoff.

Major Delivery Pathways and Loading

Data on major delivery pathways for DDT loading were obtained from PSTLA loading studies and are included in Table 18. No effort was made to assess DDT loads from direct deposition of atmospheric DDT to Puget Sound during Phase 3 sampling. As mentioned previously, loading through direct groundwater discharge was estimated from literature values, whereas loadings through other pathways were estimated from field studies specifically designed to estimate loads.

The estimated range of groundwater DDT loads is large (0.2 – 7.3 kg/yr) due primarily to differences in flows used to establish the possible range of loads. DDT load estimates in groundwater should be used with caution since they are based primarily on non-detect data and numerous assumptions, including unconfirmed assumptions about DDT mobility in groundwater.

DDT compounds were analyzed in all ten POTWs during winter and summer sampling events, yet no concentrations were detectable. Reporting limits for DDT compounds in POTW samples were generally 2 – 3 ng/l.

Table 18. Total DDT Loads (kg/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater (a)	0.2	3.8	7.3
Air Deposition	NA	NA	NA
Surface Runoff (b)	2.2	23.9	25.1
POTWs	ND	ND	ND
Ocean Exchange	NC	NC	NC

(a) Lowest and highest estimated loads
(b) Detected in commercial/industrial areas only during baseflows and in commercial, agricultural, and forest areas only during storm flows
NA=not analyzed
ND=not detected
NC=not calculated due to insufficient data

Surface runoff loads for DDT are estimated to be approximately 2.2 – 25 kg/yr for the entire Puget Sound basin based on the surface runoff study. DDT compounds were detected in all land use types during storm events except residential covers, and >99% of the loads occurred during storm flows. Loads calculated during storms generally mirrored the amount of land area for each land use cover, a somewhat confounding result since agricultural areas would be expected to have larger loads relative to land area.

A single commercial/industrial area had the only detection of DDT compounds during baseflows, and only in very low concentrations. However, this particular location had DDT concentrations an order of magnitude above all other locations – regardless of land cover – during storm flows. For each land cover, the detection frequency for DDT compounds was low and never exceeded 50%, and therefore the calculated median loads were driven by non-detected values.

DDT loads were not calculated for the ocean exchange of marine waters due to the low frequency of detection. DDT compounds were detected in only three samples at locations in the northern boundary waters, all at low concentrations (<0.4 ng/l).

The net sum of DDT loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25th, median (50th), and 75th percentile values is 2.5, 28, and 32 kg/yr, respectively. Under all of these estimates, there is a net DDT load to Puget Sound.

DDT loading from pathways that may represent *partially controllable* sources of COCs is shown as ranges (25th - 75th percentiles) and medians for each pathway in Figure 29. Each pathway represented as a contribution to the total load is displayed in Figure 30.

Total DDT loading measured from the major pathways is 2.5 – 32 kg/yr, all contributed through surface runoff and groundwater. Surface runoff accounts for the largest portion of DDT loading (77 – 88%), with the remaining amount (12 – 23%) contributed by the groundwater pathway.

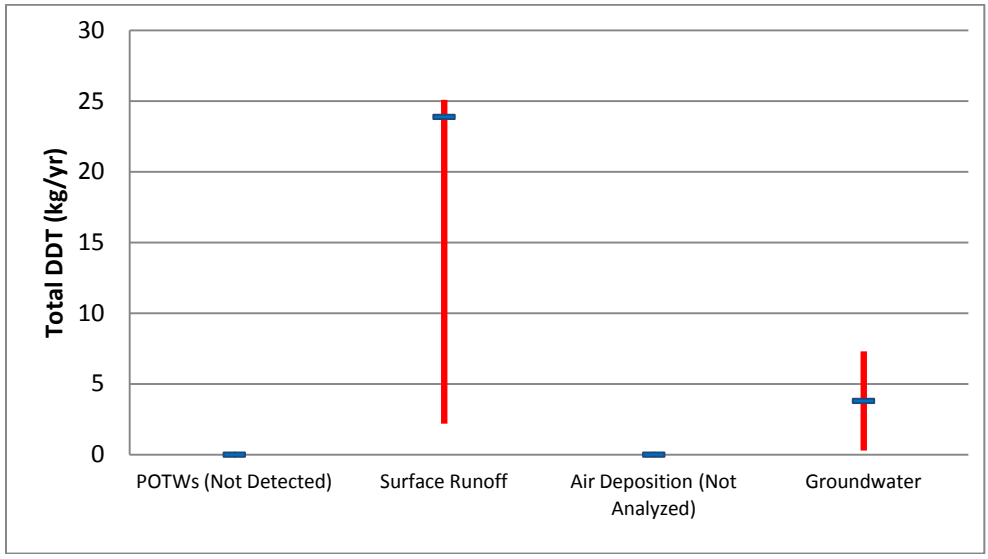


Figure 29. Range (25th - 75th percentiles) and Median Total DDT Loads to Puget Sound from Each Major Delivery Pathway.

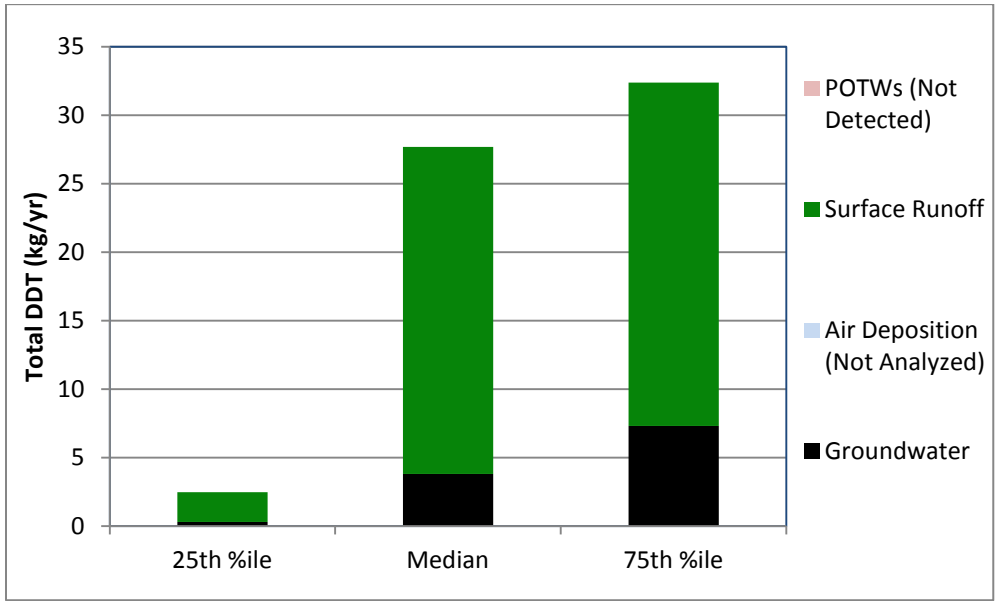


Figure 30. Total DDT Loads to Puget Sound as the Sum of Major Delivery Pathways.

Relationship Between Sources and Pathways/Loading

The conceptual model of DDT transport and fate following release appears to be rather simple compared with other COCs. Releases from primary sources likely occurred during previous decades, and soils and sediments likely serve as the major reservoirs for DDT remaining in the environment. Soil and sediment-bound PCBs are mobilized during storm flows and delivered toward Puget Sound in surface runoff. Comparatively low DDT occurrence and concentrations in agricultural soils suggest that that the bulk of DDT mobilization from agricultural soils, where the majority of DDT was likely to have been originally released, has already occurred and the large DDT reservoirs may have advanced downstream and may largely reside in Puget Sound sediments. Conversion of agricultural lands to commercial or other land uses may explain high DDT concentrations in other land uses, particularly if soils are disturbed.

It is possible that DDT is also delivered to Puget Sound through direct atmospheric deposition to marine waters, or that some of the DDT in surface waters is due to aerially deposited DDT entrained in storm runoff. Loads of direct atmospheric deposition of DDT were calculated during the Phase 1 effort, and were estimated to be 1.2 – 31 kg/yr (low to high end of range, 6.2 kg/yr as medium value) based on fluxes reported in the eastern and midwestern U.S. (0.4 – 10 ng/m^2/day; 2 pg/m^2/day as medium value).

Hazard Evaluation

The hazard evaluation indicates that DDT compounds are a Priority 1 level of concern for a range of media and receptors in both the freshwater and marine aquatic environments (Table 19).

In fresh surface waters, the 90th percentile of total DDT (sum of DDT, DDE, and DDD) concentrations (approx. 1 ug/l) is slightly higher than the acute water quality criterion, and at least 95% of the observed concentrations are above the chronic water quality criterion. More than 90% of the values from the fairly extensive dataset on DDT effects are above the 90th percentile value for observed concentrations. There are not sufficient data to evaluate DDT in marine surface waters.

The chronic freshwater water quality criterion is three orders of magnitude lower than the acute water quality criterion. This difference stems from the high bioaccumulative potential of DDT, on which the chronic criterion is indirectly based (reproductive effects in the piscivorous brown pelican), whereas the acute criterion is based on LC_{50} concentrations for numerous fish and invertebrate species (EPA, 1980).

There are no SQS for freshwater or marine sediments to compare to DDT concentrations. However, median DDT concentrations exceed the Canadian TEL, and at least >75% of the concentrations exceed the consensus-based TEC, although concentrations are well below the Canadian PEL and consensus-based PEC.

In the marine environment, median DDT concentrations in both nearshore and offshore sediments are similar to the Canadian TEL. However, a number of the nearshore sediments have much higher concentrations than those from offshore locations, and at least 10% of the DDT concentrations exceed the Canadian PEL as well as the LAET and the 2LAET (2x the LAET).

Table 19. Summary of Hazard Evaluation for DDT.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.* or *Acute WQC* or *Chronic WQC***	**Level of Concern**
	Freshwater	2,179	4%	Yes	Priority 1
	Nearshore Marine	11	0	INS[a]	U
	Offshore Marine	0	--	INS[a]	U
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. > *SQS***	**Level of Concern**
	Freshwater	365	30%	INS[b]	U
	Nearshore Marine	350	41%	INS[b]	U
	Offshore Marine	457	25%	INS[b]	U
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.***	**Level of Concern**
	Freshwater	c	≥92%	No	Priority 2
	Nearshore Marine	d	≥31%	INS[b]	U
	Offshore Marine	f	≥18%	INS[b]	U
Wildlife		**Total N**	**FOD**	**Daily Dose > *10% of Lowest Effects Dose***	**Level of Concern**
	Great Blue Heron (FW)	--	--	Yes	Priority 1
	Osprey (SW)	--	--	Yes	Priority 1
	River Otter (FW)	--	--	No	Priority 2
	Harbor Seal (SW)	--	--	No	Priority 2
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. > *NTR Criterion***	**Level of Concern**
	Freshwater	g	>68%	Yes	Priority 1
	Nearshore Marine	h	≥20%	Yes	Priority 1
	Offshore Marine	i	≥6%[j]	No	Priority 2

Yellow highlight indicates Priority 1 Level of Concern
FOD=Frequency of detection
INS[a]=Insufficient observed data available for comparison to effects data
U=Unknown level of concern due to lack of sufficient data for adequate comparison
INS[b]=Insufficient effects data available for comparison to observed data
[c] N = 83 for non-decapod invertebrates and 139 for fish tissue
[e] N = 84 for non-decapod invertebrates, N = 20 for decapods, and N = 131 for fish tissue.
[f] N = 33 for non-decapod invertebrates, N = 5 for decapods, and N = 543 for fish tissue.
[g] N range is 56 – 634 and varies for organism type, tissue type, and DDT compound
[h] N range is 48 – 491 and varies for organism type, tissue type, and DDT compound
[1] N range is 33 – 1,036 and varies for organism type, tissue type, and DDT compound
[j] FOD=0% for 4,4'-DDD in bivalve tissue

Observed DDT concentrations in freshwater invertebrates and fish are well below concentrations where effects are documented. Insufficient effects data were available to adequately compare with observed concentrations from invertebrates or fish from the marine nearshore and offshore environments.

Calculation of DDT doses for the two bird and two mammalian species evaluated indicate that only great blue heron are exposed to daily DDT doses above a concentration where at least one effect (reproductive) has been documented. However, for osprey, the calculated daily DDT dose is only slightly (<50%) below this lowest effects dose. For both of the mammalian species evaluated – river otter for freshwaters and harbor seal for marine waters – calculated daily DDT doses are more than an order of magnitude below doses where effects have been documented.

NTR criteria for DDT compounds are based on individual DDT compounds (4,4'-DDD, 4,4'-DDE, and 4,4'-DDT) rather than total DDT. For freshwater tissues, DDD and DDE 90th percentile concentrations exceed the NTR criterion for invertebrates other than bivalves. In nearshore marine waters, the observed 90th percentile DDT concentration exceeds the criterion for fish and invertebrates other than bivalves. None of the tissues have observed 90th percentile values exceeding the NTR criterion in offshore marine waters.

In general, the interquartile range of concentrations for all three compounds falls between the NTR criteria at the 17.5 g/d EPA recreational consumption rate and the 769 g/d Suquamish Tribal rate. This pattern is consistent across the observed concentrations in freshwater and in nearshore and offshore marine waters.

Regionally Important Biological-Effects Data

Total DDT (sum of DDT, DDE, and DDD) concentrations in stomach contents of outmigrant juvenile Chinook salmon were found to be elevated in fish from the Duwamish Estuary and Commencement Bay relative to the stomach content concentrations of fish from the Nisqually Estuary (Stein et al., 1995). Whole-body total DDT concentrations of juvenile Chinook salmon were found to be relatively high (over 1,000 ng/g lipid or 25 ng/g ww) in fish from the Nisqually, Duwamish, and Columbia River estuaries (Johnson et al., 2007).

Johnson et al. (2007) also found detectable levels of DDTs in stomach contents, with stomach content concentrations substantially higher in Columbia River and Grays Harbor juvenile Chinook than in Duwamish and Nisqually Estuary. They suggested that at the levels measured, DDTs are unlikely to cause adverse effect by themselves, but that they may contribute via additive or synergistic effects with other contaminants. Substantially higher levels of DDTs were found in adult Chinook salmon returning to the Duwamish River than in adult Chinook returning to Johnstone Strait, the Lower Fraser River, or the Deschutes River (Cullon et al., 2009).

An analysis of DDT concentrations in Pacific herring tissues found that concentrations from Puget Sound herring were 1.5 to 2.5 times higher than those from the Strait of Georgia (West et al., 2008), with Puget Sound concentrations ranging from 19 to 27 ng/g ww (240 to 330 ng/g lipid). Cullon et al. (2005) found similar levels of DDTs in a mixture of fishes designed to represent the diets of Puget Sound and Strait of Georgia harbor seals.

Grove and Henny (2008) did not find detections of DDT and only low detections of DDE (mean of 0.004-0.28 mg/kg ww) in river otter livers from Puget Sound. These DDE levels are much lower than those found in river otters living along the Columbia River (mean of 0.12-1.65 mg/kg ww). The prey items of harbor seals in Puget Sound are 1.6 times higher in total DDT (lipid weight) than those from the Strait of Georgia (Cullon et al., 2005).

Puget Sound Chinook, the major prey of southern resident killer whales, have higher body residues of DDTs and lower lipids compared to Chinook from the British Columbia coast (Cullon et al., 2009). Krahn et al. (2009) found the same pattern of killer-whale blubber concentrations as found for PCBs in males, mothers and calves; total DDT concentrations ranged from 1,000 to 160,000 ug/kg lipid.

PAHs

Polycyclic aromatic hydrocarbons (PAHs) are a class of compounds characterized by two or more fused aromatic rings composed of carbon and hydrogen. There are hundreds of such compounds, but most studies have focused on 16 compounds that were designated as "priority pollutants" in the federal Clean Water Act. These 16 compounds are:

Low Molecular Weight PAHs (LPAHs)

- Acenaphthene
- Acenaphthylene
- Anthracene
- Fluorene
- Naphthalene
- Phenanthrene

High Molecular Weight PAHs (HPAHs)

- Benzo(a)anthracene*
- Benzo(a)pyrene*
- Benzo(b)fluoranthene*
- Benzo(k)fluoranthene*
- Benzo(g,h,i)perylene
- Chrysene*
- Dibenzo(a,h)anthracene*
- Fluoranthene
- Indeno(1,2,3-c,d)pyrene*
- Pyrene

*Designated as probable human carcinogens by EPA (cPAHs)

LPAHs tend to be found at elevated concentrations in uncombusted fossil fuels, while HPAHs are formed during incomplete combustion of fossil fuels and other organic materials such as wood (PTI, 1991). However, source identification of PAHs found in the environment cannot

simply be distilled down to comparisons of LPAHs to HPAHs for a variety of reasons. LPAHs and HPAHs are not typically exclusive to particular sources; releases occur from diffuse sources, and degradation processes may alter PAH compositions following their release.

Most PAHs are found in mixtures, but biochemical processes and accumulation potentials, as well as toxicity of individual PAHs, may vary considerably. Creosote, a tarry substance formed as coke distillate, is used to preserve wood and has historically been a major source of PAHs in Puget Sound, particularly in areas with pole treating operations adjacent to marine waters.

In Puget Sound, PAHs are associated with liver disease and reproductive impairment of English sole, particularly in urban bays (PSAT, 2007). Concentrations in the water are difficult to measure, but limited data suggest that typical freshwater concentrations for total PAHs are in the 0.1 – 1.0 ug/l range (Appendix D-3), with marine water column concentrations slightly lower (Serdar, 2008). Total PAHs in freshwater and marine sediments are typically 100 – 1,000 ug/kg (dw) (Appendix D-4) although mean concentrations in urban bays may be up to ten-fold higher (Partridge et al., 2005).

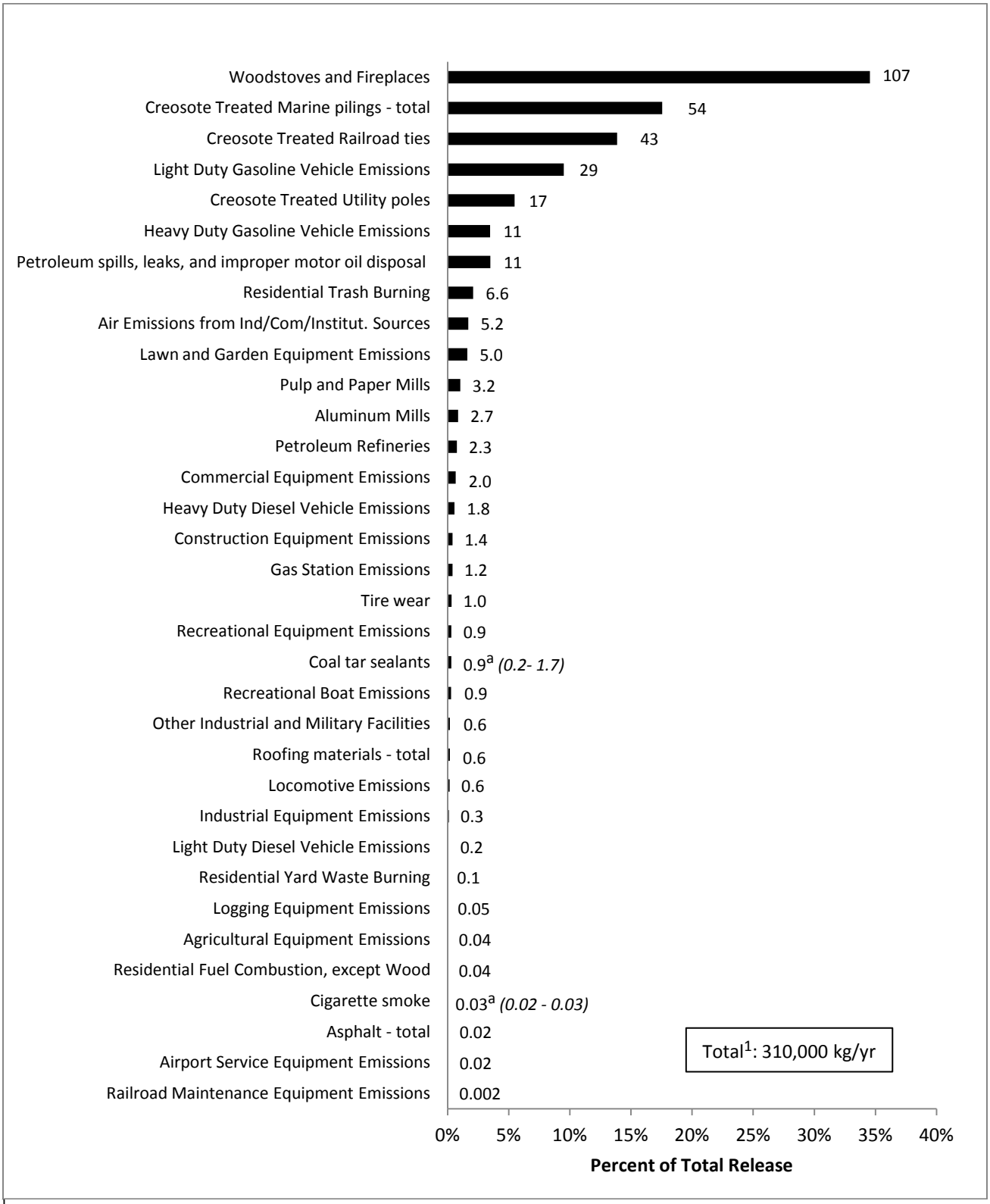
Major Releases from Primary Sources

Total annual PAH releases from sources inventoried total approximately 310,000 kg/yr in the Puget Sound basin (Figure 31). Generally speaking, the releases can be placed into two categories: combustion emissions and releases from creosote-treated wood.

Combustion releases account for most (55%) of the PAH release in the Puget Sound basin, with more than half of that amount due to woodstove and fireplace use, and smaller amounts due to residential trash burning and industrial emissions. The remainder of the combustion emissions is from petroleum fuel combustion, primarily due to gasoline use in vehicles.

Creosote-treated wood accounts for approximately one-third of the PAH release, with marine pilings (54 t/yr), railroad ties (43 t/yr), and utility poles (17 t/yr) representing the major sources.

Releases from large petroleum spills (≥ one gallon) and minor petroleum drips leaks, spillage, and improper disposal of motor oil account for PAH release of approximately 11 t/yr.



[1] Sum of best estimates. Best estimates are either the mean, mid-point, median, or most reasonable estimate for each source.

[a] Mid-point of range

Figure 31. Total PAH Release in the Puget Sound Basin (values shown are thousands kg/yr).

Major Delivery Pathways and Loading

Data on major delivery pathways for PAH loadings were obtained from PSTLA loading studies and are included in Table 20. As mentioned previously, loading through direct groundwater discharge was estimated from literature values, whereas loadings through other pathways were estimated from field studies specifically designed to estimate loads.

Table 20. PAH Loads (kg/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
	LPAH		
Groundwater (a)	7	159	311
Air Deposition	NA	NA	NA
Surface Runoff (b)	102	104	190
POTWs	3.27	8.05	34.9
Ocean Exchange	ND	ND	ND
	HPAH		
Groundwater (a)	6	124	244
Air Deposition	48.8	95.8	153
Surface Runoff (c)	25.2	36.2	50.7
POTWs	3.71	4.93	7.46
Ocean Exchange	ND	ND	ND
	cPAH		
Groundwater (a)	5	83	161
Air Deposition	20.8	43.2	69.8
Surface Runoff (d)	18.0	24.0	34.0
POTWs	NC	NC	NC
Ocean Exchange	ND	ND	ND
	Total PAH		
Groundwater (a)	13	284	555
Air Deposition (e)	48.8	95.8	153
Surface Runoff (b)	119	224	244
POTWs	7.55	18.5	45.8
Ocean Exchange	ND	ND	ND

(a) Lowest, highest, and mid-point of estimated loads
(b) Detected in commercial/industrial areas only during baseflows
(c) Detected in commercial areas only during baseflows and in commercial, residential, and agricultural areas only during storm flows
(d) Surface runoff loads based on storm flows only; not detected in any land covers during baseflows and in commercial, residential, and agricultural areas only during storm flows
(e) Total PAH based on HPAH only
NA=not analyzed
ND=not detected
NC=not calculated due to insufficient data

The estimated range of groundwater total PAH loads is large (13 – 555 kg/yr). The range in estimates is due to differences in flows used to establish the possible range of loads as well as differences in methods used to estimate representative PAH concentrations; most of the data were non-detected values. The groundwater loading calculations suggest that slightly over one-half of the groundwater loads of PAHs may be due to LPAH loads.

PAH load estimates in groundwater should be used with caution since they are based primarily on non-detect data and numerous assumptions, including unconfirmed assumptions about PAH mobility in groundwater. It is also notable that the "rule" used to sum individual PAHs to establish total PAHs (e.g. LPAH) resulted in higher values than results derived when applying summing rules used for other loading studies.

The deposition of atmospheric PAHs directly to the marine waters of Puget Sound is approximately 50 – 153 kg/yr, based on fluxes ranging from 0.012 – 0.69 ug/m^2/d. This estimate is based completely on HPAH; LPAHs were not analyzed. Median PAH fluxes were generally <0.05 ug/m^2/d, except for one location within a high-density urban (Tacoma) area including a nearby pulp mill, metal refiners, other industrial activities, and close to major roadways including interstate highways. PAH fluxes at this location were consistently an order of magnitude higher than other locations around the Puget Sound region, and were elevated six-fold above a nearby station that did not have the same air pollution influences in such close proximity. This pattern closely mirrored the deposition patterns of copper, lead, and zinc among the air sampling locations.

The total PAH load discharged from POTWs is estimated to be 6.6 – 46 kg/yr. Approximately one-half to three-quarters of the PAHs loaded from POTWs are LPAHs. Phase 1 estimates suggest that PAH loads from industrial wastewater are potentially substantial (2 – 87 kg/yr; Hart Crowser et al., 2007), although this is based on a limited dataset. The amount reported to be discharged to surface waters or transferred to POTWs from industries – as reported in the TRI – is also highly uncertain, with a high-end estimate of 90 kg PAH/yr (Ecology, 2011).

Surface water runoff loads for total PAH are estimated to be approximately 119 – 244 t/yr for the entire Puget Sound basin based on the surface runoff study, with LPAHs comprising approximately four-fifths of the load. The higher reported LPAH loads may be attributable in part to overall higher detection frequencies for LPAHs, although individual HPAH components were detected at generally higher frequencies than individual LPAH components.

In terms of land cover, PAHs are detected much more frequently in commercial/industrial areas (83%), about four times more frequently than in any other land cover. HPAH concentrations were highest in commercial areas, leading to loads calculated to be 16 – 32 kg/yr. Most of the remaining surface water PAH surface water loads were driven by a single LPAH constituent (phenanthrene) detected in 16% of the samples from forested lands, leading to a high calculated LPAH load for forests (102 – 190 kg/yr).

Most of the PAH detections occurred during storm events; PAHs were rarely detected during baseflows. Commercial/industrial areas were the only land covers where PAHs were detected during baseflow, and detection frequencies were so low (7% each for LPAH and HPAH) that baseflow loads for total PAH were ≤1 kg/yr under all scenarios used for load calculations.

PAHs were not detected in any of the marine water samples, and therefore no exchange could be calculated at the ocean boundary.

The net sum of total PAH loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25th, median (50th), and 75th percentile values is 190, 620, and 1,000 kg/yr, respectively. Under all of these estimates, there is a net PAH load to Puget Sound. However, it was not possible to assess the possibility of a net export out of Puget Sound since no PAHs were detectable in marine waters.

PAH loading from pathways that may represent *partially controllable* sources of COCs (i.e. all of the major pathways except ocean exchange) are shown as ranges (25th - 75th percentiles) and medians for each pathway in Figures 32-35. Each pathway represented as a contribution to the total load is displayed in Figures 36-39.

The surface runoff and groundwater are the largest loading pathways, accounting for a combined total of 70 – 82% of total PAH loads. Estimates at the median and 75th percentile levels suggest that approximately one-half of the total PAH loading occurs through groundwater. Air deposition accounts for 15 – 26% of the total PAH loads, while POTW loads account for 3 – 5%.

As noted previously, estimates of PAH loads from surface runoff and groundwater are driven by concentrations derived from non-detected results. Since these are the two largest reported loading pathways for PAHs, there is a large degree of uncertainty surrounding these load estimates. The authors of the present report did not attempt to modify or otherwise recalculate loads reported in the individual loading studies. However, readers are encouraged to review the loading studies to gain an understanding of the methodologies used to estimate loads where many of the sample results were below reporting limits.

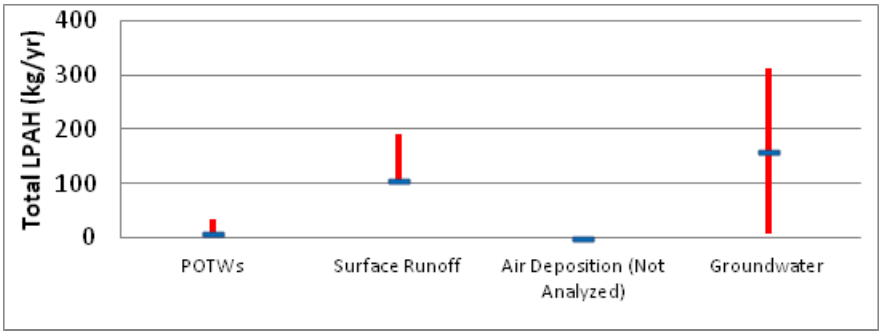


Figure 32. Range (25th - 75th percentiles) and Median LPAH Loads to Puget Sound from Each Major Delivery Pathway.

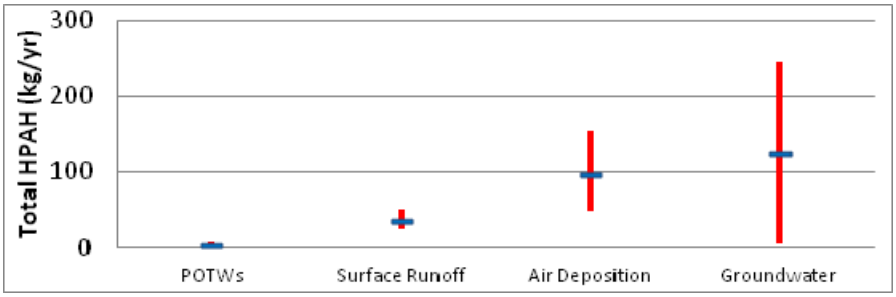


Figure 33. Range (25th - 75th percentiles) and Median HPAH Loads to Puget Sound from Each Major Delivery Pathway.

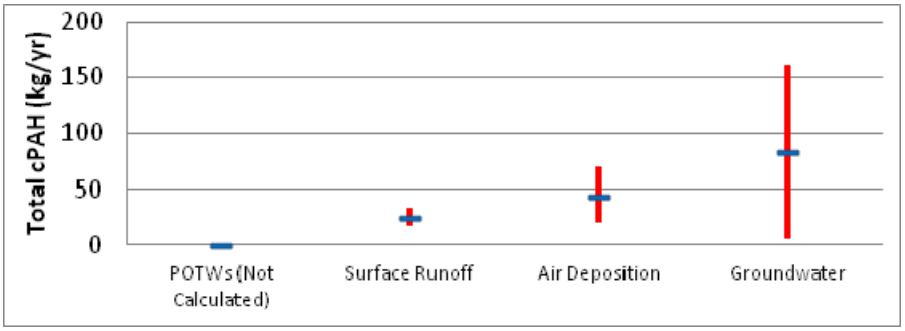


Figure 34. Range (25th - 75th percentiles) and Median cPAH Loads to Puget Sound from Each Major Delivery Pathway.

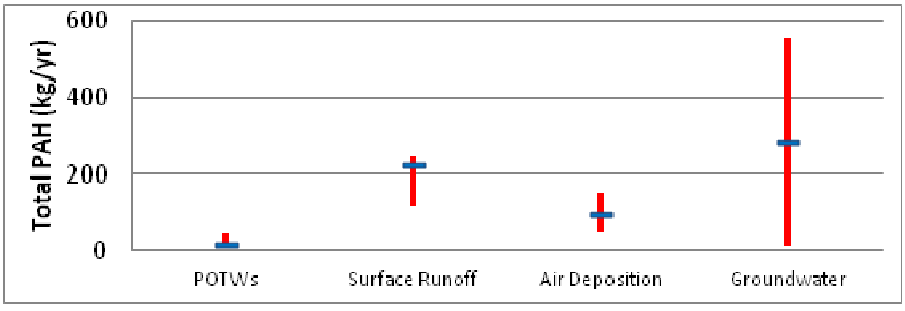


Figure 35. Range (25th - 75th percentiles) and Median Total PAH Loads to Puget Sound from Each Major Delivery Pathway.

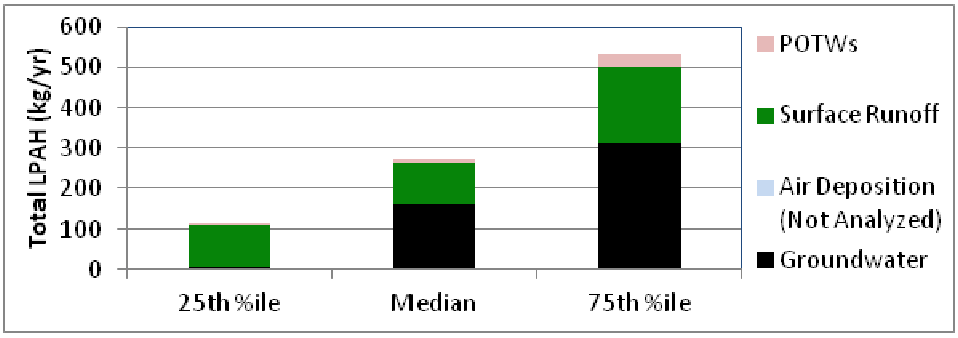


Figure 36. LPAH Loads to Puget Sound as the Sum of Major Delivery Pathways.

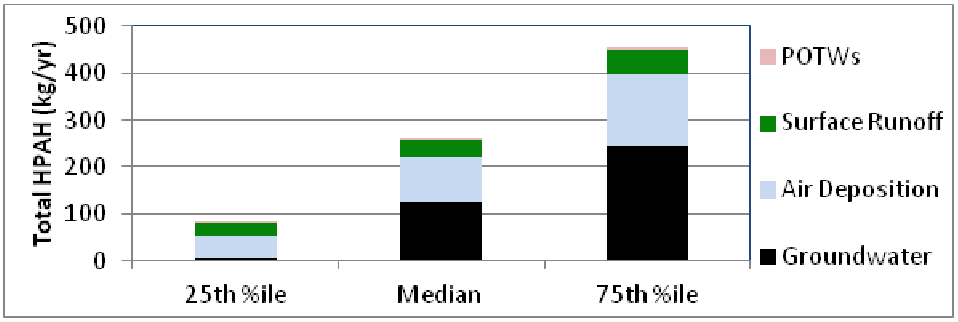


Figure 37. HPAH Loads to Puget Sound as the Sum of Major Delivery Pathways.

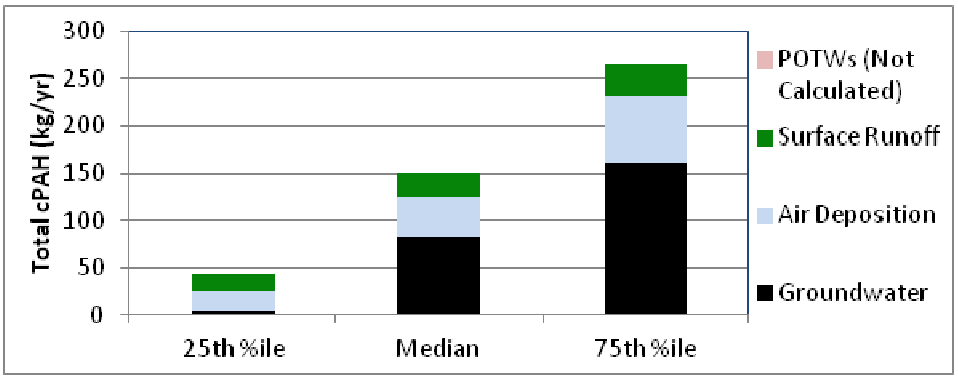


Figure 38. cPAH Loads to Puget Sound as the Sum of Major Delivery Pathways.

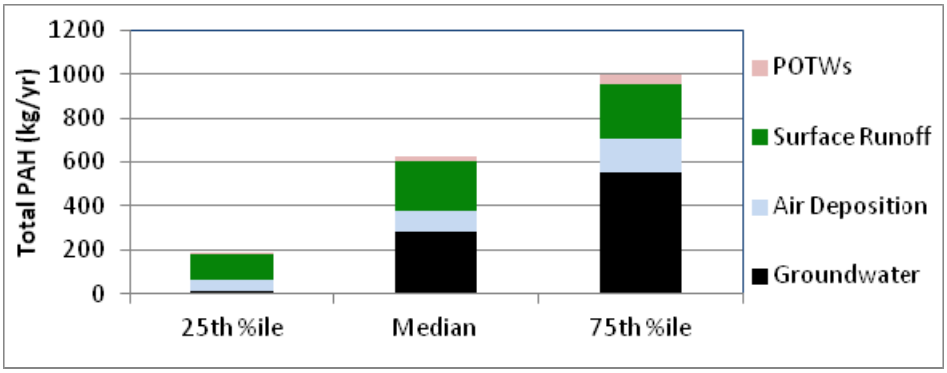


Figure 39. Total PAH Loads to Puget Sound as the Sum of Major Delivery Pathways.

Relationship Between Sources and Pathways/Loading

The conceptual model of PAH transport and fate following release is complex, and the ability to draw conclusions is hampered by the limitations of PAH loading estimates derived for Puget Sound.

The inventory of sources (Ecology, 2011) suggests that releases to air – either in particle, aerosol, or vapor form – account for two-thirds of the PAH released in the Puget Sound basin (approximately 200,000 kg/yr). Releases of chemicals to air are by nature difficult to track through a watershed, but several generalizations may be made regarding PAH releases. For instance, nearly all of the PAHs released to air originate from combustion sources and would therefore be expected to be HPAHs, the major type of PAHs formed pyrogenically.

The Air Emissions Inventory (Ecology, 2007) indicates that woodstove use is the largest single combustion source (and overall PAH source) to the Puget Sound basin (Ecology, 2011). Through analysis of anhydrosugars (markers of wood combustion), the air deposition study appears to confirm a strong signal of woodstove use during the late winter months. Nearly all of the remaining PAH combustion sources are from petroleum, such as light-and heavy-duty vehicle emissions, and from the use of other machinery using internal combustion engines.

The air deposition study measured several LPAHs and related compounds to assess PAH source signatures. The authors concluded that all of the PAHs were derived from combustion sources, originating from a mixture of petroleum and biomass (Brandenberger et al., 2010). Geographically, the widespread and relatively homogeneous distribution of PAHs among many of the sampling stations – except those in urban areas – supports a notion of a widespread regional distribution of combustion products. Air deposition sites located in urban areas showed higher PAH concentrations apparently derived from fossil fuel combustion sources, a finding supported by correlation with metals (copper and lead) associated in part with vehicle traffic.

The air deposition study appears to be consistent with the types of PAH sources inventoried in the Puget Sound basin and with other contemporary studies of PAH sources conducted on a regional level (e.g. Stein et al., 2006). However, the amounts released are three orders of magnitude higher than the amounts delivered to Puget Sound from all major loading pathways combined. If the air deposition results were extended to the entire Puget Sound watershed (approximately 750 kg/yr at the 75th percentile), and all of the deposited PAHs were entrained and delivered through surface runoff, this load would still represent <0.5% of the combustion releases. One explanation for the difference is that PAHs loads reported for surface runoff may underestimate actual loads to Puget Sound, while at the same time releases from combustion sources may be largely overestimated. However, there are no clear lines of evidence to support either supposition.

Of the remaining non-combustion sources of PAHs (130,000 kg/yr), approximately 90% is released from creosote marine pilings, railroad ties, and utility poles (Ecology, 2011). Valle et al. (2007) estimated that in the New York/New Jersey Harbor area, approximately 13% of the PAHs released over the lifetime of these products is to air, resulting in a total loss to air of 14,000 kg/yr from all three product types combined. Remaining releases occur through leaching and washout, presumably directly to marine waters for pilings. PAH leaching from rail ties and

utility poles is most likely to result in enrichment of adjacent soils (except in the case where these products are surrounded by impervious surfaces, or over-water in the case of railroad bridges). Once bound to soils, PAHs will generally remain immobile unless soils are removed or scoured through mechanical force. It is possible that some of the PAHs detected in commercial/ industrial area surface runoff during storm events were derived from utility poles or rail crossings, but the lack of major PAH components from creosote (naphthalene, acenaphthene) suggests that creosote leaching is not a major source in these instances.

Aside from PAHs released from creosote structures mounted on impervious surfaces, PAHs released from spillage of petroleum products is a potentially large source of LPAH to surface waters. An inventory of petroleum spills ≥ one gallon in the Puget Sound basin suggests that approximately 1,000 kg/yr of PAHs are released due to spillage of over 150,000 kg/yr of petroleum (Ecology, 2011). However, much larger quantities of PAHs released from small oil drips and leaks (8,500 kg/yr) and from improper disposal of used oil (1,300 kg/yr) account for the bulk of PAHs released as spills. Minor drips and leaks are likely to occur on impervious surfaces, which vastly increase the odds that PAH from this source will become entrained in runoff, some of which will find its way to surface waters. However, the results of the surface runoff study do not indicate motor oil as a major source of PAHs. Although the overall load of LPAH was higher than for HPAH, this did not appear to reflect widespread petroleum releases on impervious surfaces since the bulk of the LPAH load was from forested watersheds.

Of the POTW sources inventoried, only improper disposal of used oil is likely to be released in a constrained pathway. Most of the PAH loads discharged by POTWs is due to LPAHs, and the individual compounds frequently detected are those which are typically found at the highest concentrations in motor oil (fluoranthene, naphthalene, pyrene). This supports "down-the-drain" disposal of motor oil as a potential source of PAHs. Overall detection frequencies and concentrations of motor oil related PAHs in POTW effluent were similar between winter and summer season sampling, indicating a non-stormwater source of these PAHs and supporting the notion that improper disposal of motor oil may be a continuing PAH source to POTWs. However, there remain large differences in the quantity of PAHs released from this source (1,300 kg/yr) and the amount discharged from POTWs (8 – 46 kg/yr). At the treatment end, some quantity of PAH is presumably lost via solids removal prior to discharge, but the degree and extent of this removal is not known.

Hazard Evaluation

The hazard evaluation indicates that PAHs are a Priority 1 level of concern for freshwater sediments and for human health (Table 21). No effects data were available to adequately compare observed concentrations to tissue residue effects or effects to wildlife. The hazard evaluation indicates that levels of concern for PAHs may vary considerably depending on the media evaluated and whether PAHs are assessed individually or as groups (i.e. LPAH or HPAH).

In general, it appears that observed PAH concentrations in surface waters are much lower than effects data, although effects data are limited for freshwaters and observed data are limited for marine waters. Where both observed data and effects data were sufficient to conduct adequate comparisons, 90th percentile values of the observed concentrations were generally well below the 10th percentile values for effects concentrations.

Table 21. Summary of Hazard Evaluation for PAHs.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.* or *Acute WQC* or *Chronic WQC***	**Level of Concern**
	Freshwater	a	≥5%	No[b]/INS[c]	Priority 2/U
	Nearshore Marine	11-12	>8%	INS[d]	U
	Offshore Marine	e	<4%	No[b]/INS[f]	Priority 2/U
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. > *SQS***	**Level of Concern**
	Freshwater	g	>25%	Yes/No[h]	Priority 1/2
	Nearshore Marine	i	≥44%	No	Priority 2
	Offshore Marine	j	>34%	No	Priority 2
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.***	**Level of Concern**
	Not Analyzed				
Wildlife		**Total N**	**FOD**	**Daily Dose > *10% of Lowest Effects Dose***	**Level of Concern**
	Not Analyzed				
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. > *NTR Criterion***	**Level of Concern**
	Freshwater	k	>21%[l]	Yes/No[m]	Priority 1/2
	Nearshore Marine	n	>20%	Yes/No[o]	Priority 1/2
	Offshore Marine	p	>2%	No[b]/INS[f]	Priority 2/U

Yellow highlight indicates Priority 1 Level of Concern
FOD=Frequency of detection
[a] N range is 1,447 – 1,577 and varies for Individual PAH or PAH group
[b] No for several individual PAHs
INS[c]=Insufficient effects data available for comparison to observed data for several individual PAHs
U=Unknown level of concern due to lack of sufficient data for adequate comparison
INS[d]=Insufficient observed data available for comparison to effects data
[e] N range is 12 – 84 and varies for individual PAH or PAH group
INS[f]=Insufficient observed data available for comparison to effects data for several individual PAHs
[g] N range is 284 – 1,182 and varies for individual PAHs or PAH group
[h] No for benzo(a)anthracene; Yes for all other individual PAHs or PAH groups
[i] N range is 196 – 1,051 and varies for individual PAHs or PAH group
[j] N range is 217 – 906 and varies for individual PAHs or PAH group
[k] N range is 17 – 102 and varies for organism type, tissue type, and for individual PAHs
[l] FOD=0% for fish tissue
[m] Yes for 5 of 9 individual PAHs
[n] N range is 50 – 117 and varies for organism type, tissue type, and for individual PAHs
[o] Yes for 4 of 9 individual PAHs
[p] N range is 14 – 74 and varies for organism type, tissue type, and for individual PAHs

In freshwater sediments, for all individual PAHs (except benzo(a)anthracene), LPAH, and HPAH, the 90th percentile values of observed concentrations are above the floating percentile SQS. In many cases, more than 25% of the values exceed this threshold.

In marine sediments, none of the 90th percentiles of observed concentrations exceed the SQS. Median concentrations of LPAH in marine sediments are much closer to the lowest guideline, and the 75th percentile concentrations exceed this level (Canadian TEL). Sediments located in the nearshore environment had slightly higher LPAH concentrations than those located offshore. The 95th percentiles of nearshore LPAH concentrations exceed all of the guidelines. However, on an organic carbon-normalized basis, at least 95% of the nearshore and offshore concentrations are below the lowest guideline. HPAH concentrations in marine sediments mirror the patterns (relative to guidelines) of LPAHs, except median HPAH concentrations in both nearshore and offshore sediments exceed the lowest guideline.

Human health concerns were evaluated for nine of the 16 individual PAHs, but not for LPAH or HPAH since there are no NTR criteria for PAHs as groups. Several individual PAHs pose a Priority 1 level of concern for human health based on comparisons to the NTR criteria. For the nine PAHs evaluated for human health, benzo(a)pyrene, benzofluoranthenes (b and k), dibenzo(a,h)anthracene, and indeno(1,2,3-cd)pyrene exceed the NTR for at least one organism type in either freshwater or marine waters.

Regionally Important Biological-Effects Data

Multiple investigations have identified biomarkers of exposures to PAHs in various Puget Sound fishes. Bile and stomach content of outmigrant juvenile Chinook salmon were found to contain various PAHs (Johnson et al., 2007), demonstrating that diet pathways are important for PAH exposures. The authors suggest that the levels of exposure may result in immunosuppression and other health effects. These results expanded and confirmed previously documented PAH exposures (Stein et al., 1992; Stein et al., 1995). A dietary feeding study on juvenile Chinook documented growth and physiological responses from dietary exposures to PAHs at concentrations that were environmentally realistic in the Puget Sound (Meador et al., 2006).

Biomarkers of PAH exposures were confirmed in Puget Sound English sole, rock sole, and starry flounder collected from up to five sites in Puget Sound (Stein et al., 1992). Stein et al. (1992) found the biomarkers of exposure were related to the degree of sediment contamination. Further field study by Johnson (2000) resulted in recommended various threshold sediment concentrations of PAHs to protect English sole against liver lesions, DNA adducts in liver, and other effects. The causal relationship between elevated sediment PAH concentrations and English sole liver effects was confirmed by Myers et al. (2003). In a study of English sole from the Hylebos Waterway and Colvos Passage, Sol et al. (2008) found no correlation between PAH exposure and age and little correlation between reproductive end points and PAH exposure. However, Pacific herring embryos were found to be affected by tricyclic PAHs in weathered crude oil (Incardona et al., 2009; Carls et al., 1999).

Several laboratory studies have documented that developmental defects in fish are associated with exposures to PAHs released by weathered crude oil, notably the tricyclic-PAHs (Incardona et al., 2005; Incardona et al., 2006; Carls et al., 2008). Carls and Meador (2009)

developed a description of the oil weathering, PAH toxicity, and embryo exposures to explain the observed toxicity from PAHs in weathered oil at relatively low levels. Driscoll et al. (2010) developed a framework for describing PAH exposure as a dose to fishes in order to understand the mechanisms of exposure and toxicity.

Bis(2-ethylhexyl) phthalate

Phthalates are 87% of the 10.4 billion pounds per year world market for plasticizers, with bis(2-ethylhexyl) phthalate (a.k.a. di(2-ethylhexyl) phthalate [DEHP]) as the most common plasticizer for polyvinylchloride (PVC), constituting more than 50% of the phthalates produced (SPWG, 2007; ECB, 2008). Approximately 90-95% and possibly as much as 97% of DEHP is used to plasticize PVC products, which may contain up to 60% DEHP, although 30% may be more typical (ECB, 2008). Roughly 200-300 million pounds of DEHP are produced in the U.S. each year, with unknown amounts contained in imported products. According to ATSDR (2002), relatively little is imported (4 million pounds in 1998) or exported (14-27 million pounds per year in 1994-1998).

Phthalates are not covalently bound to PVC polymer chains and migrate out over time (Rudel and Perovich, 2009). The amount of phthalates that leach out of the PVC into air is quite variable. Factors that affect the rates of volatilization include the temperature and surface area of the material; emission rates among phthalates may also be quite variable. At a certain point the PVC undergoes glassification when it becomes stiff and brittle, and very little phthalate continues to off-gas. Once emitted, phthalates tend to adhere to dust particles rather than remain in vapor phase (ATSDR, 2002; SPWG, 2007).

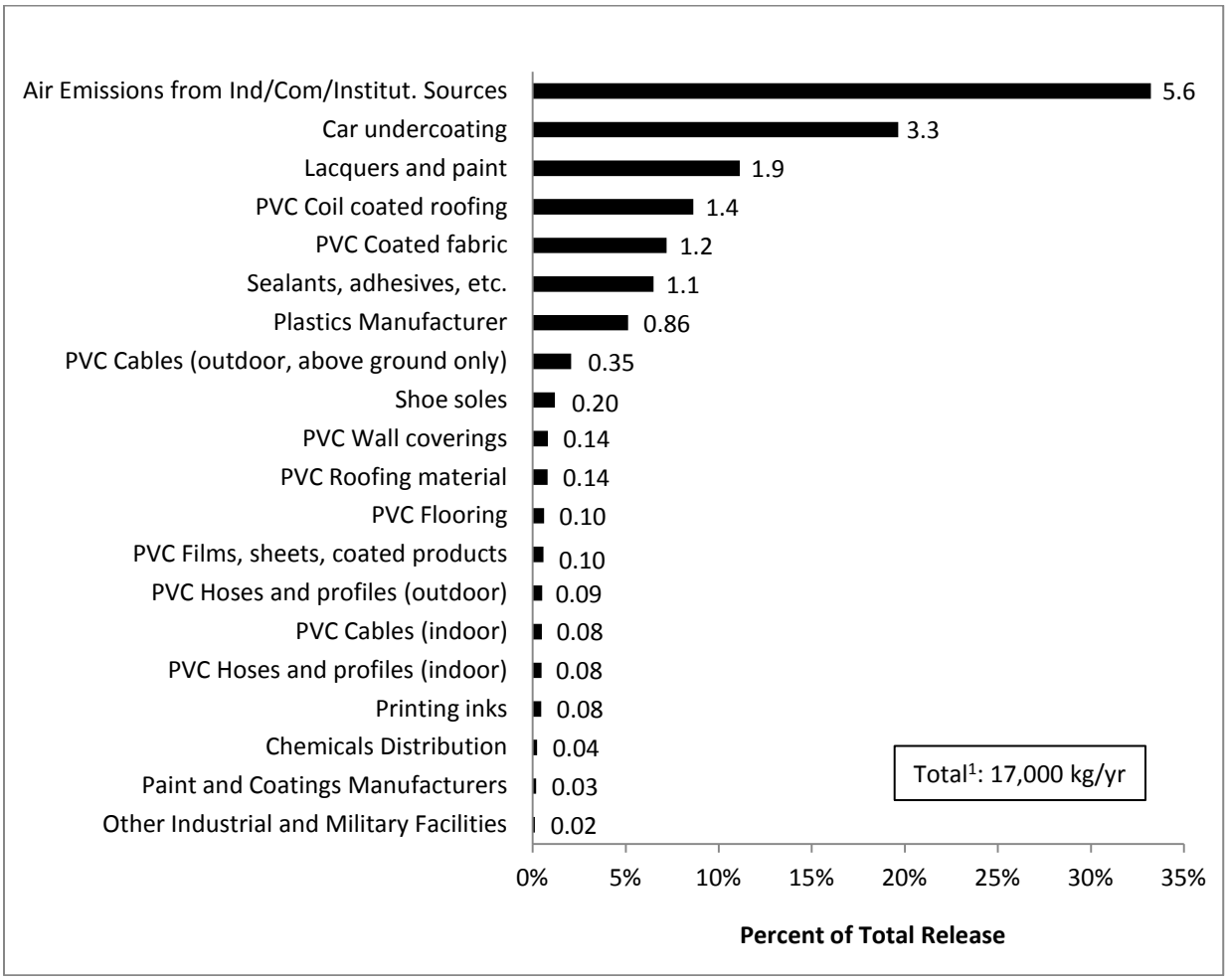
Plasticized PVC products are widely used and include exterior siding and roofing materials, automobiles, wires/cabling, advertizing banners, flooring, weather stripping, upholstery, garden hoses, swimming pool liners, footwear, clothing, food containers, tablecloths, shower curtains, rainwear, and toys. Rigid PVC products, such as pipes and windows, do not contain phthalates or other plasticizers. In Western Europe, DEHP emissions were estimated at 300 tons per year from indoor uses and 2,600 tons per year from exterior uses after measuring emissions to the air from PVC products (ATSDR, 2002). The population of Western Europe is about 400 million people, compared to about six million people in Washington State.

Non-plasticizer (non-polymer) uses of DEHP are a small percentage (<10%) of overall use. These uses include PCB replacement (dielectric fluids for electric capacitors), de-foaming agents in paper manufacturing and detergents, as well as chemical intermediates for insect repellent, cosmetics, lacquers, munitions, ceramics, printing inks, adhesives, sealants, and industrial lubricants.

Once in the environment, DEHP biodegrades in water but may accumulate in aquatic organisms to some degree (PTI, 1991). It has relatively low solubility and may resist degradation once bound to soil particles. In the aquatic environment, DEHP is nearly always detectable at concentrations >0.01 ug/l in freshwater and marine waters, and >0.01 mg/kg (dw) in sediment (PTI, 1991; Appendices D-3 and D-4).

Major Releases from Primary Sources

The inventory of phthalate sources in the Puget Sound basin suggests that total phthalate release is approximately 34,000 kg/yr for the six phthalates assessed (Ecology, 2011). Emissions from plasticized PVC products are the primary source of phthalates (SPWG, 2007), with DEHP as the dominant phthalate used as a plasticizer. In addition to releases from polymer use, DEHP releases occur through non-polymer uses, for a total annual release estimated to be 17,000 kg/yr (Figure 40).



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

Figure 40. Total DEHP Release in the Puget Sound Basin (values shown are thousands kg/yr).

DEHP release from polymer use accounts for approximately 40% of the total annual release, with large contributions from car undercoating (3,300 kg/yr), coil coated roofing (1,400 kg/yr), and coated fabric (1,200 kg/yr). Indoor polymer use accounts for approximately 5% of the loss through polymers.

Releases of DEHP from non-polymer uses include loss from lacquers, paints, sealants, adhesives, and printing inks. Combined, these account for approximately 15 - 20% of DEHP release in the Puget Sound basin. The remaining 40% (6,600 kg/yr) of DEHP releases are through industrial, commercial, and institutional point-source air emissions.

Total phthalate releases from personal care products combined are approximately 11,000 kg/yr including releases from fragrances, deodorant, nail polish, hair spray, and body lotion. According to testing data from U.S. and Swedish non-governmental organizations, diethyl phthalate (DEP) is the primary phthalate used in cosmetics and personal care products, with some products also containing di-n-butyl phthalate (DBP). However, there appears to be little if any use of DEHP in these products (DiGangi and Norin, 2002; Houlihan et al., 2002; EPA, 2009).

Major Delivery Pathways and Loading

Data on major delivery pathways for DEHP loading were obtained from PSTLA loading studies and are included in Table 22. No effort was made to assess DEHP loads from direct atmospheric deposition of DEHP to Puget Sound during Phase 3 (Brandenberger et al., 2010). As mentioned previously, loadings through direct groundwater discharge was estimated from literature values, whereas loadings through other pathways were estimated from field studies specifically designed to estimate loads.

Table 22. DEHP Loads (kg/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater (a)	14	227	440
Air Deposition	NA	NA	NA
Surface Runoff (b)	1,746	1,777	1,863
POTWs	216	439	904
Ocean Exchange	NC	NC	NC

(a) Lowest, highest, and mid-point of estimated loads
(b) Detected in residential areas only during baseflows
NA=not analyzed
NC=not calculated due to insufficient data

The estimated range of groundwater DEHP loads is large (14 – 440 kg/yr). The range in estimates is due primarily to differences in flows used to establish the possible range of loads; groundwater discharges used in the groundwater loading study ranged by an order of magnitude. DEHP load estimates in groundwater should be used with caution since they are based primarily on non-detect data and numerous assumptions, including unconfirmed assumptions about DEHP mobility in groundwater.

DEHP loads discharged from POTWs are approximately 220 – 900 kg/yr. Although field blank contamination and higher-than-desired laboratory quantitation limits for some samples resulted in uncertainty regarding some of the results, it appears that detection frequencies and concentrations between seasons (wet and dry) were not substantially different. However, seasonal first-flush concentrations were evident in agricultural sub-basins.

Surface water runoff loads for DEHP are estimated to be approximately 1,750 – 1,860 kg/yr for the entire Puget Sound basin based on the surface runoff study. DEHP was rarely detected during baseflows (3% frequency of detection) but was detected at a moderate frequency (30%) during storm flows, mostly in commercial/industrial areas. Although overall concentrations were slightly higher in commercial/industrial areas, loads generally mirror the amount of land cover represented by each land cover category; forested areas account for approximately 84% of the total DEHP load in surface runoff load to Puget Sound.

Loads of DEHP in marine waters were not calculated due to the low frequency of detection. DEHP was detected in only three samples – two in Hood Canal and one in the northern boundary waters – at low levels (≤0.06 ug/l).

The net sum of DEHP loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25th, median (50th), and 75th percentile values is 2,000, 2,400, and 3,200 kg/yr, respectively. Under all of these estimates, there is a net DEHP load to Puget Sound.

DEHP loadings from pathways that may represent *partially controllable* sources of COCs (i.e. all of the major pathways except ocean exchange) are shown as ranges (25th -75th percentiles) and medians for each pathway in Figures 41. Each pathway represented as a contribution to the total load is displayed in Figures 42.

Total DEHP loading from the major pathways assessed is 2,000 – 2,800 kg/yr. Surface runoff accounts for the largest pathway (58 – 88%), followed by POTWs (11 – 28%) and groundwater (1 – 14%).

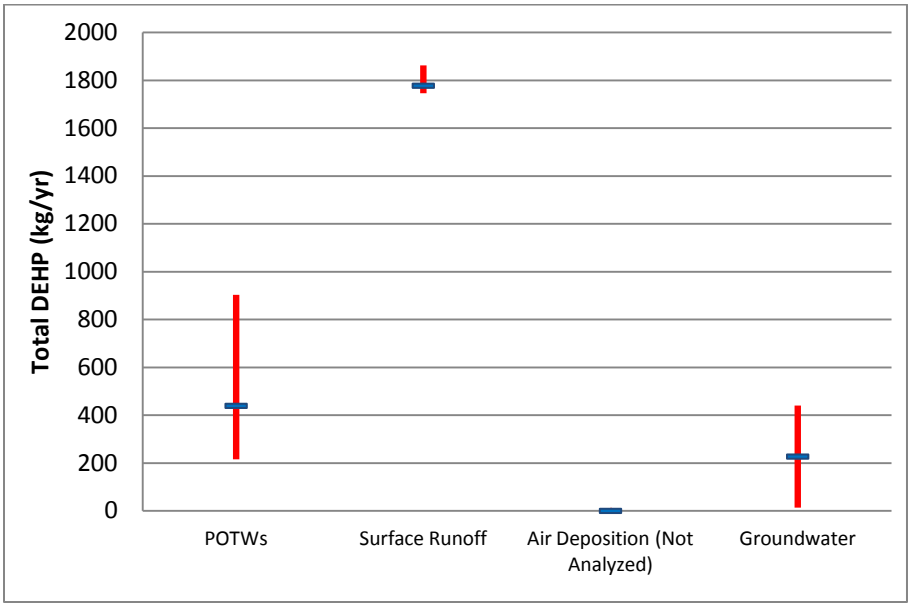


Figure 41. Range (25th - 75th percentiles) and Median DEHP Loads to Puget Sound from Each Major Delivery Pathway.

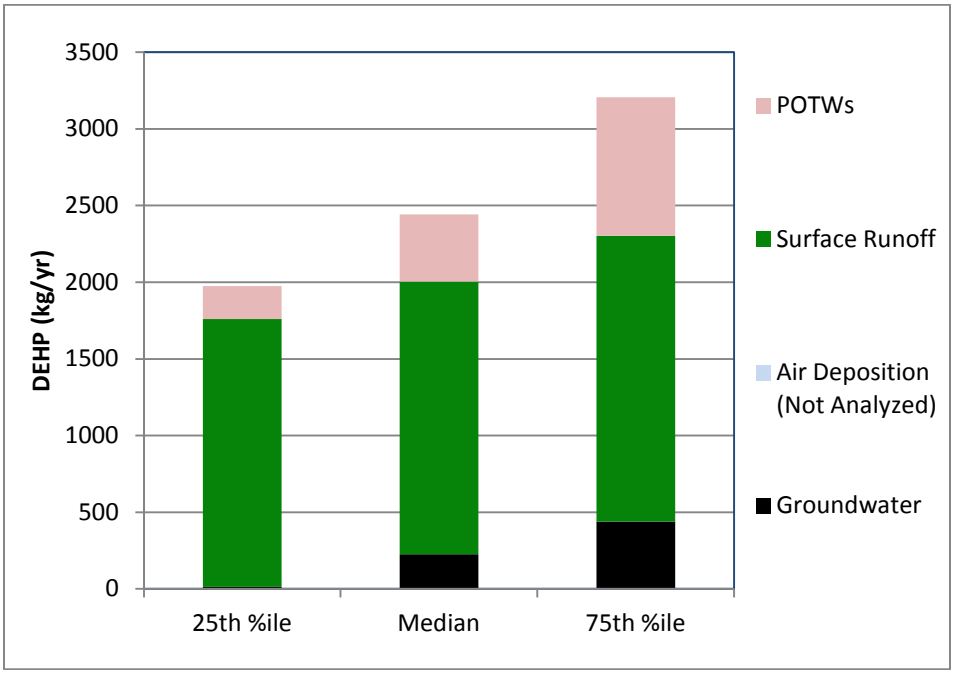


Figure 42. Total DEHP Loads to Puget Sound as the Sum of Major Delivery Pathways.

Relationship Between Sources and Pathways/Loading

The conceptual model of DEHP transport and fate following release is limited due to the low frequency of detection in surface runoff and the lack of data on DEHP in atmospheric deposition. DEHP depositions to Puget Sound of 310 to 16,000 kg/yr were estimated during the Phase 1 project, but these data were based largely on very limited sampling in an urban area (Hart Crowser et al., 2007) and may not have been representative for the Puget Sound basin. Data on atmospherically deposited DEHP loads would have proven particularly useful since the primary sources of DEHP suggest that volatilization is the principal release mechanism, followed by attachment to dust particles. In this respect, DEHP may be similar to PBDEs in their transport and fate in the environment. If DEHP follows a similar mode of fate and transport to that of PBDEs, the air deposition and POTW pathways would be expected to deliver loads of the same relative magnitude as those for surface runoff.

A portion of the DEHP emitted from both polymer and non-polymer sources is likely to occur indoors, and some fraction of these releases is presumably delivered to the sanitary sewer system. Estimates of DEHP delivered to POTWs through washing machine rinse water were conducted by the Washington Toxics Coalition and People for Puget Sound during 2009 (WTC/PPS, 2009). They estimated that roughly 960 kg/yr DEHP are delivered to Puget Sound POTWs each year due to dust-bound DEHP which becomes attached to clothing and is subsequently rinsed down the drain during the washing process.

As pointed out by the authors of the POTW loading report (Ecology and Herrera, 2010), it is difficult to draw conclusions regarding seasonal differences in COC loads from POTWs. However, it appears that POTWs discharge fairly constant DEHP loads between seasons,

suggesting the presence of a steady source of DEHP. This appears to be consistent with the indoor emissions of this compound, followed by attachment to dust particles and delivery to POTWs through sanitary drains. The inventory of sources suggests that approximately 500 kg/yr DEHP is released indoors through loss from polymers, although some of the DEHP release from non-polymer use (3,000 kg/yr) may also occur indoors. While neither the WTC/PPS report nor the present loading studies provide enough information to calculate a mass-balance of DEHP delivered to, and discharged from, POTWs, the information suggests that the amount delivered to POTWs is likely on the order of thousands of kg per year.

Some of the DEHP delivered to POTWs is likely to be from stormwater, but the contribution from this pathway may be comparatively small. Surface runoff DEHP loads in areas where stormwater is potentially diverted to POTWs (i.e. commercial/industrial and residential areas) are calculated to be approximately 120 – 230 kg/yr during storm events (Herrera, 2011). Therefore, the DEHP load in stormwater diverted to POTWs is presumed to be only a fraction of that amount.

Hazard Evaluation

The hazard evaluation indicates that DEHP is a Priority 1 level of concern for sediments and for human health (Table 23). Hazard due to tissue residue effects and effects to wildlife were not evaluated.

In fresh, nearshore marine, and offshore marine surface waters, the 90th percentile of DEHP concentrations (approx. 4 ug/l, 2 ug/l, and 3 ug/l, respectively) is two orders of magnitude below the lowest 10% of effects for freshwater, and perhaps even a greater magnitude below 10% of the effects data for marine waters, although the latter is difficult to assess due to a paucity of effects data. No water quality criteria are available for comparison.

More than 25% of the DEHP concentrations in freshwater exceed the floating percentile SQS as well as the CSL, and the median concentration (approx. 120 ug/kg dw) approaches the SQS. In marine sediments, the 90th percentiles of the nearshore and marine sediments (approx. 10,000 ug/kg organic carbon and 5,000 ug/kg organic carbon, respectively) exceed the SQS, and the 90th percentile of nearshore DEHP concentrations exceed the CSL as well.

DEHP is rarely detected in freshwater or marine fish tissue, but it is detected more frequently in bivalves and (in the case of freshwater) other invertebrates as well. The 90th percentile of DEHP concentrations in freshwater bivalves (approx. 240 ug/kg) is slightly above the NTR criterion. The nearshore marine fish tissue concentration exceeds the NTR criterion as well (at the 90th percentile level), but this is based on few data (n=8 detected concentrations). The 95th percentile concentrations in other freshwater invertebrate tissues approach the NTR criterion, and the median values for freshwater bivalves, other freshwater invertebrates, and marine nearshore bivalves all exceed the criterion adjusted to the EPA-recommended subsistence rate (142.4 g/d).

Table 23. Summary of Hazard Evaluation for DEHP.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.* or *Acute WQC* or *Chronic WQC***	**Level of Concern**
	Freshwater	1,484	84%	No	Priority 2
	Nearshore Marine	12	92%	INS	U
	Offshore Marine	84	54%	INS	U
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. > *SQS***	**Level of Concern**
	Freshwater	548	87%	Yes	Priority 1
	Nearshore Marine	513	74%	Yes	Priority 1
	Offshore Marine	474	67%	Yes	Priority 1
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.***	**Level of Concern**
	Not Analyzed				
Wildlife		**Total N**	**FOD**	**Daily Dose > *10% of Lowest Effects Dose***	**Level of Concern**
	Not Analyzed				
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. > *NTR Criterion***	**Level of Concern**
	Freshwater	a	>57%[b]	yes	Priority 1
	Nearshore Marine	c	>9%	yes	Priority 1
	Offshore Marine	d	>24%[b]	INS	U

Yellow highlight indicates Priority 1 Level of Concern
FOD=Frequency of detection
INS=Insufficient observed data available for comparison to effects data
[a] N range is 9 – 99 and varies for organism type and tissue type
[b] FOD=0% for fish tissue
[c] N range is 42 – 79 and varies for organism type and tissue type
[d] N range is 16 – 33 and varies for organism type and tissue type

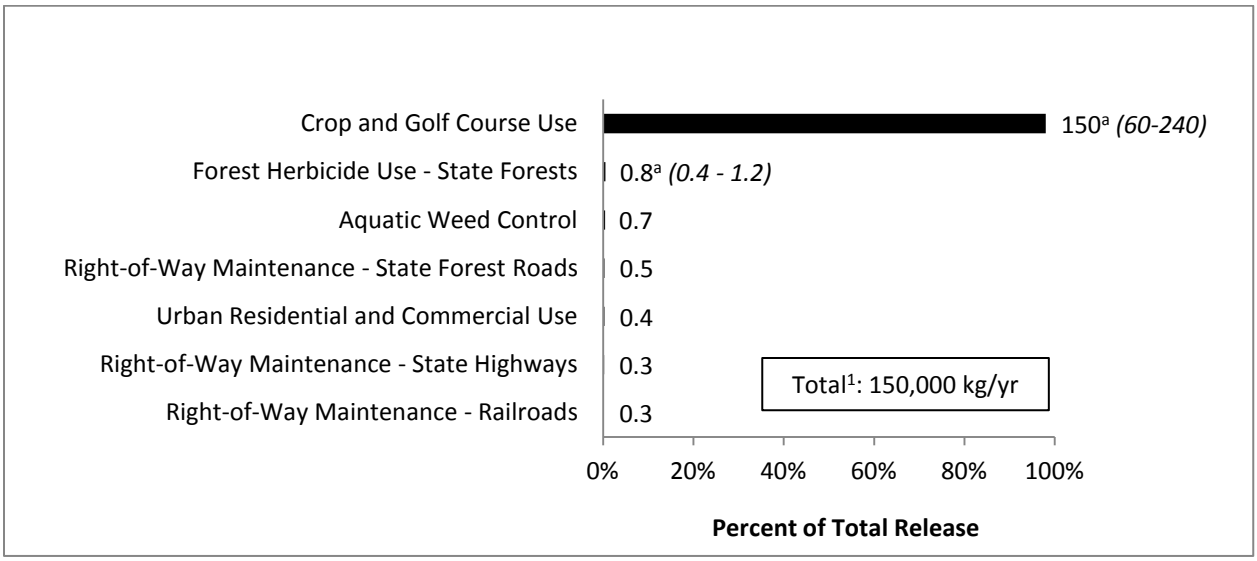
Triclopyr

Triclopyr [((3,5,6-trichloro-2-pyridinyl)oxy)acetic acid)] is a broad-leaf herbicide used primarily for rice, pasture and rangeland, rights-of-way, and turf, including home lawns and gardens (EPA, 1998). It also has limited use to control nuisance vegetation in freshwater lakes and is permitted under Ecology's Aquatic Pesticide General Permit. Triclopyr is applied as the triethylamine (TEA) salt and butoxyethyl ester (BEE) formulations and is sold under the product names Garlon® and Crossbow® for terrestrial use and Renovate® for aquatic use.

Following release in the environment, triclopyr formulations are dissociated to the triclopyr anion which is readily soluble in water. Half-lives of triclopyr formulations following terrestrial applications are generally 10 days or less. In terrestrial soils, triclopyr undergoes microbial degradation with a half-life generally less than 20 days. Once in the aquatic environment, photolysis is the major degradation mechanism and occurs rapidly, generally with a half-life of several days or less (EPA, 1998). Although triclopyr half-lives tested under laboratory conditions suggest little persistence, environmental studies conducted following applications indicate that actual half-lives may be substantially greater, perhaps on the order of 100 days or more. Due its solubility, absorption to soil particles is not a major fate process, and triclopyr does not bioaccumulate in aquatic organisms.

Major Releases from Primary Sources

Total triclopyr release from the seven application categories assessed totaled approximately 150,000 kg/yr (Figure 43). Agricultural uses of triclopyr accounted for 76 – 94% of the total release, with golf course use accounting for 5 – 22%. The remaining triclopyr use was primarily for right-of-way maintenance (0.7%), aquatic weed control (0.5%), and domestic use (0.3%). The small amounts calculated for domestic and forestry use likely underestimate actual usage rates.



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

[a] Mid-point of range

Figure 43. Total Triclopyr Release in the Puget Sound Basin (values shown are thousands kg/yr).

Major Delivery Pathways and Loading

Data on major delivery pathways for triclopyr loading were obtained from PSTLA loading studies and are included in Table 24. No effort was made to assess triclopyr loads from direct atmospheric deposition or exchange of ocean waters. Due to the low frequency of detection in POTW effluent, loads were not calculated (Ecology and Herrera, 2010). No triclopyr data were found for groundwater, and therefore no attempt was made to calculate loading from direct groundwater discharge to Puget Sound (Pitz, 2011).

Table 24. Triclopyr Loads (kg/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater	NA	NA	NA
Air Deposition	NA	NA	NA
Surface Runoff (a)	641	652	686
POTWs	NC	NC	NC
Ocean Exchange	NA	NA	NA

(a) Detected in commercial/industrial and agricultural areas only during baseflows
NA=not analyzed
NC=not calculated due to insufficient data

Surface runoff loads for triclopyr are estimated to be approximately 641 – 686 kg/yr for the entire Puget Sound basin based on the surface runoff study. Triclopyr was detected more frequently during storm flows than during baseflows, with the highest rate of detection during the fall first flush (63%). Overall detection rates were highest in commercial/industrial areas (47%), followed by residential areas (41%), agricultural areas (31%), and forests (16%). Detected concentrations varied little across different land covers, with typical levels in surface water of 0.03 ug/l. Although triclopyr was detected only in commercial/industrial and agricultural areas during baseflows, overall loads generally mirrored the amount of land cover represented by each land cover category. As a result, forested areas account for approximately 90% of the total surface runoff load to Puget Sound.

Hazard Evaluation

Triclopyr hazards were difficult to evaluate due to the lack of observed environmental data for all media except fresh surface waters (Table 25). Over 1,600 measurements of triclopyr are available for fresh surface waters, with a detection frequency of 33%. The 90^{th} percentile of these detected triclopyr values is approximately 0.1 ug/l, three orders of magnitude lower than the 10^{th} percentile of the effects data.

Table 25. Summary of Hazard Evaluation for Triclopyr.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.* or *Acute WQC* or *Chronic WQC***	**Level of Concern**
	Freshwater	1,632	33%	No	Priority 2
	Nearshore Marine	0	--	INS	U
	Offshore Marine	0	--	INS	U
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. > *SQS***	**Level of Concern**
	Freshwater	0	--	INS	U
	Nearshore Marine	0	--	INS	U
	Offshore Marine	0	--	INS	U
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.***	**Level of Concern**
	Not Analyzed				
Wildlife		**Total N**	**FOD**	**Daily Dose > *10% of Lowest Effects Dose***	**Level of Concern**
	Not Analyzed				
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. > *NTR Criterion***	**Level of Concern**
	Not Analyzed				

FOD=Frequency of detection
INS=Insufficient effects or observed data available for evaluation

Nonylphenol

Nonylphenol is a commonly detected organic chemical of particular interest due to its potential to disrupt endocrine function in aquatic organisms. While there are limited uses for the direct use of nonylphenol, it has a broad range of applications as a precursor for nonionic surfactants, particularly alkylphenol ethoxylates which are used in domestic and industrial cleaning products and emulsifiers. These include uses as domestic and commercial laundry detergents, pulp and paper processing, and as deicers and in firefighting foams and gels. Annual production in the U.S. is on the order of 200 to 300 millions of pounds per year (EPA, 2005).

The breakdown of alkylphenol ethoxylates to nonylphenol occurs largely during the sewage treatment process, although the breakdown process does not require active sewage sludges for this to occur (EPA, 2005). Nonylphenol is more resistant to further degradation than its parent compounds. It has low solubility in water and partitions to sediments where its resistance to degradation is generally increased. Based on log K_{ow} values, nonylphenol's capacity for

bioaccumulation is considered moderate, but it is generally found at low concentrations in fish tissue, possibly suggesting fish have some ability to metabolize nonylphenol (EPA, 2005).

Major Releases from Primary Sources

Total nonylphenol release in the Puget Sound basin totals approximately 180 kg/yr. All of the inventoried nonylphenol releases were from point-source air emissions. No effort was made to calculate nonylphenol releases from diffuse sources. Nonylphenol is not a required reporting requirement under the TRI, and therefore no industrial or institutional nonylphenol releases were catalogued (Ecology, 2011).

Major Delivery Pathways and Loading

Major delivery pathways for nonylphenol loading are included in Table 26. No effort was made to assess nonylphenol loads from direct atmospheric deposition. Due to the low frequencies of detection, nonylphenol was not detected in POTW effluent or in exchange of ocean waters (Ecology and Herrera, 2010; Gries and Osterberg, 2011). No nonylphenol data were found for groundwater, and therefore no attempt was made to calculate loading from direct groundwater discharge to Puget Sound (Pitz, 2011).

Table 26. Nonylphenol Loads (kg/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater	NA	NA	NA
Air Deposition	NA	NA	NA
Surface Runoff (a)	23	23	24
POTWs	NC	NC	NC
Ocean Exchange	NC	NC	NC

(a) Surface runoff loads based on storm flows only; not detected in any land covers during baseflows and in commercial/industrial areas only during storm flows
NA=not analyzed
NC=not calculated due to insufficient data

Surface water runoff loads for nonylphenol are estimated to be approximately 23 – 24 kg/yr for the entire Puget Sound basin based on the surface runoff study. There was a single detection of nonylphenol among the 126 samples analyzed in surface runoff, and therefore no patterns related to land cover types or hydrological conditions could be established.

Hazard Evaluation

Nonylphenol hazards were difficult to evaluate due to the lack of observed environmental data in some media and a lack of effects data for other media (Table 27). For fresh surface waters, however, there are ample observed data as well as effects data. The 90th percentile of observed concentrations of 4-nonylphenol (approx. 4 ug/l) is slightly below the chronic water quality criterion and several times lower than the 10th percentile of effects data; the 95th percentile of observed values exceeds the chronic criterion.

Table 27. Summary of Hazard Evaluation for 4-Nonylphenol.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.* *or* *Acute WQC* *or* *Chronic WQC*	**Level of Concern**
	Freshwater	539	26%	No	Priority 2
	Nearshore Marine	11	27%	INS[a]	U
	Offshore Marine	84	17%	No	Priority 2
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. >** *SQS*	**Level of Concern**
	Freshwater	251	12%	INS[b]	U
	Nearshore Marine	67	15%	INS[b]	U
	Offshore Marine	91	29%	INS[b]	U
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.*	**Level of Concern**
	Not Analyzed				
Wildlife		**Total N**	**FOD**	**Daily Dose >** *10% of Lowest Effects Dose*	**Level of Concern**
	Not Analyzed				
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. >** *NTR Criterion*	**Level of Concern**
	Not Analyzed				

FOD=Frequency of detection
INS[a]=Insufficient observed data available for comparison to effects data
U=Unknown level of concern due to lack of sufficient data for adequate comparison
INS[b]=Insufficient effects data available for comparison to observed data

In marine surface waters, there are few observed data of 4-nonylphenol for the nearshore (n= 11) dataset. For the offshore marine waters, there are sufficient data (n=84) for comparisons to effects and criteria. Concentrations for all marine water data are well below the water quality criteria and at least 95% of the effects data.

Of the guidelines used to assess hazards in sediments, the Canadian TEL was the only guideline with numerical values for nonylphenol; no SQS was available for freshwater or marine sediments. In freshwater sediments at least 5% of the values exceeded the TEL and over 10% of the values exceeded the TEL in marine sediments. However, the TEL was above the 95th percentile values in offshore sediments.

There are no data to assess residue effects levels in aquatic organisms, or to evaluate nonylphenol hazards to wildlife or humans. This likely reflects nonylphenol's low to moderate accumulation potential in fish and macroinvertebrates.

Oil & Grease and Petroleum Hydrocarbons

Unlike other COCs, oil & grease does not refer to one or more specific chemicals, but instead refers to a group of chemicals with loosely related chemical and physical properties. These chemicals include non-volatile hydrocarbons, vegetable oils, animal fats, waxes, soaps, greases, and related materials. The working definition of oil & grease is based on the analytical method to determine the concentration in water or soil. More specifically, oil & grease is the fraction of a sample that is extractable by n-hexane, and is sometimes referred to as hexane extractable material (HEM).

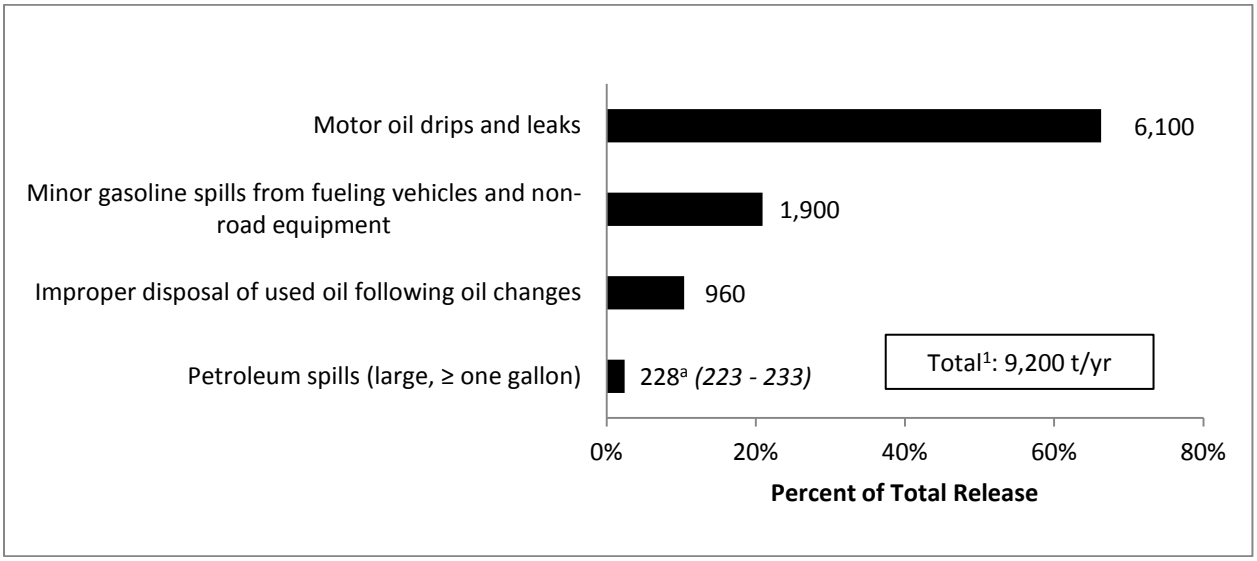
Like oil & grease, petroleum is not a specific compound, but may instead contain hundreds of chemicals, with crude oil as the ultimate source. Petroleum hydrocarbons – sometimes referred to as total petroleum hydrocarbons (TPH) – may further be divided into fractions based on the number of carbons of the major constituents. Gasoline range organics (GRO) refers to the methanol extractable fraction of organic chemicals with six to ten carbons, and generally includes a variety of gasoline alkanes, naphtha, mineral spirits, stoddard solvent, and other volatile petroleum products. Diesel range organics (DRO) are the fraction extracted with methylene chloride and have 11 – 28 carbons, and covers semi-volatile petroleum products (jet fuels through heavy fuel oils). Lube oils are typically characterized by molecules containing 29 – 35 carbons.

Major Releases from Primary Sources

The report on COC sources in the Puget Sound basin (Ecology, 2011) estimated releases of petroleum from various sources. Oil & grease releases were not estimated. Petroleum releases in the Puget Sound basin were inventoried by estimating releases from four major categories: petroleum spills ≥ one gallon, motor oil drips and leaks, minor gasoline spills, and improper disposal of used motor oil. The total quantity of petroleum released annually in the Puget Sound basin was estimated to be 9,200 metric tons (t)/yr (Figure 44). Other possible sources of petroleum including aviation fuel leaks and uncombusted oil and fuel discharged from marine engines were not estimated.

Petroleum spills ≥ one gallon were estimated to account for releases of 230 t/yr, with approximately one-half of the releases occurring in freshwaters, much of which occurs in the Lake Washington-Lake Union-Ship Canal system. Of the remaining spills, most of the volume was released to soils, followed by unknown receiving media and marine waters. Approximately one-quarter of the spill quantity was diesel fuel, followed by gasoline (approx. 10%) and jet fuel (approx. 3%), although these estimates vary depending on the source of information (see Ecology, 2011).

Although large spills may garner a host of attention, it appears that small motor oil and gasoline spills, drips, and minor leaks are responsible for approximately 87% of the total petroleum released annually in the Puget Sound basin. Improper disposal of used motor oil accounts for an additional 11% of petroleum releases inventoried.



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

[a] Mid-point of range

Figure 44. Total Petroleum Release in the Puget Sound Basin (values shown are t/yr).

Approximately 6,100 t/yr of motor oil is estimated to drip and leak from vehicle crankcases during the 58 billion kilometers travelled each year on roads in the region. Another 960 t/yr of motor oil is disposed of improperly following oil changes.

Minor gasoline spills that occur during fueling of vehicles and other motorized equipment, as well as spills that occur during the transport of portable fuel containers, lead to an estimated annual gasoline release of approximately 1,900 t, including 570 t released annually during on-road vehicle fueling at the pump. This estimate only includes liquid spillage; gasoline released through volatilization of fuel and vapor displacement during fueling was not included in this estimate.

Major Delivery Pathways and Loading

Data on major delivery pathways for oil & grease and petroleum loading were obtained from PSTLA loading studies and are included in Table 28. No effort was made to assess oil & grease or petroleum loads from direct atmospheric deposition, exchange of ocean waters, or POTWs. As mentioned previously, loading through direct groundwater discharge was estimated from literature values, whereas loadings through other pathways were estimated from field studies specifically designed to estimate loads.

Surface runoff was the only major loading pathway where oil & grease was analyzed. Loads were estimated to be approximately 8,500 – 10,600 t/yr for the entire Puget Sound basin based on the surface runoff study. Due to the low frequency of detection (<50%) in all land uses under both base and storm flow conditions, comparisons among land covers and flow conditions are not meaningful. However, seasonal first-flush elevations in oil & grease concentrations were evident for all but the forest land covers. Absolute loads calculated from surface runoff were a reflection of land cover proportions and flows, with forest lands making up the bulk of the load.

Table 28. Oil & Grease and Petroleum Loads (t/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
	Oil & Grease		
Groundwater	NA	NA	NA
Air Deposition	NA	NA	NA
Surface Runoff	8,469	8,469	10,598
POTWs	NA	NA	NA
Ocean Exchange	NA	NA	NA
	Lube Oil		
Groundwater (a)	6.03	34.2	62.4
Air Deposition	NA	NA	NA
Surface Runoff (b)	320	345	360
POTWs	NA	NA	NA
Ocean Exchange	NA	NA	NA
	Diesel Range		
Groundwater (a)	1.84	18.0	34.2
Air Deposition	NA	NA	NA
Surface Runoff	ND	ND	ND
POTWs	NA	NA	NA
Ocean Exchange	NA	NA	NA
	Gasoline Range		
Groundwater (a)	2.85	23.8	44.7
Air Deposition	NA	NA	NA
Surface Runoff	ND	ND	ND
POTWs	NA	NA	NA
Ocean Exchange	NA	NA	NA

(a) Lowest, highest, and mid-point of estimated loads
(b) Detected in agricultural areas only during baseflows
NA=not analyzed
ND=not detected

Lube oil loads were estimated for the groundwater pathway (6-62 t/yr) and for the surface runoff pathway (320 – 360 t/yr). The range of estimates for groundwater was due to differences in flows used to establish the possible range of loads; groundwater discharges used in the groundwater loading study ranged by an order of magnitude.

For surface water runoff, lube oil was detected at the highest concentrations and frequencies (75%) in commercial/industrial sub-basins during storm events. Lube oil was also detected in other land covers during storms – residential and agricultural areas displayed seasonal first-flushes – yet overall detection frequencies were low (<50%) in all but commercial/industrial land covers. Detections of lube oil were even more infrequent during baseflow sampling, where only agricultural streams had measurable lube oil.

Diesel and gasoline loads were estimated for groundwater (1.8 – 34 t/yr and 2.8 – 45 t/yr, respectively), but neither of these petroleum fractions were detected in surface water. The range of estimates for the groundwater loads was primarily due to differences in flows used to establish the possible range of loads.

The net sum of total oil & grease and petroleum loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sums for the following parameters at the respective 25th, median (50th), and 75th percentile values are: Oil and grease - 8,500, 8,500, and 10,600 t/yr; diesel – 1.8, 18, and 34 t/yr; and gasoline 2.8, 24, and 45 t/yr.

Loads for lube oil, the only form of petroleum with loading estimates available for more than one pathway, are shown in Figure 45. Total lube oil loads at the respective 25th, median (50th), and 75th percentile values are 330, 380, and 420 t/yr. Surface runoff accounts for 85 – 98% of the lube oil loads with the remaining 2 – 15% contributed by groundwater (Figure 46).

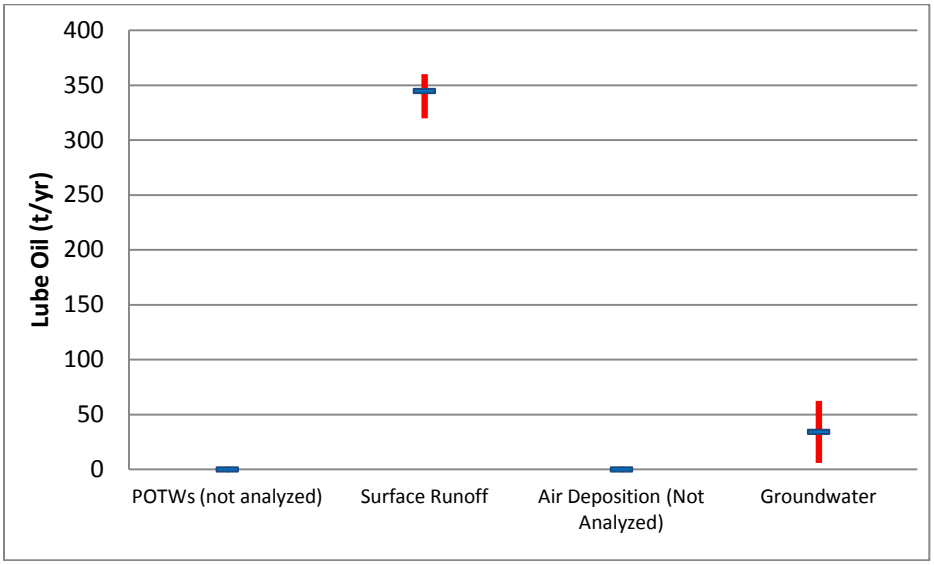


Figure 45. Range (25th - 75th percentiles) and Median Petroleum Lubricating Oil Loads to Puget Sound from Each Major Delivery Pathway.

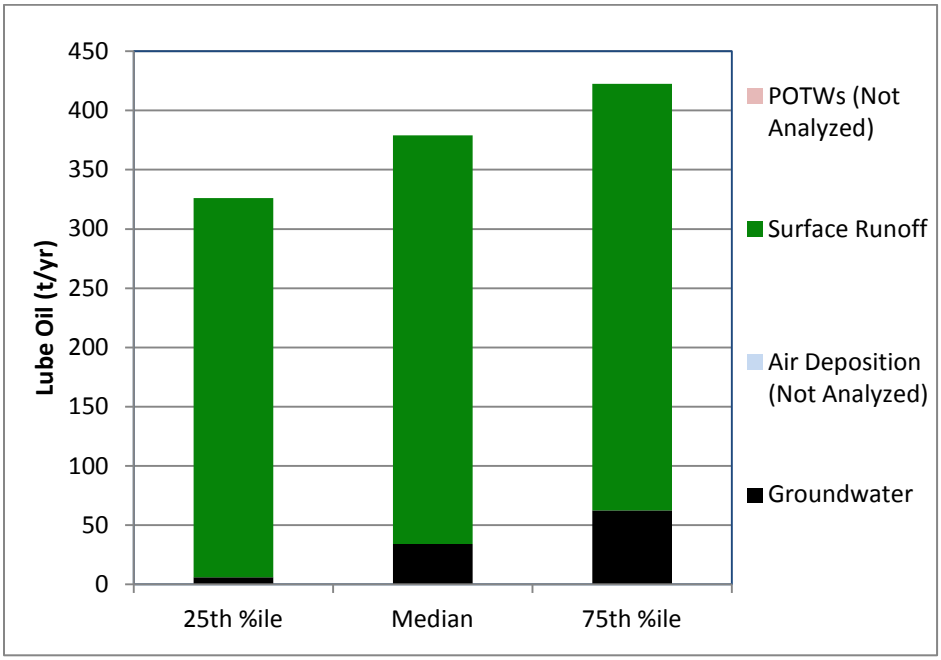


Figure 46. Petroleum Lubricating Oil Loads to Puget Sound from Each Major Delivery Pathway.

Relationship Between Sources and Pathways/Loading

The transport of oil & grease and petroleum hydrocarbons is not likely to be complex compared to many of the COCs addressed in this report. However, the lack of data in a variety of pathways, particularly the presumed major pathway (surface runoff), leaves little information on which to develop a conceptual model based on the data specific to this project.

Based on the source inventory, it appears that thousands of metric tons (t) of petroleum are released in the watershed each year. Most of the petroleum release is in the form of motor oil from engine drips and leaks (6,100 t/yr) and therefore likely to be directly to pavement. Since motor oil is only slowly degraded and is non-volatile, most would be expected to remain on the pavement until entrained by stormwater. Once entrained in stormwater, the oil may be transported directly or indirectly to surface waters, to POTWs, or become sequestered in reservoirs such as soil, detection ponds, and roadside ditches. Once bound to soil, lube oil may not be further transported unless the soil is dislodged.

Overall, it appears that approximately 6% of the motor oil released annually in the Puget Sound basin is transported to Puget Sound in surface water runoff. Diesel released on land from large spills (approximately 20 t/yr) or gasoline released during small fueling and transport spills (1,900 t/yr) was not reflected in measurable surface runoff loads. While it is possible that surface runoff estimates grossly underestimate petroleum loads due to analytical or sampling design errors, petroleum indicator PAHs (e.g. fluoranthene, naphthalene, pyrene) were also rare in surface runoff samples (Herrera, 2011), suggesting limited contribution from petroleum as a source.

Of the POTWs sources inventoried, only improper disposal of used oil (960 t/yr) is likely to be released in a constrained pathway. Petroleum was not assessed in POTWs, so there are no estimates available for this pathway. As mentioned previously in the PAH section, some of the PAHs associated with petroleum were frequently detected in POTW discharge samples, suggesting a possible petroleum source.

Hazard Evaluation

Toxicological evaluation of petroleum mixtures have historically been conducted in the aftermath of oil spills and similar events, and as a result, the majority of available petroleum toxicity data for are for crude oils. However, crude oil toxicity data are generally unsuitable for the Puget Sound basin hazard evaluation since none of the available observed environmental data are for crude oils.

A large amount of observed oil and grease data were available although these data are non-specific and do not describe a specific petroleum product; they encompass waxes, greases and other fatty acid substances from both animal, vegetable and petroleum origins. Because these environmental data are non-specific, and potentially toxic components may vary within the same concentration measured by this method; these data were considered unusable for this assessment.

Both toxicity data and environmental data were only available for four petroleum products in freshwater; heavy fuel/bunker oil, diesel fuel, gasoline, and lube oil. Toxicity data for these fours products were almost entirely based on lethal concentrations to 50% of the exposed population (LC_{50}s). Environmental data for these four products were plotted against the available toxicity data (Appendix D-3).

Only two petroleum product data results were available for marine waters and these were insufficient to estimate priority levels. No observed or effects data were available for sediment evaluation. Hazards due to tissue residue effects, effects to wildlife, and effects to human health were not evaluated.

For fresh surface waters, the median concentration for gasoline (approx. 2.5 mg/l) is above the 10^{th} percentile of effects data, and the 75^{th} percentile of gasoline concentrations exceeds the median effects concentration. For lube oil, the 90^{th} percentile of concentrations (approx. 4 mg/l) exceeds at least 95% of the effects data. For heavy fuel oil and diesel, all of the observed concentrations were two- to five-fold below effects data. In all cases, however, there were not sufficient effects data for an adequate evaluation.

Only toxicity results assessing the water soluble fraction (without free product) were used to assess these data. A more complete evaluation of petroleum would require analysis of parent and alkyl PAHs as well as issues such as phototoxicity which were beyond the scope of this assessment. In addition to the specific assessment for petroleum described here, a suite of individual and high/low molecular weight PAHs were evaluated in both the water and sediment assessments previously described above.

There are no sediment guidelines to assess the level of concern posed by petroleum in sediments, and neither effects data nor observed data are available to evaluate hazards due to residues in aquatic organisms or to evaluate petroleum hazards to wildlife or humans. Due to the uncertainties discussed above and the lack of effects data, there is a high level of uncertainty for the petroleum evaluation.

Table 29. Summary of Hazard Evaluation for Petroleum.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.* or *Acute WQC* or *Chronic WQC***	**Level of Concern**
	Freshwater	a	>8%	INS[b]	U
	Nearshore Marine	0	--	INS[c]	U
	Offshore Marine	0	--	INS[c]	U
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. > *SQS***	**Level of Concern**
	Freshwater	0	--	INS[c]	U
	Nearshore Marine	0	--	INS[c]	U
	Offshore Marine	0	--	INS[c]	U
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. > *10th %ile Effects Conc.***	**Level of Concern**
	Not Analyzed				
Wildlife		**Total N**	**FOD**	**Daily Dose > *10% of Lowest Effects Dose***	**Level of Concern**
	Not Analyzed				
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. > *NTR Criterion***	**Level of Concern**
	Not Analyzed				

FOD=Frequency of detection[a] N = 17 for heavy fuel oil, N = 295 for diesel, N = 359 for gasoline, and N = 894 for lube oil

INS[b]=Insufficient effects data available for comparison to observed data

INS[c]=Insufficient observed or effects data available for comparison

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Summary of Findings

General Considerations

Projects conducted under Phase 3 of the PSTLA which evaluated sources, loading, pathways, and relative hazards of selected COCs in the Puget Sound basin were the key sources of data used for this assessment.

The PSTLA focused on the narrow list of COCs shown below. This list was developed during Phase I of the project based on the COCs' presence in Puget Sound and their potential to cause harm, and to ensure that a broad variety of delivery pathways would be represented. There is a wide variety of chemicals in the Puget Sound basin which have the potential to cause biological and ecological harm, yet environmental data are lacking for many of them. Therefore, this assessment should be viewed as the starting point for development of a much larger toxic chemical assessment.

- Arsenic
- Cadmium
- Copper
- Lead
- Mercury
- Zinc
- Polychlorinated biphenyls (PCBs)
- Polybrominated diphenyl ethers (PBDEs)
- Polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs)
- Dichlorodiphenyltrichloroethane (DDT) and metabolites DDD and DDE
- Low molecular weight polycyclic aromatic hydrocarbons (LPAHs)
- High molecular weight polycyclic aromatic hydrocarbons (HPAHs)
- Carcinogenic polycyclic aromatic hydrocarbons (cPAHs)
- Diethylhexyl phthalate (DEHP, a.k.a. bis(2-ethylhexyl) phthalate)
- Triclopyr
- Nonylphenol
- Oil and petroleum hydrocarbons

The geographical area addressed in this *Assessment Report* includes the Puget Sound, the U.S. portions of the Straits of Georgia and Juan de Fuca, and the entire U.S. watershed for Puget Sound and the Straits (Figure 1). This is consistent with all of the land-based PSTLA loading projects (e.g. Hart Crowser et al., 2007; Envirovision et al., 2008a), except the groundwater loading analysis which excludes loads from the western Strait of Juan de Fuca.

Summary of COC Releases

A summary of the total estimated release and largest potential source for each COC is shown in Table 30. A complete list of individual sources is shown in Appendix C.

Table 30. Summary of Estimated Anthropogenic Releases of COCs in the Puget Sound Basin.

Chemical of Concern (Release Rate)	Major Sources	Modes of Release
Arsenic (0.8 t/yr)	Industrial sources	Releases to air.
	CCA-treated wood, roofing materials	Leaching from precipitation.
Cadmium (1 t/yr)	Roofing materials	Leaching from precipitation.
Copper (180 – 250 t/yr)	Urban lawn & garden use as pesticides[a]	Direct application to soil.
	Plumbing components, roof material	Leaching from precipitation or directly to POTWs.
	Brake pads	Abrasion leading to fugitive dust emission or loss directly to roadway.
	Vessel anti-fouling paint	Leaching directly to marine water.
Lead (520 t/yr)	Ammunition and hunting shot use, loss of fishing sinkers, loss of wheel weights	Release of solid metallic lead.
	Roofing materials	Leaching from precipitation.
	Aviation fuel	Combustion emissions.
Mercury (0.5 t/yr)	Thermostats, fluorescent lamps, button cells batteries, other mercury-containing material	Volatilization and leaching following disposal.
	Crematoria, industrial plants	Combustion and other air emissions.
Zinc (1,500 t/yr)	Roofing materials	Leaching from precipitation.
	Vehicle tires	Abrasion leading to fugitive dust emission or loss directly to roadway.
Total PCBs (2 t/yr)	Electrical equipment[a]	Spills and leaks.
	Residential trash burning	Combustion emissions.
	Building sealant (caulk)	Volatilization, abrasion to dust and larger particles.
Total PBDEs (0.7 t/yr)	Furniture, computer monitors, and other components of residential and commercial indoor environments	Indoor air and dust.
PCDD/Fs (9 grams TEQ/yr)	Backyard burn barrels	Combustion emissions.
Total PAHs (310 t/yr)	Woodstoves and fireplaces, light and heavy-duty vehicles	Combustion emissions.
	Creosote-treated piling, railroad ties, and utility poles	Leaching and washout, volatilization.
DEHP (17 t/yr)	Industrial, commercial, and institutional point sources	Air emissions.
	Car undercoating, roofing	Leaching, volatilization.
Triclopyr (150 t/yr)	Crop and golf course use as herbicides	Direct application to plants or soil.
Nonylphenol (0.2 t/yr)[b]	Industrial, commercial, and institutional facilities	Air emissions.
Petroleum (9,200 t/yr)	Vehicle crankcase oil	Motor oil drips, leaks, and improper disposal of used oil.
	Vehicle and off-road equipment fueling	Gasoline (minor) spillage.
DDT	NA	NA

t=metric ton (appr. 2.2 tons)
TEQ= Toxic Equivalents
NA=Not analyzed
[a]Estimate is highly uncertain
[b]Sources were not fully assessed

Information on COC releases from primary sources in the Puget Sound basin can be summarized as follows:

- Petroleum is estimated to be released in the largest quantity, followed by zinc which is the only other COC estimated to be released at a rate greater than 1,000 metric tons (t) per year. Lead, PAHs, and copper are estimated to be released at rates greater than 200 t/yr, triclopyr is released at an estimated rate over 100 t/yr, and approximately 15 t of DEHP is released annually. PCBs, cadmium, mercury, PBDEs, arsenic, nonylphenol, and PCDD/Fs are generally released at rates near one t/yr or less.

- In general, industrial, commercial, and institutional point sources do not account for large releases of COCs. Instead, a variety of diffuse sources account for a majority of the COC releases.

- Runoff and leaching of chemicals from roofing materials is potentially an important source of metals. For two metals – cadmium and zinc – release from roofing materials were estimated to account for the majority of total release, and nearly one-third of arsenic is released through roof runoff. Zinc releases from roof runoff are particularly large (>1,000 t/yr). There were also estimated to be substantial releases of copper and lead from roof materials. Leaching of metals from rooftop runoff is likely to be largely in the form of unconstrained releases. Leaching of metals from plumbing components also accounts for substantial releases of copper and zinc, but these are likely to be constrained to sanitary sewer systems and POTWs.

- Vehicle and road-related COC releases occur primarily through wear of vehicle components, combustion of fuel, and leaks of motor oil and fuel. Abrasion of brake pads account for up to one-third of the total release of copper. The second largest source of zinc was estimated to come from tire wear. Vehicle-related fuel combustion releases large quantities of COCs, accounting for about 10% of the total PAH release due to gasoline and diesel combustion, and about 5% of the total PCDD/F release, primarily due to heavy-duty diesel vehicles.

- Petroleum represents the largest COC quantity released to roadways and other impervious surfaces. Motor oil lost via drips and leaks appears to account for over 60% of the total petroleum release and likely occurs along roadways or impervious surfaces – parking lots and driveways – connected to roadways. In addition, gasoline is released at a rate of approximately 1,900 t/yr, including 570 t/yr released during on-road vehicle fueling at the pump. PAHs contained in uncombusted petroleum are also released along with the leaked petroleum at a rate of approximately 10 t/yr.

- Emissions from backyard burn barrels account for about three-quarters of the total PCDD/Fs released, and nearly all of the PCDD/F release is from combustion sources.

- Woodstoves were estimated to be the largest source of PAHs (about one-third of the total PAH release), and aside from zinc roof runoff and petroleum leakage, represent the only COC source exceeding 100 t/yr.

- About 10% of the total PCBs released in the basin was estimated to come from residential trash burning.

- Since few of the COCs assessed are typically associated with agriculture, releases from agricultural uses are generally small. Triclopyr, the only COC used exclusively as a pesticide (herbicide), is an exception with over 100 t/yr applied to crops in the Puget Sound basin. There are also some releases of metals in fertilizers applied to agricultural crops, with zinc releases being the largest (>40 t/yr).

- Copper is used in agriculture as both a pesticide and a micronutrient. Approximately 10 t/yr of copper is used as an agricultural pesticide alone, with an additional 5 t/yr used as a micronutrient. Use of copper as an urban pesticide is potentially substantial, as much as 70 t/yr by some estimates, but other estimates put it at a much lower rate (1 t/yr). The high level of uncertainty in these estimates underscores the limited information available regarding pesticide use.

- PAH releases from creosote-treated wood (railroad ties, marine pilings, and utility poles) appear to account for over one-third of the PAHs released annually in the Puget Sound basin.

- PCB leakage from electrical equipment appears to be substantial, possibly as much as 2,000 kg/yr. However, this amount may also include indoor leakage or that which is immediately cleaned up and contained, and so may overestimate actual PCB release in the Puget Sound basin by a large degree.

- PBDE and phthalate releases are difficult to assess due to the passive nature of their emissions from the materials and products in which they are used. For PBDEs, their loss via air emissions and dust particles into air and dust from commercial offices and homes, followed by subsequent release to the outdoor environment, appears to be the major release pathway. Phthalates may be released in the same manner, although releases from domestic products – including personal care products – may be more important sources. For both of these COCs, attachment to indoor dust and subsequent release to sanitary sewers may also be an important release pathway.

COC Loading and Pathways

One component of the PSTLA was to assess chemical loading from various pathways. The chemical-specific information focused on surface water runoff, atmospheric deposition, and groundwater discharge directly to marine waters, and POTWs as the major pathways for COC delivery. The flux of COCs across the Puget Sound – ocean boundary was also assessed where data were available.

For each of the pathways mentioned above, separate projects were conducted to assess loading. Descriptions of these projects are described in the introductory sections of this report. Tables 31-33 show summaries of the COC amounts loaded from each of the pathways that have *partially controllable* sources of COCs (i.e. all of the major pathways except ocean exchange).

Summaries of the relative COC contributions delivered from each of the loading pathways are shown in Figures 47 and 48.

Caution should be used interpreting Figures 47 and 48 since load estimates are incomplete for some COCs. In particular, air deposition and groundwater loading data are not available for most of the organic COCs.

Table 31. Summary of Metals Loading to Puget Sound through Major Pathways.

Metals		POTWs	Surface Runoff	Air Deposition	Ground-water*	SUM
Total Arsenic (t/yr)	25th %ile	NA	13.5	0.2	0.1	14
	Median	NA	16.9	0.4	0.4	18
	75th %ile	NA	23.4	0.5	0.8	25
Total Cadmium (t/yr)	25th %ile	NA	0.01 [a]	0.03	0.01	0.05
	Median	NA	0.01 [a]	0.05	0.22	0.28
	75th %ile	NA	0.02 [a]	0.07	0.43	0.53
Total Copper (t/yr)	25th %ile	2.5	28.4	1.9	0.1	33
	Median	4.3	35.7	2.7	2.2	45
	75th %ile	5.5	66.1	4.1	4.3	80
Total Lead (t/yr)	25th %ile	0.1	2.8	0.6	0.1	3.6
	Median	0.2	4.7	1.1	1.1	7.0
	75th %ile	0.3	7.6	1.5	2.1	12
Total Mercury (t/yr)	25th %ile	NR	0.09	0.01	0.005	0.11
	Median	0.002 [b]	0.14	0.02	0.05	0.21
	75th %ile	NR	0.24	0.03	0.09	0.37
Total Zinc (t/yr)	25th %ile	16	113	11	2	140
	Median	19	122	18	11	170
	75th %ile	24	134	26	20	200

* Lowest, highest, and mid-point of estimated loads
t=Metric ton (appr. 2.2 tons)
NA=Not analyzed
NR=Not reported
[a] Surface runoff loads based on storm flows only; not detected in any land covers during baseflows and in commercial/industrial areas only during storm flows
[b] Estimate from Phase 2 (Envirovision et al., 2008b)

Table 32. Summary of Organic Chemical Loading to Puget Sound through Major Pathways.

Organics		POTWs	Surface Runoff	Air Deposition	Ground-water*	SUM
Total PCBs (kg/yr)	25th %ile	0.1	2.5	0.7	NA	3.4
	Median	0.3	5.3	1.3	NA	6.9
	75th %ile	1.8	15.8	3.7	NA	21
Total PBDEs (kg/yr)	25th %ile	7.0	5.1	15.7	NA	28
	Median	10.6	5.7	20.3	NA	37
	75th %ile	20.7	10.0	23.8	NA	54
PCDD/Fs (g TEQ/yr)	25th %ile	NA	NA	NA	NA	NA
	Median	NA	NA	NA	NA	NA
	75th %ile	NA	NA	NA	NA	NA
Total DDT (kg/yr)	25th %ile	ND	2.2 [a]	NA	0.3	2.5
	Median	ND	23.9 [a]	NA	3.8	28
	75th %ile	ND	25.1 [a]	NA	7.3	32
LPAH (kg/yr)	25th %ile	3	102 [b]	NA	7	110
	Median	8	104 [b]	NA	159	270
	75th %ile	35	190 [b]	NA	311	540
HPAH (kg/yr)	25th %ile	4	25 [c]	49	6	84
	Median	5	36 [c]	96	124	260
	75th %ile	7	51 [c]	153	243	450
cPAH (kg/yr)	25th %ile	NC	18 [d]	21	5	44
	Median	NC	24 [d]	43	83	150
	75th %ile	NC	34 [d]	70	161	260
Total PAH (kg/yr)	25th %ile	8	119 [b]	49	13	190
	Median	18	224 [b]	96	284	620
	75th %ile	46	244 [b]	153	554	1,000
DEHP (kg/yr)	25th %ile	216	1,750 [e]	NA	14	2,000
	Median	439	1,780 [e]	NA	227	2,400
	75th %ile	904	1,860 [e]	NA	440	3,200
Triclopyr (kg/yr)	25th %ile	NC	641 [f]	NA	NA	640
	Median	NC	652 [f]	NA	NA	650
	75th %ile	NC	686 [f]	NA	NA	690
Nonylphenol (kg/yr)	25th %ile	NC	23 [g]	NA	NA	23
	Median	NC	23 [g]	NA	NA	23
	75th %ile	NC	24 [g]	NA	NA	24

* Lowest, highest, and mid-point of estimated loads.

NA=Not analyzed; ND=Not detected; NC=Not calculated due to insufficient data.

[a] Detected in commercial/industrial areas only during baseflows and in commercial/industrial, agricultural, and forest areas only during storm flows.

[b] Detected in commercial/industrial areas only during baseflows.

[c] Detected in commercial/industrial areas only during baseflows and in commercial, residential, and agricultural areas only during storm flows.

[d] Surface runoff loads based on storm flows only; not detected in any land covers during baseflows and in commercial, residential, and agricultural areas only during storm flows.

[e] Detected in residential areas only during baseflows.

[f] Detected in commercial/industrial and agricultural areas only during baseflows.

[g] Surface runoff loads based on storm flows only; not detected in any land covers during baseflows and in commercial/industrial areas only during storm flows.

Table 33. Summary of Petroleum and Oil & Grease Loading to Puget Sound through Major Pathways.

Petroleum and Oil & Grease		POTWs	Surface Runoff	Air Deposition	Ground-water*	SUM
Oil & Grease (t/yr)	25th %ile	NA	8,470	NA	NA	8,500
	Median	NA	8,470	NA	NA	8,500
	75th %ile	NA	10,600	NA	NA	11,000
Lube Oil (t/yr)	25th %ile	NA	320 [a]	NA	6	330
	Median	NA	345 [a]	NA	34	380
	75th %ile	NA	360 [a]	NA	62	420
Diesel (t/yr)	25th %ile	NA	ND	NA	2	2
	Median	NA	ND	NA	18	18
	75th %ile	NA	ND	NA	34	34
Gasoline (t/yr)	25th %ile	NA	ND	NA	3	3
	Median	NA	ND	NA	24	24
	75th %ile	NA	ND	NA	45	45

* Lowest, highest, and mid-point of estimated loads
t=Metric ton (appr. 2.2 tons)
NA=Not analyzed
ND=Not detected
[a] Detected in agricultural areas only during baseflows

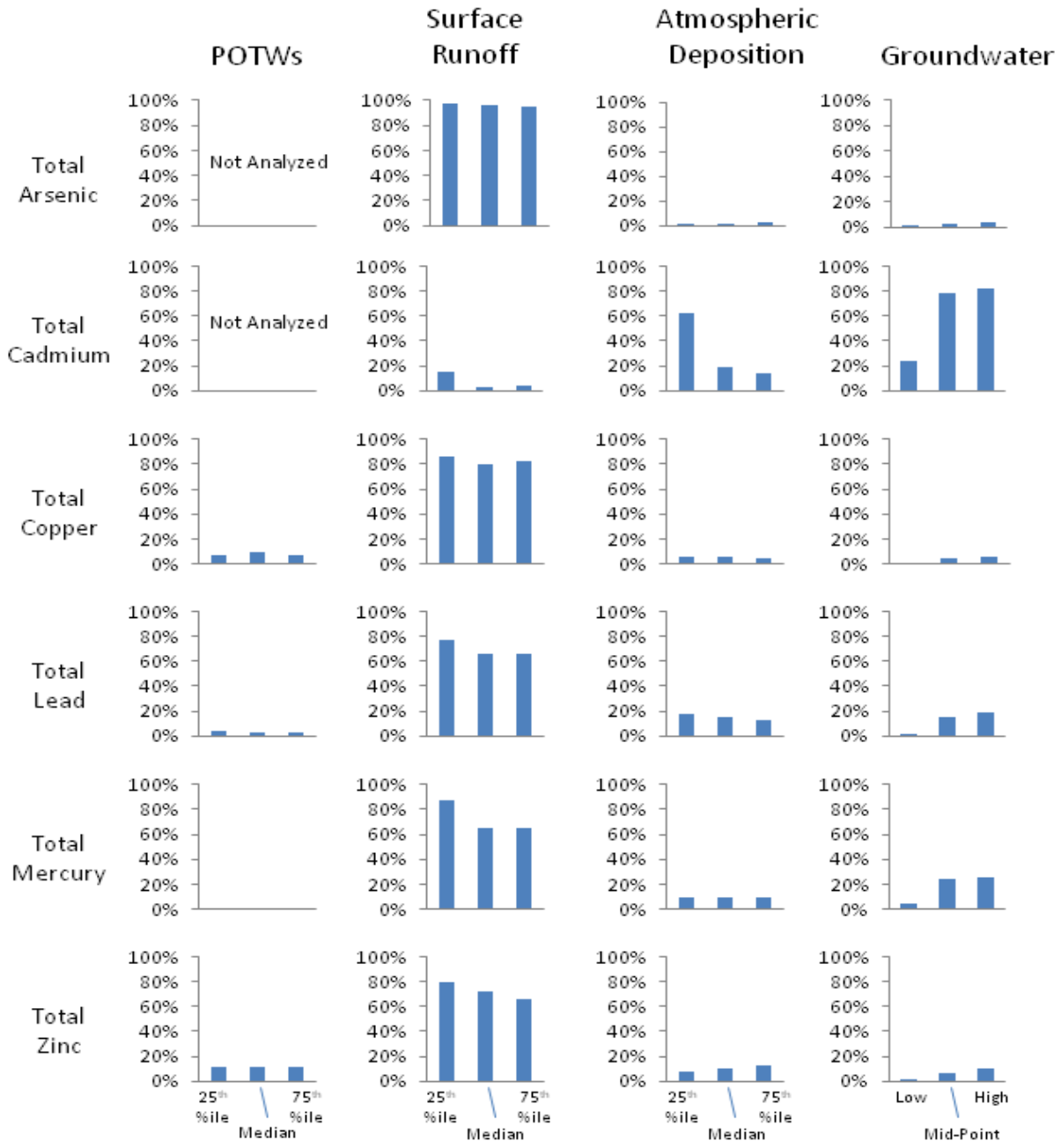

Figure 47. Percent Contribution of Major Pathways to Metals Loading in Puget Sound.

Figure 48. Percent Contribution of Major Pathways to Organic Chemical Loading in Puget Sound.

Information on the COC loading and pathways can be summarized as follows:

- Surface water runoff is the dominant pathway for all metals except cadmium. Metals are generally found at the highest concentrations in commercial/industrial and agricultural land covers, followed in decreasing order by residential and forested areas. However, forest areas account for the bulk of metals loading simply due to the comparatively high proportion of forest land cover (83% of land area) which generates the majority of the flow entering Puget Sound.

- PCBs, PBDEs, and DEHP are present in surface runoff from all land covers at very low concentrations, but tend to be highest in commercial/industrial areas, particularly during storm flows. Like most other COCs in surface runoff, the largest absolute loads are from forested areas as a result of the high proportion of forest cover and associated volume of water.

- POTW-delivered loads were comparatively small for all metals assessed. Cadmium and arsenic were not measured in POTW effluent. To some degree, loads of copper, lead, and zinc mirrored the discharge volume of POTWs relative to other annual land-based water discharge (groundwater and surface runoff) to Puget Sound (~4% of total).

- Groundwater loads directly to marine waters were estimated to constitute approximately 5-10% of the total loading for most metals. The greatest mass of metals loaded annually to Puget Sound through groundwater are from non-urban ambient areas, followed by urban ambient areas and impacted areas. The volume of groundwater discharge alone does not account for the relative contribution of groundwater loads, since groundwater accounts for only 0.2 – 2% of the total annual land-based water discharged to Puget Sound.

- Atmospheric deposition directly to the marine waters of Puget Sound is estimated to account for approximately 5 –15% of the total annual loads of copper, lead, mercury, and zinc. Lower relative loads were estimated for arsenic (≤3%), and higher loads were estimated for cadmium (14 – 62%). For all metals, the atmospheric deposition flux measured was much greater at a particular sampling location within a high-density urban area that was proximal to a nearby pulp mill, metal refiners, other industrial activities, and also close to major roadways including interstate highways. In most cases, the flux at this location was at least five-fold higher than at other locations without these urban influences.

- PBDE loads deposited directly to marine waters from the atmosphere are roughly equal to loads from surface runoff and POTWs combined. With the possible exception of HPAH, PBDEs are the only COC with the predominant load contributed by direct atmospheric deposition. In addition, PBDEs are the only COC with higher loads delivered through POTWs compared with surface runoff.

- Fluxes at the ocean boundary generally show a net export of metals out of Puget Sound. However, for cadmium and lead, there is a net import to Puget Sound from oceanic waters. Marine fluxes of cadmium and lead into Puget Sound are greater than loads from all other pathways combined.

- For most organic compounds, patterns of pathway loading are more difficult to assess than for metals due to inconsistencies in analysis among loading studies and comparatively low frequencies of detection. For instance, HPAHs were the only organic constituents analyzed in the four major pathways assessed.

- For PAHs, accurate delivery patterns and loads are difficult to assess due to infrequent detection in surface runoff. A cursory examination of loads delivered by surface runoff, atmospheric deposition, and groundwater does not reveal a clear loading pattern or principal transport pathway. However, the atmospheric deposition data appear to be the most reliable; surface runoff and groundwater loading data are derived largely from sample data with non-detect results.

- Among all of the COCs assessed, oil & grease was estimated to be delivered to Puget Sound in the largest quantity (>8,000 t/yr) with surface runoff accounting for the entire load. Although oil & grease is relatively meaningless in terms of chemical specificity – it simply refers to the fraction of a sample extractable by n-hexane – some portion of the oil & grease measured in surface runoff may be petroleum product, particularly in urban areas.

Limitations and Uncertainty of Data on COC Sources and Loads

All of the individual projects in the PSTLA contain a degree of uncertainty in the reported results, and all have limitations due to study design or due to the nature of the data collected. Limitations and uncertainty affect the usefulness of the individual projects as well as the ability to compare results among projects. Some of the most common and intractable issues are mentioned here. However, the reader is encouraged to review the individual reports in order to fully gauge uncertainty and understand how results were derived.

The projects were not designed to analyze for an identical suite of COCs. This resulted in a limited ability to fully gauge each pathway's contribution to overall loading and to compare COC quantities released among studies. For instance, air deposition and groundwater loading data are not available for most of the organic COCs, and the source inventory (Ecology, 2011) did not fully account for all major sources of zinc, nonylphenol, and petroleum releases. Appendix B shows a summary of the COCs analyzed for each project.

Much of the uncertainty surrounding the reported results for loading projects is due to sample results below reporting or detection limits (i.e. non-detects). In many cases, results were derived using datasets where more than one-half of the concentration values were reported as non-detects. There was a particularly heavy reliance on non-detects in the surface runoff and groundwater loading projects. For the assessment, this source of uncertainty is compounded by the lack of consistency in the assumptions and rules for handling data (including non-detects) among projects. It should be noted, however, that estimates of releases and loads used for this assessment are those reported in the original projects, and no attempt was made to recalculate or "normalize" results according to a common set of rules. A summary of project-by-project rules used to handle non-detects is shown in Appendix B.

It is also notable that the study conducted to assess loads from surface runoff, the major delivery pathway for most COCs, did not actually measure COC concentrations at the point of delivery to Puget Sound. The authors of the surface runoff study (Herrera, 2011) offer a detailed explanation of this and other potential sources of bias in the surface runoff project.

Relationship between Sources and Pathways/Loading

The relationships between COC sources and loading/pathways discussed in this report essentially provide a framework for the first step in developing mass-balances for COCs, from their initial release to their delivery to Puget Sound. Information provided for these conceptual models include the identification of the primary sources of COCs, mechanisms of release (e.g. combustion emission), estimates of the quantities released, COC prevalence and levels in delivery pathways, and characteristics of loading pathways (e.g. land cover) related to COC prevalence and levels.

Although the conceptual models of the relationships between sources and loading/pathways were developed on a very broad scale, some patterns appear to emerge. For instance, it appears that for most of the COCs assessed there is a one to two order of magnitude difference between the quantity released and the quantity estimated to be loaded to Puget Sound through various pathways. Notable exceptions are PCBs, triclopyr, and PAHs which have estimated releases three orders of magnitude higher than loads. However, the loading estimates for PAHs do not include the estimated release directly to marine waters from creosote-treated marine pilings.

Arsenic is the only COC with estimated loads to Puget Sound larger than releases from primary sources. This appears to be due to the natural enrichment of soils and surface runoff with arsenic, coupled with comparatively low arsenic releases from ongoing anthropogenic sources.

Estimated quantities of COCs released do not necessarily translate to equivalent loads in transport/delivery pathways due to a variety of factors affecting their behavior and fate once released in the environment. For instance, the case studies of copper mass-balance in small watersheds (Paulson et al., 2011-Draft) showed a much greater relative difference between releases and loading at the small (watershed) scale compared to the relative difference between releases and loading at the large (Puget Sound basin) scale.

In many cases, the specific COC source and the mechanism of release may have more environmental relevance then the absolute quantity released. The relationships between COC releases and their presence in specific pathways were therefore examined at finer scales than simply comparing the total annual mass released to the total annual mass loading to Puget Sound.

The following patterns reveal consistencies between releases and pathways at finer scales:

- Overall it appears that there is approximately an order of magnitude decrease between the quantity of a COC discharged to a POTW and the reported load discharged from POTWs to Puget Sound. This appears to be the case for copper, lead, and DEHP. Sources of PBDEs suggest a substantial proportion is released to POTWs as well.

- Metals (copper, lead, and zinc) and PAHs associated with vehicle sources are highly elevated in air deposition samples located near high-density urban areas and interstate highways.

- COCs which are released to air through combustion or volatilization have large load contributions from atmospheric deposition directly to marine waters compared to other pathways. PBDEs and PAHs are two primary examples. DEHP and PCDD/Fs might be expected to follow this pattern as well, but they were not measured in air deposition samples. Mercury, which is largely emitted to the air through combustion or volatilization, does not exhibit the comparatively large load from direct deposition as might be expected.

Hazard Evaluation

As noted in the introductory sections of this report, the hazards posed by different COCs are not simply associated with the quantities released to the environment or loaded to Puget Sound, but are rather more appropriately evaluated by comparing their concentrations in various environmental media to reported effects levels. To assess the relative toxic hazard posed by COCs in various media and for various receptors, the hazard evaluation assessed COCs in various media by comparing observed concentrations to data on effects or guidelines, standards, and criteria for the following categories:

- Direct hazard to aquatic life through surface water exposure
- Direct hazard to benthic organisms through sediment exposure
- Direct hazard to aquatic life based on tissue residue levels
- Hazard to wildlife based on ingestion of prey, water, and sediment
- Hazard to human health through fish/seafood consumption

The results of these comparisons were grouped into three broad "level of concern" categories: a Priority 1 level of concern, a Priority 2 level of concern, or unknown (U) level of concern due to lack of sufficient data for an assessment. Results were classified as a Priority 1 when high observed concentrations (e.g. 90^{th} percentile values) exceeded low effects concentrations (e.g. 10^{th} percentile values), selected criteria, or other threshold values. A Priority 2 level of concern was assigned in cases where high observed concentrations were below threshold values. In cases where there were not sufficient data to make a meaningful comparison, results were assigned a U. Appendix D-1 details the thresholds used for comparisons and the minimum data required for the comparisons. Table 34 provides a summary of the hazard evaluation for all of the categories and sub-categories assessed.

The hazard evaluation has several limitations that should be considered prior to acting on the results. In particular, the hazard evaluation is not a risk assessment but is instead designed to assess the *relative* level of concern of COCs *across the entire Puget Sound basin*. Although a COC may be assigned Priority 2 or U for a particular sub-category, this should not be interpreted to mean there are no hazards associated with that COC. Locally, concentration hot spots exist near major sources and may cause localized toxicity to aquatic organisms or lead to violations of standards.

In addition to the systematic methodology used to evaluate relative hazards for each COC, a review of relevant regional studies was conducted. This review was conducted to capture information on regionally important biological effects that may not have been included among the data used for the hazard evaluation. Information from these reviews was not used to assign the Priority levels summarized in Table 34, but instead was used as an additional line of evidence for assessing priorities for toxic chemical reduction and control strategies.

Table 34. Summary of the Hazard Evaluation Based on the Priority Levels of Concern for Each Sub-Category (see text for definitions of Priority levels).

COC	Surface Water			Sediment			Tissue Residue			Wildlife		Human Health			Regional Effects Data?
	Frsh.	Marine		Frsh.	Marine		Frsh.	Marine		Frsh.	Marine	Frsh.	Marine		
		nr.	off.		nr.	off.		nr.	off.				nr.	off.	
Arsenic	U	U	U	1	2	2	NA	NA	NA	NA	NA	U	U	U	No
Cadmium	2	U	U	1	2	2	NA	NA	NA	NA	NA	NA	NA	NA	No
Copper	1	1	2	1	2	2	NA	NA	NA	NA	NA	NA	NA	NA	Yes
Lead	2	U	2	2	2	2	NA	NA	NA	NA	NA	NA	NA	NA	No
Mercury	1	U	U	1	1	1	U	U	U	1	1	2	2	2	Yes
Zinc	2	U	1	1	2	2	NA	NA	NA	NA	NA	NA	NA	NA	No
PCBs	1	U	2	1	2	1	1/U	2/U	2/U	1	1	1	1	1	Yes
PBDEs	U	U	U	U	U	U	NA	NA	NA	NA	NA	NA	NA	NA	Yes
PCDD/Fs	U	U	U	U	U	U	U	U	U	1/U	2/U	1	1	1	Yes
DDT	1	U	U	U	U	U	2	U	U	1/2	1/2	1	1	2	Yes
PAHs	2/U	U	2/U	1/2	2	2	NA	NA	NA	NA	NA	1/2	1/2	2/U	Yes
DEHP	2	U	U	1	1	1	NA	NA	NA	NA	NA	1	1	U	No
Triclopyr	2	U	U	U	U	U	NA	NA	NA	NA	NA	NA	NA	NA	No
Nonylphenol	2	U	2	U	U	U	NA	NA	NA	NA	NA	NA	NA	NA	No
Petroleum	U	U	U	U	U	U	NA	NA	NA	NA	NA	NA	NA	NA	No

1=Priority 1 (highlighted in yellow)
Frsh.=freshwater
nr.=nearshore
off.=offshore
2=Priority 2
U=Unknown
NA=not analyzed

Information on the hazard evaluation and regionally important biological-effects data can be summarized as follows:

- All of the COCs except lead, PBDEs, triclopyr, nonylphenol, and petroleum were assigned a Priority 1 level of concern for at least one category. Of the COCs not assigned a Priority 1, only lead had sufficient effects data to conduct a meaningful evaluation.

- PCBs are the only COC assigned a Priority 1 in all five categories evaluated. Mercury and DDT were the only other COCs evaluated for all five categories, and they were assigned Priority 1 for four categories and three categories, respectively.

- COC concentrations in surface waters and sediments – particularly freshwater – resulted in the most COCs assigned Priority 1. Tissue residue, wildlife, and human health evaluations resulted in the fewest COCs receiving Priority 1. However, only bioaccumulative chemicals were evaluated for these latter categories, and so fewer Priority 1 assignments were expected.

- Reviews of regionally important biological-effects data showed that levels of copper, mercury, PCBs, PBDEs, PCDD/Fs, DDT, and PAHs found in the Puget Sound basin result in documented or potentially adverse effects to a variety of aquatic organisms.

Chemical-by-Chemical Summary

Arsenic

Arsenic was found to be a Priority 1 level of concern for freshwater sediments, one of the three categories for which data were sufficient for an adequate hazard evaluation. Observed data were lacking to conduct adequate evaluations for surface waters and human health.

Releases of anthropogenic arsenic are small compared to loads. The largest ongoing anthropogenic source appears to be point-source air emissions, although these are relatively small on a basin-wide scale. Loading to Puget Sound is substantial, presumably due to natural sources and possibly from historical releases, and is dominated by surface runoff. The finding that, unlike most metals, arsenic concentrations in surface runoff decrease during storm events may support the notion that a substantial portion of the arsenic in surface waters is due to natural or historic sources.

Cadmium

Cadmium was found to be a Priority 1 level of concern for freshwater sediments, one of the two categories for which data were sufficient for an adequate hazard evaluation. Observed data were lacking to conduct adequate evaluations for surface waters.

Both anthropogenic releases and loading of cadmium appear to be small. The largest ongoing source of cadmium to the environment appears to be leaching of cadmium from roofing material, constituting more than one-half of the total estimated release. Releases of cadmium from roofing

material do not appear to translate to appreciable levels in surface runoff, which represents the smallest delivery pathway.

Copper

Sufficient data were available for full hazard evaluations of copper in surface waters and sediments. Copper was found to be a Priority 1 level of concern for freshwater sediments, nearshore marine sediments, and fresh surface waters. Regionally relevant biological-effects data provide additional lines of evidence to indicate a comparatively high hazard potential for copper. These regional studies have demonstrated that dissolved copper concentrations commonly found in urban and agricultural area streams reduce olfactory function in salmonids (e.g., Hecht et al., 2007).

The source inventory (Ecology, 2011) suggests that large ongoing copper releases occur from a variety of sources including abrasion of vehicle brake pads, leaching from roofing materials, leaching of vessel anti-fouling paint, pesticide applications and micronutrient use in agricultural applications, and possibly pesticide use in urban areas. These releases result in elevated levels in surface water at locations where they are most likely to occur such as agricultural, commercial/industrial, and high-density urban areas, as well as nearshore marine waters.

Lead

Sufficient data were available for full hazard evaluations of lead in surface waters and sediments except nearshore marine waters. Lead was not a Priority 1 level of concern for any of the categories assessed. There is no indication that lead concentrations in fish and shellfish from the Puget Sound basin pose a risk to human health, although lead was not evaluated in tissue due to lack of National Toxics Rule criteria for lead. Lead remains a human health hazard through exposures from material such as lead house paint (Ecology and WDOH, 2009), but no hazard emerges from exposures that include an aquatic environment pathway.

Ongoing anthropogenic releases of lead are substantial, largely due to ammunition use, fishing sinkers, and wheel weight loss. These solid metallic lead sources and mechanisms of release appear to result in limited enrichment of the aquatic environment except in some localized instances. Surface water runoff represents the largest delivery pathway, but loads are small compared to the estimated releases from ongoing anthropogenic sources.

Mercury

Mercury was found to be a Priority 1 level of concern for fresh surface waters, freshwater and marine sediments, and all freshwater and marine wildlife species evaluated. There were not sufficient observed data to conduct adequate hazard evaluations for marine sediments or effects data to evaluate tissue residue effects. Mercury concentrations in freshwater and marine seafood were generally well below the threshold used to assign a Priority 1 level of concern for the human health evaluation. However, consumption advisories for various fish species and locations in the Puget Sound basin have been established by the Washington State Department of Health due to mercury residues in tissues.

There appears to be a variety of ongoing anthropogenic mercury releases, although a number of the regional sources have been addressed in the Mercury Chemical Action Plan (Ecology and WDOH, 2003) and it is unclear if release estimates accurately portray current releases to the environment. Mercury releases due to improper disposal of materials appear to be the largest category of ongoing anthropogenic release, followed by industrial emissions. Surface runoff is the largest delivery pathway for mercury and to some degree may reflect entrainment of mercury deposited atmospherically.

Zinc

Zinc was found to be a Priority 1 level of concern for offshore marine surface waters and for freshwater sediments. Sufficient data were available for surface water and sediment, except nearshore marine surface waters.

Zinc appears to be released at high rates from a variety of roofing materials and to a lesser degree in vehicle tire wear. The study of primary sources (Ecology, 2011) indicated that zinc releases may have been underestimated because many sources (e.g. leaching from galvanized materials) were not assessed. Surface runoff was the dominant pathway for zinc loading to Puget Sound. Zinc levels in streams from commercial/industrial areas were found to be highly elevated, possibly as a reflection of galvanized material leachate and vehicle tires where these sources are likely to be most prevalent.

Polychlorinated biphenyls (PCBs)

PCBs are a Priority 1 level of concern for all five categories assessed and for nine of the 13 sub-categories where sufficient data were available. In addition, regional data show PCB levels that may result in adverse effects to locally important fish and marine mammals. Fish consumption advisories to protect human health have also been issued for both marine and freshwaters of the Puget Sound basin.

Release estimates are highly uncertain and may overestimate the degree to which PCBs continue to be released from ongoing anthropogenic sources (Ecology, 2011). Electrical equipment (capacitors and transformers) leakage is the largest PCB source category, with loss from sealants and release from residential trash burning also making up a substantial portion. However, loading is small compared with releases and may support the notion that releases were overestimated.

Due to their persistence, PCBs that were released from historical sources and continue to be released from highly contaminated areas, such as the lower Duwamish River, continue to cycle in the aquatic environment of Puget Sound. The substantial accumulation in biological tissues may be primarily a result of legacy contamination as opposed to ongoing releases.

While PCBs remain a concern, levels appear to be declining in Puget Sound harbor seals (Noel et al., 2011) and mussels (Mearns et al., 2009). While temporal PCB trends in fish do not show a clear trend (West and O'Neill, 2007), modeling results suggest that substantial declines in English sole should be expected by 2020 at current loading rates (Pelletier and Mohamedali, 2009).

Polybrominated diphenyl ethers (PBDEs)

PBDEs are an unknown level of concern due to a lack of environmental effects data, criteria, and guidelines for PBDEs. However, the lack of effects data used in the hazard evaluation methodology should not be interpreted as a low level of concern since a growing body of evidence suggests environmental concentrations may cause adverse effects to humans (Ecology and WDOH, 2006), marine animals (Ross, 2006), and birds (Fernie et al., 2009). The historical record is generally not adequate to assess PBDE trends in the Puget Sound environment since PBDEs have not typically been included in sampling investigations prior to the beginning of the 21st century.

Cessation of production through voluntary actions and bans since the mid-2000s has removed major PBDE formulations from new consumer products such as mattresses, televisions, computers, and residential upholstered furniture. However, much of the PBDEs produced historically may remain in consumer products and commercial office products and these potentially represent substantial diffuse ongoing sources.

PBDE loading patterns are different than for other COCs assessed. Direct atmospheric deposition represents the largest delivery pathway, followed by POTWs and surface runoff. The high proportion of PBDE loading through atmospheric deposition and POTWs appears to be consistent with the major sources and release mechanisms.

Polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans (PCDD/Fs)

PCDD/Fs are a Priority 1 level of concern for a species representing a freshwater mammal (river otter) and for human health due to residue levels in freshwater and marine seafood. In addition, regional data show that Puget Sound harbor seal and southern resident killer whale prey items have higher PCDD/Fs compared to the same prey from the Strait of Georgia and the British Columbia coast. There were not sufficient data to conduct hazard evaluations for surface waters or sediments, or to evaluate tissue residue effects.

In the Puget Sound region, the major historical sources of PCDD/Fs – use of elemental chlorine in pulp bleaching, pentachlorophenol wood treatment operations, and combustion of saltwater-infused hog fuel – have been eliminated to a large extent (EPA, 1991; Yake et al., 1998; EPA, 2006), and ongoing releases are from combustion sources such as backyard burn barrels. Accumulation in biota is likely to be mainly a result of historical releases which continue to cycle in the aquatic environment, although no loading analyses were conducted to corroborate the small releases estimated for the Puget Sound basin.

Dichlorodiphenyltrichloroethane (DDT) and metabolites DDD and DDE

DDT compounds were found to be a Priority 1 level of concern for fresh surface waters, the freshwater and saltwater bird species evaluated, and human health. There were not sufficient observed or effects data to conduct hazard evaluations for marine surface waters, sediments (fresh and marine), or direct effects based on marine tissue residues. Regional data show high levels in accumulation for a variety of Puget Sound fish and marine mammals, including evidence to show apparent links between high levels in top-level, marine-mammal predators and their prey items.

Although a ban on DDT use in the United States has been in effect for decades, DDT compounds continue to exceed numerous documented effects levels due to their persistence, particularly in freshwater. DDT also persists in tissues of aquatic biota due to its highly bioaccumulative nature, but concentrations in Puget Sound basin fish are generally low, particularly when compared to watersheds with intensive agricultural use outside of the basin, such as watersheds in eastern Washington (e.g. Schneider and Coots, 2006; Johnson et al., 2010b). There are no apparent ongoing anthropogenic releases of DDT in the Puget Sound basin, and overall loading to Puget Sound appears to be low. DDT compounds will likely persist in the aquatic environment due to mobilization of DDT-bound soil particles and continued cycling in the aquatic environment due to historical releases.

Polycyclic aromatic hydrocarbons (PAHs)

PAHs are a Priority 1 level of concern for freshwater sediments and human health. There were not sufficient observed data to conduct adequate hazard evaluations for all individual PAHs in surface waters or for human health due to residues in offshore marine seafood. Localized areas with high degrees of PAH-contaminated sediments have also been a historical problem in Puget Sound, and resulting liver lesions in English sole have been a well-documented pathology associated with exposure to these sediments (Malins et al., 1987; Landahl et al., 1990; Myers et al., 1990). Recent evidence suggests that risks of liver lesions dramatically decrease when PAH-contaminated sediments are capped or removed (PSAT, 2007), and liver disease in English sole is currently being proposed as a Puget Sound-wide indicator of ecosystem health (PSP, 2011b).

There appears to be large, ongoing anthropogenic releases of PAHs in the Puget Sound basin. Generally speaking, PAH sources may be broken down into two categories: combustion emissions and releases from creosote-treated materials. PAH loads to Puget Sound are three orders of magnitude lower than estimated releases, but these loading estimates do not take PAH releases from creosote-treated pilings directly to marine waters into account. Groundwater appears to be the largest delivery pathway for PAHs, but the groundwater loading estimates are based largely on estimates derived from non-detects and should be viewed with caution. Estimated PAH loads through surface runoff are comparatively small due to the infrequency at which PAHs were detected in surface water samples.

To some degree, the lack of detectable PAHs in surface runoff may reflect the major sources and release mechanism. Few of the major sources would be expected to release PAHs directly to impervious surfaces, with the possible exception of PAH releases through motor oil loss.

Bis (2-ethylhexyl) phthalate (DEHP)

DEHP is a Priority 1 level of concern for freshwater sediments and human health. There were not sufficient observed data to conduct adequate hazard evaluations for marine surface waters or human health due to residues in offshore marine seafood.

There appears to be large ongoing anthropogenic DEHP releases in the Puget Sound basin. In addition to air emissions from point sources, DEHP is released from a variety of materials, particularly those containing PVC, although DEHP may be released from some non-polymer products as well. Surface runoff is the largest delivery pathway, although DEHP was not measured in air deposition samples. The major DEHP sources and mechanisms of release suggest that atmospheric deposition may be a major pathway. DEHP loads from POTW discharge are also relatively high and may indicate loss through indoor consumer products, and subsequent rinsing down the drain is another important delivery pathway.

Triclopyr

Triclopyr was evaluated only for hazards in fresh surface waters and was assigned a Priority 2 level of concern. There were not sufficient observed or effects data to conduct adequate hazard evaluations for marine surface waters or sediments.

Unlike other COCs evaluated, triclopyr's only intended use is as an herbicide, and therefore its usage equates to environmental release. Major uses in the Puget Sound basin appear to be from crop and golf course use, with minor applications to road and railroad right-of-ways and for limited aquatic weed control. Surface runoff was the only delivery pathway where loads were calculated; loads were estimated to be two orders of magnitude below release estimates.

Nonylphenol

Nonylphenol was evaluated only for hazards in fresh surface waters and marine offshore waters; it was assigned a Priority 2 level of concern for both. There were not sufficient observed or effects data to conduct adequate hazard evaluations for nearshore marine surface waters or sediments. Nonylphenol is not highly toxic, but instead may exert effects at the sub-lethal level, and has documented endocrine-disrupting effects (King County, 2007). Compared with most other COCs, little is known about nonylphenol sources and effects in the Puget Sound basin.

There were limited release and loading estimates calculated for nonylphenol. Sources of nonylphenol were not fully assessed in the study of primary sources (Ecology, 2011), and loading was calculated only for surface runoff. Although a major source of nonylphenol is reported to be the breakdown of alkylphenol ethoxylates during the sewage treatment process (EPA, 2005), it was not detected frequently enough in POTW effluent to calculate loads from this pathway.

Oil and petroleum hydrocarbons

Petroleum was not fully evaluated for hazards in surface waters or sediments due to a lack of effects data (fresh surface waters) or observed data (sediments, marine surface waters). Evidence suggests that weathered crude oil has specific toxic effects to marine organisms, such as cardiac impairment and other effects in fish (Incardona et al., 2005; Incardona et al., 2006; Carls et al., 2008). However, these effects are due to specific components of weathered oil, most notably PAHs. The complexities of evaluating effects from petroleum exposure are discussed in the report section dealing specifically with petroleum.

The source inventory indicates that petroleum is released to the Puget Sound basin in much larger quantities than other COCs addressed, and the loading studies indicate it is loaded in the largest quantity. Most of the petroleum release is in the form of motor oil from engine drips and leaks and therefore likely to be directly to pavement. The large lube oil loads in surface water runoff may be a reflection of these releases. However, the large estimated releases of gasoline due to small fueling and transport spills did not translate to measurable loads in surface runoff.

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Prioritizing Chemicals of Concern and Recommendations to Fill Data Needs

Prioritizing COCs

A lines-of-evidence approach was used to move further toward a goal of deciding how best to prioritize actions and resources for controlling toxic chemicals in the Puget Sound basin. This lines-of-evidence approach considers information on the four major components of the assessment (sources, loading, pathways, and the relative hazards), but the approach mostly relies on COC sources and the relative hazards posed by COCs as determined by the hazard evaluation and review of other regional studies.

This approach adopts the rationale that chemicals with the greatest potential to elicit toxic effects at existing concentrations should be an important factor in determining the priority for source control efforts. Priority was also given to COCs associated with large opportunities for source control. This reflects the extent to which there are existing regulatory actions to control releases, such as bans, management of materials, or other permanent actions which reduce releases to the environment. Given the uncertainty associated with individual estimates of releases or loadings, this lines-of-evidence approach provides a supportable rationale for establishing relative priorities for control actions.

Opportunities for source control are considered large where the major sources of a COC have not been addressed by control actions; where some of the major sources have been addressed, opportunities may be considered medium. In cases where actions have been implemented to control and reduce all or most of the major sources and this appears to have resulted in low rates of loading to Puget Sound, the opportunities for controlling a COC are considered small. This assessment relies principally on the *Sources Report* (Ecology, 2011) with limited input by Ecology staff and management to gauge the opportunities for source control; this assessment was not intended to be a detailed review of management initiatives. Table 35 summarizes major sources for each COC and possible opportunities for reducing those sources.

Based on the lines-of-evidence approach, copper, PAHs, DEHP, and petroleum sources were rated as have the highest priority for early actions. The reasoning for this determination is as follows:

- A substantial portion of the fresh and marine water copper data observed basin-wide falls within concentrations where effects have been documented (including reduced olfactory function in salmonids). Copper is released in large quantities from a variety of sources which appear to translate to substantial loads to the Puget Sound ecosystem. The use of copper in pesticide applications and the release of copper from roofing materials are sources which warrant further investigation. In addition, the effectiveness of recent legislation to limit copper in brake pads and vessel anti-fouling paint should be evaluated.
- A number of individual PAHs surpass (do not meet) freshwater sediment guidelines and human health criteria. In addition, a variety of studies have demonstrated links between PAH

exposure and adverse effects to regionally relevant aquatic species. There appear to be numerous opportunities for control actions, primarily for combustion sources and for creosote-treated wood.

- Observed DEHP concentrations in both freshwater and marine environments exceed (do not meet) criteria for protection of benthic species and human health. Substantial amounts of DEHP are released in the Puget Sound basin, much of which occurs initially through releases to air from off-gassing of plasticized polymers and point-source air emissions. Several non-polymer uses of DEHP may also provide opportunities for source reduction.
- The relative hazard posed by petroleum in the Puget Sound basin was not able to be evaluated due primarily to the lack of biological-effects data and the absence of criteria to protect aquatic organisms, wildlife, or human health. However, some of the COCs addressed in this assessment are components of petroleum and may be released in substantial quantities along with the release of petroleum. In particular, substantial releases of PAHs are estimated to be released from petroleum. The major sources of petroleum are diffuse, such as motor oil drips and leaks and minor gasoline spillage during vehicle fueling, and therefore offer ample opportunities for reduction efforts.

Several COCs were found to be a Priority 1 level of concern based on the hazard evaluation but were not determined to be among the highest priorities for reduction actions since the major sources have been addressed through regulatory programs or other efforts. For instance, mercury poses a relatively high hazard to freshwater and marine aquatic organisms and wildlife based on doses calculated from observed data. However, many of the historical regional sources of mercury to the Puget Sound basin have been eliminated or are being addressed by the Mercury Chemical Action Plan (Ecology and WDOH, 2003). Similarly, PCBs are a Priority 1 level of concern for all hazard evaluation categories, but PCBs have been banned for decades, the major sources (use in electrical equipment) are highly regulated, and current loads to Puget Sound appear to be small.

Although the systematic prioritization approach identified four COCs for early actions, other factors should be considered to determine the need and feasibility for developing control and reduction strategies for other COCs. For instance, PBDEs are ubiquitous environmental contaminants, and although voluntary actions and bans have removed major PBDE formulations from new consumer products, much of the PBDEs produced historically may remain in consumer products and commercial office products and these potentially represent substantial diffuse ongoing sources. The hazard evaluation was not able to adequately assess the relative hazards associated with PBDEs due to a lack of environmental standards, although there is evidence in the available literature to suggest this COC may pose a hazard at observed concentrations.

Additional research is needed to assess the relative hazards posed by PBDEs and other COCs for which there are only limited environmental data. By the same token, COCs with limited source information should be further evaluated to assess additional opportunities for source control. Of the COCs addressed in this report, PBDEs and nonylphenol were the COCs that should receive top attention for further research on potential hazard as well as possible opportunities for source control.

Table 35. Summary of Possible Actions to Reduce COCs in the Environment.

COC	Opportunities for Source Control	Major Ongoing Anthropogenic Sources	Possible Actions for Reductions
Arsenic	Medium	Industrial air emissions	Maintain existing permit controls.
		CCA-treated wood leaching	Continue ban for most non-structural uses.
		Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
Cadmium	Medium	Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
Copper	Large	Pesticides use on urban lawns and gardens	More data needed on actual pesticide use.
		Residential plumbing component leaching	Continue to implement Lead and Copper Rule.
		Brake pad abrasion	Continue to implement legislation enacted to reduce source.
		Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
		Vessel anti-fouling paint leaching	Continue to implement legislation enacted to reduce source.
Lead	Small	Ammunition and hunting shot use	Implement CAP and enforce existing regulations.
		Loss of fishing sinkers and wheel weights	Implement CAP and enforce existing regulations.
		Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
		Aviation fuel combustion	Implement CAP and enforce existing regulations.
Mercury	Medium	Consumer product improper disposal	Continue to implement CAP and enforce existing regulations.
		Crematoria and industrial air emissions	Continue existing permit limits.
Zinc	Large	Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
		Vehicle tire abrasion	Investigate source where it poses local concern.
Total PCBs	Small	Electrical equipment spills and leakage	Continue programs for management and disposal.
		Residential trash burning	Continue enforcing existing ban.
		Building sealant (caulk) volatilization and abrasion	Investigate source where it poses local concern.
Total PBDEs	Medium	Furniture, computer monitors, and other components of residential and commercial indoor environments	Enforce ban on new products but consider control actions to reduce the release from existing products.
PCDD/Fs	Small	Backyard burn barrels	Continue enforcing existing ban.
Total DDT	Small	None apparent	Investigate source where it poses local concern.
Total PAHs	Large	Woodstoves and fireplace combustion emissions	Continue change out programs, investigate catalysts/capture devices, promote alternatives to wood heat.
		Vehicle combustion emissions	Anti-idling programs, continue/expand engine retrofits for private section engines, enforce existing vehicle controls.
		Creosote-treated piling, railroad ties, and utility poles	Control actions needed, gather information to identify highest priority areas.
DEHP	Large	Polymer (primarily PVC) off-gassing	Gather additional information on extent of releases.
		Industrial, commercial, and institutional air emissions	Maintain existing permit controls.
		Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
Triclopyr	Medium	Herbicide use on crops and golf courses	More data needed on pesticide use.
Nonylphenol	Unknown	Industrial, commercial, and institutional air emissions	More information needed on emissions from these sources and unidentified releases.
Petroleum	Large	Motor oil drips and leaks Used motor oil improper disposal	Expand existing education/workshop programs.
		Gasoline spillage (minor) during fueling	Possible opportunity for source control, but more data needed on extent of releases.

Bold=Recommended as priority for near-term actions based on lines-of-evidence approach.

Strategies to control toxic chemicals in the environment may be accomplished using two general approaches: control at the source and control along the pathway. Source control strategies aimed at limiting or eliminating the initial release of chemicals may be achieved through education, chemical alternatives, release prevention technologies, or banning the use of specific chemical products. Control of a chemical once it has been released in the environment is typically more difficult and expensive, involving the use of a management actions (e.g. timing of pesticide application) or physical and technological resources (e.g. grass-lined ditches along roadways, wastewater treatment plants).

Prevention is the preferred option for controlling toxic chemicals in the environment, but source control options are not always feasible or necessary. While finding, reducing and eliminating primary releases of COCs at their source is critical to a clean and sustainable Puget Sound, so too is ensuring compliance with hazardous waste regulations, inspecting permitting facilities to ensure air and water quality, responding to spills, and cleaning up toxic messes when COCs are mismanaged.

The following recommendations provide a mixture of possible source and pathway control priorities for the target list of COCs addressed in this report.

General Recommendations

- Use results from the Puget Sound Toxics Loading Analysis (PSTLA) to help develop a long-term strategy to reduce toxic threats to Puget Sound. This strategy should include a comprehensive list of actions to prevent and manage chemical releases in conjunction with cleanup actions to reduce overall contaminant levels.

- Develop a list of specific control actions or source/pathway investigations to support control actions that may be incorporated into Ecology's long-term strategy for control of toxic chemicals under the National Estuary Program. These actions or investigations should be consistent with the findings and broader recommendations presented in this report. Examples of specific actions or investigations might be (1) adopting Low Impact Development Best Management Practices in commercial/industrial areas or (2) assessing the effectiveness of piling removal programs to reduce PAHs in the aquatic environment.

- Couple source control actions with effectiveness monitoring to assess if and how source control actions are actually reducing contaminant levels. This information is needed for an adaptive management framework to evaluate which actions should continue or be discontinued in favor of more effective actions.

Specific Recommendations

- Roofing materials appear to be an important source of metals and possibly DEHP in the Puget Sound basin. Monitoring should be conducted to further evaluate the release of these contaminants from roofing materials.

- Reduce copper loads to freshwater streams and marine waters, and assess the impacts of these efforts. The impact of recently passed legislation limiting copper and other metals in brake pads will not likely be known for at least a decade. In the meantime, the impact of copper released from pesticide/micronutrient use and vessel bottom paint should be investigated. One of the largest sources of copper is potentially from the urban use of copper in agricultural products by homeowners. Better information should be collected to evaluate the importance of these releases.

- Strategies to control the release of petroleum should be a high priority. Results of the *Sources* study (Ecology, 2011) indicate that over 9,000 metric tons of petroleum is released annually in the Puget Sound basin. Petroleum is generally released to impervious surfaces, enhancing its capacity to become mobilized in stormwater. There appears to be considerable opportunities for controlling sources of lube oil and gasoline since they are primarily released from crankcase drips and leaks and from minor spillage during fueling operations. The importance of these sources should be further evaluated.

- Strengthen existing programs to remove creosote pilings and bulkheads from the aquatic environment. Information analyzed on PAH sources and loading suggests creosote pilings may account for the largest overall PAH release to surface waters. The feasibility and need to remove creosote-treated rail ties that are over water or adjacent to sensitive aquatic areas should also be evaluated.

- Evaluate the effectiveness of actions implemented to reduce the amount of mercury released to the environment. Multiple programs and agencies have been involved with mercury disposal and recycling programs since the initiation of the Mercury Chemical Action Plan in 2003 (Ecology and WDOH, 2003). Organizing and streamlining information would make it easier to track ongoing releases to the environment and end-of-life data for mercury-containing materials.

- Evaluate the factors that appear to be effectively attenuating metals from roof and road runoff in low-density residential areas, and consider how to apply these factors in commercial/ industrial areas where there appears to be little attenuation between sources and streams. If warranted, consider applying these attenuation mechanisms to high-density residential areas.

- Conduct inspections once every three years at those businesses in Washington that routinely handle large amounts of COCs. Washington is failing to find and resolve environmental threats from millions of pounds of hazardous waste in the Puget Sound basin. Hazardous wastes are toxic, flammable, or reactive, and when mismanaged, they contaminate soil, air, and water. Ten years ago, hazardous waste inspectors found serious environmental threats at 27% of businesses; the current rate is 63% (Darin Rice, Ecology Hazardous Waste and Toxics Reduction Program, written communication). Additional resources are needed to reduce the environmental threat rate to 30% by 2015, resulting in less contamination to soil, air, and water.

Data Needs

PSTLA was a four-year effort primarily focused on assessing chemical loads to Puget Sound from all of the major delivery pathways (groundwater, air deposition, surface water runoff, publicly-owned treatment works, and ocean exchange). This assessment will be useful in putting other scientific studies and action priorities into perspective, and possibly helping to shape new ones. However, there appears to be little value in pursuing further refinements to basin-wide loading assessments for the chemicals already addressed. This does not preclude the need for additional loading refinements for particular pathways or basin-wide modeling exercises, but it appears that current data needs should be conducted at a finer resolution to focus specific actions. These smaller scales may be geographical (e.g. watersheds, specific land covers, urban bays), specific pathways (e.g. stormwater), related to specific sources (e.g. the contribution of emissions sources to chemicals in stream runoff), or assessments of hazards (e.g. local hazard evaluation).

The following recommendations are provided to fill these finer-scale data needs:

- Characterize the factors that lead to high COC concentrations in streams draining commercial/industrial and agricultural areas. Assess runoff in high-density urban areas and, if warranted, assess the factors leading to high COC concentrations as well.

- Collect information on agricultural and urban usage of copper-based products in the Puget Sound basin.

- Evaluate concentrations of cadmium, copper, lead, zinc, and DEHP released from various roofing materials.

- For any work conducted to assess PAHs and petroleum hydrocarbons in surface runoff or POTWs, include sampling and analytical methods better suited to detection of these chemicals.

- Stormwater discharges directly to Puget Sound or to major rivers near their mouths should be assessed for chemicals to evaluate the importance of this loading pathway. Although the surface runoff study (Herrera, 2011) theoretically encompassed these conveyances, they were likely underestimated since high-density urban areas were under-represented in the study. Much of the information required for such an assessment may soon be available through data collection and reporting requirements of the Phase 1 municipal stormwater permit. A detailed analysis of this dataset should be conducted.

- Incorporate the data collected under PSTLA into the Puget Sound Box Model for the purpose of evaluating reductions needed to meet the Puget Sound “dashboard indicators” and other appropriate environmental targets. In addition to PCBs, selected metals, PBDEs, and PAHs appear to be good candidates for modeling since there are ample opportunities for control actions and the model may be able to predict conditions needed to meet reduction targets.

- More information is needed to help distinguish natural and legacy sources of contaminants in environmental pathways such as surface water runoff. This will help gauge the feasibility and effectiveness of actions taken to reduce releases of chemicals from contemporary anthropogenic (human-caused) releases.

- Continue to identify and assess chemicals that may be more detrimental to the Puget Sound ecosystem than the COCs addressed in PSTLA studies. Current-use pesticides, pharmaceuticals, and personal care products are examples of chemicals that are delivered to fresh and marine surface waters of the Puget Sound basin, yet their potential for effects is poorly understood (Lubliner et al., 2010).

- In general, industrial, commercial, and institutional point sources do not account for large releases of COCs. Instead, a variety of diffuse (nonpoint) sources account for a majority of the COC releases. However, it will be important to ensure that both the results of this *Assessment Report* and data on existing prevention and management controls help guide future actions and investments on Puget Sound clean-up and restoration work.

  Lack of investment in existing programs designed to safely manage COCs produced by commerce can let otherwise controlled and contained COCs "out of their bottle," where they become a threat to Puget Sound. For example, Washington ranks near the bottom of states in the U.S. for safe hazardous waste management (Darin Rice, Ecology Hazardous Waste and Toxics Reduction Program, written communication). Hazardous wastes are persistent, toxic, flammable, or reactive, and when mismanaged, they contaminate land, air, and water. Environmental threats are posed from millions of pounds of hazardous waste and hazardous products in the Puget Sound basin. Conducting fewer hazardous waste inspections results in more violations that directly contaminate land and water. So it is not surprising that as state inspection resources have diminished, the chance of finding spills of COCs and other significant environmental threats are at historic highs (Darin Rice, Ecology Hazardous Waste and Toxics Reduction Program, written communication).

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Appendices

Appendix A. Summary of the Puget Sound Toxics Loading Analysis (PSTLA) Projects

Project	Ref	Preparer	Status	Type of Study
Phase 1				
Initial Estimate of Toxic Chemical Loadings to Puget Sound	Hart Crowser et al., 2007	Hart Crowser, Ecology, EPA, Partnership	Completed - 2007	Loading estimates. Simple model using available data.
Phase 2				
Improved Estimates of Loadings from Surface Runoff and Roadways Addendum 1 (related to oil and petroleum) Addendum 2 (related to loading calculation method)	Envirovision et al., 2008a Ecology, 2009 Herrera, 2010	EnviroVision, Herrera, Ecology Ecology Herrera	Completed - 2008 Addendum 1 - 2009 Addendum 2 - 2010	Loading estimates. Simple model using available data. Clarification of oil and petroleum definitions. Revised and improved methodology for estimating runoff volumes.
Improved Estimates of Loadings from Dischargers of Municipal and Industrial Wastewater	Envirovision et al., 2008b	EnviroVision, Herrera, Ecology	Completed - 2008	Loading estimates. Simple model using available data.
Sediment Flux/Puget Sound Sediments Bioaccumulation Model – Derived Concentrations for Toxics	Ecology and Environment, 2009	Ecology and Environment	Completed - 2009	Criteria Evaluation. Bioaccumulation model using available data.
Identification and Evaluation of Water Column Data for Puget Sound and Its Ocean Boundary	Serdar, 2008	Ecology	Completed - 2008	Inventory and evaluation of existing data.
Studies to Support a Human Health Risk Assessment		EPA/Kissinger	Status unknown	Studies using available information to assess risks to human health from ingestion of toxicants in seafood.
Development of Simple Numerical Models – The Long-Term Fate and Bioaccumulation of Polychlorinated Biphenyls in Puget Sound	Pelletier, and Mohamedali, 2009	Ecology	Completed - 2009	Fate and bioaccumulation model for PCBs. Complex model using available data.
A Toxics-Focused Biological Observing System for Puget Sound	Johnson et al., 2010a	NOAA, UC Davis, WDFW	Completed - 2010	Proposal to monitor toxicants. Based on review of existing data.
Phase 3				
Characterize Toxic Chemical Loadings via Surface Runoff	Herrera, 2011	Herrera, Ecology	Completed - 2011	Sampling and loading estimates. Simple model using newly acquired field data.
Modeling Surface Runoff in Two Pilot Watersheds	Under Development	Under Development	Under development	Under Development
Study of Atmospheric Deposition of Air Toxics to the Waters of Puget Sound	Brandenberger et al., 2010	Battelle, Ecology	Completed - 2010 (PCBs complete 2011)	Sampling and loading estimates. Simple model using newly acquired field data.
Characterization of Toxic Chemicals in Marine Waters and Selected Tributaries to Puget Sound	Gries and Osterberg, 2011	Ecology	Completed - 2011	Sampling and loading estimates. Simple model using newly acquired field data.
Refine Numerical Model of Toxics in Puget Sound and Evaluate Pollution Reduction Scenarios	Under Development	Under Development	Under development	Under Development
Priority Pollutant Scans of Ten POTWs	Ecology and Herrera, 2010	Ecology, Herrera	Completed - 2011	Sampling and loading estimates. Simple model using newly acquired field data.
Primary Sources of Selected Toxic Chemicals and Quantities Released in the Puget Sound Basin	Ecology, 2011	Ecology	Completed - 2011	Inventory of chemical releases using available data.
Pharmaceuticals and Personal Care Products in Wastewater Treatment Systems	Lubliner et al., 2010	Ecology, EPA	Completed - 2010	Evaluation of POTW treatment efficacy and sampling. Evaluation based on newly acquired field data.
Persistent Organic Pollutants in Three Guilds of Pelagic Marine Species from the Puget Sound	West, et al., 2011a and b; Noel et al., 2011	WDFW	Completed - 2011	Assessment of bioaccumulative chemicals in plankton, fish, and harbor seals. Based on newly acquired field data
Toxic Chemical Loadings via Groundwater Discharge Directly to Puget Sound	Pitz, 2011	Ecology	Completed - 2011	Loading estimates. Simple model using available data.
Assessment Report	Present Report	Ecology, King County DNR	Completed - 2011	Synthesis of existing PSTLA loading and sources information, hazard evaluation

Appendix B. Chemicals Analyzed for Loading Studies and Methods Used to Handle Non-Detects

Table B-1. Chemicals Analyzed for Loading Studies.

Chemical Class	Loading Study				
Chemical	Surface Runoff [a]	Atm. Dep. [b]	POTWs [c]	Ocean Exch. [d]	Ground-water [e]
Metals					
Aluminum	X				
Arsenic	**X**	**X**		**X**	**X**
Barium	X				
Beryllium	X				
Cadmium	**X**	**X**		**X**	**X**
Cobalt	X				
Copper	**X**	**X**	**X**	**X**	**X**
Lead	**X**	**X**	**X**	**X**	**X**
Manganese	X				
Mercury	**X**	**X**			**X**
Monomethyl mercury		X			
Nickel	X				
Selenium	X				
Thallium	X				
Tin	X				
Zinc	**X**	**X**	**X**	**X**	**X**
Polychlorinated Biphenyls (PCBs)					
21 "NOAA Status & Trends" Congeners [f]		**X**			
209 PCB Congeners	**X**		**X**	**X**	
Polybrominated Diphenyl Ethers (PBDEs)					
14 PBDE Congeners [g]		**X**			
38 PBDE Congeners [h]	**X**		**X**	**X**	
Polychlorinated Dibenzodioxins and Furans (PCDD/Fs)					
Total TCDD					**X**
Total TCDF					**X**

Table B-1 (Cont'd). Chemicals Analyzed for Loading Studies.

Chemical Class		Loading Study				
	Chemical	Surface Runoff [a]	Atm. Dep.[b]	POTWs[c]	Ocean Exch.[d]	Ground-water[e]
Chlorinated Pesticides						
	2,4'-DDD	**X**		**X**	**X**	**X**
	2,4'-DDE	**X**		**X**	**X**	**X**
	2,4'-DDT	**X**		**X**	**X**	**X**
	4,4'-DDD	**X**		**X**	**X**	**X**
	4,4'-DDE	**X**		**X**	**X**	**X**
	4,4'-DDT	**X**		**X**	**X**	**X**
	Aldrin	X		X	X	
	alpha-BHC	X		X	X	
	beta-BHC	X		X	X	
	delta-BHC	X		X	X	
	gamma-BHC (Lindane)	X		X	X	
	Chlorpyriphos	X		X	X	
	cis-Chlordane	X		X	X	
	trans-Chlordane	X		X	X	
	Chlordane	X		X	X	
	Dacthal (DCPA)	X		X	X	
	DDMU			X		
	Dieldrin	X		X	X	
	Endosulfan I	X		X	X	
	Endosulfan II	X		X	X	
	Endosulfan sulfate	X		X	X	
	Endrin	X		X	X	
	Endrin Aldehyde	X		X	X	
	Endrin Ketone	X		X	X	
	Heptachlor	X		X	X	
	Heptachlor epoxide	X		X	X	
	Hexachlorobenzene	X		X	X	
	Methoxychlor	X		X	X	
	Mirex	X		X	X	
	cis-Nonachlor	X		X	X	
	trans-Nonachlor	X		X	X	
	Oxychlordane	X		X	X	
	Toxaphene	X		X	X	

Table B-1 (Cont'd). Chemicals Analyzed for Loading Studies.

Chemical Class	Chemical	Loading Study: Surface Runoff [a]	Atm. Dep. [b]	POTWs [c]	Ocean Exch. [d]	Ground-water [e]
Low Molecular Weight Polycyclic Aromatic Hydrocarbons (LPAHs)						
	Acenaphthene	**X**		**X**	**X**	**X**
	Acenaphthylene	**X**		**X**	**X**	**X**
	Anthracene	**X**	**X**	**X**	**X**	**X**
	Fluorene	**X**		**X**	**X**	**X**
	Naphthalene	**X**		**X**	**X**	**X**
	Phenanthrene	**X**	**X**	**X**	**X**	**X**
High Molecular Weight Polycyclic Aromatic Hydrocarbons (HPAHs)						
	Benzo(a)anthracene*	**X**	**X**	**X**	**X**	**X**
	Benzo(a)pyrene*	**X**	**X**	**X**	**X**	**X**
	Benzo(b)fluoranthene*	**X**	**X**	**X**	**X**	**X**
	Benzo(g,h,i)perylene	**X**	**X**	**X**	**X**	**X**
	Benzo(k)fluoranthene*	**X**	**X**	**X**	**X**	**X**
	Chrysene*	**X**	**X**	**X**	**X**	**X**
	Dibenzo(a,h)anthracene*	**X**	**X**	**X**	**X**	**X**
	Fluoranthene	**X**	**X**	**X**	**X**	**X**
	Indeno(1,2,3-cd)pyrene*	**X**	**X**	**X**	**X**	**X**
	Pyrene	**X**	**X**	**X**	**X**	**X**
Phthalate Esters						
	Bis(2-ethylhexyl)phthalate	**X**		**X**	**X**	**X**
	Butylbenzylphthalate	X		X	X	
	Diethylphthalate	X		X	X	
	Dimethylphthalate	X		X	X	
	Di-N-butylphthalate	X		X	X	
	Di-N-octylphalate	X		X	X	
Herbicides						
	2,3,4,5-Tetrachlorophenol	X		X		
	2,3,4,6-Tetrachlorophenol	X		X		
	2,4,5-T	X		X		
	2,4,5-TP (Silvex)	X		X		
	2,4,5-Trichlorophenol	X		X	X	
	2,4,6-Trichlorophenol	X		X	X	
	2,4-D	X		X		
	2,4-DB	X		X		
	3,5-Dichlorobenzoic acid	X		X		
	Acifluorfen	X		X		

Table B-1 (Cont'd). Chemicals Analyzed for Loading Studies.

Chemical Class		Loading Study				
	Chemical	Surface Runoff [a]	Atm. Dep.[b]	POTWs[c]	Ocean Exch.[d]	Ground-water[e]
Herbicides						
	Bentazon	X		X		
	Bromoxynil	X		X		
	Clopyralid	X		X		
	Dicamba I	X		X		
	Dichlorprop	X		X		
	Diclofop-methyl	X		X		
	Dinoseb	X		X		
	Ioxynil	X		X		
	MCPA	X		X		
	MCPP (Mecoprop)	X		X		
	Pentachloroanisole	X		X	X	
	Pentachlorophenol	X		X	X	
	Picloram	X		X		
	Triclopyr	**X**		**X**		
Semivolatile Organics						
	1,2,4,-Trichlorobenzene	X		X	X	
	1,2-Dichlorobenzene	X		X	X	
	1,2-Diphenylhydrazine	X			X	
	1,3-Dichlorobenzene	X		X	X	
	1,4-Dichlorobenzene	X		X	X	
	1,7-Dimethylphenanthrene		X			
	1-Methylnaphthalene	X		X	X	
	2,2'-Oxybis[1-chloropropane]				X	
	2,4-Dichlorophenol	X		X	X	
	2,4-Dimethylphenol	X		X	X	
	2,4-Dinitrophenol	X		X	X	
	2,4-Dinitrotoluene	X		X	X	
	2,6-Dimethylphenanthrene		X			
	2,6-Dinitrotoluene			X	X	
	2-Chloronaphthalene	X		X	X	
	2-Chlorophenol	X		X	X	
	2-Methylnaphthalene	X		X	X	
	2-Methylphenol			X	X	
	2-Nitroaniline	X		X	X	
	2-Nitrophenol	X		X	X	

Table B-1 (Cont'd). Chemicals Analyzed for Loading Studies

Chemical Class	Chemical	Loading Study				
		Surface Runoff [a]	Atm. Dep.[b]	POTWs[c]	Ocean Exch.[d]	Ground-water[e]
Semivolatile Organics						
	3,6-Dimethylphenanthrene		X			
	3,3'-Dichlorobenzidine	X		X	X	
	3B-Coprostanol			X	X	
	3-Nitroaniline	X		X	X	
	4,6-Dinitro-2-methylphenol	X		X	X	
	4-Bromophenylphenylether	X		X	X	
	4-Chloro-3-methylphenol	X		X	X	
	4-Chloroaniline	X		X	X	
	4-Chlorophenyl-Phenylether	X		X	X	
	4-Methylphenol	X		X	X	
	4-Nitroaniline	X		X	X	
	4-Nitrophenol	X		X	X	
	4-Nonylphenol	**X**		**X**	**X**	
	Benzoic acid			X	X	
	Benzyl alcohol			X	X	
	bis(2-Chloroethoxy)methane	X		X	X	
	bis(2-Chloroethyl)ether	X		X	X	
	Bisphenol A	X		X	X	
	Caffeine	X		X	X	
	Carbazole	X		X	X	
	Cholesterol	X		X	X	
	Dibenzofuran	X		X	X	
	Ethanol, 2-chloro, phosphate (3:1)	X		X	X	
	Hexachlorobutadiene	X		X	X	
	Hexachlorocyclopentadiene	X		X	X	
	Hexachloroethane	X		X	X	
	Isophorone	X		X	X	
	Nitrobenzene	X		X	X	
	N-Nitrosodimethylamine	X		X		
	N-Nitrosodi-N-propylamine	X		X	X	
	N-Nitrosodiphenylamine	X		X	X	
	Perylene		X			
	Phenol	X		X	X	
	Retene	X	X	X	X	
	Triclosan	X		X	X	
	Triethylcitrate	X		X	X	

Table B-1 (Cont'd). Chemicals Analyzed for Loading Studies

Chemical Class	Chemical	Loading Study: Surface Runoff [a]	Atm. Dep.[b]	POTWs[c]	Ocean Exch.[d]	Ground-water[e]
Oil & Grease and Petroleum Hydrocarbons						
	Oil & Grease	**X**				
	TPH-Gas range	**X**				**X**
	TPH-Diesel range	**X**				**X**
	TPH-Lube oil range	**X**				**X**
Anhydrosugars						
	Galactosan		X			
	Levoglucosan		X			
	Mannosan		X			
Perfluorinated Compounds						
	Perfluorodecanoate			X		
	Perfluoroheptanoate			X		
	Perfluorohexanoate			X		
	Perfluorononanoate			X		
	Perfluorooctanoate			X		
	Perfluorooctane sulfonate			X		
	Perfluoropentanoate			X		
	Perfluorohexane sulfonate			X		
	Perfluorobutanoate			X		
	Perfluorobutane sulfonate			X		
	Perfluorooctane sulfonamide			X		
	Perfluoroundecanoate			X		
	Perfluorododecanoate			X		

Bolded and Shaded cells indicate Chemicals of Concern

[a] Herrera, 2011

[b] Brandenberger et al., 2010

[c] Ecology and Herrera, 2010

[d] Gries and Osterberg, 2011

[e] Pitz, 2011

[f] 21 "NOAA Status & Trends" Congeners = PCB-8, -18, -28, -44, -52, -66, -77, -101, -105, -118, 126, -128, -138, -153, -170, -180, -187, -195, -200, -206, and -209

[g] 14 PBDE Congeners = PBDE-17, -28, -47, -66, -71, -85, -99, -100, -138, -153, -154, -183, -190, -209

[h] 38 PBDE Congeners = PBDE-7, -10, -15, -17, -28, -30, -47, -49, -66, -71, -77, -85, -99, -100, -119, -126, -138, -139, -140, -153, -154, -156/159, -171, -180, -183, -184, -191, -196, -197/204, -201, -203, -205, -206, -207, -208, -209

[*] Carcinogenic PAHs (cPAHs)

Table B-2. Methods Used to Derive Representative Chemical of Concern (COC) Concentrations Where Sample Results Include Non-Detects.

Loading Study	Scenario and Substitution Methods for Non-Detects (NDs)		Rules for Summing Constituents for Groups (e.g. PAHs, PCBs)	Possible Bias as a Result of Method	COCs Where Substitution Method Was Used
	All of data set NDs	Part of data set NDs			
Surface Runoff (Herrera, 2011)	Maximum RL used and the final derived values were presented as "<" and flagged with a "U"	Where ≥ 50% of results were ND, ½ MRL assigned to NDs and final value flagged as "E" Where < 50% of results were ND, ½ MRL assigned to NDs with no flag for final value	Only detected results were summed (zero assigned to NDs) Where all results were ND, the highest MRL was used to represent the sum	Substitution of ND with ½ MRL appears to be reasonable estimate in cases where up to 70% results are ND [a]. At higher rates of ND, this substitution method may yield conservative results (biased high) For summed parameters, the procedure used yields minimum or near-minimum possible values	Arsenic and copper were 0% ND (detected in 100% of samples) ½ MRL assigned to NDs for Lead, mercury, zinc, total PCBs, and total PBDEs were <50% ND (detected in ≥ 50% and < 100% of samples) ½ MRL assigned to NDs for cadmium, total PAHs, cPAH, LPAH, HPAH, DEHP, triclopyr, nonylphenol, and lube oil were ≥ 50% ND (detected in < 50% of samples)
Atmospheric Deposition (Brandenberger et al., 2010)	Not applicable	For all parameters except PCBs, MDLs assigned to NDs For PCBs, zero assigned to NDs	For all parameters except PCBs, detected results and NDs (assigned MDLs) were summed For PCBs, only detected results were summed (zero assigned to NDs)	For all parameters except PCBs, the procedure used yields maximum possible values For PCBs, the procedure used yields minimum possible values	MDLs assigned to NDs for PBDEs Zero assigned to NDs for PCBs
POTWs (Ecology and Herrera, 2010)	No attempt was made to derive representative concentration where FOD < 50%	Where n ≥ 10 and FOD ≥ 50%, ROS used to calculate representative concentration Where n < 10 and FOD ≥ 65%, ½ MRL assigned to NDs	Only detected results were summed (zero assigned to NDs) Where all results were ND, the highest MRL was used to represent the sum	Substitution of ND with ½ MRL appears to be reasonable estimate in cases where up to 50% results are ND [a]. This substitution procedure was not used at higher FODs. ROS method was found to yield similar results when compared to substitution of ND with ½ MRL. For summed parameters, the procedure used yields minimum or near-minimum possible values	Copper, lead, and zinc were 0% ND (detected in 100% of samples) ROS used for some PBDE congeners, some individual PAHs, and DEHP ½ MRL assigned to NDs for some PCB congeners Representative concentrations not calculated for DDT compounds, some individual PAHs, triclopyr, nonylphenol

Table B-2 (Cont'd). Methods Used to Derive Representative Chemical of Concern (COC) Concentrations Where Sample Results Include Non-Detects.

Loading Study	Scenario and Substitution Methods for Non-Detects (NDs)		Rules for Summing Constituents for Groups (e.g. PAHs, PCBs)	Possible Bias as a Result of Method	COCs Where Substitution Method Was Used
	All of data set NDs	Part of data set NDs			
Ocean Exchange (Gries and Osterberg, 2011)	No attempt was made to derive representative concentrations where all of data set were NDs	When most of the samples had detected results, only detected values were used to calculate representative concentrations When few of the samples had detected results, ½ RL assigned to NDs	Only detected results were summed (zero assigned to NDs) Where all results were ND, the highest MRL was used to represent the sum	Using only detected concentrations yields maximum possible values Substitution of ND with ½ MRL appears to be reasonable estimate in cases where up to 70% results are ND [a]. At higher rates of ND, this substitution method may yield conservation results (biased high) For summed parameters, the procedure used yields minimum or near-minimum possible values	Arsenic, cadmium, copper, and zinc were 0% ND (detected in 100% of samples) Only detected values were used to calculate representative lead concentrations ½ MRL assigned to NDs for PCBs and PBDEs Representative concentrations not calculated for DDT compounds, PAHs, DEHP, nonylphenol
Groundwater (Pitz, 2011)	Two methods were used: (1) ½ RL assigned to NDs, and (2) the minimum RL of the data set was assigned to NDs	Two methods were used: (1) ½ RL assigned to NDs, and (2) the minimum RL of the data set was assigned to NDs	All values were summed after values for individual chemicals were generated using the ND substitution procedures (½ RL or minimum RL)	Results appear to be biased low when comparing to results generated from using only detected values. Assignment of the minimum RL to NDs generates the most downward bias.	All procedures applied to all COCs

ND=non-detected
RL=reporting limit
MRL=maximum reporting limit
MDL=method detection limit
FOD=frequency of detection
ROS=regression on order statistics

[a] Antweiler, R.C. and H.E Taylor, 2008. Evaluation of statistical treatments of left-censored environmental data using coincident uncensored data sets: I. Summary statistics. Environmental Science and Technology 42: 3732-3728.

Appendix C. Summary of Release Estimates for All COCs (from Ecology, 2011)

Table C-1. Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
Arsenic	**TOTAL**					**0.79 (0 - 1.7)**	**t/yr**	**100%**
Arsenic	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Unknown release	Unknown form	Air	0.28	t/yr	36% (0% - 100%)
Arsenic	Pesticides and Wood Preservation	CCA-treated wood	Leaching	Solubilized in water	Soil, Surface water	0.27 (0.04 - 0.5)	t/yr	34% (0% - 100%)
Arsenic	Buildings and Grounds	Roofing materials - asphalt shingle	Leaching, Corrosion	Solubilized in water, Particulate matter in water	Surface water, POTWs, Groundwater	0.15 (0 - 0.84)	t/yr	19% (0% - 100%)
Arsenic	Miscellaneous Material Use	Fertilizers	Direct application to soil	Solid, Liquid	Soil	0.06	t/yr	8% (0% - 100%)
Arsenic	Industrial and Institutional Point Sources	Wood-Treatment Facility	Unknown release	Unknown form	Water	0.01	t/yr	2% (0% - 100%)
Arsenic	Buildings and Grounds	Residential Fuel Use, except Wood	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	t/yr	1% (0% - 100%)
Cadmium	**TOTAL**					**0.96 (0.84 - 1.2)**	**t/yr**	**100%**
Cadmium	Buildings and Grounds	Roofing materials - total	Leaching, Corrosion	Solubilized in water, Particulate matter in water	Surface water, POTWs, Groundwater	0.59 (0.5 - 0.7)	t/yr	61% (53% - 68%)
Cadmium	Miscellaneous Material Use	Fertilizers	Direct application to soil	Solid, Liquid	Soil	0.26	t/yr	27% (22% - 31%)
Cadmium	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Fugitive air release	Dust, Vapor	Air	0.06	t/yr	6% (5% - 7%)
Cadmium	Vehicles and Roads	Tire wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	0.03 (<0.01 - 0.06)	t/yr	3% (<1% - 6%)
Cadmium	Vehicles and Roads	Brake pad wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	0.03 (<0.01 - 0.06)	t/yr	1% (<1% - 7%)
Cadmium	Buildings and Grounds	Residential Fuel Use, except Wood	Dispersal of dust following wear	Particulate matter, Fugitive dust	air	0.01	t/yr	<1%
Cadmium	Buildings and Grounds	Woodstoves and Fireplaces	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	t/yr	<1%

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
Cadmium	Non-Point Combustion Sources	Locomotive Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	<0.01	t/yr	<1%
Cadmium	Industrial and Institutional Point Sources	Specialty Glass Manufacturer	Fugitive air release, Combustion	Dust, Vapor, Aerosols, Particulate matter	Air	<0.01	t/yr	<1%
Copper	**TOTAL**					**180 - 250 (120 - 390)**	**t/yr**	**100.0%**
Copper	Buildings and Grounds	Urban lawn & garden use of pesticides	Direct application to soil or vegetation	Solid, Liquid	Soil, Vegetation	1.1 - 73	t/yr	0.6% - 29% (0.3% - 38%)
Copper	Buildings and Grounds	Plumbing fixtures, pipes, and solder	Leaching	Solubilized in water	POTWs	39 (8.6 - 130)	t/yr	16% - 22% (4% - 45%)
Copper	Vehicles and Roads	Brake pad wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	37	t/yr	15% - 21% (10% - 31%)
Copper	Buildings and Grounds	Roofing materials - total	Leaching, Corrosion	Solubilized in water, Particulate matter in water	Surface water, POTWs, Groundwater	27 (12 - 43)	t/yr	11% - 16% (3% - 29%)
Copper	Industrial and Institutional Point Sources	Army Base	Unknown release	Unknown form	Unknown	25	t/yr	10% - 14% (6% - 21%)
Copper	Pesticides and Wood Preservation	Antifouling paint - total	Leaching, Ablation	Solubilized in water, Particulate matter in water	Marine surface water, Marine sediment	23 (12 - 54)	t/yr	9% - 13% (3% - 34%)
Copper	Pesticides and Wood Preservation	Agricultural use of pesticides - total	Direct application to soil or vegetation	Solid, Liquid	Soil, Vegetation	10	t/yr	4% - 6% (2% - 8%)
Copper	Miscellaneous Material Use	Micronutrients	Direct application to soil	Solid, Liquid	Soil	5.4	t/yr	2% - 3% (1% - 5%)
Copper	Industrial and Institutional Point Sources	Naval Shipyard	Fugitive air release, Undefined release to surface water	Dust, Vapor, Undefined form released to surface water	Air, Surface water, Other	5.1	t/yr	2% - 3% (1% - 4%)
Copper	Vehicles and Roads	Tire wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	1.9 (0.02 - 5.4)	t/yr	<1% - 1% (<1% - 4%)
Copper	Pesticides and Wood Preservation	Aquatic-use algaecides in pools, fountains, spas, etc.	Direct application to water in contained pools (swimming pools, fountains, etc.)	Solid, Liquid	POTWs, Soils	1.5	t/yr	<1% - 1% (<1% - 1%)

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
Copper	Industrial and Institutional Point Sources	Other Industrial and Military Facilities	Unknown release	Unknown form	Unknown	0.83	t/yr	<1%
Copper	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Fugitive air release, Combustion	Dust, Vapor, Aerosols, Particulate matter	Air	0.44	t/yr	<1%
Copper	Industrial and Institutional Point Sources	Metal Foundries	Fugitive air release	Dust, Vapor	Air	0.22	t/yr	<1%
Copper	Pesticides and Wood Preservation	CCA-treated wood	Leaching	Solubilized in water	Soil, Surface water	0.06 (0.04 - 0.08)	t/yr	<1%
Lead	**TOTAL**					**520 (150 - 1,000)**	**t/yr**	**100%**
Lead	Outdoor Product Use	Ammunition, Hunting shot use	Intentional loss	Soilid metal	Soil, Surface water	370 (27 - 820)	t/yr	72% (13% - 87%)
Lead	Industrial and Institutional Point Sources	Army Base	Unknown release	Unknown form	Unknown	39	t/yr	8% (4% - 25%)
Lead	Outdoor Product Use	Fishing sinker loss	Unintentional loss	Soilid metal	Surface water, Aquatic sediment	36 (32 - 54)	t/yr	7% (3% - 31%)
Lead	Vehicles and Roads	Wheel weight loss	Unintentional loss	Solid metal	Impervious surfaces, Roadside areas	28 (20 - 29)	t/yr	5% (2% - 18%)
Lead	Buildings and Grounds	Roofing materials - total	Leaching, Corrosion	Solubilized in water, Particulate matter in water	Surface water, POTWs, Groundwater	18 (15 - 20)	t/yr	3% (2% - 12%)
Lead	Non-Point Combustion Sources	Aviation fuel combustion	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	16	t/yr	3% (2% - 10%)
Lead	Industrial and Institutional Point Sources	Various Industrial Facilities, not including pulp mills	Unknown release	Unknown form	Unknown	2.3	t/yr	<1% (<1% - 2%)
Lead	Industrial and Institutional Point Sources	Naval Shipyard	Unknown release	Unknown form	Unknown	1.8	t/yr	<1% (<1% - 1%)
Lead	Vehicles and Roads	Brake pad wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	2.6 (0.04 - 13)	t/yr	<1% (<1% - 8%)
Lead	Vehicles and Roads	Tire wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	1.2 (0.01 - 1.8)	t/yr	<1% (<1% - 1%)
Lead	Industrial and Institutional Point Sources	Pulp and Paper Mills	Undefined release to surface water	Undefined form released to surface water	Water	0.66	t/yr	<1%
Lead	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Fugitive air release, Combustion	Dust, Vapor, Aerosols, Particulate matter	Air	0.53	t/yr	<1%
Lead	Buildings and Grounds	Plumbing fixtures, pipes, and solder	Leaching	Solubilized in water	POTWs	0.21 (0.2 - 0.9)	t/yr	<1%

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
Lead	Miscellaneous Material Use	Fertilizers	Direct application to soil	Solid, Liquid	Soil	0.04	t/yr	<1%
Lead	Buildings and Grounds	Residential Fuel Use, except Wood	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.02	t/yr	<1%
Lead	Non-Point Combustion Sources	Locomotive Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	t/yr	<1%
Mercury	**TOTAL**					**0.54 (0.47 - 0.61)**	**t/yr**	**100%**
Mercury	Miscellaneous Material Use	Thermostat Disposal	Volatilization, Leaching, Washout	Vapor, Liquid, Particle-bound	Air, Groundwater, Soil	0.13 (0.11 - 0.16)	t/yr	24% (20% - 31%)
Mercury	Miscellaneous Material Use	Fluorescent Lamp Disposal	Volatilization	Vapor	Air	0.10	t/yr	18% (16% - 20%)
Mercury	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Unknown release	Unknown form	Air	0.05	t/yr	9% (8% - 11%)
Mercury	Industrial and Institutional Point Sources	Crematoria Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.05 (0.02 - 0.07)	t/yr	9% (4% - 12%)
Mercury	Industrial and Institutional Point Sources	Cement Plants	Volatilization, Combustion	Vapor, Aerosols, Particulate matter	Air	0.04	t/yr	8% (7% - 9%)
Mercury	Miscellaneous Material Use	Auto Convenience Switch Disposal	Volatilization, Leaching, Washout	Vapor, Liquid, Particle-bound	Air, Groundwater, Soil	0.04 (0.02 - 0.06)	t/yr	7% (4% - 12%)
Mercury	Industrial and Institutional Point Sources	Petroleum Refineries	Volatilization, Combustion	Vapor, Aerosols, Particulate matter	Air	0.03	t/yr	6% (5% - 7%)
Mercury	Personal Care Products	Dental Amalgam Excretion	Human Excretion	Excrement	POTWs, Groundwater	0.02	t/yr	4% (3% - 4%)
Mercury	Miscellaneous Material Use	Button Cell Batteries	Volatilization, Leaching, Washout	Vapor, Liquid, Particle-bound	Air, Groundwater, Soil	0.02	t/yr	4% (3% - 4%)
Mercury	Industrial and Institutional Point Sources	Steel Mills	Unknown release	Unknown form	Unknown	0.02	t/yr	3%
Mercury	Industrial and Institutional Point Sources	Residual Fuel Oil Combustion	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	t/yr	2%
Mercury	Personal Care Products	Dental Office Amalgam Waste	Wastewater	Liquid	POTWs, Groundwater	0.01	t/yr	2%
Mercury	Buildings and Grounds	Residential Fuel Use, except Wood	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	t/yr	1%
Mercury	Industrial and Institutional Point Sources	Pulp and Paper Mills	Undefined release to surface water	Undefined form released to surface water	Surface water	0.01	t/yr	1%
Mercury	Miscellaneous Material Use	Thermometers (Household)	Volatilization, Leaching, Washout	Vapor, Liquid, Particle-bound	Air, Groundwater, Soil	<0.01	t/yr	<1%

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
Mercury	Miscellaneous Material Use	Fertilizers	Direct application to soil	Solid, Liquid	Soil	<0.01	t/yr	<1%
Mercury	Industrial and Institutional Point Sources	Other Industrial and Military Facilities	Unknown release	Unknown form	Unknown	<0.01	t/yr	<1%
Mercury	Vehicles and Roads	Gasoline and Diesel Combustion	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	<0.01	t/yr	<1%
Zinc	**TOTAL**					**1,500 (300 - 3,200)**	**t/yr**	**100%**
Zinc	Buildings and Grounds	Roofing materials - total	Leaching, Corrosion	Solubilized in water, Particulate matter in water	Surface water, POTWs, Groundwater	1,330 (210 - 2,800)	t/yr	87% (37% - 97%)
Zinc	Vehicles and Roads	Tire wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	82 (4.8 - 150)	t/yr	5% (<1% - 33%)
Zinc	Miscellaneous Material Use	Fertilizers and Micronutrients	Direct application to soil	Solid, Liquid	Soil	41	t/yr	3% (1% - 13%)
Zinc	Buildings and Grounds	Plumbing fixtures, pipes, and solder	Leaching	Solubilized in water	POTWs	30 (20 - 93)	t/yr	2% (<1% - 25%)
Zinc	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Volatilization, Fugitive air release, Combustion	Vapor, Dust, Aerosols, Particulate matter	Air	12	t/yr	<1% (<1% - 4%)
Zinc	Vehicles and Roads	Motor oil leaks and improper disposal				7.9 (5.7 - 8.9)	t/yr	<1% (<1% - 3%)
Zinc	Vehicles and Roads	Brake pad wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	7.1 (0.22 - 44)	t/yr	<1% (<1% - 13%)
Zinc	Industrial and Institutional Point Sources	Steel Mills	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	4.3	t/yr	<1% (<1% - 1%)
Zinc	Industrial and Institutional Point Sources	Pulp and Paper Mills	Undefined release to surface water	Undefined form released to surface water	Surface water	3.7	t/yr	<1% (<1% - 1%)
Zinc	Industrial and Institutional Point Sources	Naval Shipyard	Fugitive air release	Dust, Vapor	Air	1.8	t/yr	<1%
Zinc	Industrial and Institutional Point Sources	Petroleum Refineries	Unknown release	Unknown form	Unknown	1.1	t/yr	<1%
Zinc	Industrial and Institutional Point Sources	Other Industrial and Military Facilities	Unknown release	Unknown form	Unknown	0.77	t/yr	<1%
Zinc	Industrial and Institutional Point Sources	Steel Galvanizers	Unknown release	Unknown form	Unknown	0.73	t/yr	<1%
PCBs	**TOTAL**					**2,100 (1,500 - 2,800)**	**kg/yr**	**100%**
PCBs	Miscellaneous Material Use	Large capacitors	Leakage	Liquid	Soil, Impervious surfaces	1,100	kg/yr	52% (40% - 75%)
PCBs	Miscellaneous Material Use	Small capacitors	Leakage	Liquid	Soil, Impervious surfaces	500 (1 - 1,000)	kg/yr	24% (<1% - 41%)

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
PCBs	Buildings and Grounds	Residential Trash Burning	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	280	kg/yr	13% (10% - 19%)
PCBs	Miscellaneous Material Use	Transformers	Leakage	Liquid	Soil, Impervious surfaces	130 (7 - 250)	kg/yr	6% (<1% - 15%)
PCBs	Buildings and Grounds	Sealants (Caulking)	Volatilization, Abrasion and fragmentation from weathering	Vapor, Sorption to dust particles	Air, Fugitive dust	110 (71 - 140)	kg/yr	5% (3% - 9%)
PBDEs	**TOTAL**					**680 (220 - 2,300)**	**kg/yr**	**100%**
PBDEs	Buildings and Grounds	Indoor office space air	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	430 (120 - 750)	kg/yr	64% (7% - 88%)
PBDEs	Buildings and Grounds	Indoor residential dust	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	160 (100 - 320)	kg/yr	23% (5% - 72%)
PBDEs	Buildings and Grounds	Indoor office space dust	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	78 (<0.01 - 1,200)	kg/yr	12% (<1% - 84%)
PBDEs	Buildings and Grounds	Indoor residential air	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	9.5 (0.6 - 18)	kg/yr	1% (<1% - 8%)
PCDD/Fs	**TOTAL**					**9.4**	**g TEQ/yr**	**100%**
PCDD/Fs	Buildings and Grounds	Backyard Burn Barrels	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	7.3	g TEQ/yr	77%
PCDD/Fs	Vehicles and Roads	Heavy Duty Diesel Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.81	g TEQ/yr	9%
PCDD/Fs	Industrial and Institutional Point Sources	Pulp and Paper Mills	Combustion, Undefined release to surface water	Aerosols, Vapor, Undefined form released to surface water	Air, Surface water	0.49	g TEQ/yr	5%
PCDD/Fs	Buildings and Grounds	Woodstoves and Fireplaces	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.38	g TEQ/yr	4%
PCDD/Fs	Non-Point Combustion Sources	Construction Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.18	g TEQ/yr	2%
PCDD/Fs	Vehicles and Roads	Light Duty Gasoline Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.08	g TEQ/yr	<1%
PCDD/Fs	Industrial and Institutional Point Sources	Cement Plants	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.05	g TEQ/yr	<1%

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
PCDD/Fs	Vehicles and Roads	Light Duty Diesel Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.04	g TEQ/yr	<1%
PCDD/Fs	Non-Point Combustion Sources	Industrial Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.03	g TEQ/yr	<1%
PCDD/Fs	Non-Point Combustion Sources	Commercial Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.02	g TEQ/yr	<1%
PCDD/Fs	Industrial and Institutional Point Sources	Petroleum Refineries	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.02	g TEQ/yr	<1%
PCDD/Fs	Industrial and Institutional Point Sources	Other Industrial and Military Facilities	Unknown release	Unknown form	Air, Surface water	0.02	g TEQ/yr	<1%
PCDD/Fs	Non-Point Combustion Sources	Recreational Boat Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	g TEQ/yr	<1%
PCDD/Fs	Buildings and Grounds	Lawn and Garden Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	g TEQ/yr	<1%
PCDD/Fs	Non-Point Combustion Sources	Agricultural Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	g TEQ/yr	<1%
PCDD/Fs	Non-Point Combustion Sources	Logging Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	<0.01	g TEQ/yr	<1%
PCDD/Fs	Vehicles and Roads	Heavy Duty Gasoline Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	<0.01	g TEQ/yr	<1%
PCDD/Fs	Non-Point Combustion Sources	Airport Service Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	<0.01	g TEQ/yr	<1%
PCDD/Fs	Non-Point Combustion Sources	Recreational Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	<0.01	g TEQ/yr	<1%
PCDD/Fs	Industrial and Institutional Point Sources	Wood-Treatment Facility	Unknown release	Unknown form	Unknown	<0.01	g TEQ/yr	<1%
PAH	**TOTAL**					**310**	**t/yr**	**100%**
PAH	Buildings and Grounds	Woodstoves and Fireplaces	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	110	t/yr	34%
PAH	Pesticides and Wood Preservation	Creosote Treated Marine pilings - total	Leaching, Washout, Volatilization	Solubilized in water, Vapor	Surface water, Air	54	t/yr	18%
PAH	Pesticides and Wood Preservation	Creosote Treated Railroad ties	Leaching, Washout, Volatilization	Solubilized in water, Vapor	Soil, Air, Surface water	43	t/yr	14%
PAH	Vehicles and Roads	Light Duty Gasoline Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	29	t/yr	10%

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
PAH	Pesticides and Wood Preservation	Creosote Treated Utility poles	Leaching, Washout, Volatilization	Solubilized in water, Vapor	Soil, Air, Surface water	17	t/yr	6%
PAH	Vehicles and Roads	Heavy Duty Gasoline Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	11	t/yr	3%
PAH	Vehicles and Roads	Petroleum spills, leaks, and improper motor oil disposal	Leakage, Spillage, Direct release, Improper disposal	Liquid	Impervious surfaces, Soils, Stormwater, POTWs, Landfills	11	t/yr	3%
PAH	Buildings and Grounds	Residential Trash Burning	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	6.5	t/yr	2%
PAH	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Combustion, Volatilization	Aerosols, Vapor, Particulate matter	Air	5.2	t/yr	2%
PAH	Buildings and Grounds	Lawn and Garden Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	5.0	t/yr	2%
PAH	Industrial and Institutional Point Sources	Pulp and Paper Mills	Volatilization, Fugitive air release, Combustion	Vapor, Dust, Aerosols, Particulate matter	Air	3.2	t/yr	1%
PAH	Industrial and Institutional Point Sources	Aluminum Mills	Volatilization, Fugitive air release, Combustion	Vapor, Dust, Aerosols, Particulate matter	Air	2.7	t/yr	<1%
PAH	Industrial and Institutional Point Sources	Petroleum Refineries	Fugitive air release, Volatilization, Combustion	Dust, Vapor, Aerosols, Particulate matter	Air	2.3	t/yr	<1%
PAH	Non-Point Combustion Sources	Commercial Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	2.0	t/yr	<1%
PAH	Vehicles and Roads	Heavy Duty Diesel Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	1.8	t/yr	<1%
PAH	Non-Point Combustion Sources	Construction Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	1.3	t/yr	<1%
PAH	Vehicles and Roads	Gas Station Emissions	Volatilization	Vapor	Air	1.2	t/yr	<1%
PAH	Vehicles and Roads	Tire wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	0.98	t/yr	<1%
PAH	Non-Point Combustion Sources	Recreational Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.94	t/yr	<1%

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
PAH	Vehicles and Roads	Coal tar sealants	Leaching, Abrasion	Solubilized in water, Particulate matter in water	stormwater, fugitive air, dust	0.92 (0.17 - 1.7)	t/yr	<1%
PAH	Non-Point Combustion Sources	Recreational Boat Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.86	t/yr	<1%
PAH	Industrial and Institutional Point Sources	Other Industrial and Military Facilities	Unknown release	Unknown form	Air, Surface water, Other	0.58	t/yr	<1%
PAH	Buildings and Grounds	Roofing materials - total	Leaching	Particulate matter, Solubilized in water	Surface water, POTWs	0.57	t/yr	<1%
PAH	Non-Point Combustion Sources	Locomotive Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.49	t/yr	<1%
PAH	Non-Point Combustion Sources	Industrial Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.30	t/yr	<1%
PAH	Vehicles and Roads	Light Duty Diesel Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.21	t/yr	<1%
PAH	Buildings and Grounds	Residential Yard Waste Burning	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.15	t/yr	<1%
PAH	Non-Point Combustion Sources	Logging Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.05	t/yr	<1%
PAH	Non-Point Combustion Sources	Agricultural Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.04	t/yr	<1%
PAH	Buildings and Grounds	Residential Fuel Use, except Wood	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.04	t/yr	<1%
PAH	Non-Point Combustion Sources	Cigarette smoke	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.03 (0.02 - 0.03)	t/yr	<1%

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
PAH	Vehicles and Roads	Asphalt - total	Leaching, Abrasion	Solubilized in water, Particulate matter in water	stormwater, fugitive air, dust	0.02	t/yr	<1%
PAH	Non-Point Combustion Sources	Airport Service Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.02	t/yr	<1%
PAH	Non-Point Combustion Sources	Railroad Maintenance Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	<0.01	t/yr	<1%
Phthalates	**TOTAL**					**34**	**t/yr**	**100%**
Phthalates	Personal Care Products	Fragrance	Washout, Volatilization	Liquid, Vapor	POTWs, Groundwater, Air	11	t/yr	32%
Phthalates	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Unknown release	Unknown form	Air	9.6	t/yr	28%
Phthalates	Vehicles and Roads	Car undercoating	Washout, Vapor	Liquid, Vapor	Surface water, Soil, Air	3.3	t/yr	10%
Phthalates	Miscellaneous Material Use	Lacquers and paint	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	1.9	t/yr	5%
Phthalates	Buildings and Grounds	PVC Coil coated roofing	Leaching, Volatilization	Liquid, Vapor	Surface water, POTWs, Air	1.5	t/yr	4%
Phthalates	Personal Care Products	Nail polish	Washout, Volatilization	Liquid, Vapor	POTWs, Groundwater, Air	1.4	t/yr	4%
Phthalates	Miscellaneous Material Use	PVC Coated fabric	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	1.2	t/yr	4%
Phthalates	Miscellaneous Material Use	Sealants, adhesives, etc.	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	1.1	t/yr	3%
Phthalates	Industrial and Institutional Point Sources	Plastics Manufacturer	Volatilization	Vapor	Air	0.86	t/yr	3%
Phthalates	Personal Care Products	Hair spray (aerosol and pump spray)	Washout, Volatilization	Liquid, Vapor	POTWs, Groundwater, Air	0.4	t/yr	1%
Phthalates	Miscellaneous Material Use	PVC Cables (outdoor, above ground)	Volatilization	Vapor, Sorption to soil	Air	0.35	t/yr	1%
Phthalates	Personal Care Products	Deodorant (solid)	Washout, Volatilization	Liquid, Vapor	POTWs, Groundwater, Air	0.29	t/yr	<1%
Phthalates	Miscellaneous Material Use	Shoe soles	Abrasion	Dust particles	Surface runoff, POTWs, Fugitive dust	0.2	t/yr	<1%
Phthalates	Buildings and Grounds	PVC Wall coverings	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	0.14	t/yr	<1%
Phthalates	Buildings and Grounds	PVC Roofing material	Leaching, Volatilization	Solubilized in water, Vapor	Surface water, POTWs, Air	0.14	t/yr	<1%
Phthalates	Buildings and Grounds	PVC Flooring	Volatilization, Abrasion	Vapor, Dust particles	Air, Fugitive dust	0.1	t/yr	<1%
Phthalates	Miscellaneous Material Use	PVC Films, sheets, coated products	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	0.1	t/yr	<1%

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
Phthalates	Buildings and Grounds	PVC Hoses and profiles (outdoor)	Volatilization	Vapor	Air	0.09	t/yr	<1%
Phthalates	Buildings and Grounds	PVC Cables (indoor)	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	0.08	t/yr	<1%
Phthalates	Buildings and Grounds	PVC Hoses and profiles (indoor)	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	0.08	t/yr	<1%
Phthalates	Miscellaneous Material Use	Printing inks	Washout, Volatilization	Liquid, Vapor	POTWs, Groundwater, Air	0.08	t/yr	<1%
Phthalates	Industrial and Institutional Point Sources	Boat Manufacturer	Volatilization	Vapor	Air	0.05	t/yr	<1%
Phthalates	Industrial and Institutional Point Sources	Chemicals Distribution	Volatilization	Vapor	Air	0.04	t/yr	<1%
Phthalates	Industrial and Institutional Point Sources	Paint and Coatings Manufacturers	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	0.03	t/yr	<1%
Phthalates	Industrial and Institutional Point Sources	Other Industrial and Military Facilities	Volatilization	Vapor	Air	0.02	t/yr	<1%
Triclopyr	**TOTAL**					**150 (63 - 240)**	**t/yr**	**100%**
Triclopyr	Pesticides and Wood Preservation	Crop and Golf Course Use	Direct application to vegetation	Liquid	Vegetation and soils	150 (60 - 240)	t/yr	98% (95% - 99%)
Triclopyr	Pesticides and Wood Preservation	Forest Herbicide Use - State Forests	Direct application to vegetation	Liquid	Vegetation and soils	0.8 (0.4 - 1.2)	t/yr	<1% (<1% - 1%)
Triclopyr	Pesticides and Wood Preservation	Aquatic Weed Control	Direct application to surface water	Liquid or granular	Surface water	0.68	t/yr	<1% (<1% - 2%)
Triclopyr	Pesticides and Wood Preservation	Right-of-Way Maintenance--State Forests	Direct application to vegetation	Liquid	Vegetation and soils	0.5	t/yr	<1%
Triclopyr	Buildings and Grounds	Urban lawn & garden use of pesticides	Direct application to soil or vegetation	Liquid	Soil, Vegetation	0.43	t/yr	<1%
Triclopyr	Vehicles and Roads	Right-of-Way Maintenance - State Highways	Direct application to vegetation	Liquid	Vegetation and soils	0.3	t/yr	<1%
Triclopyr	Vehicles and Roads	Right-of-Way Maintenance - Railroads	Direct application to vegetation	Liquid	Vegetation and soils	0.1	t/yr	<1%
Nonylphenol	**TOTAL**					**0.18**	**t/yr**	**100%**
Nonylphenol	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Unknown release	Unknown form	Air	0.18	t/yr	100%

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
Petroleum	**TOTAL**					**9,300**	**t/yr**	**100%**
Petroleum	Vehicles and Roads	Motor oil drips and leaks	Leakage	Liquid	Impervious surfaces	6,100	t/yr	66%
Petroleum	Vehicles and Roads	Minor gasoline spills from fueling vehicles and non-road equipment	Spillage	Liquid	Impervious surfaces, Soils	1,900	t/yr	21%
Petroleum	Vehicles and Roads	Improper disposal of used oil following oil changes	Direct release, Improper disposal	Liquid	Stormwater, Soils, POTWs, Landfills	960	t/yr	10%
Petroleum	Miscellaneous Material Use	Petroleum spills (large)	Spillage	Liquid	Surface water, Soil, Impervious surfaces	228 (223 - 233)	t/yr	3% (2% - 3%)

Appendix D. Hazard Evaluation Summary

Appendix D-1. Description of Methodology and Data Assessed (see the following pages)

The following sections of Appendix D are available only online as links to this *Assessment Report:* www.ecy.wa.gov/biblio/1103055.html

Appendix D-2. Hazard Evaluation – Results of Prioritization

Appendix D-3. Hazard Evaluation – Plots of Observed and Effects Data for Surface Waters

Appendix D-4. Hazard Evaluation – Plots of Observed Data and Threshold Values for Sediments

Appendix D-5. Hazard Evaluation – Plots of Observed and Effects Data for Tissue Residues

Appendix D-6. Hazard Evaluation – Plots of Estimated and Effects Doses for Wildlife

Appendix D-7. Hazard Evaluation – Plots of Observed Data and Criteria for Human Health

Appendix D-8. Hazard Evaluation – Summary Statistics for Environmental (Observed) Data

Appendix D-9. Hazard Evaluation – Water Effects Summary Data

Appendix D-10. Hazard Evaluation – Sediment Guidelines

Appendix D-11. Hazard Evaluation – Tissue Residue Effects Data

Appendix D-12. Hazard Evaluation – Wildlife Effects Data

Appendix D-13. Hazard Evaluation – ECOTOX QA Summary

Description of Contents for Appendix D

Appendix D-1
Description of the methodology and data assessed for the hazard evaluation. Includes a narrative summary of the results.

Appendix D-2
Tables showing results for the hazard evaluation. Each table shows a summary of whether the observed concentrations exceed threshold values, and notes on the data used for the comparisons.

Appendix D-3
Plots comparing observed environmental concentrations to effects concentrations for surface water.

Appendix D-4
Plots comparing observed environmental concentrations to guidelines for sediment.

Appendix D-5
Plots comparing observed environmental concentrations to effects concentrations for tissue residue.

Appendix D-6
Plots comparing calculated environmental doses to effects doses for wildlife.

Appendix D-7
Plots comparing observed environmental tissue concentrations to criteria for human health.

Appendix D-8
Tables showing summary statistics for the observed environmental concentrations used in the hazard evaluation.

Appendix D-9
Folder containing tables with summaries of ECOTOX data used in the hazard evaluation (the petroleum effects data are not from ECOTOX). A file containing ECOTOX codes is also included in this folder.

Appendix D-10
Table showing guidelines and other threshold values for sediment.

Appendix D-11
Tables showing summaries of the tissue residue effects for Lower Willamette River and the Lower Duwamish River Remedial Investigations

Appendix D-12
Folder containing tables with summaries of wildlife effects data. A list of references reviewed for the wildlife evaluation is also included in this folder.

Appendix D-13
Folder containing tables with summaries of the quality assurance (QA) review of the ECOTOX data and units.

Appendix D-1

Hazard Evaluation for Chemicals of Concern in the Puget Sound Basin – Description of Methodology and Data Assessed

Introduction

This chapter outlines the methods and results of the assessment conducted to estimate the relative hazard posed by exposure to the chemicals of concern (COCs) assessed in the Puget Sound Toxics Loading Analysis (PSTLA) studies. The primary purpose of this assessment was to provide a general overview of the potential for these COCs to cause adverse effects (to aquatic life and select wildlife species), and to a lesser extent, human health effects through seafood consumption within the Puget Sound basin. The assessment described here was not intended to identify specific ecological effects or quantify risk. The primary purpose of the assessment was to provide an additional weight of evidence (WOE), along with the loadings and sources information (documented in the main body of this report), to prioritize COCs for further action.

The large scale regional focus was intended to evaluate COCs at a broad level. A key goal of this effort was to provide information to help prioritize COCs based on their potential to cause adverse effects. The assessment used some of the chemical concentration data generated by the PSTLA studies discussed in this report; however, readily available environmental data for water, sediment and tissue from other sources were the primary basis of this prioritization process. To estimate the potential for effects, environmental data were compared to readily available toxicity data obtained primarily from established databases, sources and regulations. The outcome of this process was used to establish a general "priority" for management of each of the COCs.

This assessment included the following evaluations:

- Direct hazard to aquatic life through surface water exposure
- Direct hazard to benthic organisms through sediment exposure
- Direct hazard to aquatic life based on tissue residue levels
- Hazard to wildlife based on ingestion of prey, water and sediment
- Hazard to human health through fish/seafood consumption

Although some elements of the "Risk Assessment" process were applied to the hazard prioritization presented here, this effort is not intended to serves as a risk assessment. Conducting such an assessment for the Puget Sound region was beyond the scope of this effort. The remainder of this section describes the process used to acquire both the observed environmental data and toxicity data, in addition to the assumptions used to access and use this information. The section also documents the methods used to conduct the effects prioritization, including a description of the process used to determine the priority for each COC. Finally, the results of the assessment (organized by COC) and a discussion of the uncertainty and limitations associated with this process are presented.

Methods

The following sections describe the process used to identify and acquire both the observed environmental data and the toxicity data used for this assessment. Also described is the approach used to assess effects to wildlife, including selection of representative species ("receptors") and appropriate COCs, derivation of daily doses (to estimate toxicity from ingestion pathways) and the model used to estimate exposure to COCs by wildlife. Lastly, the processes used to summarize the environmental and effects data, determine the potential for effects and priority are described.

Environmental Data Collection

Environmental data, collected from a variety of sources, were used to estimate the general range of possible exposure concentrations to COCs. With the exception of a few specific sources, data collection was limited to readily available public databases and only those data collected between January 1, 2000 and July 2010 were considered "recent" and included. When available, surface water, sediment and tissue data were collected from the sources outlined in Table 1. Due to the different purposes for which some of these data were collected, not all sources included data for all matrices in both fresh water and marine environments.

Table 1. Summary of data sources and data types used in this assessment. All data were accessed from their respective sources in July 2010.

Data Source	Matrix					
	Water		Sediment		Tissue	
	Fresh	Marine	Fresh	Marine	Fresh	Marine
Ecology's EIM System [1]	X	X	X	X	X	X
King County's LIMS[2]	X	X	X	X	X	X
US Geological Survey [3]	X	N/A	See Footnote[3]	N/A	X	N/A
Puget Sound Ambient Monitoring Program	N/A	N/A	N/A	N/A	N/A	X
Regional EMAP [4]	N/A	N/A	N/A	X	N/A	X
ENNVEST Study [5]	N/A	N/A	N/A	N/A	N/A	X
Toxics Loading Studies [6]	X	X	N/A	N/A	N/A	X
WDFW[7]	N/A	N/A	N/A	N/A	N/A	X

N/A – data not available

1 - EIM – Ecology's Environmental Information Management System

2- LIMS - Laboratory Information Management System, King County data not previously submitted to EIM

3 - USGS data obtained from online database. Sediment data were obtained but not used; USGS only analyzes the <63µ sediment fraction, which is not comparable to the remainder of the data used in this assessment.

4 - Includes NOAA's mussel watch data (Valerie Partridge, Environmental Assessment Program, written communication, 2010).

5- US Department of Defense (Johnston, R.K. 2007)

6 - Includes Ecology's Ocean Exchange/River Mouth Loading study, Fish Tissue Assessment and Surface Runoff studies discussed in this document.

7 – Washington Department of Fish and Wildlife (West et al. 2011).

The datasets described in Table 1 were combined into a composite database. This process required a number of "rules" and assumptions to ensure that data were consistently formatted (similar naming conventions, units etc.) and in a chemical form appropriate for later comparison to effect concentrations. Table 2 outlines the key rules and assumptions used to combine and process the environmental data.

Table 2. Summary of rules and assumptions used to acquire and summarize observed environmental data.

All Data
• The COC list was expanded to include all forms of these chemicals.
• Data from all sources were standardized to reflect parameter and qualifier names as defined by EIM. When no EIM parameter (for certain co-eluting PBDE congeners) was available the closest match was selected.
• Only data collected between January 1, 2000 and July, 2010 were acquired.
• All data were standardized to common units.
• **Qualified** "B" qualified data and "estimated data" were included. Data with the <u>following qualifiers were not included</u> in the assessment: ▪ "Rejected Data" ▪ The following "U" qualified non-detect data - "U", "U?", "UJ", "UJG", "UJK"," UJL".
• **Summing** - Polychlorinated biphenyl's (PCBs), polycyclic aromatic hydrocarbons (PAHs) and dioxins in all matrices were summed based on SMS rules: ▪ For summed compounds, only compounds detected in a sample were summed.
• **PAHS** ▪ LPAHs include naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene ▪ HPAHs include fluoranthene, pyrene, benz(a)anthracene, chrysene, total benzofluoranthenes (B, J and K), Benzo(a)pyrene, indeno(1,2,3-c,d)pyrene, dibenzo(a,h)anthracene, and benzo(g,h,i)perylene. ▪ cPAHs were not summed for any media because standards and toxicity information for the sum of cPAHs whereas not available for water, sediment, tissue, or in the NTR.
• **Petroleum Compounds** – data for the following petroleum related compounds were identified and collected: TPH as heavy fuel oil, Diesel range TPH, Gasoline range TPH, Lube oil range TPH.
Sediment Data
• Organic carbon (OC) normalization was conducted for marine sediment data when the corresponding sediment quality value was OC-normalized. Otherwise, all sediment data were dry-weight normalized.
• USGS freshwater sediment data were presented as the chemical concentration in the <63 µm fraction. These sediment data were not used due to incompatibility with the majority of the available sediment data.
Tissue Data
• Tissue data were grouped into common tissue type designations for the tissue residue, wildlife and human health assessments. For example, mussel tissue data labeled as "somatic" and "visceral" were categorized as "whole body no shell". Whole body tissue data labeled as "no-gut", "no exoskeleton" were classified as "whole body". Fillet data classified as "skin on", "no skin" were combined and classified as "fillet". Lipid-normalized tissue data were not used in this assessment due to the inconsistencies and availability of lipid data for all tissue concentrations.

The majority of data used in the assessment were obtained from EIM, followed by the King County LIMs. The remaining datasets were relatively small in comparison, but were included

because the parameters and matrices measured complemented the EIM and LIMS datasets. While it is recognized that there may be other sources of environmental data (e.g., various research publications, NOAA, USFW) that could have been included in this assessment, due to the scope and timeline associated with this task, it was necessary to focus on the largest and most readily available electronic sources of primarily ambient data that did not require significant data review or re-entry. Since the intent of this effort was to better understand general regional conditions and not identify "hot spots", these data are assumed to provide reasonable estimates of exposure.

A review of the NOAA database (http://response.restoration.noaa.gov/type_subtopic_entry.php?RECORD_KEY%28entry_subtopic_type%29=entry_id,subtopic_id,type_id&entry_id(entry_subtopic_type)=751&subtopic_id(entry_subtopic_type)=5&type_id(entry_subtopic_type)=1) indicated that a significant proportion the applicable data were already included in EIM and LIMs and much of the data had been collected before 2000. A large proportion of the remaining data were primarily collected from the Duwamish River CERCLA site. It was decided that inclusion of these data in the assessment would possibly skew the results.

The primary goal of this assessment was to provide a high level summary evaluation of relative hazard; therefore, the data were differentiated into three course spatial scales; (1) freshwater, (2) marine nearshore, and (3) marine offshore. The freshwater to nearshore boundary was defined by the original data. If data were classified as "freshwater" by the original data source, they were retained as such; if classified as marine/saltwater the data were further parsed between near and offshore. Nearshore was defined as all marine areas less than 10 meters deep (based on MLLW); offshore was defined as all other marine areas.

Data records not meeting the intent of the nearshore/offshore designation were adjusted accordingly (e.g., marine locators >10m in depth in estuaries like the Duwamish were classified as nearshore). Some sample depths varied due to tidal influences. However, relatively few samples were attributed close enough to the 10m depth (e.g. 9m or 11m) to potentially fall into a different marine area based on depth of tide at the time of sampling.

Comparison of sediment data to sediment guidelines (described below) required that the marine sediment data for nonionic/non-polar organic chemicals be organic carbon (OC) -normalized. Dry-weight concentrations were used for marine sediment samples when OC was outside the range of 0.5 to 3.0%. No associated OC data were available for approximately 35% of the sediment samples. To utilize these samples, these data were OC-normalized using the mean nearshore (2.22%) or mean offshore (1.74%) OC percentages from the remaining sediment results. The process of correcting sediment data for OC resulted in some differences in the total number of measurements (N) for OC and dry weight normalized data presented in the summary tables and figures. For example, for a given COC the N for dry weight-based measurements may be 5, while the N for the OC-normalized measurements is 1.

Tissue data were grouped differently depending on which assessment was being conducted. Tissue samples were segregated into fresh, nearshore and offshore samples based on the location they were collected. Table 3 attributes tissue to freshwater or marine species for informational purposes.

Table 3. Fish and invertebrate tissues and their groupings for the assessments using tissue data.

Common name	Latin Name	Fresh vs Marine	Tissue Assessment	Human Health Assessment	Wildlife Assessment
Asian clam	*Corbicula fluminea*	FW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Bay ghost shrimp	*Neotrypaea californiensis*	SW	Decapod	Other invertebrate	not included
Bay mussel	*Mytilus trossulus*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Bent-nose macoma	*Macoma nasuta*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Black bullhead	*Ameiurus melas*	FW	Fish	Fish	Fish
Black crappie	*Pomoxis nigromaculatus*	FW	Fish	Fish	Fish
Blackmouth (Resident) Chinook salmon	*Oncorhynchus tshawytscha*	SW	Fish	Fish	Fish
Blue mussel	*Mytilus edulis*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Bluegill	*Lepomis macrochirus*	FW	Fish	Fish	Fish
Brook trout	*Salvelinus fontinalis*	FW	Fish	Fish	Fish
Brown bullhead	*Ameiurus nebulosus*	FW	Fish	Fish	Fish
Brown Rockfish	*Sebastes auriculatus*	SW	Fish	Fish	Fish
Brown trout	*Salmo trutta*	FW	Fish	Fish	Fish
Bull trout	*Salvelinus confluentus*	FW	Fish	Fish	Fish
Butter clam	*Saxidomus giganteus*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate

Table 3. Fish and invertebrate tissues and their groupings for the assessments using tissue data.

Common name	Latin Name	Fresh vs Marine	Tissue Assessment	Human Health Assessment	Wildlife Assessment
California mussel	*Mytilus californianus*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Catworm genus	*Nephtys*	SW	Bivalve + other invertebrates	Other invertebrate	not included
Channel catfish	*Ictalurus punctatus*	FW	Fish	Fish	Fish
Chinook salmon	*Oncorhynchus tshawytscha*	SW	Fish	Fish	Fish
Chum salmon	*Oncorhynchus keta*	SW	Fish	Fish	Fish
Coho salmon	*Oncorhynchus kisutch*	SW	Fish	Fish	Fish
Common carp	*Cyprinus carpio*	FW	Fish	Fish	Fish
Copper rockfish	*Sebastes caurinis*	SW	Fish	Fish	Fish
Cutthroat trout	*Oncorhynchus clarkii*	FW	Fish	Fish	Fish
Dabs	*Pleuronectidae*	SW	Fish	Fish	Fish
Dock shrimp	*Pandalus danae*	SW	Decapod	Other invertebrate	not included
Dungeness crab	*Cancer magister*	SW	Decapod	Other invertebrate	Invertebrate
English sole	*Parophrys vetulus*	SW	Fish	Fish	Fish
Fat gaper	*Tresus capax*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Flathead sole	*Hippoglossoides elassodon*	SW	Fish	Fish	Fish
Gaper clam	*Tresus sp.*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Graceful rock crab	*Cancer gracilis*	SW	Decapod	Other invertebrate	Invertebrate
Hake	*Merluccius*	SW	Fish	Fish	Fish

Table 3. Fish and invertebrate tissues and their groupings for the assessments using tissue data.

Common name	Latin Name	Fresh vs Marine	Tissue Assessment	Human Health Assessment	Wildlife Assessment
	productus				
Japanese littleneck	*Tapes philippinarum*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Kokanee	*Oncorhynchus nerka*	FW	Fish	Fish	Fish
Largemouth bass	*Micropterus salmoides*	FW	Fish	Fish	Fish
Largescale sucker	*Catostomus macrocheilus*	FW	Fish	Fish	Fish
Lingcod	*Ophiodon elongatus*	SW	Fish	Fish	Fish
Longnose sucker	*Catostomus catostomus*	FW	Fish	Fish	Fish
Lumbriculus oligochaete	*Lumbriculus*	FW	Bivalve + other invertebrates	Other invertebrate	not included
Macoma clams	*Macoma sp.*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Mediterranean mussel	*Mytilus galloprovincialis*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Melita amphipods	*Melitidae*	SW	Decapod	Other invertebrate	not included
Milky venus	*Compsomyax subdiaphana*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Mountain whitefish	*Prosopium williamsoni*	FW	Fish	Fish	Fish
Northern pikeminnow	*Ptychocheilus oregonensis*	FW	Fish	Fish	Fish
Pacific Dover sole	*Microstomus*	SW	Fish	Fish	Fish

Table 3. Fish and invertebrate tissues and their groupings for the assessments using tissue data.

Common name	Latin Name	Fresh vs Marine	Tissue Assessment	Human Health Assessment	Wildlife Assessment
	pacificus				
Pacific geoduck	*Panopea abrupta*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Pacific herring	*Clupea pallasii*	SW	Fish	Fish	Fish
Pacific littleneck	*Protothaca staminea*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Pacific oyster	*Crassostrea gigas*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Pacific staghorn sculpin	*Leptocottus armatus*	SW	Fish	Fish	Fish
Pacific Tomcod	*Microgadus proximus*	SW	Fish	Fish	Fish
Peamouth	*Mylocheilus caurinus*	FW	Fish	Fish	Fish
Pile perch	*Rhacochilus vacca*	SW	Fish	Fish	Fish
Pink salmon	*Oncorhynchus gorbuscha*	SW	Fish	Fish	Fish
Pollock	*Theragra chalcogramma*	SW	Fish	Fish	Fish
Prickly sculpin	*Cottus asper*	SW	Fish	Fish	Fish
Pumpkinseed sunfish	*Lepomis gibbosus*	FW	Fish	Fish	Fish
Purple mahogany-clam	*Nuttallia obscurata*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Pygmy whitefish	*Prosopium coulterii*	FW	Fish	Fish	Fish
Quillback rockfish	*Sebastes maliger*	SW	Fish	Fish	Fish
Rainbow trout	*Oncorhynchus mykiss*	FW	Fish	Fish	Fish

Table 3. Fish and invertebrate tissues and their groupings for the assessments using tissue data.

Common name	Latin Name	Fresh vs Marine	Tissue Assessment	Human Health Assessment	Wildlife Assessment
Ratfish	*Hydrolagus colliei*	FW	Fish	Fish	Fish
Red rock crab	*Cancer productus*	SW	Decapod	Other invertebrate	Invertebrate
Redside shiner	*Richardsonius balteatus*	FW	Fish	Fish	Fish
Reticulate sculpin	*Cottus perplexus*	FW	Fish	Fish	Fish
Rock bass	*Ambloplites rupestris*	FW	Fish	Fish	Fish
Rock sole	*Lepidopsetta bilineata*	SW	Fish	Fish	Fish
Sand sole	*Psettichthys melanostictus*	SW	Fish	Fish	Fish
Scorpion fishes (Order)	Scorpaeniformes	SW	Fish	Fish	not included
Sea cucumber	*Molpadia intermedia*	SW	Bivalve + other invertebrates	Other invertebrate	Invertebrate
Shiner perch	*Cymatogaster aggregata*	SW	Fish	Fish	Fish
Signal crayfish	*Pacifastacus leniusculus*	FW	Decapod	Other invertebrate	Invertebrate
Sixgill shark	*Hexanchus griseus*	SW	Fish	Fish	not included
Slender sole	*Eopsetta exilis*	SW	Fish	Fish	Fish
Smallmouth bass	*Micropterus dolomieui*	FW	Fish	Fish	Fish
Softshell clam	*Mya arenaria*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Spot prawn	*Pandalus platyceros*	SW	Decapod	Other invertebrate	not included

Table 3. Fish and invertebrate tissues and their groupings for the assessments using tissue data.

Common name	Latin Name	Fresh vs Marine	Tissue Assessment	Human Health Assessment	Wildlife Assessment
Staghorn Sculpin	*Leptocottus armatus*	SW	Fish	Fish	Fish
Starry flounder	*Platichthys stellatus*	SW	Fish	Fish	Fish
Striped seaperch	*Embiotoca lateralis*	SW	Fish	Fish	Fish
Western Pearlshell	*Margaritifera falcata*	FW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Yellow perch	*Perca flavescens*	FW	Fish	Fish	Fish

Summary Statistics

All environmental data were combined by matrix (water, sediment, tissue) and spatial location (freshwater, marine near- and offshore). Tissues were also grouped into "bivalve", "fish" and "other invertebrate" categories for the human health assessment. Both fillet and whole-body samples were used for the human health assessment. For the tissue residue assessment, tissues were divided into "fish"; "non-decapod invertebrates", and "decapods" (crabs and shrimps); all tissue residue comparisons were based on whole-body tissue concentrations. Summary statistics (min, max, mean, median, total number of samples, and frequency of detection) for these data were calculated using MSAccess and Total Access Statistics. Summary statistics for each matrix are presented in Appendix E.

Identification of Effects Concentrations

Unless noted otherwise, all toxicity data used in this assessment were obtained from readily available databases. A literature search was conducted to identify studies of contaminant impacts to northwest regional species which may not have been included in available toxicity databases. In many cases the regional data were not dose-response effects data and inappropriate to directly compare with observed environmental concentrations (i.e. data were lipid normalized, study included multiple chemical exposures, field based studies, etc.). These data are primarily discussed as an additional WOE when evaluating the overall hazard for each COC. The following sections describe the process used to obtain the effects data and any assumptions used in their selection.

Surface Water – Direct Effects to Aquatic Life

To determine the potential for effects to aquatic life from direct exposure to COCs in surface water, relevant effects concentrations were identified to compare with the observed environmental data. EPA's ECOTOXicology (ECOTOX) database (http://cfpub.epa.gov/ecotox) was the primary source of surface water effects data. The "Advanced Database Query" option in ECOTOX was used to obtain the majority of the data which allows for selection of specific taxonomic, chemical, result, condition, publication, and report formats to suit the project needs.

The following rules were used to identify the appropriate toxicological effects data.

- Both aquatic plant and animal data were included in the search process (animal data were accessed in July 2010; plant data were accessed and added in June 2011).
- Effect concentrations classified as EC0, LC0, NOEC, NOEL, and NR-ZERO were not included because they were considered "no effect" results.
- Concentration units based on area (e.g., AI kg/ha, ae kg/ha), or any unit other than volume were excluded. Molar-type units (i.e., M, uM, nM) were converted to ug/L.
- The following endpoint types were included from the ECOTOX database: Lethal Concentration (LC)/Lethal Dose (LD), Effect Concentration (EC)/Effect Dose (ED), Lowest Observable Effect Concentration (LOEC), Lowest Observable Effect Level (LOEL), and NR-LETH (Near Lethal) values, and all effect measurements for both fresh and saltwater organisms. Endpoint types such as bioaccumulation factor, inhibition concentration, and time to mortality were not used. Bioaccumulation was addressed to some degree in the tissue,

wildlife and human health assessments described below. A detailed evaluation of the potential for effects associated with bioaccumulation was beyond the scope of this assessment.

- Washington State water quality criteria were included for comparison when available. Concentration types were selected to match the water quality criteria where available. Thus, for most metals, effects associated with the dissolved concentration type were used for comparison to freshwater and marine observed data. Total and dissolved mercury were used due to the different forms used by the acute and chronic WQC. Formulation type (F) was excluded for COCs except triclopyr, DDTs, and PCBs.
- If "NR" (not reported) was the result for concentration or media type (freshwater or saltwater), then that effect value was excluded.

A summary of the data derived from the ECOTOX database used in this assessment can be found in Appendix D-9.

Although efforts are made by EPA to accurately represent toxicity data in the ECOTOX database, the data are not thoroughly vetted through a detailed quality control process. It was beyond this effort's capacity to review each of the thousands of original papers and documents from which ECOTOX was derived. However, to provide additional confidence in the quality of the ECOTOX data, 125 randomly selected documents were obtained and reviewed for accuracy and correspondence with ECOTOX. These papers represented approximately 5% of those identified by this assessment.

To evaluate the accuracy of molar unit conversions by ECOTOX, 25 papers were reviewed. Molar units were found to be converted correctly by 24 of the 25 randomly selected papers. One paper did not measure metallic zinc as reported by ECOTOX, it was instead evaluating zinc pyrithione, an organic zinc antifouling compound. If this paper is considered as reporting error the "unit error" rate is 3%.

To evaluate the ability of the ECOTOX database to accurately represent the data presented in the original source, 100 journal articles were reviewed representing 821 individual toxicity values. The review resulted in identification of 171 values that were incorrectly represented by ECOTOX (20% error rate) and would have an impact on the outcome of the assessment. A number of other errors were identified (e.g., misclassification of effects types and test species), but they did not impact the outcome of this assessment. The majority of errors were associated with use of the salt concentration of a COC to represent the effect concentration, rather than the active ingredient concentration.

Other common errors were associated with the classification of NOEC values as effect concentrations and the use of mixture concentrations to represent a single chemical exposure. Use of the salt concentration as the effect concentration rather than the active ingredient would likely underestimate the potential for effects, while use of NOEC values would likely overestimate the potential. A summary of the results of the ECOTOX QA/QC process can be found in Appendix D-13.

Sediment – Direct Effects to Benthic Organisms

To determine potential hazard to benthic organisms from direct exposure to sediment COCs, relevant sediment guidelines and thresholds were identified for comparison with the observed environmental data. The primary standards and guidelines used in this assessment were the Washington State Marine Sediment Management Standards (SMS) and the Floating Percentile (FP) based freshwater sediment guidelines developed by Ecology (Avocet Consulting 2003; Avocet Consulting and SAIC 2002; RESET 2009). A number of other sediment guidelines were also used to provide additional context to assess the sediment data. Because this effort was not exhaustive, not all available guidelines were included.

Three sets each of freshwater and marine sediment guidelines (total of 6 sets of guidelines) were selected for comparison to observed sediment concentrations. It is acknowledged that sediment pore water may be an additional important route of exposure for benthic organisms; however, readily accessible pore water toxicity data and observed pore water concentration data for the Puget Sound regional were not available. In addition, variability in the methods used to extract and analyze pore water makes comparison across studies challenging.

Marine sediment data were compared to the following guidelines/standards:

- The Washington State SMS (Chapter 173-204 WAC), which consist of two levels, a "Sediment Quality Standard" (SQS) and "Cleanup Screening Level" (CSL) and the "Apparent Effects Thresholds" (AETs) including the "Lowest AET" (LAET) and the "Second Lowest AET" (2-LAET)
- The Canadian Marine Sediment Guidelines (CCME 2001) which consist of a "threshold effect level" (TEL) and a "probable effects level" (PEL).

The SMS SQS was the primary standard used to evaluate the marine sediment data; the remainder of the guidelines presented and described here were intended to provide additional context and included as part of the WOE discussion.

The SMS SQS represents the concentration below which no adverse effects to biological resources are expected; the CSL is less stringent and corresponds to the concentration at which minor adverse effects to biological resources are expected. For comparison to the SMS, all nonionic/nonpolar organic compounds were normalized to percent total organic carbon (TOC) content. However, if TOC content was outside the range considered appropriate for normalization, (i.e., less than 0.5 or greater that 3.0 percent), these data were only compared with the Puget Sound AETs. An AET represents the chemical concentration above which adverse biological effects have been demonstrated to always occur. The LAET was used as the equivalent of the SQS, and the 2LAET was used to represent the CSL.

The Canadian Marine Sediment Guidelines consist of two thresholds. The TEL represents the concentration below which adverse biological effects are expected to rarely occur, while the PEL defines the level above which adverse effects are expected to frequently occur. The TELs and PELs represent three effect ranges:

- The minimal effect range within which adverse effects rarely occur (i.e., fewer than 25% of samples have adverse effects occur below the TEL)
- The possible effect range within which adverse effects occasionally occur (i.e., the range between the TEL and PEL)
- The probable effect range within which adverse biological effects frequently occur (i.e., more than 50% of samples have adverse effects above the PEL) (CCME 2001).

The freshwater sediment data were compared to three sets of sediment guidelines. Ecology's FP based freshwater sediment guidelines Avocet Consulting 2003; Avocet Consulting and SAIC 2002; RESET 2009, the Canadian Freshwater Sediment Guidelines (CCME 2001, Smith et al. 1996), and the Consensus-based Guidelines (MacDonald et al. 2000) were used; all guidelines consist of a set of two thresholds.

The 2003 Draft Washington FP guidelines obtained from Ecology's EIM database were used in this assessment and include the "Sediment Quality Standard" (FP-SQS) and a "Cleanup Screening Value" (FP-CSL). The FP-SQS was the primary threshold used to evaluate the freshwater sediment data; the remainder of the thresholds/guidelines described here were used to provide additional context and included as part of the WOE discussion. The FP thresholds were developed based on bioassay hit definitions from Washington's marine SQS and the CSL. The FP thresholds include a third guideline based on a statistically significant difference (STAT); the STAT guideline was not used in this prioritization process.

The FP SQS defines a biological effect when the difference between the mortality rate in the test and control is greater than 10%, when the growth test/control ratio is less than 0.8 and when the decrease in Microtox® luminescence test/control ratio is less than 0.85. The FP CSL defines a biological effect when the test results for the same bioassays are greater than 25%, less than 0.7, and less than 0.75, respectively. A more detailed description of the derivation of these thresholds can be found in Avocet Consulting (2003) and Avocet Consulting and SAIC (2002).

Similar to the marine guidelines described above, the Canadian Freshwater Sediment guidelines (CCME 2001) consist of a "Threshold Effect Level" or TEL that represents the concentration below which effects are infrequently observed and a "Probable Effects Level" or PEL, which represents the concentration above which effects are frequently observed.

The Consensus Based Freshwater Sediment Guidelines (MacDonald et al. 2000) consist of a "Threshold Effects Concentration" or TEC, the level below which effects are not expected and a "Probable Effects Concentration" or PEC, the level above which effects are expected. In this context the term "consensus" does not mean agreement among scientists on the best guideline, but rather that a variety of sediment quality guidelines from different sources were combined to generate the thresholds. A more detailed discussion of the derivation of all of these sediment thresholds (except the FP's) can be found in Wenning et al. 2005. A summary of the sediment guidelines used in this assessment are presented in Appendix D-10,

Tissue – Direct Effects to Aquatic Life

To determine the potential for effects to aquatic life via waterborne or dietary exposure to the COCs, relevant tissue residue effect concentrations were identified for comparison with the observed environmental tissue residue data described above. Initially, the Environmental Residue Effects Database (ERED) (http://el.erdc.usace.army.mil/ered/) was identified as the source of effects data for the tissue assessment.

Due to the complexities associated with tissue residue data and concern for using data appropriate for this assessment, 25% of the 100 original papers on which the ERED data were based were reviewed for accuracy. Review of the original literature resulted in an unacceptable error rate (~50%). Numerous errors were identified and included use of the dose concentration to represent the effect concentration, a value not represented by a statistically significant effect, and incorrect values (e.g., paper did not measure concentration in tissue, a lipid normalized value presented as a wet weight value). Due to the high error rate, use of the ERED database would require review of all original data sources, which was beyond the scope of this project. A summary of the results of the ERED QA/QC process can be found in Appendix D-13.

As an alternative, two regional efforts that evaluated tissue residue effects were identified. Both the Lower Duwamish Waterway Group and the Lower Willamette Group recently completed final and draft, Remedial Investigation Baseline Ecological Risk Assessments respectively, which included review and evaluation of tissue residue effects data (Windward 2010, Windward 2009). Both groups identified numerous tissue residue effects concentrations that were subsequently reviewed by toxicologists from various agencies and groups. Since these data had been previously reviewed and vetted by numerous professionals, they were identified as a readily available reliable source of tissue residue effects data for this assessment.

The tissue residue effects concentrations are intended to estimate the direct effect of a COC on an organism via waterborne or dietary exposure. Although considerable effort has been expended over the years to relate tissue metal residues to effects, with the exception of selenium, mercury and tributyl-tin, these efforts have achieved only limited success (Adams et al. 2010).

Due to the disparate physical/chemical characteristics of metals, their environmental presence in multiple forms and states, the fact that some are essential micronutrients and some are controlled by metabolic processes, metals and inorganics as a group continue to be more toxicologically challenging than organics when trying to apply the tissue residue approach. Residue approaches for metals require detailed consideration of metal specific and species specific details and determination of the toxicologically active fraction of the total body/organ tissue residue (Luoma and Rainbow 2005 in McCarty 2010).

The development of the Biotic Ligand Model (BLM) for some metals demonstrates the successful use of a tissue residue based approach for metals that can be used when the target organ and receptors have been identified and the amount of metal necessary to produce toxicity has been established. However, this is not necessarily the case for whole-body tissue residue concentrations for most metals. Aquatic organisms use a variety of storage, detoxification and excretion mechanisms to address metal exposure. As a result, measuring the total metal in an organism provides limited information regarding the biologically active metal concentrations

within an organism (Adams et al. 2010; Meador et al. 2008). Due to the complexity of this type of assessment, a meaningful evaluation of metal tissue residues requires a much more detailed evaluation of the available toxicity data than could be conducted here. Therefore, tissue residue data for metals were not evaluated in this hazard prioritization process.

Although there are similar limitations in the use of tissue residue effects concentrations for organic chemicals, the relationships for some compounds (e.g., bioaccumulative and persistent) are much more established (Meador et al. 2008). Thus, the tissue assessment was restricted to bioaccumulative organic compounds that are not readily metabolized and for which data (effects and observed environmental) were available (mercury, PCBs, PBDEs, DDTs, and dioxins). When available, whole-body tissue residue effects data for fish, non-decapod invertebrates and decapods were used in the assessment. All of the available tissue residue effects data were based on wet weight concentrations. Where possible, a qualitative assessment of regionally based lipid normalized effects thresholds are discussed as part of the WOE discussion. A summary of the effects data used in this assessment can be found in Appendix D-11.

Wildlife

Four wildlife receptors were selected for evaluation by this assessment; harbor seal, osprey, river otter and great blue heron (A detailed description of species selection is described in the subsequent section on wildlife receptor and COC selection). The wildlife effects thresholds are based on the daily dose (mg chemical/kg-body weight/day) of a COC known to cause adverse effects to test species of birds or mammals. There are no state or federal standards to evaluate contaminants in wildlife; wildlife effects doses were obtained from published dose-response studies. These studies typically expose test animals to a COC through ingestion of food or water containing known contaminant concentrations and observe any effects on growth, reproduction, development or survival.

Effect doses for this assessment were obtained from multiple publications, some being compilations of effect doses from EPA efforts and included the Great Lakes Water Quality Initiative Criteria Documents (EPA 1995), the Ecological Soil Screening Levels (EPA 2007), Toxicological Benchmarks for Wildlife (Sample et al. 1996), the Draft Lower Willamette River Remedial Investigation Baseline Ecological Risk Assessment (Windward 2009) and the Lower Duwamish Waterway Remedial Investigation Baseline Ecological Risk Assessment (Windward 2010). Individual publications supplemented these sources as available. Effects doses were applied as they were presented in the source documents unless errors of interpretation were found (e.g., the dose causing an effect is incorrect, typographical errors, unit conversion errors). Safety or uncertainty factors were not used to estimate potentially hazardous levels in the assessment.

Due to data availability issues, it is common to use data from published dose-response studies conducted on test species (birds or mammals) other than those of interest in an assessment. All daily doses associated with either bird or mammal species were grouped for use with the appropriate bird or mammal receptor identified for this study. For the effects daily doses calculated directly from published data, safety factors were not applied to adjust for interspecies, lowest effect to no effect value, or any other uncertainty. This decision was made because there is no knowledge of which direction, and to what magnitude uncertainty would be biased. For

example, the sensitivity of one species to chemical exposure may be greater or lesser than another. Until each species is tested, it is impossible to predict which species will be more sensitive and the degree of difference in sensitivity between species. In addition, safety factors are not standardized and therefore, when applied, are not consistent in magnitude.

A number of field studies have documented bioaccumulation of PCBs and dioxins/furans in Puget Sound harbor seals and orcas, conducted biopsies and examined immune suppression in these organisms (Ross et al. 1995, de Swart et al. 1996, Ross et al. 1996, Ross et al. 2004, Levin et al. 2005, Cullon et al. 2009). However, to date, a dose-response study has not been conducted to provide the necessary information to develop a toxicity-based daily ingestion dose for marine mammals. Because these results are not dose-response studies, they could not be used in the quantitative part of this assessment. They are instead discussed qualitatively as part of the WOE discussion. Published research on wildlife exposure and effects to COCs conducted in or near Puget Sound are summarized in the Results Section. A summary of the quantitative effects data used in the wildlife assessment can be found in Appendix D-12.

Human Health

The effect threshold used for the human health assessment was based on the National Toxics Rule (NTR) 40CFR§131.36. A number of national and regional fish consumption rates (Table 5) were calculated using the same methods to provide additional perspective and because the NTR specified rate is considered under protective for certain populations and ethnic groups such as Asians, Pacific Islanders and Native Americans. The tissue thresholds were derived by back calculating intake rates from existing water quality criteria using the applicable bioconcentration factors (BCFs) presented in the original water quality criteria development documents. COCs not part of the NTR were not assessed for human health hazards, e.g. lead, triclopyr, and the sum of cPAHs although individual PAHs were prioritized.

Table 5. List of consumption limits assessed in this document.

Guidance/Reference	Rate
NTR Standard Rate(64 FR 61184)	6.5 gm/day
EPA Recreational Rate (EPA 2000)	17.5 gm/day
EPA Subsistence Rate (EPA 2000)	142.4 gm/day
Tulalip Tribal/King County Asian Pacific Islander Rate (Toy 1996; Sechena 1999)	242.5 gm/day
Suquamish Tribal Rate (Suquamish Tribe 2000)	769 gm/day

There are no applicable human health standards for sediment. Both the Washington Sediment Management Standards and the Model Toxics Control Act (MTCA) use site specific assessments of exposure to assess human health risk; however, this process was not practical to conduct on a Puget Sound regional scale. Potential effects associated with exposure to sediment through recreational, shell fishing, or beach use would require parameterizing a human health risk assessment which was beyond the scope of this project. Similarly, water was not evaluated because recreational or consumptive water uses would require developing regional estimates of lifetime human water exposures which was also beyond the scope of this project.

Petroleum

Total petroleum hydrocarbons (TPH) is a term used to describe a large family of chemical compounds that originate from crude oils, coal tars, oil shales, and similar materials. The specific composition of petroleum products varies depending upon (1) the source of the crude oil and (2) the refining practices used to produce the product. TPH and "TPH gasoline" represent a mixture of petroleum compounds and serve as coarse estimates of the presence of the individual constituents that may cause toxicity to aquatic organisms. The ECOTOX database used for the water assessment only includes data for individual chemical components and does not include toxicity data for complex mixture compounds such as TPH. As a result, it was necessary to use a slightly different approach to assess TPH mixtures.

Toxicological evaluation of petroleum mixtures have historically been conducted in the aftermath of oil spills and similar events. Sources consulted for toxicity information on TPH and petroleum products were API, 1994; Barron, et al. 1999a; Barron, et al. 1999b; Tsvetnenko, 1998; and Woodward et al. 1983. The majority of the available toxicity data were for crude oils; however, these studies were deemed unsuitable since none of the available observed environmental data were for crude oils.

Based on availability of both observed and effects data, four classifications of petroleum products from the available environmental data were evaluated: heavy fuel/bunker oil, diesel fuel, gasoline, lube oil. Only toxicity results assessing the water soluble fraction (without free product) were used to assess these data. A more complete evaluation of petroleum would require analysis of parent and alkyl PAHs as well as issues such as phototoxicity which were beyond the scope of this assessment. In addition to the specific assessment for petroleum described here, a suite of individual and high/low molecular weight PAHs were evaluated in both the water and sediment assessments previously described above.

Selection of Wildlife Receptors and COCs

The following section describes the process used to select wildlife species and COCs evaluated in the wildlife assessment. When conducting a wildlife effects assessment, it is necessary to select a finite number of species or "receptors". In part, this is due to the impracticality of trying to assess effects to all wildlife species that reside in the Puget Sound region.

Receptors are generally assumed to be conservative representatives of other species with similar diets, trophic status and biology. A bird and a mammal species were evaluated in freshwater habitats and marine habitats. Two bird (osprey and great blue heron) and two mammal species (harbor seal and river otter) were selected based on their position as top level predators, a diet consisting primarily of fish, and their use of different feeding strategies, all of which make them at greatest risk of exposure. In addition, a substantial amount of ecological information has been published about these species allowing reasonable exposure modeling assumptions.

The four selected receptors are not intended to represent all wildlife species that may be exposed to COCs in the Puget Sound area. However, these receptors do represent other piscivorous

species which tend to bioaccumulate chemicals to a greater degree than lower trophic level species.

The great blue heron was selected as the freshwater bird species because it is a common, piscivorous bird in Puget Sound freshwater and marine habitats. Some populations exclusively reside and feed in freshwater habitats as evidenced by large, established heron rookery sites on lakes and rivers. This large wading bird consumes fish of a variety of sizes and species including large predatory fish such as largemouth bass and trout. Also, some are year-round residents of this region. The river otter was selected as the freshwater mammal species because it is a common mammal in freshwater habitats and consumes primarily fish (EPA 1993). There is also evidence that river otter in Washington State accumulate mercury, PCBs, and dioxins/furans (Grove and Henny 2008). Although river otter may migrate between marine and freshwater habitats, some are exclusive to freshwater habitats. For the purposes of this assessment, it was assumed that both the great blue heron and river otter consume 100% of their prey from freshwater habitats and reside year-round in the Puget Sound region.

Osprey and harbor seal were selected as marine habitat receptors. The osprey was selected over the bald eagle, another common top predator of fish, for a number of reasons. Osprey primarily consume fish as compared to the bald eagle with consumes a large portion of birds and mammals; the osprey is much smaller than the bald eagle, making their relative ingestion rate and exposure higher. Osprey consistently hunt by diving or grasping prey from the water, whereas the bald eagle may scavenge for food, exclusively hunt salmon during salmon runs, or hunt for birds. Osprey migrate south during the winter, but breed in western Washington.

The harbor seal was selected to represent an aquatic marine mammal because it is a top predator feeding exclusively on aquatic prey, primarily fish, and has been reported to have elevated tissue concentrations of PCBs. The harbor seal was selected over the orca because the harbor seal is a smaller mammal with relatively higher ingestion rate and exposure, and because much more ecological information is available for the harbor seal. For the purposes of this assessment, it was assumed that the diet of both the osprey and harbor seal was 100% fish and that they reside only in Puget Sound.

The COCs evaluated by the wildlife assessment were limited to bioaccumulative compounds for which sufficient effects and environmental data were available and included mercury, PCBs, DDTs, and dioxins/furans. Many of the COCs are neither detected in fish (wildlife prey) nor bioaccumulative. PAHs are metabolized by fish, and therefore are not bioconcentrated (or analytically detected very frequently). Triclopyr is broken down quickly in the environment, and is not bioaccumulative. Bis(2-ethylhexyl)phthalate (DEHP) was infrequently detected (<10% FOD) in only two species of fish collected from nearshore habitats. Nonylphenol is also not considered to be bioaccumulative. Although nonylphenol is a potential endocrine disruptor, describing the potential endocrine disruption of nonylphenol to wildlife populations has never been done before.

Metals have naturally high FOD in the environment; however, the toxicity of some metals is complicated by their need as essential trace minerals. At the same time, many metals are moderately bioaccumulative and toxic to wildlife. Lead poisoning of birds exposed to lead shot and fishing weights has been a long standing concern. Although lead shot was banned for use in

waterfowl hunting in 1991, it continues to be used in upland game hunting, posing risk to non-waterfowl bird species (USGS 2009). Because this assessment is focused on the aquatic environment, assessment of upland exposure of birds to lead shot was outside the scope of this assessment and not evaluated. Based on the chemical characteristics of these COCs, it was assumed that the potential for chronic wildlife exposure was very low and effort was focused on the more persistent and bioaccumulative COCs.

Exposure Model for Wildlife Receptors

Daily doses of COCs were estimated using a simple exposure model that included a body weight normalized sum of daily food intake, drinking water intake and incidental sediment ingestion during foraging and other behaviors. This is represented by the following algorithm:

$$\text{TDD} = \frac{[C_w * IR_w] + [C_s * IR_s] + [C_f * IR_f]}{BW}$$

Where:

TDD = Total daily dose (mg/kg-BW/d)

C_w = 95% UCL of mean chemical concentration in water (mg/L)

C_s = 95% UCL of mean chemical concentration in sediment (mg/kg)

C_f = 95% UCL of mean chemical concentration in food (mg/kg)

IR_w = Ingestion rate of water (L/day)

IR_s = Incidental ingestion rate of sediment (kg/day)

IR_f = Ingestion rate of food (kg/day)

BW = Body weight (kg)

The 95% upper confidence limit of the mean (95% UCL) concentration of COCs in water, sediment and food was used to estimate the amount of each COC ingested by wildlife. The datasets from which the 95% UCL statistic was calculated were the same as those used in the water, sediment, and tissue assessments with the exception of the fish tissue dataset. The 95% UCL fish tissue concentrations are summarized in Appendix D-8. The diet of each receptor was conservatively assumed to be 100% fish. Although a proportion of the receptor's actual diet may include invertebrates, the 95% UCL fish tissue concentration is higher than the invertebrate tissue concentration. Therefore, assuming a diet of 100% fish results in a conservative approach, and meets the screening goal of this wildlife assessment. Data for all fish species, with the exception of six gill sharks, were assumed to be prey. A number of sources were reviewed to characterize the harbor seal (Cullon et al. 2005, EPA 1993), osprey (EPA 1993), great blue heron (EPA 1993, Butler 1992, Alexander 1977), and river otter (EPA 1993) diets.

The wildlife receptors used in this assessment prey on a wide variety of species; based on published information on their prey consumption, none show consistent preference for some prey species over others. Because prey size data were unavailable for observed concentrations, size was not a criterion used to screen fish tissue data for inclusion in the assessment.

The daily food ingestion rates were estimated using the allometric equations of Nagy (1987) which relate food ingestion rate to body weight. These equations are:

Birds: $FI = 0.0582 * BW^{0.651}$

Mammals: $FI = 0.0687 * BW^{0.822}$

Where:

FI = food ingestion rate (kg/day dry weight)

BW = body weight (kg)

Calder and Braun (1983) also use this type of relationship to estimate water ingestion rates.

These allometric equations were used to estimate drinking water intake for receptors and are presented below.

Birds: $WI = 0.0598 * BW^{0.67}$

Mammals: $WI = 0.099 * BW^{0.90}$

Where:

WI = water intake (L)

BW = body weight (Kg)

Sediment ingestion rates have not been empirically measured in wildlife studies, but have been estimated using acid-insoluble ash measurement in scat or digestive tracts of animals. Beyer et al. (1994) used ash measurements in scat to estimate the relative proportion of inorganic solids (i.e. sediments and soils) in the diet of multiple wildlife species. None of the four wildlife receptors were subjects of the Beyer et al. study; however, these data are useful to develop rough estimates of sediment ingestion rates for species that share similar feeding strategies. Empirical measurement of sediment ingestion rate is challenging and difficult to model due to ecological variability between and within species. Thus, the sediment ingestion rates were established using best professional judgment and relied heavily on the Beyer et al. (1994) which published estimates varying from <2% to 9% of the daily food ingestion rate for mammals[1] and <2% to 30% of total food ingestion rate for birds[2].

Since total exposure is sensitive to sediment ingestion, separate daily doses for the species evaluated in this assessment were estimated assuming a low and high sediment ingestion rate based on their similarities in feeding strategy and foraging habitat to species from Beyer et al. (1994). Best professional judgment was used to select a low and a high sediment ingestion rate intended to bound the range of realistic potential sediment ingestion rates for each receptor (Table 6).

Where possible, the body weight assumed for each receptor was based on local information summarized in Table 6. The average body weight for the smaller sex, if applicable, was used in the model, because food intake for smaller-bodied animals is proportionately greater than for the larger-bodied cohorts resulting in a larger daily dose and a more conservative estimate. Body weights and ingestion rates for adult life stages were applied to represent the majority of the animal's reproductive lifetime. However, effects dose studies included dosing of immature

[1] Only four species of mammals were included in the study and only one, the raccoon, forages in aquatic habitat.

[2] Most species of birds in the study were either shorebirds or herbivorous birds. Sediment ingestion rates for dabbling and diving ducks ranged from <2 to 3% of food ingestion.

individuals. Therefore, the effects doses reflect the most sensitive lifestage and should be protective of younger lifestages.

Table 6. Wildlife body weights and ingestion rate assumptions.

Receptor	Body weight (kg)	Source	Food IR (kg/day dw)	Sediment IR (% of Food IR)	Water IR (L/day)
Osprey	1.45	EPA 1993	0.075	1 (4)	0.078
Harbor Seal	77.0	Assuncao et al. 2007	0.985	2 (4)	1.098
Great Blue Heron	2.1	Simpson 1984 as cited in Butler 1992	0.094	2 (4)	0.098
River Otter	7.9	EPA 1993	0.376	4.5 (9.0)	0.636

IR = ingestion rate. Two different sediment ingestion rates were applied for each receptor while holding all other variables constant because this variable is the greatest source of uncertainty in the daily dose model. The higher rate is in parentheses.
Food ingestion rates are from Nagy (1987); sediment ingestion rates are from Beyer et al. (1994); water ingestion rates are from Calder and Braun (1983).

Hazard Assessment

As previously discussed, the intent of this assessment was to provide a general, high level overview of the potential for the COCs to cause deleterious effects in the matrices evaluated. To conduct the quantitative portion of the hazard evaluation, the observed environmental data were compared to the respective effects concentrations. It should be noted that these comparisons are based on single chemical exposures in a single matrix (water, sediment and tissue) and do not account for any effects associated with exposure to chemical mixtures or other physical stressors or conditions (i.e., temperature, dissolved oxygen, hardness etc.) that may be present and influence bioavailability. Sufficient data for both effects and observed environmental concentrations were not available to evaluate the priority for every COC for each type of evaluation.

Water – Direct Effects to Aquatic Life

To assess potential effects to aquatic life through direct exposure to surface waters, observed surface water concentrations for COCs were presented as box plots of percentiles (5^{th}, 10^{th}, 25^{th}, 50^{th}, 75^{th}, 90^{th} and 95^{th} %iles) plotted adjacent to box plots of percentiles representing the available effects concentrations in a series of figures. If water quality criteria (WQC) were available, these values were also presented. In some cases (e.g., DDT and mercury), the WQCs are well below the available effect concentrations.

Some WQC, particularly those for bioaccumulative chemicals, are derived based on a "final residue value" and the potential to bioaccumulate which typically results in a value that is lower than effects concentrations based on direct exposure. In some cases (e.g., nonylphenol) chronic effects data are limited and the chronic WQC is derived using an acute to chronic ratio (ACR). Use of the ACR can also result in a value that may be below effects concentrations presented in ECOTOX. For a more detailed description of how WQC were derived for these COCs see http://water.epa.gov/scitech/swguidance/waterquality/standards/criteria/aqlife/index.cfm. It is also important to note that water quality criteria are not derived to protect aquatic organisms

through the bioaccumulation pathway. The tissue residue assessment described below was intended to help address this pathway for a select group of COCs.

The total number of analytical measurements and number of observed detected concentrations are also presented on each figure. Most freshwater and marine (near- and offshore) data were presented on the separate figures; results for some COCs for which data were limited are combined into a single figure. If a COC was not detected (or measured), or effects data were not available, a figure was not prepared. All data were plotted on a log scale.

Sediment – Direct Effects to Benthic Organisms

To assess the potential for effects to benthic organisms, sediment COC concentrations were presented as box plots of percentiles plotted against the sediment guidelines (represented as horizontal lines on each figure). The standard or guideline use as the primarily comparison is presented as a solid red line, while the remainder of the guidelines provided for additional context are presented as dotted blue lines. When appropriate, based on the guidelines used for comparison, both dry weight and OC normalized concentrations were presented. In general, separate graphics were generated for marine and freshwater sediment data; results for some COCs for which there were limited data are combined into a single figure. If a COC was not detected (or measured), or effects data were not available, a figure was not prepared. All data were plotted on a log scale.

Tissue Residues – Direct Effects to Aquatic Life

The tissue assessment was similar to that described above for water. Observed tissue residue concentrations were presented as box plots of percentiles along with the available tissue residue effects concentrations; all data were plotted on a log scale. Graphics were generated for whole-body tissue types (fish, non-decapod invertebrates, and decapods) where both effects and observed environmental data were available. Marine and freshwater tissue data were graphed separately. If a COC was not detected, or effects data were not available, a figure was not prepared. As discussed above, the tissue residue assessment was limited to four PBTs (DDTs, dioxins, PCBs and mercury). Although PBDEs were not assessed quantitatively, the current effects literature and regional studies are reviewed in the WOE discussion.

Wildlife

The wildlife assessment compared literature-based daily effects doses for birds and mammals to the estimated daily doses of COCs for the four receptors (great blue heron, osprey, river otter and harbor seal). The literature-based daily effects doses were rank ordered and plotted against the estimated daily doses; data were plotted separately for birds and mammals. Two estimated daily doses were calculated for each receptor based on a low and a high estimate of sediment ingestion rates. Both estimates are shown on the figures.

Observed PCBs, DDTs and dioxins/furans data were assessed as sums. PCBs were assessed as Aroclor® sums for tissue and sediment and as PCB congener sums in water because too few Aroclor® detections occurred in the observed water data. DDT sums included DDT, DDE, and

DDD isomers. Dioxins and furans were converted to TEQs and summed to a total TEQ. The effects doses included individual Aroclors®, or DDTs, DDEs, and DDDs and their mixtures. Dioxin and furan effects doses were treated the same as observed data by converting to TEQs and summing to a total TEQ.

Human Health

The human health assessment utilized the National Toxics Rule (NTR) (40CFR§131.36) to establish default assumptions of body weight, toxicity and daily fish/shellfish consumption rates. The NTR uses a national average fish tissue consumption rate of 6.5 gm/day; this consumption rate was used to calculate a screening threshold and used as the primary basis of this assessment. However, this rate is suspected to be under-protective of various other users and ethnic groups (EPA 2007).

To account for different groups who may consume fish/shellfish at higher rates, five consumption rates were also used to derive alternative levels for informational purposes. Two consumption rates (242 gm/day King County API (Sechena 1999) and 243 gm/day Tulalip Tribe (Toy 1996) were almost identical so they were averaged to create one consumption scenario of 242.5 gm/day. In addition to the NTR standard rate and the average of the King County API/Tulalip rate, the EPA recommended recreational and subsistence consumption rates (EAP 2000) and the Suquamish ingestion rate (Suquamish Tribe 2000) were also used. The levels calculated based on these additional consumption rates are presented to provide additional context and are discussed as an additional WOE.

This methodology was used to evaluate bivalve, fish (whole-body and filet) and other invertebrate tissue data for human health consumption risks. The NTR is the only regulatory standard in Washington State applicable for human health risks related to consumption of surface water. However the surface water standards in the NTR are predominantly influenced by bioaccumulation by fresh water fish tissue and not the consumption of water alone. Consumption and/or dermal exposure to water alone would require development of a human health risk assessment for the entire Puget Sound region. A water risk assessment would need to include an estimate of freshwater exposure point concentrations or probabilistic estimates of exposure; such an evaluation was deemed to be beyond the scope of this assessment.

Tissue criteria were backcalculated from the NTR based water quality criteria as shown below in Table 7. The bioconcentration factors (BCFs) shown are from the original criteria documentation. These BCFs and the applicable fresh and marine water quality criteria were used along with the original 6.5 gm/day and modified consumption rates to derive tissue concentrations deemed protective at a range of tissue consumption rates as shown in Table 5.

Table 7. Summary of NTR water quality criteria and bioconcentration factors (BCF) used in the human health assessment.

COC	BCF	NTR Freshwater Human Health Criteria - Water and Organisms (µg/L)	NTR Marine Water Human Health Criteria - Organisms Only (µg/L)
Arsenic	44	0.018	0.14
Mercury (estuarine)	3765	n/a	0.15
Mercury (freshwater)	5500	0.14	n/a
Bis(2-Ethylhexyl)Phthalate	130	1.8	5.9
2,3,7,8-TCDD (Dioxin)	5000	0.000000013	1.4E-08
Total Polychlorinated Biphenyls (PCBs)	31,200	0.00017	0.00017
4,4'-DDT	53,600	0.00059	0.00059
4,4'-DDE	53,600	0.00059	0.00059
4,4'-DDD	53,600	0.00083	0.00084
Anthracene	30	9600	110,000
Benzo(a)Anthracene	30	0.0028	0.031
Benzo(a)Pyrene	30	0.0028	0.031
Benzo(b)Fluoranthene	30	0.0028	0.031
Benzo(k)Fluoranthene	30	0.0028	0.031
Dibenzo(a,h)Anthracene	30	0.0028	0.031
Fluoranthene	1150	300	370
Fluorene	30	1300	14,000
Indeno(1,2,3-cd)Pyrene	30	0.0028	0.031
Pyrene	30	960	11,000

To account for potential rounding errors and changes in estimates of cancer toxicity since the NTR was adopted; cancer slope factors or reference doses were first derived from the NTR calculations for both freshwater and marine waters. These slope factors, or dose for mercury, were then used in the following formulas to derive tissue thresholds.
Following is the calculation of the human health criterion for freshwater organisms along with 2L of drinking water per day consumption:

$$\text{Fresh} - \text{HH} = \frac{\text{RF x BW x } (1{,}000\,\frac{\mu g}{mg})}{q1 * \text{x } [\text{WC} + (\text{FC x BCF})]}$$

Where:

Fresh-HH = Freshwater criterion in ug/L
RF = Risk Factor = 1 x 10 (-6)
BW = Body Weight = 70 kg
q1* = Cancer slope factor/toxicity (Hg only), chemical specific
WC = Water Consumption = 2 L/day
FC = Fish and Shellfish Consumption varied according to Table 5
BCF = Bioconcentration Factor

Following is the calculation of the human health criterion for marine water organism only consumption:

$$\text{Marine} - \text{HH} = \frac{\text{RF x BW x } (1{,}000\,\frac{\mu g}{mg})}{\text{q1} * \text{x FC x BCF}}$$

Where:

Marine-HH = Marine criterion in ug/L
RF = Risk Factor = 1 x 10 (-6)
BW = Body Weight = 70 kg
q1* = Cancer slope factor/toxicity (Hg only), chemical specific
FC = Fish and Shellfish Consumption varied according to Table 5
BCF = Bioconcentration Factor

For COCs not included in the NTR, a hazard evaluation was not conducted to evaluate their hazard level for the same reasons that water and sediment hazard evaluation were not conducted.

Petroleum

As previously discussed, due to issues associated with the non-specific nature of petroleum product measurements in surface waters, this COC was evaluated using a slightly different process than the remainder of the COCs in water. Petroleum in freshwater was evaluated using data associated with the release of products to water (spills).

Most available toxicity data were for crude oils, which were not considered applicable for this assessment because no crude oil environmental data were available. Similarly, a large amount of observed oil and grease data were available from the EIM and King County LIMS databases. However, these data do not describe a specific petroleum product; they encompass waxes, greases and other fatty acid substances from both animal, vegetable and petroleum origins. Because these environmental data are non-specific, and potentially toxic components may vary within the same concentration measured by this method; these data were considered unusable for this assessment. Thus, toxicity data and environmental data were both only available for four petroleum products: heavy fuel/bunker oil, diesel fuel, gasoline, and lube oil. Toxicity data for these fours products were almost entirely based on lethal concentrations to 50% of the exposed population (LC50s).

Environmental data for these four products were plotted against the available freshwater toxicity data. Only two petroleum product data results were available for marine waters and these were insufficient to estimate hazard priority levels.

Determination of Priority

Due to the broad screening nature of this assessment, in addition to uncertainties associated with the available data (see Uncertainty Section), a conservative approach was used to determine the potential hazard posed by each COC. While an effort was made to use a consistent approach to classify the priority for each component of the assessment, due to the nature and availability of the data used and variability of methods used for each component, there are differences between some of the approaches.

The reader is urged to use caution when comparing priorities across matrices and type of assessment. The potential for ecological effects for COCs classified as Priority 1 is much higher than that for Priority 2. However, the specific type of ecological effect is not defined for Priority 1 COCs and can vary from mortality to more subtle impacts like physiological changes. The following sections describe the process by which the potential for effects was determined for each element of the assessment. Table 8 summarizes the thresholds used to define priority levels and sufficiency of data.

Water – Direct Effects to Aquatic Life

A conservative approach was used to determine the potential for effects to aquatic life from direct exposure to surface waters. If the 90th %ile of the observed environmental concentrations of a COC was above the 10th %ile concentration of the effect concentrations, the COC was classified as "Priority 1". If the 90th %ile of the observed environmental concentrations was less than the 10th %ile of the effect concentrations, the COC was classified as "Priority 2". If there were insufficient data (<15 effects or <50 observed environmental values) to assess a particular COC, it was classified as "Unknown" or "U". A classification of Priority 2 or "U" is not intended to suggest that this COC is not important. It is assumed that all of these COCs are priorities at some level.

In freshwaters, hardness can have significant influence on the toxicity of metals. Due to the large volume of data used for this assessment it was not practical to evaluate the hardness concentrations associated with each observed or effect metal concentration. The reader is cautioned to take this factor into account when evaluating the findings of the freshwater metals assessment. The WQC were calculated for metals with hardness-based standards using a hardness value of 25 mg/L-CaO_3 which is approximately the average freshwater hardness in Western Washington.

Where available, data from regional studies and other readily available thresholds were used as an additional WOE to qualitatively assess each COC. In general, the available regional data for the direct water exposure were limited to copper and PAHs.

Sediment – Direct Effects to Benthic Organisms

To determine the potential for effects to benthic organisms from direct sediment exposure, the 90th %ile concentration for each COC was compared to the marine SQS or freshwater FP-SQS, in addition to the other sediment guidelines described above.

A process similar to that described above for water was used to assess COC priorities for sediment. If the 90th %ile of the observed sediment concentrations was above the marine SQS or the freshwater FP-SQS the COC was classified as Priority 1; if the 90th %ile concentration was less than the sediment standard/guideline is was classified as Priority 2. COCs for which there were insufficient data (effects or observed concentrations data) to assess were classified as "Unknown" or "U". Sediment COCs were classified as "U" if a FP-SQS or SQS value was not available or if the number of observed environmental concentrations was low (<30). As previously discussed, classification of a COC as a Priority 2 or "U" does not indicate the COC is not potentially important.

It is assumed that all of these COCs are priorities at some level. Where available, data from regional studies and other readily available thresholds were used as an additional WOE to qualitatively assess each COC. In general, the available regional data for the direct sediment exposure were limited to PAHs and PCBs.

The level of uncertainty associated with data availability for each COC was also summarized. Data uncertainty was based on the total number of measurements and the availability of sediment thresholds for each COC. As indicated for water above, this assessment assumes that the available environmental data are representative of the overall region; however, the lower the number of measurements for each COC, the greater the uncertainty that these data are representative.

Tissue –Direct Effects to Aquatic Life

The approach used to determine priorities in the tissue assessment was the same as that described above for water. The data for this element of the assessment were very limited; when interpreting the tissue assessment results, the reader is also encouraged to take into account the amount and type of both tissue residue effects and observed data available. A COC was classified as "Priority 2" if the 90^{th} %ile observed concentration was below the 10^{th} %ile effects concentration. A "U" or "Unknown" priority indicates there was insufficient effects data (<5 effects or >20 observed values) to allow assessment.

Where available, data from regional studies and other readily available thresholds were used as an additional WOE to qualitatively assess each COC. In general, the available regional data for tissue residue related impacts was limited to PAHs and PCBs.

Wildlife

A COC was classified as "Priority 1" when the estimated daily dose was greater than or within 0.1 times the lowest effect dose. Because there are far fewer published effects doses for wildlife than aquatic life, there is greater uncertainty in estimating the lowest effect threshold. For this reason, a COC was classified as "Priority 2" if its estimated daily dose was less than an order of magnitude (i.e., a factor of 10) of the lowest effect dose. A "U" or "Unknown" priority indicates there was insufficient effects data (<5 effects values) to allow assessment. Only one published effects dose was available for PBDEs in birds (Fernie et al. 2011), and none for mammals. Thus, a discussion of the estimated total PBDE daily doses for the avian receptors compared to this published effects dose is included in the WOE discussion in lieu of the full quantitative evaluation.

Human Health

If the 90^{th} %ile of the observed tissue concentrations exceeded the NTR screening criteria the COC was classified at Priority 1; if the NTR value was below the 90^{th} %ile concentration, the COC was classified as Priority 2. Some COCs had many environmental measurements for one tissue type, but few of other tissue types. In these instances, best professional judgment was used to determine the adequacy of all the tissue data within the fresh, near, or offshore

environment to develop a hazard ranking for the COC in tissue on the whole. As discussed above, classification as a Priority 2 does not indicate that a COC is not a priority. It is assumed that all COCs are priorities at some level. Results of the comparison to other consumption rate data were used as an additional WOE and discussed qualitatively below. Most COCs were found at comparable concentrations across the bivalve, fish, and other invertebrate tissue groups suggesting that prioritization rankings are relatively robust regardless of tissue type evaluated.

Table 8. Thresholds used to define priorities and sufficiency of data.

Line of Evidence	Threshold for Priority	Threshold for Sufficient Data
Surface Water	90th%ile Observed Conc. > *10th %ile Effects Conc.* *or* *Acute WQC or Chronic WQC*	n ≥ 15 for Effects Data n ≥ 50 for Observed Data
Sediment	90th%ile Observed Conc. > *SQS*	n ≥ 100 for Observed Data
Tissue Residue Effects	90th%ile Observed Conc. > *10% of 10th%ile Effects Conc.*	n ≥ 5 for Effects Data n ≥ 20 for Observed Data
Wildlife	Daily Dose >*10% of Lowest Effects Dose*	n ≥ 5 for Effects Data
Human Health	90th%ile Observed Conc. >*NTR Criterion*	Best Professional Judgment

Results and Discussion

The following sections provide an overview of the availability of both the environmental occurrence data and effects data in addition to the outcome of the individual effects assessments and a summary of the overall outcome of the assessment.

Environmental and Effects Data Availability

Water

Surface water data were used to evaluate direct effects to aquatic life, and in the wildlife assessment as appropriate to estimate exposure through water ingestion. The number of measurements for COCs in freshwater ranged from over 5000 for copper to less than 10 for dioxins. With the exception of PCB and PBDE congeners, dioxins/furans and nonylphenol, there were more than 1200 measurements for each COC in freshwater.

Relative to freshwater data, the number of measurements in marine (near- and offshore) surface waters was significantly less; nearshore data were most limited. With the exception of copper, there were less than 50 measurements (often less than 15) for each COC in nearshore waters. In general, there were less than 100 measurements for each COC in offshore waters. Marine data were not available for petroleum, triclopyr and dioxins. The water data are summarized in Appendix D-8.

The relatively low number of measurements for many of the COCs in marine waters suggests that the spatial coverage for these data is limited and these data may not be representative of the region.

Effects data were considered "sufficient" when a range of values (at least 15 effect concentrations) representing a variety of endpoint types other than mortality were available. It is generally uncommon for ambient concentrations of COCs to be present at levels that would cause mortality; organisms are typically exposed to concentrations likely to cause more subtle effects (e.g., effects to growth and reproduction). If the effects data were limited to mortality based endpoints, it is possible that priority rank may have been underestimated because the potential for effects other than mortality could not be evaluated. For example, most of the available effects data for PCBs in marine water were based on mortality; and this COC was classified as Priority 2. However, exposure to low levels of PCBs can result in food web biomagnification and cause effects to the immune systems of higher trophic level organisms. As such, it should be noted that direct water exposure is not the most sensitive approach to assess this COC.

In freshwater, there were sufficient effects data for most of the COCs with the exception of some individual PAHs, dioxins and PBDEs. Effects data for these COCs were limited, particularly for endpoints other than mortality.

Effects data for aquatic life in marine waters were sufficient for some COCs, but were limited (especially non-mortality effect data) for a number of COCs. In marine waters effects data for arsenic, cadmium, PCBs, PBDEs and a number of individual PAHs were limited.

Sediment

Sediment data were used to evaluate direct effects to benthic organisms and were also incorporated into the wildlife assessment where appropriate to estimate exposure from incidental sediment ingestion. Sediment data were available for all COCs with the exception of triclopyr and petroleum. All sediment data are summarized in Appendix D-8. With the exception of PCB and PBDE congeners, nonylphenol and a few individual PAHs, there were more than 300 measurements for each COC in both freshwater and marine (near- and offshore) sediments. Of the 3 sets of freshwater sediment guidelines used in this assessment, threshold values were available for the majority of sediment COCs. Only one set of freshwater guidelines was available for a number of individual PAHs, LPAHs, HPAHs and dioxin/furans; two sets of guidelines were available for DDT and some individual PAHs and only a single threshold was available for nonylphenol.

Of the 3 sets of marine sediment guidelines used in this assessment, threshold values were available for the majority of sediment COCs. However, only 2 sets of guidelines were available for some individual PAHs and one set for dioxins/furans; only one sediment guideline was available for nonylphenol.

Tissue

Tissue data were used to evaluate direct effects to aquatic life (limited to PBTs) and wildlife (select organic chemicals only) and to assess human health (select organic chemicals, mercury and arsenic only). The different assessments required the various combinations of tissue types. A summary of the tissue data used for each assessment type can be found in Appendix D-8. The number of measurements for some COCs and tissue types was very low and limited the degree to which these chemicals could be assessed.

Effects data to evaluate the direct impact of tissue residues on aquatic life were very limited for most COCs, particularly for marine fish; effects data for these tissue types were not available for mercury, dioxins/furans and DDTs. The number of tissue residue effects concentrations for some tissue types and COCs was often less than 5. The lack of tissue residue effects data poses limitations on this element of the assessment. As a result, the reader is cautioned to evaluate the available data when interpreting these results.

Wildlife

Generally, the available environmental data were sufficient to meet the needs of the wildlife assessment. Far greater uncertainty exists with the effects data due to the limited number of dose-response data for wildlife species from published studies. The low number of effect dose-response data available for birds and mammals poses limitations on the assessment, particularly for dioxins/furans. The lack of dose-response data for PBDEs precludes an estimation of potential adverse effects to wildlife from exposure to this COC.

Human Health

A number of known or potentially bioaccumulative COCs were not evaluated in the human health portion of this assessment because they are not part of the human health standards in the NTR. These chemicals include:

1. Acenaphthene
2. Acenaphthylene
3. Inorganic arsenic
4. Benzo(g,h,i)perylene
5. Cadmium
6. Chrysene
7. Lead
8. Nonylphenol
9. Phenanthrene
10. Polybrominated diphenyl ethers (PBDEs)

Generally, environmental data were sufficient to meet the needs of this assessment. All chemicals without NTR criteria were classified as “unknown” due to the lack of standards against which to compare.

Effects Prioritization

The results of the quantitative assessments for water, sediment, tissue media and effects on wildlife and human health are presented in Appendix D-2. The remainder of this section summarizes the results for each COC, including any additional evidence provided by studies of regional significance.

Metals

With the exception of mercury, the assessment of metals was limited to water, sediment and human health. The availability of NTR criteria for metals limited the assessment to mercury and arsenic. Due to data availability and scope limitations, metals were not evaluated in wildlife and tissue residue.

Arsenic

Water

Freshwater

Arsenic was classified as "U" because there were limited effects data for dissolved arsenic. The 90th %ile observed freshwater arsenic concentration is below both the acute and chronic WQC.

Marine Water

Arsenic in nearshore and offshore waters was classified as "U" because no effect data were available for arsenic in marine waters.

Sediment

Freshwater Sediment

Arsenic was classified as Priority 1 based on comparison of the 90th %ile observed sediment concentration to the FP-SQS. The 50th %ile observed sediment arsenic concentrations is below all additional guidelines used in this assessment, suggesting that only the highest concentrations are of greatest concern.

Marine Sediment

Arsenic was classified as Priority 2 based on comparison of the 90th %ile observed near and offshore sediment copper concentrations to the SMS SQS. The 90th %ile observed arsenic concentrations in both near and offshore sediment are below additional guidelines evaluated except for the TEL.

Tissue

Arsenic was not assessed in tissues.

Wildlife

Arsenic was not assessed for wildlife.

Human Health

The NTR is based on inorganic arsenic however; most of the environmental data were for total arsenic which includes less toxic organic forms like arsenobetaine. Both inorganic and total arsenic exceeded the NTR by several orders of magnitude; however, the inorganic dataset was extremely limited with only 15 measurements in all tissue type. Thus, arsenic was classified as "U" in both fresh and marine tissue.

Cadmium

Water

Freshwater

Cadmium was classified as Priority 2 because the 90$^{th\ \%}$ile observed freshwater concentration is below the 10th %ile of the effects data. The 95%ile observed cadmium concentration was above the chronic WQC.

Marine Water

There were insufficient observed data to prioritize cadmium in nearshore and offshore marine waters and it was categorized as "U".

Sediment

Freshwater Sediment

Cadmium was classified as Priority 1 based on comparison of the 90th %ile observed sediment concentration to the FP-SQS. The 50th %ile observed sediment cadmium concentration is below all additional guidelines used in this assessment, suggesting that only the highest concentrations are of concern.

Marine Sediment

Cadmium was classified as Priority 2 based on comparison of the 90th %ile observed near and offshore sediment concentrations to the SMS SQS. The 90th %ile observed concentration of cadmium in both near and offshore sediment are below all of the additional guidelines evaluated.

Tissue

Cadmium was not assessed in tissue.

Wildlife

Cadmium was not assessed for wildlife.

Human Health

Cadmium was not assessed for human health because it is not included in the NTR.

Copper

Water

Freshwater

Copper in freshwater was classified as Priority 1 because the 90th %ile copper concentration exceeded both the 10th %ile effects concentration and the chronic copper WQC. The 95th %ile observed copper concentration also exceeds the acute WQC.

Marine Water

Copper in offshore waters was classified as Priority 2 because the 90th %ile observed offshore concentration is below the 10th %ile concentration of the effects data. Copper in nearshore waters was classified as Priority 1 because the 90th %ile observed nearshore concentration is above the 10th %ile effect concentration. In addition the acute and chronic water quality criteria exceeded the 90th %ile observed concentration. The 95th %ile observed offshore copper concentration was above the chronic WQC.

Sediment

Freshwater Sediment

Copper was classified as Priority 1 based on the comparison of the 90th %ile observed sediment concentration to the FP-SQS. The 50th %ile observed sediment copper concentration is below all additional guidelines used in this assessment, suggesting that only the highest concentrations are of greatest concern.

Marine Sediment

Copper was classified as Priority 2 based on comparison of the 90th %ile observed near and offshore sediment concentrations to the SMS SQS. The 90th %ile observed copper concentrations in both near and offshore sediment are below all of the additional guidelines evaluated.

Tissue

Copper was not assessed in tissue.

Wildlife

Copper was not assessed for wildlife.

Human Health

Copper was not assessed for human health because it is not included in the NTR.

Additional Evidence from Regional Studies

Aquatic Life

Copper is one of the most far-reaching potential priority toxicants in the Puget Sound region. This is due to its ability to alter the sensory capacity and behavior of a wide variety of aquatic organisms. A number of researchers have documented effects on regional species. Tierney et al. (2010) reviewed over 150 papers and found that avoidance behaviors were common in a variety of fresh and salt water fishes at less the 1 ug/L to concentrations ranging up to 20-30 ug/L.

Tested species included coho and Chinook salmon, as well as rainbow trout and golden shiner. Hecht et al. (2007) compiled a similar body of evidence for the disruptive effects of copper on juvenile salmonids. They used US EPA methodologies to calculate benchmark concentrations predicted to represent 10% and 50% reductions in chemosensory response at 0.18 ug/L and 2.1 ug/L respectively. These concentrations bracket a variety of other regional primary literature sources which confirm that the environmentally relevant range of <1.0 ug/L to 5.0 ug/L copper adversely impacts a variety of Puget Sound basin fish, particularly salmonids. Similar neurologic impacts were found by Linbo et al. (2006) on the mechanosensory lateral line of fish.

Sandahl et al. (2004) found copper concentrations of 4.4 ug/L produced sublethal neurotoxicity in coho salmon. In this laboratory study, copper reduced the ability of coho salmon to detect the natural oderants taurcholic acid and L-serine. Further study by Sandahl et al. (2007) confirmed that concentrations as low as 2 ug/L copper are not only affecting the neurologic systems of fish but also alter their behavioral responses to alarm pheromones. Baldwin et al. (2003) also found olfactory inhibition at the comparable, environmentally relevant, concentration of 2.3 ug/L.

Hansen et al. (1999) produced a seminal work which was utilized by several of the subsequent reviews discussed above. In it they documented Chinook salmon avoidance behaviors at concentrations as low as 0.7 ug/L dissolved copper. However, Chinook also failed to avoid concentrations >44 ug/L due to the extensive neural saturation. This window of affect potentially contributes to mortality from prolonged copper exposure or impairment of olfactory dependent behaviors such as homing.

Additional local studies by McIntyre et al. (2008) found that water hardness had little impact on copper's ability to alter olfactory function in coho salmon despite water hardness being a variable influencing the Washington State water quality standards. These regional reviews and studies provide an additional line of evidence suggesting that copper is a very important toxicant at concentrations well within the range found it the Puget Sound regional environment.

Lead

Water

Freshwater

Lead was classified as Priority 2 because the 90th %ile observed freshwater concentration is below the 10th %ile of the effects data.

Marine Water

Lead in offshore marine waters was classified as Priority 2 because the 90th %ile observed concentrations are below the 10th %ile of the effects data. In nearshore marine waters, lead was classified as "U" due to the insufficient amount of observed data.

Sediment

Freshwater Sediment

Lead was classified as Priority 2 because the 90th %ile observed freshwater sediment concentration is below the FP-SQS.

Marine Sediment

Lead was classified as Priority 2 based on the comparison of the 90th %ile observed near and offshore sediment concentrations to the SMS SQS. The 90th %ile observed lead concentrations in both near and offshore sediment are below all of the additional guidelines evaluated.

Tissue

Lead was not assessed in tissue.

Wildlife

Lead was not assessed for wildlife.

Human Health

Lead was not assessed for human health because it is not included in the NTR.

Water

Freshwater

The 90th %ile mercury concentration was below the 10th %ile of the effects concentration, and also exceeded the chronic WQC. Mercury in freshwater was classified as Priority 2.

Marine Water

Mercury in nearshore and offshore waters was classified as "U" because there were a limited number of environmental measurements (n = 13 and 7, respectively).

Sediment

Freshwater Sediment

Mercury was classified as a Priority 1 COC based on the comparison of the 90th %ile observed sediment concentration to the FP-SQS. The 50th %ile observed sediment lead concentrations is below all additional guidelines used in this assessment, suggesting that only the highest concentrations are of greatest concern.

Marine Sediment

Mercury was classified as Priority 1 based on comparison of the 90th %ile observed near and offshore sediment concentrations to the SMS SQS. The 90th %ile observed mercury concentrations in both near and offshore sediments are below all of the additional guidelines evaluated.

Tissue

Freshwater

Mercury is classified as "U" for freshwater non-decapod invertebrates and fish because insufficient observed and effects data are available for the assessment.

Marine Water

Mercury is classified as "U" for all three tissue types in near and offshore marine waters mainly due to a lack of effects concentrations. No marine effects concentrations were available for non-decapod invertebrates and fish and only one marine decapod effect concentration was available. There are observed concentrations for all tissue types although only five for offshore decapods.

Wildlife

Mercury was classified as Priority 1 for all four wildlife receptors because the estimated daily doses are at or above the lowest effect doses. Estimated daily doses of mercury to great blue heron and river otter are above 2 or more effects doses, whereas those of osprey and harbor seal are at or just above the lowest effect dose. Generally, mercury is estimated to bioaccumulate more in receptors living in freshwater habitats than marine habitats around Puget Sound.

Human Health

Freshwater

Mercury was classified as Priority 2 because the 90th percentile tissue concentration for bivalves, fish and other invertebrates did not exceed the NTR.

Marine Water

Mercury was classified as Priority 2 because the 90th percentile tissue concentration for bivalves, fish and other invertebrates did not exceed the NTR.

Additional Evidence from Regional Studies

Wildlife

Regional evaluations of mercury residues in Puget Sound wildlife in the Puget Sound are limited. Johnson et al. (2009) measured mercury concentrations in osprey eggs from the Lower Duwamish River in 2003 and again in 2006/2007. These data demonstrated that mercury concentrations in osprey eggs decreased between these sample periods.

Grove and Henny (2008) measured contaminants in the livers of river otter carcasses collected by trappers in western Oregon and western Washington, including Puget Sound. Mercury liver concentrations were higher in adult river otters from Puget Sound (mean of 7.89 mg/kg dry) than those from the northwest Washington area (mean of 5.85 mg/kg dry weight), located just east of Puget Sound and including the greater Seattle urban area. Mercury levels in Willamette River otter livers and coastal Oregon were slightly higher (mean of 9.2-9.3 mg/kg dw), but similar to Puget Sound levels. This study demonstrates that river otters living in the Puget Sound area are exposed to and bioaccumulate mercury.

Zinc

Water

Freshwater

Zinc was classified as Priority 2 because the 90th %ile observed freshwater concentration is below the 10th %ile of the effects data.

Marine Water

Zinc in offshore waters was classified as Priority 1 because the 90th %ile observed nearshore zinc concentrations is above the 10th %ile concentration of the effects data. Insufficient observed data for zinc in nearshore waters was available; zinc in nearshore waters was classified as "U".

Sediment

Freshwater Sediment

Zinc was classified as Priority 1 based on the comparison of the 90th %ile observed sediment concentration to the FP-SQS. The 50th %ile observed sediment zinc concentration was below all additional guidelines used in this assessment, suggesting that only the highest concentrations are of greatest concern.

Marine Sediment

Zinc was classified as Priority 2 based on comparison of the 90th %ile observed near and offshore sediment concentrations to the SMS SQS. The 90th %ile observed sediment zinc concentration is above the TEL; the 90th %ile observed nearshore concentration was below the TEL.

Tissue

Zinc was not assessed in tissue.

Wildlife

Zinc was not assessed for wildlife.

Human Health

Zinc was not assessed for human health because it is not included the NTR.

PCBs

The potential for effects associated with PCB exposure was assessed for all media and pathways (water, sediment, tissue, human health and wildlife). Observed environmental PCB concentrations were assessed as the sum of Aroclors® and the sum of congeners. In general, the majority of the available toxicity data were based on individual Aroclor® exposures. It was not practical to compare observed individual Aroclor® and congener data to available effects data for individual compounds. Caution is advised in the use of Aroclor® data; these data may not be optimal due to shifts in the congener composition associated with weathering. It is also important to note that the PCB WQC is not protective of aquatic life through the bioaccumulation pathway.

Water

Freshwater

The 90th %ile observed concentrations of both PCB congeners and Aroclors® were below the 10th %ile concentration of the available effects data. However, the 90th %ile PCB Aroclor® concentration exceeded the chronic WQC. PCB Aroclors® were classified as Priority 1; PCB congeners were classified as Priority 2.

Marine Water

Extremely limited PCB concentration data are available for marine nearshore waters, with only 11 PCB Aroclor® concentrations and no PCB Congener data available; PCBs in nearshore waters were classified as "U". Similarly, PCB Aroclor® data in offshore marine waters were not available. However, sufficient PCB Congener data were available in marine offshore waters, where the 90th %ile concentration of total PCB congeners was below the 10th %ile of the available effects concentrations; PCB congeners in offshore marine waters were classified as Priority 2.

Sediment

Freshwater Sediment

PCB Aroclors® in freshwater sediments were classified as Priority 1. The 90th %ile observed concentrations of both PCB congeners and Aroclors® were above the FP-SQS, in addition to 3 of the 5 additional guidelines evaluated. Only the PEC fell above the 90th %ile concentration of both PCB congeners and Aroclors®. With the exception of the TEL, the 50th %ile PCB Aroclor® concentration was below all remaining guidelines evaluated. In general, this suggests that areas with the highest concentrations are of concern. Sediment congener data were limited (n=26); as such they were classified as "U".

Marine Sediment

PCB Aroclors® in marine offshore sediment were classified as Priority 1 because the 90th %ile OC normalized PCB Aroclor® concentrations in marine offshore sediments exceeded the marine SQS. The 90th %ile concentrations of OC normalized PCB Congeners in offshore sediments and PCB Aroclors® in nearshore sediments did not exceed the SQS, resulting in a Priority 2 classification. Insufficient data were available to evaluate PCB congeners in nearshore sediments.

Tissue

Freshwater

PCB Aroclors® and PCB congeners in freshwater non-decapod invertebrates are classified as Priority 1 because the 90th %ile observed concentrations are higher than the 10th %ile of the effects data. All other freshwater tissues are classified as "U" for both PCB Aroclors® and

congeners due to the limited number of effects values and/or environmental concentrations. There were no observed data available for PCB congeners in decapods.

Marine
PCB Aroclors® and congeners in nearshore decapods are classified as Priority 2 because the 90th %ile observed concentration is below the 10th %ile effects concentration. PCB Aroclors® and congeners in nearshore non-decapod invertebrates are also classified as Priority 2. PCBs in fish are classified as "U" due to the limited amount of available effects data.

PCB Aroclors® and congeners in offshore decapods and fish were classified as "U" due to a lack of effects values or insufficient quantity of observed concentrations. PCB Aroclors® in offshore non-decapod invertebrates were classified as Priority 2 but PCB congeners are classified as "U" because of insufficient numbers of observed and effect concentrations in non-decapod invertebrates (< 20 and < 5 respectively).

Wildlife

PCBs were classified as Priority 1 for all four wildlife receptors because the estimated daily doses are more than 10 times lower than the lowest effect doses. Generally, PCBs are estimated to bioaccumulate to a greater degree in receptors living in marine habitats than freshwater habitats around Puget Sound. Estimated daily PCB doses to osprey, river otter and harbor seal are above several effects doses. Those of the great blue heron hover near the three lowest effect doses.

Human Health

PCBs were classified as a Priority 1 human health concern; multiple freshwater and near- and offshore tissues types exceeded the NTR PCB concentration standard. The range of observed PCB concentrations analyzed by Aroclor® and congener methods vary from one another. This variability is likely the result of multiple projects using different analytical methods for different suspected levels of contamination.

Additional Evidence from Regional Studies

Aquatic Life

PCBs have been detected in outmigrant juvenile salmon (Johnson et al., 2007) from multiple northwest estuaries and hatcheries, including three in the Puget Sound. Whole-body juvenile Chinook salmon from the Duwamish River contained the highest PCB concentration (103 ng/g wet weight or 3100 ng/g lipid) of any of the locations tested. Johnson et al. (2007) note that this concentration is higher than NOAA's estimated threshold for adverse health effects of 2400 ng/g lipid. Separately, juvenile salmonid PCB exposures were documented as occurring via food source by an analysis of stomach content of outmigrants at three locations in Puget Sound (Stein et al. 1995). Meador et al. (2010) found that PCB tissue concentrations in outmigrant juvenile Chinook from the Duwamish estuary varied by time and location within the estuary, suggesting that localized heterogeneity of sediment concentrations may substantially impact accumulation in fishes.

PCB concentrations in adult Puget Sound Chinook salmon tissues were found to be 3 – 5 times higher than those measured in six other populations of Chinook salmon on the West Coast of North America (O'Neill and West 2009). O'Neill and West note that these elevated tissue concentrations have resulted in consumption advisories, and have implications for the viability of these fish and southern resident killer whales. Cullon et al. (2009) found PCBs in adult Chinook returning to the Duwamish River, as well as in Puget Sound Chinook smolts.

PCBs concentrations in Puget Sound herring and Puget Sound flatfish have also been evaluated. Puget Sound herring were found to contain 3 to 9 times higher concentrations of PCBs than herring from the Strait of Georgia, with Puget Sound whole-body concentrations ranging from about 120 to 160 ng/g wet weight (West et al. 2008). Analyses of various biomarkers of pollution exposures in benthic flatfish were shown to successfully differentiate between sites with differing degrees of sediment contamination (Stein et al. 1992). Cullon et al. (2005) also found about seven times higher levels of PCBs in a mixture of fishes designed to represent the diet of Puget Sound harbor seals than in a similar mixture of fish designed to represent the diet of harbor seals from the Strait of Georgia. Sol et al. (2008) found a statistically significant correlation between PCB concentrations in English sole livers and two biological effects parameters.

Wildlife

A number of studies have been conducted in the Puget Sound region investigating exposure and/or effects of PCBs and other persistent and bioaccumulative contaminants on wildlife, particularly marine mammals. The salient information on PCBs in wildlife is summarized here.

Johnson et al. (2009) measured PCB concentrations in osprey eggs from the Lower Duwamish River and compared them to those sampled from the upper Willamette River. Total PCB residues were significantly higher in Lower Duwamish River osprey eggs (geometric mean = 897 ug/kg wet weight) compared to those from the Willamette River (geometric mean = 182 ug/kg ww). These results demonstrate that adult osprey bioaccumulation and maternal transfer of PCBs is occurring in osprey nesting in PCB contaminated areas of Puget Sound. This study also compared egg residues over time and determined that PCB concentrations in osprey eggs from the Lower Duwamish River had decreased 53% between 2003 and 2007.

Grove and Henny (2008) also demonstrated the bioaccumulation of PCBs in river otter livers from Puget Sound. The Puget Sound area river otters accumulated more PCBs (as total PCBs) than otters from other areas in western Washington.

PCBs and other organochlorines have been shown to cause immunosuppression, thyroid disruption and possibly cancer in harbor seals (Tabuchi et al. 2006, Ylitalo et al. 2005; Simms et al. 2000; Ross et al. 1996; Ross et al. 1995; de Swart et al. 1996; de Swart et al. 1995; Van Loveren et al. 1994). Vitamin A disruption has also been observed in harbor seal pups found on the Washington State coast whose mothers contained high PCB residues in their blubber (Simms et al. 2000). This effect on seal pups is suspected to result from exposure to contaminated milk.

There is substantial evidence that Puget Sound harbor seals and killer whales are bioaccumulating PCBs at very high concentrations in their blubber. The prey items of Puget Sound harbor seals were measured to have 7 times higher concentrations of PCBs than prey from Strait of Georgia on a lipid basis (Cullon et al. 2005), which corresponds to PCB concentrations measured in harbor seal blubber.

PCB tissue concentrations are often reported as dioxin toxicity equivalents (i.e. TEQs) which represent the toxicity of dioxin-like PCBs, dioxins and furans relative to the most toxic dioxin - 2,3,7,8-TCDD. Puget Sound harbor seals have significantly higher TEQs in blubber (158 ng/kg lipid weight) compared to seals from the Strait of Georgia (33 ng/kg lipid weight) (Ross et al. 2004); the TEQ contribution was greater from PCBs than dioxins and furans. Levin et al. (2005) also found that the majority of TEQs in harbor seal pups (from southern B.C.) were from PCBs, not dioxins and furans.

Ross et al. (2000) reported measured mean total PCB concentrations in transient and Southern resident male killer whales of 251 and 146 mg/kg -lipid, respectively. The authors concluded these marine mammals are among most contaminated in the world. Further research on the northern, southern and transient killer whale communities have discovered that males bioaccumulate more PCBs than females; female offload a portion of their tissue burden to their young through maternal transfer (Krahn et al. 2007; Krahn et al. 2009). PCB concentrations in the blubber of mothers decrease temporarily during nursing and can reach concentrations below those in their calves. Mothers initiate bioaccumulation again after calves are weaned. Total PCB concentrations in southern resident killer whales range from about 5,000 to 180,000 µg /kg lipid. For all but three recent mothers, the measured concentrations exceed a marine mammal threshold for blubber concentrations (17,000 µg /kg lipid) (Krahn et al. 2007).

Although environmental concentrations of PCBs are gradually declining, one modeled estimate of southern resident killer whale recovery projects that blubber concentrations will not decrease to the marine mammal threshold until 2063 (Hickie et al. 2007). The reviewed studies suggest that marine mammals in Puget Sound are accumulating PCBs in their blubber to very high concentrations. The results of the quantitative assessment are congruent and classify PCBs as Priority 1.

PBDEs

The PBDE assessment was limited due to the lack of effects data, guidelines or criteria. While PBDEs have been measured in a variety of media, appropriate effects data were insufficient to fully asses this COC.

Water

Surface water data for PBDEs in freshwater (n=255) and marine offshore waters (n=126) were available; however, appropriate effects data were not available in the ECOTOX database, nor is there a WQC for PBDEs. Due to the lack of effects data, PBDEs in both fresh and marine waters were classified as “U”.

Sediment

PBDEs in freshwater and marine sediments were classified as "U". A limited number of observed concentration data for PBDEs in marine (n=46) and freshwater (n=77) sediments were available. However sediment guidelines are not available for PBDEs in either fresh or marine sediments.

Tissue

PBDEs were not assessed in tissue due to a lack of effects thresholds.

Wildlife

PBDEs were not assessed quantitatively in wildlife due to a lack of effects doses. See the Additional Evidence from Regional Studies below for a discussion of available effects information.

Human Health

PBDEs are prioritized as unknown, "U" because NTR criteria are not available.

Additional Evidence from Regional Studies

Aquatic Life

Sloan et al. (2010) detected PBDEs in outmigrant Chinook salmon tissue and stomach content from four sites in Puget Sound. Concentrations in wild outmigrant juveniles were higher than in hatchery fish. PBDE concentrations in Puget Sound juvenile fishes ranged from 67 to 13,000 µg /kg lipid, which was generally comparable to those measured in the Lower Columbia River and Estuary. Sloan et al. (2010) conclude that PBDEs may be contributing to reduced health and fitness in outmigrant juvenile Chinook salmon.

PBDEs have also been detected in adult Chinook salmon returning to the Duwamish River; however, they were not detected in adult Chinook returning to the Johnstone Strait, Lower Fraser River, or the Deschutes River (Cullon et al. 2009). Lema et al. (2008) demonstrated that dietary exposures of certain PBDEs by adult fathead minnows alter thyroid status and thyroid hormone-regulated gene transcription. Arkoosh et al. (2010) found that juvenile Chinook salmon exposed to moderate doses of PBDEs through their diet may be at increased risk of disease relative to those exposed to higher or lower doses of PBDEs in their diet potentially indicative of a complex U-shaped dose response curve for PBDEs in Chinook salmon. PBDE levels in a mixture of fishes designed to represent the diet of Puget Sound harbor seals were found to be about four to five times higher than in a similar mixture of fish designed to represent the diet of harbor seals from the Strait of Georgia (Cullon et al. 2005).

Wildlife

Because little information on PBDE toxicity to wildlife is available and a quantitative assessment could not be conducted, the publications available are reviewed here regardless of their geographic location. Few studies have been conducted examining effects of PBDEs on birds.

The studies reviewed indicate that PBDEs impact the reproduction and endocrine system similarly to PCBs. Fernie et al. 2005 showed that American kestrel egg injection and oral gavage exposure to PBDE congeners caused hepatic oxidative stress and altered thyroid hormone and vitamin A concentrations and glutathione metabolism. Exposure to PBDE congener 71 for 75 days adversely impacted courtship and mating behavior of American kestrels (Fernie et al. 2008). These birds also displayed significant delays in clutch initiation and produced smaller eggs (Fernie et al. 2009). Eggshell thinning and reduced hatching success also resulted.

A study of species sensitivity to PBDEs (PBDE-71) observed that pentabrominated diphenyl ether (Penta BDE) exposure to eggs at 0.01 to 20 mg/kg caused decreased pipping and hatching success in American kestrels but not chickens or Mallard ducks (McKernan et al. 2009). Species sensitivity was concluded to be Mallard ducks <chickens <American kestrels.

Total PBDE concentrations in osprey eggs and nestling plasma were significantly lower in birds from the Lower Duwamish River (eggs: 321 ug/kg ww; plasma: 6 ppb ww) compared to (Johnson et al. 2009) those from the upper Willamette River (eggs: 897 ug/kg ww; plasma: 22 ug/kg ww). The total PBDE concentrations in the osprey eggs did not change significantly between 2003 and 2007. Reproductive failure was observed in four of nine nests in the Lower Duwamish area. A small dataset from this study suggests that some nestlings may have experienced immunosuppression. However, the results were inconclusive due to the small sample size.

One study (Fernie et al. 2011) was acquired for which a dietary effect dose could be determined. Fernie et al. (2011) exposed American kestrels to PBDEs (Hexa-BDE) via dietary exposure and measured reproductive parameters. Adult kestrels exposed to 0.51 mg/kg/d PBDE, an environmentally relevant dose, through their diet displayed less courtship behaviors, earlier egg laying, a greater clutch size and smaller eggs; however, there were no significant differences in the fledging or hatching rates compared to control.

To compare exposures of birds feeding in Puget Sound, a PBDE daily dose was estimated using the same methods as the other COCs for wildlife. At the higher sediment ingestion rate (4%), the estimated daily dose of total PBDEs for the osprey is 0.006 mg/kg/d. The estimated daily dose for the great blue heron is also 0.006 mg/kg/d. If the threshold from Fernie et al. 2011 is considered an effect dose, piscivorous birds in the Puget Sound watershed are estimated to experience lower exposure by approximately a factor of 100.

Compared to birds, a larger but still limited number of publications exist on the effects of PBDEs in mammals. Rodent exposure studies have demonstrated thyroid hormone disruption (Hallgren et al. 2001, Zhou et al. 2002), developmental neurotoxic and behavioral effects (Ericksson et al. 2001, Viberg et al. 2003a, Viberg et al. 2003b). A study of grey seal pups and

juveniles observed a relationship between circulating thyroid hormones, transport proteins and PBDE uptake (Hall et al. 2003).

Similar to PCBs, there is evidence of PBDE bioaccumulation in the blubber of marine mammals at high concentrations. However, absolute total PBDEs concentrations appear to be lower than total PCBs. Cullon et al. (2005) measured PBDE concentrations 5 times higher in harbor seal prey from Puget Sound than the Strait of Georgia; however, the mean PBDE concentration was 5 times lower than that measured for PCBs. Krahn et al. (2009) and Rayne et al. (2004) found the same pattern of killer whale blubber concentrations in males, mothers and calves as they found for PCBs with males having the highest concentrations and females experiencing fluctuations due to maternal transfer. Krahn et al. (2005) measured total PBDE concentrations in killer whale blubber ranging from 680 to 15,000 ug/kg lipid. Mean PBDE concentrations in northern male killer whale blubber have been found to be significantly lower (203 ug/kg lw) than those of southern resident (942 ug/kg lw) and transient males (1015 ug/kg lw).

Although a full quantitative effects assessment was not conducted for PBDE exposure to wildlife, published research demonstrates that PBDEs are bioaccumulating to high concentrations in Puget Sound marine mammals. This coupled with the growing evidence that PBDE exposure can cause thyroid and developmental effects in mammals strongly suggest that PBDEs should be classified Priority 1.

Dioxins and Furans

Both observed environmental concentrations and effects data for dioxins and furans (PCDD/Fs) were limited. As a result the assessment was limited to evaluation of potential effects to wildlife and human health.

Water

Observed surface water data for PCDD/Fs were limited to 7 measurements in freshwater for 3 compounds (1,2,3,4,7,8 HxCDD, 1,2,3,7,8 PeCDD and 2,3,7,8 TCDD). No observed data were available for marine waters; effects data for these compounds were very limited. Due to the lack of data to assess these compounds, dioxins and furans in surface waters were classified as "U".

Sediment

PCDD/Fs in freshwater and marine sediments were classified as "U". A moderate number (n >700) of observed concentration data for PCDD/Fs in marine and freshwater sediments were available. However, FP SQS and SQS values are not available for PCDD/Fs. The 90^{th} %ile observed freshwater sediment concentration exceeded both the PEL and TEL; while the 50^{th} %ile concentration was below both the PEL and TEL. The 90^{th} %ile observed nearshore marine sediment concentration was just above the PEL; while the 50^{th} %ile concentration was above the TEL. The 90^{th} %ile observed offshore was below the PEL, but above the TEL.

Tissue

PCDD/Fs were not evaluated in freshwater or marine tissues due to the lack of effects data or the lack of observed concentrations. No observed PCDD/Fs concentrations are available in freshwater or marine tissues.

Wildlife

PCDD/Fs were classified as Priority 1 for the great blue heron and river otter because the estimated daily dose was greater than 0.1 times the lowest effects dose. For harbor seal, PCDD/Fs were classified as Priority 2 because adequate effects data were available and the estimated daily doses are more than 10 times lower than the lowest effects dose. The osprey daily doses are estimated to be almost 1,000 times lower than the lowest effects dose; however, substantial uncertainty exists around the effects of PCDD/Fs on birds so the resulting classification was "U". The estimated daily doses for great blue heron are close to one of the two existing effects doses for birds, so the assumption was made that reproductive effects were likely occurring.

Human Health

2,3,7,8-Tetrachlorodibenzo-p-dioxin (2,3,7,8 TCDD) was classified as Priority 1; NTR criteria were not available for other PCDD/Fs. The 90^{th} %ile of the observed 2,3,7,8 TCDD concentrations exceeded the NTR criteria for all three tissue groups (bivalves, fish and other invertebrates). Tissue from both near and offshore areas exceeded the NTR criteria for one or more tissue types, although sample sizes were generally smaller than for freshwater tissues.

Additional Evidence from Regional Studies

Aquatic Life

Generally similar concentrations of PCDD/Fs were found in adult Chinook salmon returning to the Duwamish River, Johnstone Strait, Lower Fraser River, and the Deschutes River (Cullon et al. 2009).

Wildlife

PCDD/Fs TEQ residues in river otter livers from Puget Sound contributed one third to the total TEQs (Grove and Henny, 2008) indicating that PCBs accumulate in river otter livers to a concentration that is twice as toxic as dioxins and furans.

Studies in Puget Sound of harbor seal and southern resident killer whale prey items have shown that prey of these marine mammals are higher in dioxins and furans compared to the same prey from the Strait of Georgia and British Columbia coast (Cullon et al. 2005, Cullon et al. 2009). PCBs in Harbor seal prey were 3-4 times higher on a lipid basis than prey from the Strait of Georgia (Cullon et al. 2005). However, Ross et al. (2000) found that dioxin and furan concentrations in killer whale blubber were much lower than PCBs and there were no differences

between whales from the northern and southern resident and transient communities. This was suspected to be due to metabolic removal of dioxins and furans.

DDTs and metabolites

Observed environmental data for DDTs were available for water, sediment and tissue; data in marine waters were very limited. Effects data were available for water, marine sediment, tissue residue, wildlife and human health. Freshwater sediment guidelines were not available.

Water

Freshwater

The 90th %ile observed concentration of DDTs and metabolites was below the 10th %ile of the available effects data; however, the 90th %ile concentration was above both the acute and chronic DDT WQC. DDTs in freshwater were classified as Priority 1.

Marine

DDTs were measured, but not detected, in a limited number of samples (n=11) in marine nearshore waters; there were no DDT measurements in offshore waters. DDTs in marine waters were classified as "U".

Sediment

Freshwater Sediment

A FP-SQS is not available for DDT in freshwater sediment; as such this COC was classified as "U". The 90th %ile observed DDT freshwater sediment concentration was well below both the PEL and PEC, while the 50th %ile concentration was above both the TEC and TEL.

Marine Sediment

A SQS is not available for DDT; as such, this COC was classified as "U". The 90th %ile observed concentration in nearshore marine sediment was above the PEL, 2LAET, LAET and TEL; the 50th %ile concentration was below all four of these guidelines. In offshore sediments, both the 90th %ile and 50th %ile were below the PEL, 2LAET and LAET; and above the TEL.

Tissue

Freshwater

Total DDTs are classified as Priority 2 in freshwater non-decapod and fish tissue because the 90th %ile concentration is below the 10th %ile effects data. DDTs could not be assessed in freshwater decapods due to a lack of observed concentrations.

Marine

Total DDTs are classified as "U" in all marine nearshore and offshore tissues due to insufficient (< 5 values) effects data.

Wildlife

DDT and metabolites were assessed as the sum of DDT, DDE, and DDD isomers and were classified as Priority 1 for great blue heron and osprey because the estimated daily doses are greater than 0.1 times the lowest effect dose. The mammals, river otter and harbor seal, were both classified as Priority 2 because the estimated daily doses are more than 10 times lower than the lowest effect dose.

Human Health

4,4' DDT was classified as Priority 2 in all freshwater and offshore tissues types. In the nearshore area, the 90th %ile concentration of 4,4' DDT for both fish and other invertebrates exceeded the NTR threshold and were classified as Priority 1. For the DDT metabolite 4,4' DDE, only the 90th %ile of other invertebrate tissues in freshwater exceeded the NTR threshold and was classified as Priority 1. All tissues in near and offshore areas were classified as Priority 2. For the DDT metabolite 4,4' DDD, other invertebrates tissues in freshwater were classified as Priority 1, while all other areas and tissues were classified as Priority 2.

Additional Evidence from Regional Studies

Aquatic Life

Total DDT (sum of DDT, DDE, and DDD) concentrations in stomach contents of outmigrant juvenile Chinook salmon from the Duwamish Estuary and Commencement Bay were found to be elevated relative to the stomach content concentrations of fish from the Nisqually Estuary (Stein et al. 1995). Whole-body total DDT (sum of DDT, DDE, and DDD) concentrations in juvenile Chinook salmon were found to be relatively high (over 1000 ng/g lipid or 25 ng/g wet weight) in fish from the Nisqually, Duwamish and Columbia River Estuaries (Johnson et al. 2007).

Johnson et al. (2007) also found detectable levels of DDTs in stomach contents, with stomach content concentrations substantially higher in Columbia River and Grays Harbor juvenile Chinook than in Duwamish and Nisqually Estuary juvenile Chinook. Johnson et al. (2007) suggest that at the observed levels, DDTs are unlikely to cause adverse effect by themselves; however, they may contribute via additive or synergistic effects with other contaminants. Substantially higher levels of DDTs were found in adult Chinook salmon returning to the Duwamish River than in adult Chinook returning to the Johnstone Strait, Lower Fraser River, or Deschutes River (Cullon et al. 2009).

Analysis of DDT concentrations in Pacific herring indicated that concentrations from Puget Sound herring were 1.5 to 2.5 times higher than those from Strait of Georgia (West et al. 2008), with Puget Sound concentrations ranging from 19 to 27 ng/g wet weight (240 to 330 ng/g lipid).

Cullon et al. (2005) found similar levels of DDTs in a mixture of fishes designed to represent the diets of Puget Sound and Strait of Georgia harbor seals.

Wildlife

Grove and Henny (2008) evaluated DDTs in river otters. They did not detect DDT and detected only low concentrations of DDE (mean of 0.004-0.28 mg/kg ww) in river otter livers from Puget Sound which were much lower than those found in animals residing near the Columbia River (mean of 0.12-1.65 mg/kg ww).

Lipid based concentrations of total DDT concentrations in Harbor seal prey in Puget Sound are 1.6 times higher than those from the Strait of Georgia (Cullon et al. 2005). Puget Sound Chinook, the primary prey of southern resident killer whales, have higher body residues of DDTs and lower lipids compared to Chinook from British Columbia coast (Cullon et al. 2009). Krahn et al. (2009) found the same pattern of killer whale blubber concentrations as found for PCBs in males, mothers and calves. That is, males have the highest DDT concentrations in their blubber and female blubber concentrations vary with their maternity status due to maternal transfer. Total DDT concentrations in killer whales ranged from 1,000 to 160,000 ug/kg lipid.

PAHs

PAHs were evaluated in water, sediment and for human health.

Water

The majority of the toxicity data available for PAHs in surface waters is based on individual PAHs. As such, Total PAHs, HPAH and LPAH were not directly evaluated here.

Freshwater

While there were sufficient observed concentration data (N>1500 measurements) for the individual PAHs evaluated here, there were limited effects data for a number of PAH compounds. The 90th %ile observed concentrations of acenaphthene, anthracene, benzo(a)pyrene, fluoranthene, fluorene, naphthalene, phenanthrene, and pyrene were below the 10th %ile of the available effects concentrations; these COC s were classified as Priority 2. Effects data were insufficient to evaluate benzo(a)anthracene, benzo(k)fluoranthene, benzo(g,h,i)perylene, chrysene, and dibenzo(a,h)anthracene; these COCs were classified as "U".

Marine Water

Observed concentrations of individual PAHs in nearshore waters were very limited (N=12). Due to the lack of sufficient measured concentrations, in addition to the limited availability of effects data, individual PAHs in nearshore waters were classified as "U".

While there were sufficient observed nearshore marine concentration data for some individual PAHs, marine effects data were limited for a number of COCs. The 90th %ile observed concentrations of acenaphthene, fluoranthene, naphthalene and phenanthrene in nearshore waters

were below the 10th %ile of the available effects data; these COCs were classified as "U". The remainder of the individual PAHs in nearshore waters (anthracene, benzo(a)anthracene, benzo(a)pyrene, benzo(k)fluoranthene, chrysene, dibenzo(a,h,)anthracene, fluorene and pyrene) were classified as "U" due to insufficient data to fully assess these COCs. There were insufficient data to evaluate individual PAHS in offshore waters; these COCs were classified as "U".

Sediment

Freshwater Sediment

Both LPAH and HPAHs in freshwater sediments were classified as Priority 1. The 90th %ile observed LPAH concentration was also above the FP-CSL; however, the 75th %ile concentration was below both the FP SQS and FP CSL. The 90th %ile HPAH concentration was below the FP-CSL and the 75th %ile concentration was below .both the FP-CSL and FP-SQS. These data suggest that only some of the highest detected concentrations are likely to be of concern. With the exception of benzo(a)anthracene which was classified as Priority 2, all of the individual PAHs evaluate were classified as Priority 1.

Marine Sediment

Both LPAH and HPAH in near and offshore sediments were classified as Priority 2; the 90th %ile observed concentrations were below the SQS. All of the individual PAHS in near and offshore sediments were also classified as Priority 2.

Tissue

Tissue residue concentrations of PAHs were not evaluated because these chemicals typically to not accumulate in the tissue of vertebrates and are rapidly metabolized in fish. It was beyond the scope of this effort to evaluate PAHs in those invertebrates that are not capable of metabolizing PAHs.

Wildlife

PAHs were not evaluated because these chemicals typically do not bioaccumulate in wildlife.

Human Health

Five if the nine individual PAHS (benzo(a)pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene, dibenzo(a,h)anthracene, and indeno(1,2,3-cd)pyrene) evaluated in freshwater tissue exceeded the NTR criteria and were classified as Priority 1. Concentrations of the remaining (anthracene, fluorene, fluoranthene, and pyrene) PAHs evaluated were below the NTR criteria and classified as Priority 2.

The same pattern of Priority classification was observed in nearshore tissues as was found for the freshwater tissues. Tissue data from nearshore waters was limited; as a result, five of the individual PAHS were classified as "U" (benzo(a)pyrene, benzo(b)fluoranthene,

benzo(k)fluoranthene, dibenzo(a,h)anthracene, and indeno(1,2,3-cd)pyrene). The remainder of offshore marine tissue was classified as Priority 2 for human consumption.

Additional Evidence from Regional Studies

Aquatic Life

Multiple investigations have identified biomarkers of PAH exposure in various Puget Sound fishes.

Bile and stomach content of outmigrant juvenile Chinook salmon were found to contain various PAHs (Johnson et al. 2007), demonstrating that diet pathways are important PAH exposure pathways. The authors suggest that exposure may result in immunosuppression and other health effects. These results expanded and confirmed previously documented PAH exposures (Stein et al. 1992; Stein et al. 1995). A dietary feeding study on juvenile Chinook documented growth and physiological responses from dietary exposures to PAHs at concentrations that were environmentally realistic in the Puget Sound (Meador et al. 2006).

Biomarkers of PAH exposure were confirmed in Puget Sound English sole, rock sole, and starry flounder collected from up to five sites in Puget Sound (Stein et al. 1992). Stein et al. found that biomarkers of exposure were related to the degree of sediment contamination. Further field study (Johnson 2000) resulted in recommended a sediment threshold of 1000 ppb total PAHs to protect English sole against liver lesions, DNA adducts in liver, and other effects. The causal relationship between elevated sediment PAH concentrations and English sole liver effects was confirmed by Meyers et al. (2003). In a study of English sole from the Hylebos Waterway and Colvos Passage, Sol et al. (2008) found no correlation between PAH exposure and age and little correlation between reproductive end points and PAH exposure. Pacific herring embryos were found to be affected by tricyclic PAHs in weathered crude oil (Incardona et al. 2009; Carls et al. 1999).

Several laboratory studies have documented that developmental defects in fish are associated with exposure to PAHs released by the weathered crude oil, notably the tricyclic-PAHs (Incardona et al. 2005; Incardona et al. 2006; Carls et al. 2008). Carls and Meador (2009) developed a description of the oil weathering, PAH toxicity, and embryo exposures to explain the observed toxicity of PAHs in weathered oil at relatively low levels. Driscoll et al. (2010) developed a framework for describing PAH exposure as a dose to fishes in order to understand the mechanisms of exposure and toxicity.

Bis(2-Ethylhexyl) Phthalate

The assessment of bis(2-ethylhexyl) phthalate was limited to water and sediment.

Water

The 90th %ile observed concentration of bis(2-ethylhexyl) phthalate in freshwater was below the 10th %ile of the available effects data; this COC was classified as Priority 2. Insufficient observed and effects data for bis(2-ethylhexyl) phthalate in marine nearshore waters were

available; as such it was classified as "U". The 90th %ile observed concentration of bis(2-ethylhexyl) phthalate in offshore marine water was below the 10th %ile of the available effects data; as such it was classified as Priority 2.

Sediment

The 90th %ile observed concentration of bis(2-ethylhexyl) phthalate in freshwater sediments was above the FP-SQS and the FP-CSL; this COC was classified as Priority 1. The OC-normalized 90th %ile concentrations in both marine nearshore and offshore sediments exceeded the marine SQS, resulting in Priority 1 classification.

Tissue

Bis(2-ethylhexyl)phthalate tissue concentrations were not evaluated because phthalates typically do not accumulate to a significant degree in tissues.

Wildlife

Bis(2-ethylhexyl) phthalate was not evaluated.

Human Health

The 90th %ile of the observed freshwater bivalve tissue concentration of bis(2-ethylhexyl) phthalate exceeded the NTR criteria. While there were a number of measurements (>100) for the other tissue types in marine and freshwaters, there were too few detections to calculate a 90th percentile. Thus for freshwater, bis(2-ethylhexyl)phthalate was classified as Priority 1, while for marine tissues bis(2-ethylhexyl)phthalate was classified as priority "U".

Triclopyr

The assessment of triclopyr was limited to water.

Water

The 90th %ile observed triclopyr concentration in freshwater was below the 10th %ile concentration of the available effects concentrations and was classified as Priority 2. No observed triclopyr data were available in marine nearshore or offshore waters, resulting in a classification of "U".

Sediment

No observed concentrations of triclopyr are available in freshwater or marine sediments. This COC was classified as "U" in sediments.

Tissue

Tissue concentrations were not evaluated because these types of pesticides typically do not accumulate in tissues.

Wildlife

Triclopyr was not evaluated because it is not bioaccumulative in wildlife.

Human Health

Triclopyr is not listed in the NTR and was not evaluated.

Nonylphenol

The assessment of nonylphenol was limited to water and sediment. Effects data are limited for this COC.

Water

Nonylphenol was classified as Priority 2 in freshwater because the 90th %ile concentration is below the acute and chronic WQC and below the 10th %ile of the available effects data. However, the 95th %ile observed concentration was above the chronic WQC for nonylphenol. Nonylphenol was classified as "U" in marine nearshore water because insufficient effects and observed data were available. Nonylphenol was classified as Priority 2 in offshore marine water because the 90th %ile concentration is below the acute and chronic WQC and below the 10th %ile of the available effects data.

Sediment

Nonylphenol was classified as "U" in sediments because no FP-SQS is available for freshwater sediments and no SQS or CSL are available for marine sediments.

Tissue

Nonylphenol tissue concentrations were not evaluated because this COC does not typically bioaccumulate to a significant degree in tissues.

Wildlife

Nonylphenol was not evaluated because it is not bioaccumulative in wildlife.

Human Health

Nonylphenol is not listed on the NTR and was not evaluated.

Petroleum

Water

All four of the individual petroleum products were classified as "U" because the number of effect concentrations for each product was insufficient (< 15) and the number of observed concentrations for heavy fuel/bunker oil was insufficient (< 50).

Sediment

Petroleum was not assessed in sediment because there are no sediment guidelines nor observed sediment data.

Tissue

Petroleum was not assessed for tissue residues because there are no effects nor observed tissue data.

Wildlife

Petroleum could not be evaluated for wildlife because there are no effects data.

Human Health

Petroleum was not evaluated because it is not listed on the NTR.

Combined Prioritization for all Elements of the Assessment

The specific COCs evaluated in the quantitative assessments varied; a summary of the media and pathways evaluated for each COC is presented in Table 9. The chemicals assessed in this report were placed into three groups based on the likelihood that they may currently be causing widespread environmental effects (Table 10). This grouping was based on a review of the individual priority classification for each line of evidence evaluated.

COCs with multiple Priority 1 and different lines of evidence

Those chemicals with two or more Priority 1 classifications for the different lines of evidence were categorized as "Multiple Priority 1". These chemicals represent the COCs with the most compelling evidence that they may be causing widespread environmental effects in the Puget Sound region. Chemicals in "Multiple Priority 1" are likely to warrant action to reduce the potential for widespread environmental affects.

COCs with a single Priority 1 or line of evidence

Those chemicals with one priority 1 classification were placed in "Single Priority 1". These COCs represent those with strong evidence that they may be causing widespread environmental

effects in the Puget Sound region, but where the evidence is limited to one line evidence from the many evaluated. Chemicals in "Single Priority 1" are likely to warrant action to reduce the potential for widespread environmental affects.

COCs with no Priority 1 and no additional lines of evidence

COCs that were not classified in any media for pathway were categorized as "No Priority 1". These COCs represent those with ongoing concern about their effects, but for which limited evidence is available to indicate they may be causing widespread environmental impacts in the Puget Sound region. Some "No Priority 1" COCs were not evaluated for some lines of evidence due to limited availability of observed data and/or the lack of effects data. Chemicals in "No Priority 1" may warrant action based on existing concerns that the current assessment was unable to capture and the extent and nature of the potential effects from these chemicals.

Table 9. Summary of COCs Assessed.

COC	Water	Sediment	Tissue	Wildlife	Human Health
Arsenic	X	X	NA	NA	X[1]
Cadmium	X	X	NA	NA	NA
Copper	X	X	NA	NA	NA
Lead	X	X	NA	NA	NA
Mercury	X	X	X	X	X
Zinc	X	X	NA	NA	NA
PCBs[2]	X	X	X	X	X
PBDEs	NA	NA	NA	X[3]	NA
PCDD/Fs[4]	X	NA	NA	X	X
DDT and Metabolites[5]	X	X	X	X	X
LPAHs[6]	NA	X	NA	NA	NA
HPAH[6]	NA	X	NA	NA	NA
Acenaphthene	X	X	NA	NA	NA
Anthracene	X	X	NA	NA	X
Benzo(a) anthracene	X	X	NA	NA	X
Benzo(a)pyrene	X	X	NA	NA	X
Benzo(b) fluoranthene[7]	NA	X	NA	NA	X
Benzo(k) fluoranthene[7]	X	X	NA	NA	X
Chrysene	X	X	NA	NA	NA
Dibenzo(a,h) anthracene	X	X	NA	NA	X
Fluoranthene	X	X	NA	NA	X
Fluorene	X	X	NA	NA	X
Indeno(1,2,3)pyrene	X	X	NA	NA	X
Naphthalene	X	X	NA	NA	NA
Phenanthrene	X	X	NA	NA	NA
Pyrene	X	X	NA	NA	X
Bis(2-Ethylhexyl) Phthalate	X	X	NA	NA	X
Triclopyr	X	NA	NA	NA	NA
Nonylphenol	X	NA	NA	NA	NA
Petroleum - Heavy Fuel Oil	X	NA	NA	NA	NA
Petroleum - Diesel Fuel Oil	X	NA	NA	NA	NA
Petroleum - Gasoline	X	NA	NA	NA	NA
Petroleum - Lube Oil	X	NA	NA	NA	NA

NA - not evaluated in the hazard assessment for one or more reasons.
[1] Specifically the inorganic arsenic form was assessed for human health.
[2] Assessed as individual Aroclors and/or congeners or total PCBs.
[3] In the WOE discussion for PBDEs, limited effects data are discussed in the context of estimated daily doses for wildlife.
[4] Assessed as individual congeners, 2,3,7,8-TCDD or TEQs.
[5] Assessed as DDT, DDE, and DDD or as a sum.
[6] LPAHs and HPAHs are assessed as individual PAHs in all assessments except sediment.
[7] Assessed as part of total benzofluoranthenes in sediment assessment.

Table 10. Overall chemical groupings based on evaluation of all lines of evidence, including regional studies.

Multiple Priority 1 Classifications	Single Priority 1 Classifications		No Priority 1 Classifications
Copper	Arsenic*	Chrysene*	Lead*
Mercury*	Cadmium*	Fluoranthene*	Benzo(a)anthracene*
Zinc*	PBDEs*	Fluorene*	Triclopyr*
PCBs*	LPAHs	Naphthalene*	Nonylphenol*
Dioxins/Furans*	HPAHs	Phenanthrene*	Petroleum – Diesel*
DDT/DDE/DDD*	Anthracene*	Pyrene*	Petroleum – Heavy Fuel Oil*
Bis(2-ethylhexyl)phthalate*	Benzo(ghi)perylene*		Petroleum – Gasoline*
Benzo(a)pyrene*	Acenaphthene*		Petroleum – Lube Oil*
Dibenzo(ah)anthracene*	Acenaphthylene		
Indeno(123-cd)pyrene*			
Benzo(b)fluoranthene*			
Benzo(k)fluoranthene*			

Notes: COCs with an "*" were not prioritized in at least one media or pathway due to insufficient data (observed or effect data). It is important to note that not all COCs were evaluated in all media or pathways; the reader is encouraged to review Table 9 which summarizes the assessments that were conducted on each COC.

Uncertainty

While this hazard evaluation provides a broad general overview of the potential for the COCs evaluated to cause adverse ecological and human health effects, a number of uncertainties associated with the assessment process should be taken into consideration when interpreting the results. The following sections highlight the key uncertainties associated with the various elements of the assessment.

Environmental Data

This assessment included collection of a large number of environmental measurements for COCs in sediment, water and tissue from throughout the region. The EIM database likely includes the majority of observed ambient environmental data available for the Puget Sound region. Combined with the additional data included in this assessment, the dataset likely represents a reasonable representation of conditions in the Puget Sound region for many of the COCs. However, as previously indicated, there are likely data associated with special studies and research that have not been incorporated into readily accessible databases that were not included here.

Due to the broad nature of this assessment, the appropriateness of the analytical detection limits for the available environmental data were not evaluated. It is not anticipated that detection limits are a significant source of uncertainty for most COCs. However, for some COCs, such as PCB Aroclors® in surface waters, insufficient detection limits are a likely significant source of uncertainty. While there were a relatively large number of measurements for PCB Aroclors® (N>1200 in freshwater) the FOD was very low (3.5%). Based on a comparison to the much higher FOD for PCB congeners in water (58%), it is likely that Aroclor® measurements in water may represent an underestimate of the observed water concentration of this COC and PCBs as congeners better represents PCB concentrations and thus priority.

The assessment methodology is focused on COCs, which due to high concentrations (90^{th} %ile) in some areas may pose a threat to Puget Sound. Including estimates of non-detected concentrations (e.g. detection limits, ½ detection limits, or zero) would result in lowering the 90^{th} %ile for infrequently detected compounds, although it would not likely influence 90^{th} %iles for frequently detected chemicals. Thus, commonly measured but rarely detected COCs such as DDT/DDD/DDE in water would be less likely to rank as Priority 1, while the rank of commonly detected COCs such as zinc in water would be unchanged. By considering only detected concentrations, some COCs with low FODs may be conservatively included in Priority 1.

For the Human Health priority classifications, the greatest source of uncertainty is associated with COCs that were not evaluated because there are no NTR criteria for these chemicals; the priority for these COCS is unknown. An additional uncertainty is associated with the exposure pathways that were not assessed here (e.g., dermal water exposures as well as air, inhalation, and dust exposures). These various other exposure pathways result in an additive exposure to COCs.

Ecological Data

The wildlife assessment required a number of ecological parameters to estimate daily doses of COC s for wildlife receptors. The most important of these parameters are the food and sediment ingestion rates because bioaccumulation is driven mainly by prey and sediment ingestion. Although a model was applied to estimate prey ingestion rates, the uncertainty associated with this parameter is small relative to the sediment ingestion rates. This is due to the significant relationship between body size and food ingestion rate. However, sediment ingestion is less predictable from body size and is more dependent on feeding strategies and foraging habitat. The total daily dose estimated using the upper and lower sediment ingestion rates for each receptor in the wildlife assessment demonstrated that there is little relative sensitivity to this parameter. Thus, the high uncertainty associated with sediment ingestion rates appears to have minimal impact on the results of this assessment.

Effects Data

Due to the variety of effects data used for this assessment there are a number of uncertainties that should be considered when interpreting the results. The water assessment relied primarily on the effect data obtained from the ECOTOX database. While an effort was made to screen out effects data that were inappropriate for use, the sheer volume of effect concentrations precluded a detailed review of these data. The QA/QC process used to evaluate the accuracy of the ECOTOX data base suggests that the use of these data is likely a source of uncertainty. It is unclear if the errors associated with the ECOTOX database would result in an over- or underestimate of the potential for effects to occur.

Some of the greatest uncertainty in the surface water assessment was associated with lack of effects data for some COCs, particularly for marine organisms and some dissolved metals. Uncertainties associated with the effects data used for the surface water assessment include, but are not limited to differences in the following variables: species sensitivity, exposure conditions (water quality - hardness and pH, light regime, temperature, feeding regime if any, chemical form of the COC and whether the test was static or flow through), test duration, appropriateness of endpoints evaluated and type of endpoint. These differences make comparability of effects data challenging at best.

An additional source of uncertainty was the lack of non-mortality based effects data for some COCs; this was especially an issue for some of the marine COCs. Only evaluating effects associated with COC exposure at concentrations that cause mortality may underestimate the potential for these COCs to cause more subtle impacts (e.g., growth, reproduction etc.) to some aquatic organisms. Uncertainty is also associated with the comparison of surface water concentrations of bioaccumulative COCs (PCBs, mercury, DDTs) to effects data based on direct water exposure only. This comparison does not account for indirect impacts associated with bioaccumulation; the potential for adverse effects is likely underestimated for these COCs. The reader is encouraged to evaluate the types of effects data available for each COC when drawing conclusions about these results.

Hardness can have a significant influence on the toxicity of metals in freshwater; however, due to the large volume of data evaluated here it was not practical to account for study specific hardness. Due to the large number of effects data that were available for most metals, it is not expected that this would have a significant impact on the outcome of the assessment. In general, hardness in the Puget Sound region tends to be low; therefore, it is unlikely that the effects data would have significantly underestimated metal toxicity.

The sediment assessment relied on a variety of established sediment guidelines thresholds. While these guidelines have been reviewed by others and some have undergone regulatory scrutiny, there are still some uncertainties associated with their use. For some COCs the concentration of OC and sulfides in the sediment can have a significant influence on bioavailability and toxicity. While some of the thresholds used here incorporated organic carbon, most did not account for site specific conditions that could influence bioavailability and toxicity. This assumption could have resulted in both an over- or under estimate of the potential for effects. The sediment guidelines were developed based on impacts to benthic organisms. Therefore, they do not provide a direct assessment of how sediment associated COCs can indirectly impact other aquatic organisms through bioaccumulation.

There are significant uncertainties associated with the effects data used for the tissue assessment. While use of tissue residues to assess toxicity can be a useful tool in some cases, care must be taken when using these data to estimate the potential for effects, particularly when used in a screening approach. In many cases, the available tissue residue data are not based on experiments designed to directly relate tissue residue to an effect and as a result they lack dose response data. An additional limitation results from the way much of the residue effects data are reported; unlike water based toxicity data which is reported in the context of a dose response, relatively little of the tissue residue data is reported as such (Meador et al. 2008).

For some organisms and COCs, lipid content can be an important factor in interpreting the toxic response. Due to the nature of this assessment and the limited availability of data, lipid content was not incorporated into the data interpretation.

In addition, there is also significant uncertainty associated with the number and type of tissue residue effect concentrations available for some COCs and tissue types; in some cases there were only one or two tissue residue effects levels available for a COC and tissue type. Some of the tissue residue effects data were limited to data only for mortality endpoints; which likely underestimated the prioritization for these COCs and tissue types. Typically, elevated tissue levels of bioaccumulative compounds will result in more subtle effects to growth or reproduction before they cause mortality. These factors contribute to the high level of uncertainty associated with the tissue residue assessment.

The petroleum effects concentrations were limited to fuel and lubricating oils. It is suspected that actual toxicity can vary substantially based on product additives, weathering, and phototoxicity. Additionally, the studies that served as a source of effects data utilized various water-product mixtures. For the purposes of this assessment only dissolved fractions were utilized. However, there is anecdotal evidence that product spills are common and sheens of product can often be found in the environment in selected areas. Waters with free product are

potentially more toxic and/or present physical hazards to organisms. These issues create a high level of uncertainty for the petroleum assessment.

As previously discussed, the limited number of published effect doses for the wildlife receptors assessed here poses the greatest level of uncertainty in assessing potential effects in wildlife. In comparison to the volume of effects data available for aquatic life, the data available for wildlife may seem very weak; however, it should be noted that the dose-response studies for wildlife are frequently very comprehensive in examining effects. In comparison to a 30-day aquatic study that measures lethal doses, wildlife dose-response studies often include exposure over a reproductive cycle and examine everything from changes in adult body weight to egg size, time to hatch, female menses, and fertility of offspring. All these observations get lumped into broad categories. Considering the relative number of effects doses available for wildlife, the greatest uncertainty is associated with the dioxin/furan effects doses, particularly for birds.

Only about one-half of the COCs in this assessment were also listed in the NTR. The remaining chemicals were not evaluated, and thus pose a significant uncertainty regarding the potential for these COCs to cause human health effects. As previously discussed, none of the COCs were evaluated to estimate potential risk from water ingestion or dermal exposure. The same issue applies to sediment exposures, as there are no adopted and published standards for human exposure to sediments. Systemic human toxicity was also not evaluated for any of the COCs.

Data Gaps

It was assumed that the data identified for this assessment would be relatively representative of the region. However, due to the broad nature of this effort, the spatial distribution of data for a given COC within an assessment area (freshwater, near and offshore areas) is not well understood. This is a key gap in the current analytical approach when making broad generalizations regarding the priority on a sound wide basis.

For some matrices and spatial areas (e.g., freshwater metals where N was >3000) the assumption that the data are representative is likely true; however, data were limited for some COCs in some matrices and spatial areas (e.g., offshore COCs where many Ns were <20). This is not to suggest that thousands of measurements are necessary to provide a reasonable estimate of priority; in some cases other factors can be taken into account to increase the confidence of this assumption. For example, since the source of many COCs to offshore waters is via upland or nearshore inputs it can be reasonably assumed that if a COC is not Priority 1 in the nearshore it is not likely to be of Priority 1 in the offshore marine waters. There may be some exceptions to this logic; for example, bioaccumulative and hydrophobic chemicals tend to be present in freshwater streams at relatively low levels, but are subsequently biomagnified to high levels in upper trophic level organisms.

Potential gaps in data availability and spatial distribution should be taken into account when interpreting these data and using them to prioritize future efforts. A spatial analysis of existing data to identify regions or areas with high and low data densities would be a first step in understanding the magnitude and distribution of environmental data gaps.

As presented in the uncertainty discussion above, there were limited effects data for a number of COCs and matrices. This data gap is particularly significant for a number COCs in marine matrices. In water, there were limited effects data for many of the COCs evaluated; in some cases the available data were primarily based on mortality effects. This data gap limited the ability to fully assess the potential for these COCs to cause effects in marine waters and was compounded by the limited number of marine surface water measurements, particularly nearshore waters, for many COCs where the number of measurements was typically less than 15. Very few surface water PCB measurements in both marine and freshwaters were available; this limited the ability to fully assess this COC in this matrix.

In comparison to surface water, the sediment dataset (both effects and observed) was more robust. The most significant sediment data gap was the lack of multiple sediment guidelines for some COCs and a lack of nonylphenol environmental data. Only one guideline was available to evaluate nonylphenol in both marine and freshwater sediments. Only one set of guidelines were available for HPAH and LPAH and a number of individual PAHs in freshwater sediments and for dioxins/furans in both freshwater and marine sediments. Sediment guidelines were also lacking for triclopyr and PBDEs.

There were significant data gaps associated with the tissue assessment. Tissue residue effects data were limited for a number of the COCs and tissue types evaluated. In some cases there were less than five tissue residue effect values (e.g., mercury in marine tissues; PCBs in freshwater decapods and marine fish; dioxins/furans in all tissue types except freshwater fish; DDTs in marine tissues). There were very few measurements of dioxins/furans in tissue; no data were available for freshwater tissue types. There were limited PBDE tissue data and no readily available effects data for this COC. These data gaps limited the ability to fully assess the potential effects of these COCs.

As previously indicated, the lack of NTR criteria limited the number of COCs that could be evaluated for human health effects. Although there were invertebrate inorganic arsenic data, there were no inorganic arsenic data for freshwater or marine fish, which limited the ability to fully assess the human health priority for this COC. Additionally triclopyr, PBDEs and the sum of cPAHs are not part of the NTR and could not be prioritized.

The limited amount of effects data for PBDEs limited the ability to assess them in tissue and wildlife.

Recommendations for Further Assessment

- The spatial distribution of data used in this assessment was not assessed beyond the general habitat types (freshwater, marine near and offshore). The data for some COCS in all matrices are not likely evenly distributed throughout the region. As a result it is unknown if the data used in this assessment are truly representative of the region or only limited areas. This is of greatest concern for those COCs for which data were limited (see data gaps listed above). To fully understand if the priority identified for a COC applies on a regional basis, or is limited to a discrete area, additional data analysis that examines the spatial distribution

of the data could be done. This analysis would provide additional information to focus future hazard reduction efforts to areas of greatest concern.

- Marine surface water data were limited for a number of COCs. Expansion of the number and type of parameters measured by ambient surface water monitoring programs would provide a means to fill this environmental data gap. The parameter list should focus on COCs that were Priority 1 in freshwaters; in addition to those likely to be bioaccumulated in the marine food web (e.g., nonylphenol, mercury, PCBs, HPAHs, PBDEs, and DDTs).

- Petroleum product data was not available for marine waters and the degree to which individual PAHs represent spilled and degraded petroleum products is unknown. Future investigations targeting a more complete suite of alkylated PAHS, other PAH and petroleum products, and their degradates could more fully characterize petroleum hazards.

- Sediment data for nonylphenol, particularly in marine sediment was limited. Sediment data for PCB congeners and PBDEs was also limited, particularly in marine nearshore areas. PCB Aroclor data is likely sufficient to identify priority; however additional PBDE and nonylphenol data would improve the ability to determine the priority for these COCs

- Tissue data for inorganic arsenic were very limited. Collection of inorganic arsenic data in edible seafood tissue would provide more certainty in determining human health priority for this COC.

- Effects data to assess surface waters were limited for a number of COCs, particularly for effects other than mortality. It is possible that additional data may be identified through a literature search and review for those COCs that are of greatest concern (this was beyond the scope of the current assessment). Additional effects data for some COCs would provide better certainty in determining the priority.

- Sediment guidelines were not available or limited for a number of COCs (PBDEs, dioxins, and nonylphenol). It is possible that additional effects data may be identified through a literature search. Additional effects data for these COCs would provide better certainty in determining priority.

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Appendix E. Glossary, Acronyms, and Abbreviations

Glossary

Ambient area: Un-impacted area; away from point sources of contamination.

Anthropogenic: Human-caused.

Aquatic life: Aquatic life refers to any organism which spends all of, the majority of, or significant portions of its life stage in water.

Attenuation: The reduction in the concentration, mass, toxicity, mobility, or volume of a chemical due to chemical, biological, or physical processes.

Baseflow: Groundwater discharge to a surface stream or river. The component of total streamflow that originates from direct groundwater discharges to a stream.

Basin: A drainage area or watershed in which all land and water areas drain or flow toward a central collector such as a stream, river, or lake at a lower elevation.

Benthic: Bottom-dwelling.

Bioaccumulative chemicals: Chemicals that build up and become concentrated in organisms.

Bivalves: Mollusks having a shell consisting of two hinged valves. Examples are clams, mussels, and oysters.

Built-up roof: A general term that can be applied to many flat, membrane, or torch-down roof types.

Catch basin: Large underground container for the collection of sediment and other debris from stormwater run-off. Designed to catch or collect the dirt and debris, and prevents it from entering surface water.

Congener: In chemistry, congeners are related chemicals. For example, polychlorinated biphenyls (PCBs) are a group of 209 related chemicals that are called congeners.

Constrained systems: Physical systems that generally do not allow chemicals to become mobilized in other environmental compartments or environmental pathways (e.g., sanitary sewer systems).

Dry season: In this study, May through September.

Effects data: In this study and specifically for the hazard evaluation, data from toxicity testing or derived otherwise to indicate chemical concentrations where effects are elicited.

Effluent: An outflowing of water from a natural body of water or from a man-made structure. For example, the treated outflow from a wastewater treatment plant.

Enrichment: Natural or human-caused enhancement of chemical concentrations in environmental media such as water or soils.

Exceeds (or is above) guidelines or standards: Does not meet guidelines or standards.

First flush: A rain event following an extended dry period. Runoff from a first flush can contain elevated concentrations of contaminants.

Flux: Amount that flows through a unit area in a unit of time.

Guilds: In this study, groups distinguished by their trophic level.

Hazard evaluation: A screening-level assessment of relative toxic hazards posed by chemicals at observed concentrations.

Land cover types: In this study, commercial/industrial, residential, agricultural, and forests.

Load pathways: In this study, groundwater, air deposition, surface water runoff, publicly-owned treatment works, and ocean exchange.

Loading: The input of pollutants into a waterbody.

Marine water: Saltwater

Micronutrient: Nutrients required by organisms at very low concentrations.

Nutrient: Substance such as carbon, nitrogen, and phosphorus used by organisms to live and grow. Too many nutrients in the water can promote algal blooms and rob the water of oxygen vital to aquatic organisms.

Observed data: In this study and specifically for the hazard evaluation, data on chemical concentrations in various media sampled since January 1, 2000.

Ocean boundary: Approximate area or space – theoretically defined by a vertical plane, a series of planes, or polygons – where Pacific Ocean waters are exchanged with the waters of Puget Sound.

Ocean exchange: The flux of Pacific Ocean waters and the waters of Puget Sound across the ocean boundary.

Organics: Shorthand for organic chemicals (those which contain carbon).

Parameter: Water quality constituent being measured (analyte). A physical, chemical, or biological property whose values determine environmental characteristics or behavior.

Pelagic: Open-water.

Personal care products (or toiletries): Products used for personal hygiene or beautification. Personal care includes products as diverse as chapstick, colognes, cotton swabs, deodorant, eye liner, facial tissue, hair clippers, lipstick, lotion, makeup, mouthwash, nail files, pomade, perfumes, personal lubricant, razors, shampoo, shaving cream, skin cream, toilet paper, cleansing pads and wipes, lip gloss, toothbrushes, and toothpaste, to give a few examples.

Piscivorous: Fish-eating.

Point source: Sources of pollution that discharge at a specific location from stacks, pipes, outfalls, or conveyance channels to a surface water.

Salmonid: Any fish that belong to the family *Salmonidae*. Basically, any species of salmon, trout, or char.

Sediment: Solid fragmented material (soil and organic matter) that is transported and deposited by water and covered with water (example, river or lake bottom).

Sill: A relatively shallow area of the seabed.

Source: For the purpose of the present project, the term *source* is strictly defined as: the object or activity from which a COC is initially released to environmental media (air, water, or soil) or released in a form which can be mobilized and transported in an environmental pathway.

Stormwater: The portion of precipitation that does not naturally percolate into the ground or evaporate but instead runs off roads, pavement, and roofs during rainfall or snow melt. Stormwater can also come from hard or saturated grass surfaces such as lawns, pastures, playfields, and from gravel roads and parking lots.

Study Area: The geographical study area for this *Assessments* study is Puget Sound, the U.S. portions of the Straits of Georgia and Juan de Fuca, and the entire U.S. watershed for Puget Sound and the Straits.

Surface runoff: In this study, surface runoff is broadly defined to include stormwater, nonpoint source overland flow, and groundwater discharge to surface waters that flow into marine waters.

Toxicant: Toxic contaminant.

Toxics: Shorthand for toxic chemical.

Water column: In a waterbody, a conceptual cylinder of water extending from the top of the sediment layer to the surface of the water.

Watershed: Basin. A drainage area in which all land and water areas drain or flow toward a central collector such as a stream, river, or lake at a lower elevation.

Wet season: In this study, October through April.

Acronyms and Abbreviations

a.k.a.	also known as
ABS	acrylonitrile butadiene styrene
AOP	Air Operating Permit
API	American Petroleum Institute or [King County] Asian and Pacific Islander
ATSDR	Agency for Toxic Substances and Disease Registry
BEE	butoxyethyl ester
CAP	Chemical Action Plan
CCA	chromated copper arsenate
COC	chemical of concern
cPAH	carcinogenic polycyclic aromatic hydrocarbon
CSL	cleanup screening level
DBP	di-n-butyl phthalate
DDT	dichlorodiphenyltrichloroethane
DEHP	di(2-ethylhexyl) phthalate [a.k.a. bis(2-ethylhexyl) phthalate]
DEP	diethyl phthalate
ECB	European Chemicals Bureau
Ecology	Washington State Department of Ecology
EPA	U.S. Environmental Protection Agency
Hg	mercury
HPAH	high-molecular weight polycyclic aromatic hydrocarbon
LAET	lowest apparent effects threshold
LCR	Lead and Copper Rule

LPAH	low-molecular weight polycyclic aromatic hydrocarbon
N	number
NOAA	National Oceanic Atmospheric Administration
NTR	National Toxics Rule
OC	organic carbon
PAH	polycyclic aromatic hydrocarbon
PBDE	polybrominated diphenyl ether
PBT	persistent, bioaccumulative, and toxic substance
PCB	polychlorinated biphenyl
PCDD/F	polychlorinated dibenzo-*p*-dioxin and dibenzofuran
PEC	probable effects concentration
PEL	probable effects level
POP	persistent organic pollutant
POTW	Publicly-owned treatment works
PSP	Puget Sound Partnership
PSTLA	Puget Sound Toxics Loading Analysis
PVC	polyvinyl chloride
SQS	Sediment Quality Standards
TEA	triethylamine
TEC	threshold effects concentration
TEL	thresholds effects level
TEQ	toxic equivalent
TRI	Toxics Release Inventory
UC	University of California
USGS	U.S. Geological Survey
WAC	Washington Administrative Code
WDFW	Washington Department of Fish and Wildlife
WDOH	Washington State Department of Health
WQC	water quality criteria

Units of Measurement

°C	degrees Celsius
d	day
dw	dry weight
g	gram, a unit of mass
kg	kilograms, a unit of mass equal to 1,000 grams.
kg/d	kilograms per day
km	kilometer, a unit of length equal to 1,000 meters.
m	meter
mg	milligrams
mg/kg	milligrams per kilogram (parts per million)
mg/l	milligrams per liter (parts per million)
ng/g	nanograms per gram (parts per billion)
ng/kg	nanograms per kilogram (parts per trillion)
ng/l	nanograms per liter (parts per trillion)
pg/l	picograms per liter (parts per quadrillion)

t	tonne (metric ton, equal to 1,000 kg); appr. 2.2 tons
t/yr	tonnes (metric tons) per year
ug/kg	micrograms per kilogram (parts per billion)
ug/l	micrograms per liter (parts per billion)
ww	wet weight

Persistent Organic Pollutants in Feed and Rainbow Trout from Selected Trout Hatcheries

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Persistent Organic Pollutants in Feed and Rainbow Trout from Selected Trout Hatcheries

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April 2006

Waterbody numbers: Statewide

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Abstract

During 2005, persistent organic pollutants (POPs) were analyzed in fish feed and catchable rainbow trout from ten Washington Department of Fish and Wildlife (WDFW) hatcheries and the fish purchased by WDFW from one private hatchery operator. Fish originating from the same hatchery populations were also sampled approximately 2½ months following planting into unpolluted lakes in order to assess contaminant depuration or uptake. All feed and tissue (fillet) samples were analyzed for a variety of chlorinated pesticides, polychlorinated biphenyls (PCBs), a select group of polybrominated diphenyl ethers (PBDEs), and lipid content. A subset of feed and tissue samples was also analyzed for polychlorinated dioxins and furans (PCDD/Fs).

Feed samples had the following mean wet weight concentrations: ΣPCBs[1] – 13.8 ng/g, ΣDDT – 8.2 ng/g, ΣPBDEs – <0.25 ng/g, PCDD/F toxic equivalent – 0.75 pg/g. Fish tissue samples had the following respective mean wet weight concentrations in hatchery and planted rainbow trout: ΣPCBs – 13.0 and 3.1 ng/g, ΣDDT – 3.9 and 8.8 ng/g, ΣPBDEs – 0.66 ng/g for both. PCDD/F toxic equivalent averaged 0.032 pg/g in hatchery fish but was not analyzed in trout collected from lakes.

Other pesticides found in feed (f), hatchery fish (h), and planted fish (p) were: DDMU (f,h,p), dieldrin (f,h,p), hexachlorobenzene (f,h,p), pentachloroanisole (f,h,p), trans-nonachlor (f,h), cis-chlordane (f,h), trans-chlordane (f), methoxychlor (f), and toxaphene (f).

Results suggest that some portion of POP concentrations in trout from unpolluted waters may originate from hatcheries. In addition, some catchable trout contain POP concentrations above regulatory criteria when they are planted in lakes.

[1] Σ = Total

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Staff with the Washington Department of Fish and Wildlife:

- John Kerwin, Hatchery Division Manager, helped conceive and develop this project
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- District biologists provided valuable suggestions and information on lakes and their trout populations
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Introduction

Recent reports have indicated that commercially farmed salmon, hatchery-raised trout, and the feed used to grow them may contain polychlorinated biphenyls (PCBs) and other persistent organic pollutants (POPs). For instance, Hites et al. (2004) showed that salmon raised in net-pens had substantially higher PCBs than those caught wild, presumably due to PCB-contaminated feed. Carline et al. (2004) found that concentrations of PCBs in hatchery rainbow trout (*Oncorhynchus mykiss*) fillets were correlated to concentrations in feed, and nearly all the body burden was due to PCBs in the diet. Other investigations have revealed detectable concentrations of dioxins, dieldrin, and endrin as well as PCBs in hatchery broodstock salmon and trout (Millard et al., 2004). In Pennsylvania, PCB contamination of edible tissues accumulated through dietary uptake in hatcheries exceeded thresholds for issuance of consumption advisories (Carline et al., 2004).

Currently there is no statewide program in Washington to evaluate toxic chemicals in hatchery feed or hatchery fish. At the same time, low levels of POPs in fish from lakes and streams across the state are being detected at an increasing rate (e.g., Seiders 2003; Seiders and Kinney, 2004) due to increased sampling coverage and better analytical detection limits. These waterbodies are often added to the list of impaired waters as required by the federal Clean Water Act section 303(d), and subsequently require a plan to control or clean up the contaminants. Many of the POPs found in fish tissue (e.g., PCBs, dioxins) are ubiquitous environmental contaminants and may be found globally through atmospheric deposition, historical releases, or food-web cycling. Fish may accumulate low concentrations of these chemicals through one or more of these pathways, although it is nearly impossible to distinguish and quantify these diffuse sources, and control and clean-up is often unrealistic. Due to recent data, however, contamination stemming from hatcheries is now considered a possible source of POPs in fish.

Study Description

Catchable rainbow trout – fish approximately six inches or more released into lakes and streams just prior to the opening of fishing season – were sampled from ten Washington Department of Fish and Wildlife (WDFW) hatcheries. Approximately 2½ months following planting, samples from un-mixed hatchery populations were sampled from stocked lakes. All feed and tissue (fillet) samples were analyzed for a variety of chlorinated pesticides, PCB aroclors, a select group of polybrominated diphenyl ethers (PBDEs), and lipid content. A subset of feed and tissue samples was also analyzed for polychlorinated dioxins and furans (PCDD/Fs). Specific project objectives were to:

- Measure concentrations of POPs in catchable rainbow trout released to lakes by WDFW.
- Measure concentrations of POPs in feed used to raise catchable rainbow trout in WDFW hatcheries to assess the correlation between diet and contaminant burdens in fish tissue.
- Estimate the degree of contaminant depuration or uptake in catchable rainbow trout following their release into lakes.

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Methods

Study Design

Fish feed and catchable rainbow trout were sampled from ten WDFW hatcheries during March 29 - April 5, 2005. Hatchery selection was made based on consultation with John Kerwin, Hatchery Division Manager with the WDFW Fish Program. Figure 1 shows locations of hatcheries. All ten hatcheries use well or spring water for hatching and rearing, although Tucannon River water is used in the final six-month rearing phase at the Tucannon Hatchery. Vancouver and Puyallup Hatchery personnel have also observed surface runoff entering hatcheries during rainy periods, but the extent of the exposure to fish is minor. POPs are much less likely to be present in groundwater and surface water due to their low solubility and immobility in soils.

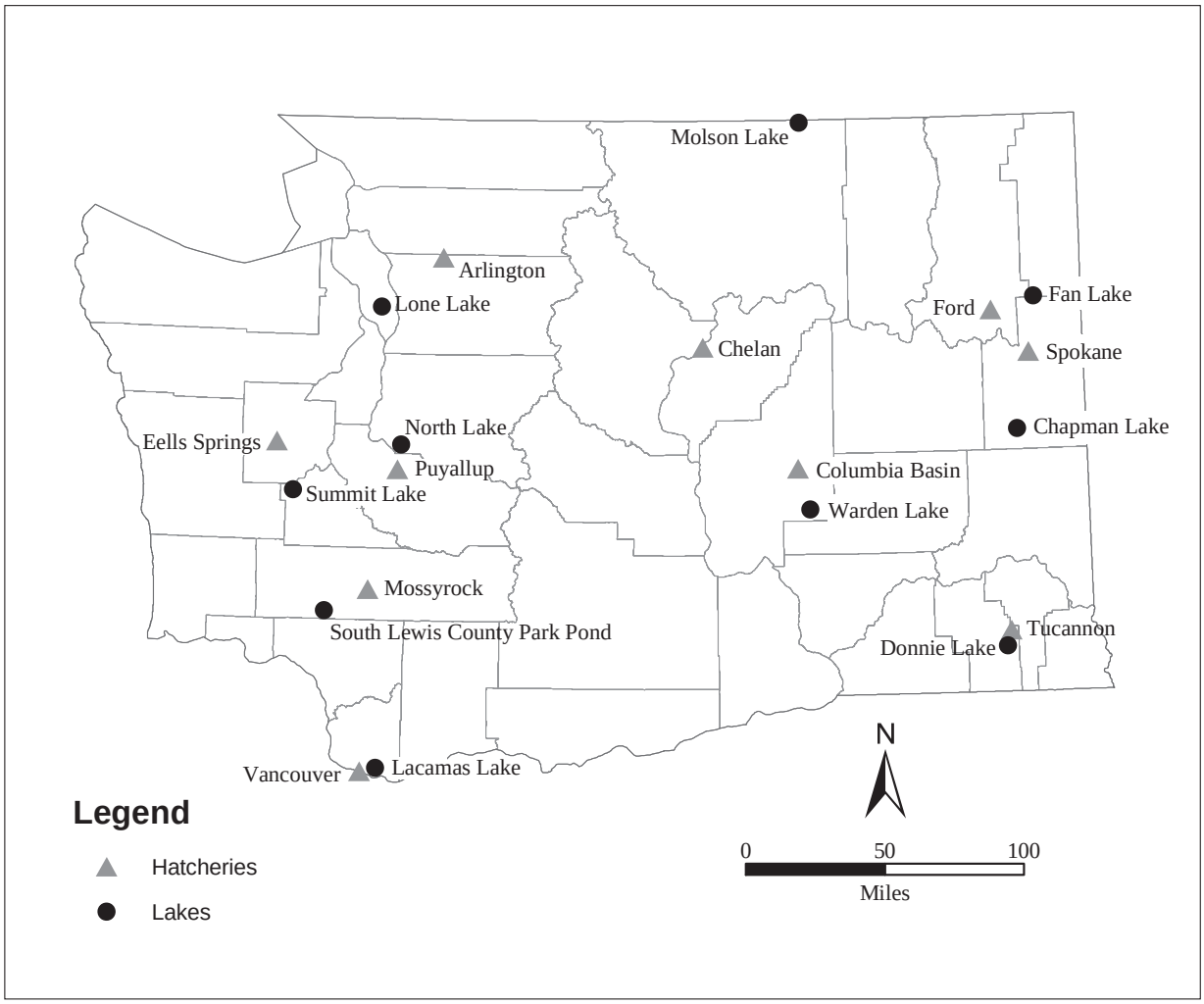

Figure 1. Locations of Hatcheries and Lakes Sampled for the 2005 Study of Persistent Organic Pollutants in Hatchery Feed and Hatchery Fish.

Hatchery personnel were interviewed about the feed used, schedule for changes in feed size and type, weight growth obtained using the sampled feed, hatchery water source, planting schedules, and other pertinent information related to the project. Feed samples consisted of material being fed to the trout at the time of sampling. In most cases, fish had been on the feed sampled for at least four months during which they had gained 50% - 80% of their mass (Table 1).

Table 1. Feed Analyzed from WDFW Hatcheries.

Hatchery	Feed Sampled	Pellet Size (mm)	Manufacturer	Period of use	Weight Gain
Arlington	Rangen	3.2	Rangen, Inc. Buhl, ID	6 mo.	76%
Chelan	Silver Cup Trout	3.0	Nelson & Sons, Inc., Murray, UT	6 mo.	76%
Columbia Basin	Silver Cup Fish Feed	3.2	Nelson & Sons, Inc., Murray, UT	6 mo.	80%
Eells Springs	Rangen	4.0	Rangen, Inc. Buhl, ID	4 mo.(a)	56%
Ford	Orient	4.0	Skretting, Vancouver, B.C.	na	50%
Mossyrock	Silver Cup Salmon	3.0	Nelson & Sons, Inc., Murray, UT	10 mo.	na
Puyallup	EWOS Vita	3.0	EWOS, Surrey, B.C.	na	na
Spokane	Silver Cup Fish Feed	3.2	Nelson & Sons, Inc., Murray, UT	na	78%
Tucannon	EWOS Pacific	3.0	EWOS, Surrey, B.C.	1 mo.(b)	70% (c)
Vancouver	Rangen	4.0	Rangen, Inc. Buhl, ID	6 mo.	82%

(a) fish fed Rangen since fry stage
(b) fish fed EWOS 2.0 for preceding 4 months, and EWOS 1.2 for 2 months prior to that
(c) weight gain during diet of EWOS 1.2, EWOS 2.0, and EWOS 3.0
na - not available

Ten rainbow trout specimens from each hatchery were randomly selected for sampling. They were from the general catchable populations which were in the process of being planted or were planned to be stocked within the subsequent weeks.

Ten triploid rainbow trout from Troutlodge, a private facility that supplies trout to WDFW, were provided by WDFW staff. Triploid trout are fish with three sets of chromosomes produced by pressure-treating the newly dividing fertilized eggs. Since they are sterile, more energy is used for somatic growth than gamete production, and the resulting triploid trout is larger than diploid fish of the same age. No feed samples or post-plant fish associated with Troutlodge were sampled for this study.

Lakes selected for sampling were based on the following criteria:

1. No known contaminant sources and low potential for appreciable contamination
2. Little or no natural rainbow trout production
3. Rainbow trout originating from a single hatchery planted between late-March and mid-April, 2005
4. Geographically dispersed to reflect a variety of ecosystem types, water chemistry, aquatic environments, and regions of the state containing differing preponderance of land use types.

Few of the approximately 380 lakes stocked annually with catchable trout have any contaminant data. Therefore, criterion 1 was assumed to be met unless a potential contaminant source was obvious. District WDFW biologists were interviewed to satisfy criterion 2. Criterion 3 was the most difficult to meet due to the common practice of multiple plantings of fish from different hatcheries. Lakes are also often planted at various intervals throughout the spring, which would yield uncertainties in fish residence periods; these lakes were avoided. Planting reports provided weekly by WDFW were reviewed in order to find lakes with single hatchery plants and fish residence times of approximately 2½ months. Criterion 4 was easily satisfied due to the geographic separation of the hatcheries and their associated lakes. Table 2 lists lakes where rainbow trout were sampled and their hatcheries of origin.

Table 2. Lakes Sampled and 2005 Rainbow Trout Plants.

Lake	County	Area (hect.)	Mean Depth (meters)	2005 Stock Date	Number	Mean Weight (grams)	Hatchery
Lone	Island	41	2.7	30-Mar	4,060	114	Arlington
				18-Apr	593*	649	Troutlodge
Molson	Okanogan	9.3	1.8	12-Apr	4,160	142	Chelan
Warden	Grant	81	8.2	15-Mar	12,723	116	Columbia Basin
				18-Mar	11,596	108	
				5-Apr	685	123	
Summit	Thurston	214	16	28-Mar	400	1,746	Eells Springs
				29-Mar	402	1,681	
				14-Apr	10,048	142	
				15-Apr	646	825	
				22-Apr	15,097	138	
				25-Apr	9,810	153	
				26-Apr	5,056	142	
Fan	Pend Oreille	32	7.6	22-Mar	3,021	86	Ford
S. Lewis Co. Park Pond	Lewis	4.5	2.7	14-Apr	3,043	134	Mossyrock
North	King	23	4.3	20-Apr	8,500	114	Puyallup
Chapman	Spokane	61	20	15-Mar	5,925	91	Spokane
Donnie	Columbia	0.4	0.9	14-Apr	420	108	Tucannon
Lacamas	Clark	129	7.3	7-Mar	2,000	227	Vancouver
				8-Apr	3,000	267	
				26-Apr	4,000	197	

*Triploid fish

Sampling Procedures

Hatchery feed and pre-plant catchable rainbow trout samples were collected with assistance of hatchery staff. Fish averaged 235 mm total length and 152 gm in weight (excluding Troutlodge samples). Feed samples were placed directly in 1-liter organics-free glass jars with Teflon lid liners and certificates of analysis. Fish from hatcheries were killed with a blow to the skull, double-wrapped in aluminum foil, sealed in zip-lock polyethylene bags, and transported on ice to Ecology headquarters where they were weighed and measured prior to being stored frozen at –20° C.

Rainbow trout from lakes were collected by hook-and-line or electrofishing. Following capture, fish were observed for signs confirming previous hatchery residence. Specimens were then killed with a blow to the skull, weighed to the nearest gram and measured to the nearest millimeter, assigned a sample number, double-wrapped in aluminum foil, placed in zip-lock polyethylene bags, and transported on ice to Ecology headquarters where they were stored frozen at –20° C. Rainbow trout collected from lakes averaged 270 mm total length and 211 gm in weight.

When ready for processing, fish were partially thawed then scales were removed for aging by WDFW. Composite samples of homogenate tissue were prepared by methods described by EPA and the Washington State Toxics Monitoring Program (EPA, 2000; Seiders, 2003). Briefly, fish were scaled, skin-on fillets removed, and equal mass aliquots of tissue were homogenized with three passes through a Kitchen-Aid food processor for each composite. Homogenates were placed in a 4-oz organics-free glass jar with Teflon lid liner and certificate of analysis and stored frozen.

All resection was done with non-corrosive stainless steel implements on a clean aluminum foil surface. Persons preparing samples wore non-talc polyethylene or nitrile gloves changed between samples. Resection and homogenizing equipment was cleaned using Liquinox® detergent and hot tap water, followed by rinses with deionized water, pesticide grade acetone, and pesticide grade hexane, then air-dried in a fume hood before use.

Laboratory Analysis and Data Quality

Sample Preparation

Feed

Feed samples were first Soxhlet extracted using 1:1 methylene chloride/hexane, then solvent exchanged into hexane and adjusted to 10 ml. Extracts were split, half for PCB/chlorinated pesticide and half for PBDE and lipid analysis.

Extracts for PCB and chlorinated pesticide analysis were eluted through 2 gm micro Florisil® columns first with 100% hexane and collected as the “0% Florisil fraction”, followed by elution with 1:1 hexane/preserved diethyl ether, collected as the “50% Florisil fraction”. When the

"0% Florisil fractions" were solvent-reduced, the remaining extracts were as much as 50% lipids, unsuitable for gas chromatography (GC) analysis, and were therefore added to the "50% Florisil fraction". The combined extracts were then back-extracted with acetonitrile to remove lipids and re-eluted through 2 gm micro Florisil® columns with 100% hexane ("0% Florisil fraction") and 1:1 hexane/preserved diethyl ether ("50% Florisil fraction"). Each fraction was solvent-exchanged to iso-octane and concentrated to 1 ml. One-half of the "50% Florisil fraction" and the "0% Florisil fraction" were treated with concentrated sulfuric acid prior to analysis. The remainder of the "50% fraction" was analyzed without acid treatment.

Fish Tissue

Tissue samples were first Soxhlet extracted using 1:1 methylene chloride/hexane, then solvent exchanged into hexane and adjusted to 10 ml. Extracts were split, half for PCB/chlorinated pesticide and half for PBDE and lipid analysis.

Extracts for some PCB analyses (sample nos. 05248100 – 05248109) were eluted through 2 gm micro Florisil® columns with 100% hexane, solvent-exchanged to iso-octane, and concentrated to 1 ml. Extracts were treated with concentrated sulfuric acid prior to analysis.

For chlorinated pesticide analysis and some PCB analyses (sample nos. 05144080 – 05144090), extracts were eluted through 2 gm micro Florisil® columns with 100% hexane and collected as the "0% Florisil fractions", followed by elution with 1:1 hexane/preserved diethyl ether. The hexane/ether fractions were adjusted to 5 ml and back-extracted with acetonitrile to remove lipids and re-eluted through 2 gm micro Florisil® columns with 1:1 hexane/preserved diethyl ether, and collected as the "50% Florisil fraction". Each fraction was solvent-exchanged to iso-octane and concentrated to 1 ml. One-half of the "50% Florisil fraction" and the "0% Florisil fraction" were treated with concentrated sulfuric acid prior to analysis. The remainder of the "50% fraction" was analyzed without acid treatment.

Analyses for PCBs, chlorinated pesticides, and PBDEs were conducted at the Manchester Environmental Laboratory (MEL) using dual column GC/ECD. Sample preparation and analysis methods were modifications of EPA SW-846 Methods 3540, 3620, and 8081/8082.

Samples for PCDDs/PCDFs were analyzed at Pacific Rim Laboratories, Inc. (Surrey, B.C.) using high resolution GC/MS isotope dilution methodology of EPA Method 1613B. Percent lipid was analyzed gravimetrically at MEL. The complete list of analytes is in Appendix B.

Data Quality

Overall quality of the data was fair. Precision for Aroclor analysis was 11% relative percent difference. Analysis was also performed with a high degree of precision for DDT compounds (14%), other chlorinated pesticides (17%), PBDEs (17%), and PCDD/Fs (13%).

One standard reference material was analyzed along with feed and tissue; NIST 1974b – Organics in Frozen Mussel Tissue (https://srmors.nist.gov/tables/view_table.cfm?table=109-2.htm). Chlorinated pesticide analytes, including DDT compounds, were only 67% of certified concentrations on average. Total PCBs were 82% of the reference concentration. These results indicate a possible low bias for these analyte groups.

Data Analysis

PCB, DDT, PBDE, and lipid concentrations were compared between hatchery and lake rainbow trout using the Wilcoxon signed-rank test, a non-parametric equivalent of the paired t-test (Zar, 1984). Spearmann ranked correlations among contaminant groups and sample types were done using SYSTAT 9.01 software program (SPSS, 1998). Non-detected values were treated as zero for statistical tests to avoid misinterpretation of comparisons between hatchery tissue samples and the lake tissue samples, which had different detection limits for the same analytes.

Non-detects were also treated as zero for samples analyzed in duplicate. Therefore, values presented as the mean of duplicate analyses may be biased low. The complete set of chemistry data is in Appendix C.

Results

Field Observations

Physical observations of rainbow trout collected from lakes indicated that they originated from hatcheries sampled earlier in the year. Most specimens had gnawed pectoral fins, or dorsal and caudal fin erosion. Scale annuli patterns also indicated fish were from the year's stock of catchables (John Sneva, WDFW, written communication, 8/10/2005). This physical evidence, coupled with local knowledge from WDFW District Biologists and WDFW plant records, supports the conclusion that the trout had been raised as catchables from known hatcheries.

Rainbow trout collected from lakes in June were larger on average than just prior to their release from hatcheries (Table 3). Increases in total length averaged 15%, and weight gain was 39% on average. Mean condition factors in hatchery and planted trout were 1.11 and 1.02, respectively. Condition factors fell below 1.0 in half the lakes, suggesting food supply was limited. Gut contents were not examined, but aside from the fin erosion mentioned previously, the fish collected from lakes appeared healthy and took bait and lures readily in most cases.

Contaminants in Feed and Fish

PCBs, DDT, and PBDEs

Most feed and fish tissue samples contained measurable concentrations of PCBs (Table 4). Aroclor-1254 was the most commonly detected, followed by 1260, 1242, and 1248; none of the other Aroclors were detected.

All samples contained DDT compounds, with 4,4'-DDE comprising 74% of the ΣDDT on average. All but one of the tissue samples contained low levels of PBDEs; none of the feed samples had detectable PBDEs. PBDE-47 was the most common congener detected, followed by 99, 71/100, and 138/209.

Mean concentrations of lipids in feed were high (16.8%) compared to tissue. This high fat diet resulted in high lipid levels in hatchery rainbow trout fillet tissue (mean of 3.2%). Although the catchable rainbow trout increased in size following planting, it appears that muscle lipid was depleted to meet their energy requirements, with an average 60% decrease in lipid content (1.2%).

Table 3. Length and Weight of Rainbow Trout Collected from Hatcheries and Lakes.

Hatchery/Lake	2005 Collection Date	N	Total Length (mm, mean ± SD)	Weight (gm, mean ± SD)	Condition Factor (mean ± SD)
Arlington Hatchery	29-Mar	10	245 ± 16	165 ± 34	1.11 ± 0.09
Lone Lake	16-Jun	10	306 ± 12	334 ± 37	1.17 ± 0.06
Chelan Hatchery	5-Apr	10	253 ± 16	178 ± 37	1.08 ± 0.11
Molson Lake	13-Jun	10	296 ± 20	303 ± 59	1.16 ± 0.08
Columbia Basin Hatchery	5-Apr	10	230 ± 15	140 ± 31	1.15 ± 0.08
Warden Lake	9-Jun	10	251 ± 10	147 ± 23	0.93 ± 0.08
Eells Springs Hatchery	1-Apr	10	230 ± 8	142 ± 19	1.16 ± 0.07
Summit Lake	13-Jun	7	259 ± 13	160 ± 16	0.92 ± 0.12
Ford Hatchery	4-Apr	10	197 ± 15	83 ± 15	1.09 ± 0.06
Fan Lake	14-Jun	8	290 ± 14	271 ± 47	1.11 ± 0.15
Mossyrock Hatchery	5-Apr	10	260 ± 14	190 ± 31	1.08 ± 0.06
S. Lewis Co. Park Pond	14-Jun	8	259 ± 10	176 ± 24	1.01 ± 0.07
Puyallup Hatchery	1-Apr	10	218 ± 20	111 ± 31	1.04 ± 0.11
North Lake	13-Jun	10	245 ± 12	141 ± 21	0.96 ± 0.08
Spokane Hatchery	4-Apr	10	210 ± 13	98 ± 21	1.04 ± 0.08
Chapman Lake	15-Jun	4	243 ± 10	125 ± 13	0.87 ± 0.04
Tucannon Hatchery	4-Apr	10	206 ± 18	108 ± 30	1.21 ± 0.06
Donnie Lake	16-Jun	10	254 ± 19	145 ± 33	0.87 ± 0.07
Vancouver Hatchery	5-Apr	10	298 ± 24	303 ± 91	1.12 ± 0.10
Lacamas Lake	17-Jun	9	285 ± 14	249 ± 37	1.07 ± 0.06
Troutlodge Hatchery	4-Apr	10	374 ± 22	678 ± 133	1.29 ± 0.19

Condition Factor = $(W[g] \times 100/L[cm]^3)$
N = number
SD = standard deviation

Table 4. Lipid, ΣPCB, ΣDDT, and ΣPBDE Concentrations in Feed and Rainbow Trout Fillet Tissue (ng/g, ww).

Sample Type/Location	% Lipid	ΣPCB	ΣDDT	ΣPBDE
Hatchery Feed				
Arlington	18.19	**13.8**	**6.3**	U(0.25-1.2)
Chelan	13.75	**34.8**	**9.4**	U(0.25-1.2)
Columbia Basin	14.47	**11.6**	**6.3**	U(0.24-1.2)
Eells Spring	12.70	**12.5**	**5.9**	U(0.24-1.2)
Ford*	25.85	U(2.5)	**3.7**	U(0.25-1.2)
Mossyrock	19.64	**27.6**	**11.0**	U(0.25-1.2)
Puyallup*	16.14	U(2.5)	**6.6**	U(0.25-1.2)
Spokane	15.79	**16.4**	**5.9**	U(0.25-1.2)
Tucannon	15.01	**8.2**	**21**	U(0.25-1.2)
Vancouver	16.08	**13.3**	**5.8**	U(0.25-1.2)
Hatchery Rainbows				
Arlington	3.97	**12.1**	**4.8**	**0.64**
Chelan	3.05	**67**	**4.1**	**1.09 J**
Columbia Basin	4.10	**18.5**	**6.5**	**0.90 J**
Eells Spring*	2.42	U(2.4)	**2.7**	**0.52**
Ford	2.35	U(2.5)	**2.5**	**0.24 J**
Mossyrock	2.69	**15.8**	**3.9**	**0.89 J**
Puyallup	3.07	U(2.3)	**2.4**	**0.24**
Spokane	2.48	**11.7**	**2.9**	**1.10 J**
Tucannon	3.69	U(2.4)	**5.3**	**0.27**
Vancouver*	4.00	**4.8**	**4.0**	**0.71 J**
Troutlodge	5.39	**14.4**	**5.7**	**0.84 J**
Planted Rainbows				
Lone Lake*	1.67	U(4.8)	**1.9**	**0.96 J**
Molson Lake	2.05	**8.6**	**5.8**	U(0.49-2.4)
Warden Lake	0.61	U(4.9)	**3.7**	**0.46 J**
Summit Lake*	0.40	**5.0**	**3.2**	**0.56**
Fan Lake	2.66	U(5.0)	**57**	**0.40 J**
South Lewis Co. Park Pond	0.77	**5.9**	**2.4**	**1.33 J**
North Lake	0.65	U(4.9)	**2.9**	**1.23 J**
Chapman Lake	0.44	**11.8**	**5.0**	**1.01 NJ**
Donnie Lake	1.29	U(5.0)	**3.6**	**0.25 J**
Lacamas Lake	1.50	U(5.0)	**3.0**	**0.42 J**

ΣPCB = the sum of detected Aroclors
ΣDDT = the sum of detected 4,4' and 2,4' homologues of DDD, DDE, and DDT
ΣPBDE = the sum of detected PBDE congeners analyzed
Detected concentrations in **bold**
* Samples analyzed in duplicate. Results shown are mean of laboratory analyses.
U - The analyte was not detected at or above the reported result
J - The analyte was positively identified. The associated numerical value is an estimate.
NJ - There is evidence that the analyte is present. The associated numerical result is an estimate.

Lipid catabolism in muscle and consequent contaminant mobilization may explain higher mean ΣPCB in hatchery fish compared to fish from lakes (13 and 3.1 ng/g, respectively; Figure 2). Differences between contaminant concentrations in hatchery and planted rainbow trout appear to be regulated by more than lipid decreases, however. Mean ΣDDT concentrations in tissue were higher in lakes (8.8 ng/g) compared to tissues from hatchery fish (3.9 ng/g), although concentrations were nearly identical when the outlier from Fan Lake was removed.

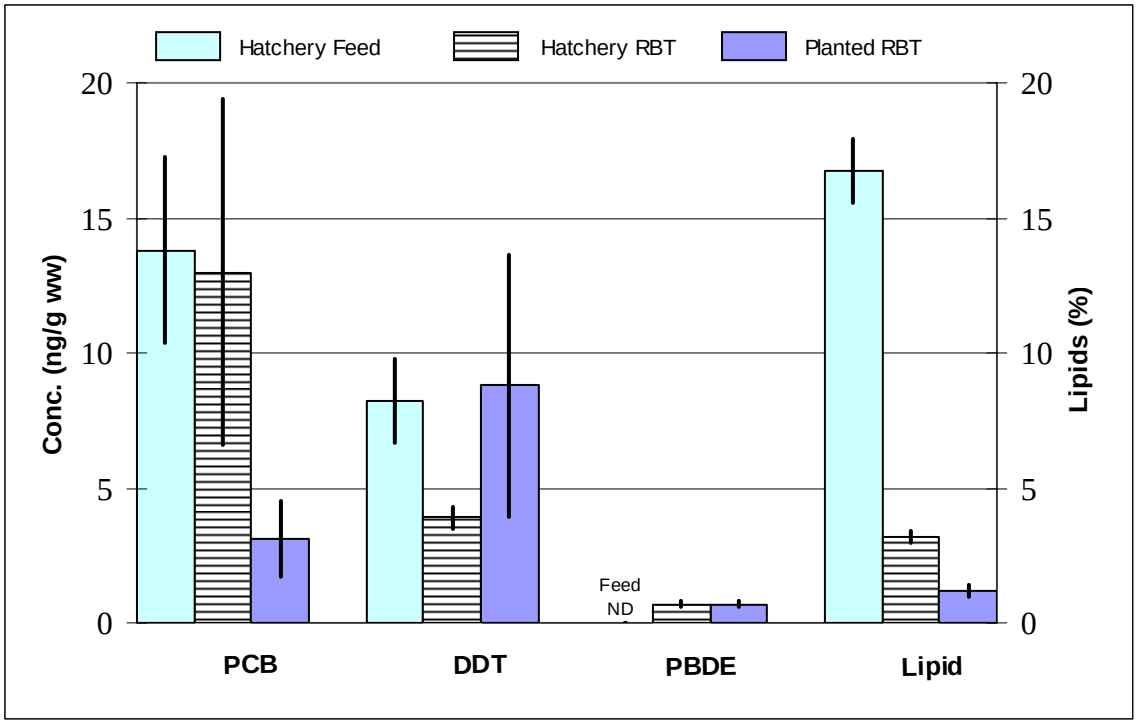

Figure 2. Mean (SE) Concentrations of ΣPCBs, ΣDDT, ΣPBDE, and Percent Lipid in Hatchery Feed, Hatchery Rainbow Trout, and Planted Rainbow Trout.

Neither ΣPCB nor ΣDDT were significantly different in hatchery fish compared to planted fish (Wilcoxon signed-rank test, α=0.05). There was also no significant difference in PBDEs between groups, although this was not surprising since mean concentrations were identical (0.66 ng/g, respectively).

PCBs tended to be highest in hatchery fish whose food had comparatively high PCB concentrations. For example, the Chelan Hatchery had high ΣPCB in both feed and fish, while Ford and Puyallup Hatcheries had no detectable PCBs in either feed or fish. However, this pattern was not true for ΣDDT and ΣPBDE in hatchery samples.

To examine possible relationships among sample types and contaminants, a ranked correlation analysis was performed on major variables in hatchery and lake samples (Table 5). There were no strong correlations between contaminants in hatchery fish and their lake counterparts. With the exception of lipids and ΣDDT in hatchery fish, lipids were not highly correlated with ΣPCB, ΣDDT, or ΣPBDE, a somewhat surprising finding but lending support to the notion that factors other than lipid may be the primary determinants in contaminant residue levels, particularly for DDT compounds and PBDEs.

Table 5. Spearman Ranked Correlation Matrix of Major Variables in Feed and Tissue Samples.

	Hatch. Feed Lipid									
Hatch. Feed ΣPCB	-0.182	Hatch. Feed ΣPCB								
Hatch. Feed ΣDDT	-0.159	0.226	Hatch. Feed ΣDDT							
Hatch. Fish Lipid	-0.139	-0.012	0.256	Hatch. Fish Lipid						
Hatch. Fish ΣPCB	-0.156	**0.753**	0.280	0.356	Hatch. Fish ΣPCB					
Hatch. Fish ΣDDT	-0.333	0.280	0.402	**0.721**	**0.563**	Hatch. Fish ΣDDT				
Hatch. Fish ΣPBDE	-0.365	**0.817**	0.086	0.170	**0.822**	0.426	Hatch. Fish ΣPBDE			
Planted Fish Lipid	0.467	0.030	-0.030	0.042	0.125	0.127	-0.213	Planted Fish Lipid		
Planted Fish ΣPCB	-0.307	**0.753**	0.131	**-0.519**	0.405	-0.171	**0.685**	-0.294	Planted Fish ΣPCB	
Planted Fish ΣDDT	-0.285	-0.109	-0.293	-0.382	0.019	-0.042	0.182	0.176	0.246	Planted Fish ΣDDT
Planted Fish ΣPBDE	0.382	0.103	0.055	-0.103	-0.006	-0.394	0.043	**-0.539**	0.184	**-0.636**

Spearman correlation coefficients ≥|0.500| in **bold**

Dioxins and Furans

Four samples each of hatchery feed and rainbow trout tissue were analyzed for 2,3,7,8-substituted PCDDs and PCDFs (Table 6). Toxic equivalents (TEQs) were calculated using toxic equivalency factors (TEFs) proposed by Van den Berg et al., 1998.

Table 6. TEQ Concentrations (pg/g, ww) in Four Feed and Rainbow Trout Samples and Percent TEQ Contribution by Congener.

	Vancouver	Mossyrock	Ford*	Spokane
Hatchery Feed				
TEQ	0.562	1.226	0.028	1.194
2,3,7,8-TCDD	33%	30%	0%	16%
1,2,3,7,8-PeCDD	38%	44%	0%	53%
1,2,3,4,7,8-HxCDD	0%	0%	0%	0%
1,2,3,6,7,8-HxCDD	4%	4%	0%	4%
1,2,3,7,8,9-HxCDD	0%	3%	0%	3%
1,2,3,4,6,7,8-HpCDD	2%	1%	18%	1%
OCDD	0%	0%	1%	0%
2,3,7,8-TCDF	9%	7%	71%	12%
1,2,3,7,8-PeCDF	2%	2%	10%	1%
2,3,4,7,8-PeCDF	11%	9%	0%	10%
1,2,3,4,7,8-HxCDF	0%	0%	0%	0%
1,2,3,6,7,8-HxCDF	0%	0%	0%	0%
2,3,4,6,7,8-HxCDF	0%	0%	0%	0%
1,2,3,7,8,9-HxCDF	0%	0%	0%	0%
1,2,3,4,6,7,8-HpCDF	0%	0%	0%	0%
1,2,3,4,7,8,9-HpCDF	0%	0%	0%	0%
OCDF	0%	0%	0%	0%
Hatchery Rainbows				
TEQ	0.053	0.041	0.012	0.024
2,3,7,8-TCDD	0%	0%	0%	0%
1,2,3,7,8-PeCDD	0%	0%	0%	0%
1,2,3,4,7,8-HxCDD	0%	0%	0%	0%
1,2,3,6,7,8-HxCDD	0%	0%	0%	0%
1,2,3,7,8,9-HxCDD	0%	0%	0%	0%
1,2,3,4,6,7,8-HpCDD	0%	0%	0%	0%
OCDD	0%	0%	0%	0%
2,3,7,8-TCDF	35%	68%	35%	68%
1,2,3,7,8-PeCDF	11%	17%	0%	0%
2,3,4,7,8-PeCDF	39%	0%	0%	0%
1,2,3,4,7,8-HxCDF	13%	12%	56%	28%
1,2,3,6,7,8-HxCDF	0%	0%	0%	0%
2,3,4,6,7,8-HxCDF	0%	0%	0%	0%
1,2,3,7,8,9-HxCDF	0%	0%	0%	0%
1,2,3,4,6,7,8-HpCDF	2%	2%	9%	3%
1,2,3,4,7,8,9-HpCDF	0%	0%	0%	0%
OCDF	0%	0%	0%	0%

* Samples analyzed in duplicate. Results shown are mean of laboratory analyses.

TEQs were much higher in hatchery feed compared to rainbow trout, with the exception of Ford Hatchery feed which had a TEQ up to 40 times lower than the other hatcheries. Like other contaminants, it appears that TEQ concentrations were not determined primarily by lipid content. Congeners contributing to TEQ were markedly different between high and low TEQ samples, with tetra- and penta-substituted dioxin congeners providing approximately 70% of the toxicity. In contrast, the Ford feed sample had none of these congeners detected.

Congener patterns were even more distinct when feed and fish tissue samples were compared. None of the tissue samples contained detectable concentrations of PCDDs. Most of the toxicity in tissue samples was derived from tetra-, penta-, and hexa-substituted furan congeners.

Other Chlorinated Pesticides

In addition to DDT compounds, nine chlorinated pesticides or breakdown products were detected in feed samples, six were detected in hatchery rainbow trout, and four were found in trout collected from lakes (Figure 3). DDMU, like its parent DDT, was detected in all hatchery feed samples. Although DDMU was much higher in Fan Lake trout (4.4 ng/g) compared to other lakes, feed from the originating hatchery (Ford) had the lowest concentration (0.2 ng/g), and Ford Hatchery rainbow trout did not have a detectable DDMU concentration.

Dieldrin had an unusual detection pattern among samples; it was detected in only 10% (Mossyrock, one of ten) feed samples, and 20% of lake fish tissue samples, but was found in 70% of the hatchery tissue samples. It should be noted, however, that detection limits for dieldrin in feed samples were approximately three-to-five times higher than in tissue samples (see Appendix C). Hexachlorobenzene was found in only one sample from planted rainbow trout (Summit Lake, 0.7 ng/g) but not in fish from the original population at Eells Springs Hatchery. The comparatively high concentration and lack of apparent link to the hatchery suggests these fish may have accumulated hexachlorobenzene from a local source in Summit Lake.

Pentachloroanisole, a degradation product of pentachlorophenol, was the only additional compound detected in planted rainbow tissue. Concentration of pentachloroanisole doubled in Lacamas Lake fish compared with the original hatchery population, suggesting a possible local source. Tucannon hatchery rainbows also apparently accumulated pentachloroanisole from the hatchery feed, although residues in the hatchery fish did not persist while the fish were in a (Donnie) lake environment.

Nonachlor and cis-chlordane, two components of commercial-grade chlordane, were found in at least half of the hatchery feed and fish samples. Another chlordane component, trans-chlordane, was found in 40% of feed samples. Two other chlorinated pesticides, toxaphene and methoxychlor, were found in 30% and 20% of feed samples, respectively.

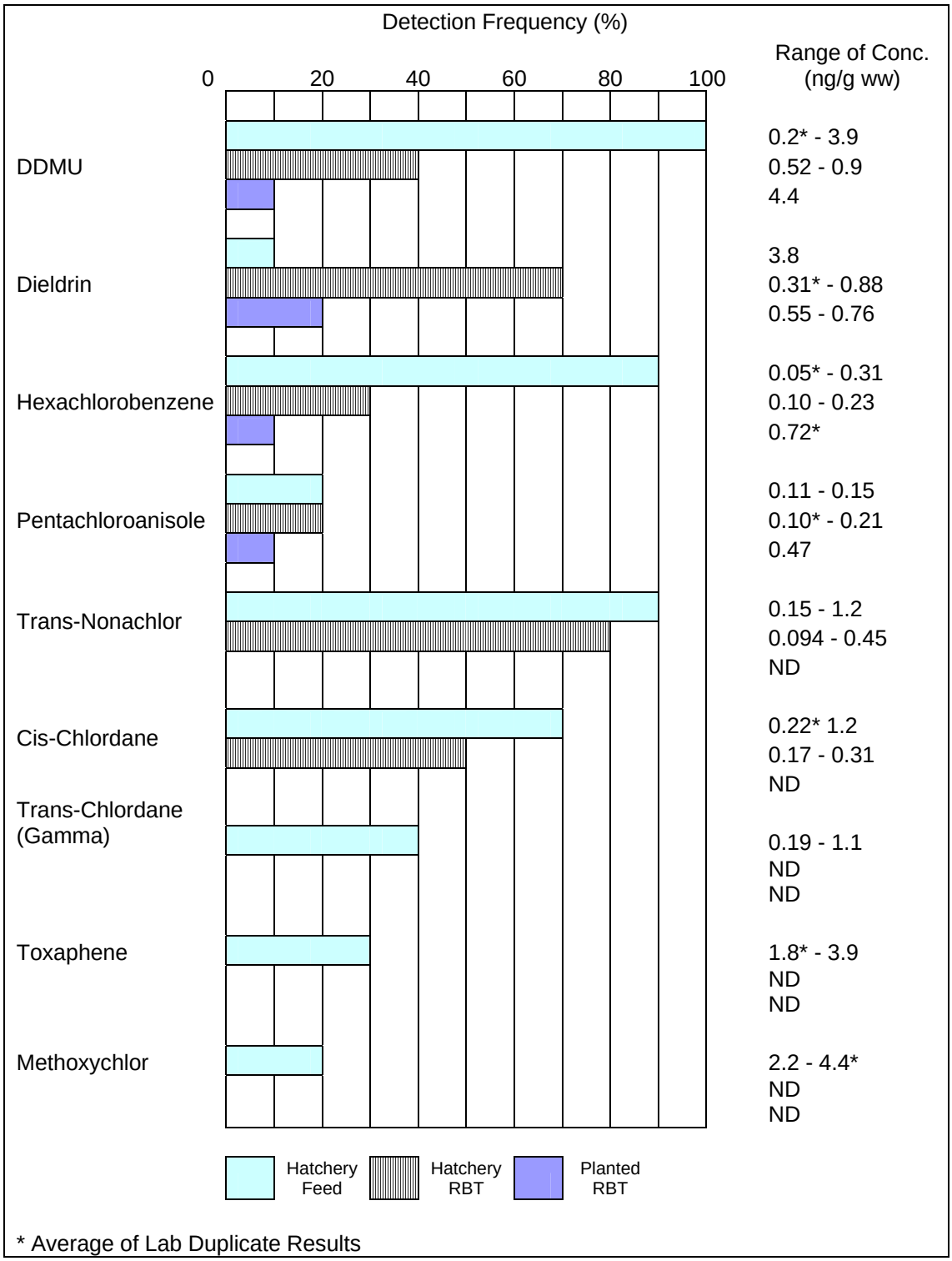

Figure 3. Detection Frequency of Chlorinated Pesticides (Excluding DDT Compounds) and Range of Detected Concentrations.

Discussion

Feed Ingredients and Residue Tolerances

The four brands of feed analyzed during the present 2005 survey have a wide range of contaminant levels. Rainbow trout collected at hatcheries gained approximately 50-80% of their final weight on these feeds, and may have gained most of their initial weight on starter feeds with similar ingredients.

Feed sack labels obtained at the time of sampling indicated that minimum amounts of crude protein (40-45%) were similar among feeds, as were maximum amounts of crude fiber (1.5-5%) and ash (9-12%). Crude fat was the other major component, constituting 10-24% of the weight which generally showed good agreement with lipid analysis in the present study (r^2=0.64).

In terms of contaminant residues, the origin of the lipids in the feed is probably a much greater concern than their percent by weight. All of the feeds sampled advertise fish oil as a major ingredient which is likely to be the major source of PCBs, chlorinated pesticides, PBDEs, PCDD/Fs, and other lipophilic contaminants (Jacobs et al., 2002).

The exact source of the fish oil and fish meal, as well as other major ingredients in hatchery feed (wheat flour, soybean oil, blood meal, feather meal, and poultry by-product), may change from batch to batch, depending on the availability and cost of the raw products. Fishmeal may derive largely from anchovy, menhaden, capelin, and herring, and fish oil is a by-product of the fishmeal manufacturing industry.

The negative correlation between lipids and major contaminants in feed is another indication that the source of fish oil is probably a more important determinant in contaminant residues than lipid content by weight. However, contaminant concentrations in feed apparently play a large role in accumulation by fish, probably due to the high rate of weight conversion from feed to fish (>80%). This is demonstrated by the strong correlation between ΣPCB in hatchery feed and hatchery fish (r=0.75), and a weaker yet positive correlation for ΣDDT (r=0.40).

In the case of the Tucannon hatchery, where the fish are raised in water from the Tucannon River for six months prior to stocking, fish weight gain actually exceeded the weight of feed provided (conversion of 163%) due to availability of natural prey items.

The only domestic regulation concerning POP contaminants is a federal PCB residue tolerance of 2,000 ng/g for feed components of animal origin (21 CFR 109.30). The Canadian Food Inspection Agency has an “actionable level” of 2,000 ng/g for PCBs, but it only applies to fish oil destined for animal feed (CFIA, 2003). In 2001, the European Union (EU) established maximum tolerable levels of PCDD/F TEQs in fish in various fish products and feeds. The maximum levels for fish feed and fish oil for use other than direct human consumption is 2.25 and 6.0 pg TEQ/g, respectively (Directive 2001/102/EC).

Table 7 shows a summary of major contaminants in hatchery feeds and commercial aquaculture feeds for salmonids. Care should be used interpreting the available data since information on the intended species is often not included. For instance, feed for commercial salmon aquaculture typically contains higher fish oil content than trout feed, and some species, such as catfish and tilapia, may be fed entirely vegetable-based feeds.

Table 7. Contaminant Concentrations in Various Hatchery and Commercial Aquaculture Feeds and Applicable Regulations (mean concentrations unless otherwise noted).

Source	ΣPCB (ng/g ww)	ΣDDT (ng/g ww)	ΣPBDE (ng/g ww)	Dioxin TEQ (pg/g ww)	Lipid (%)	Ref.
Contaminant Levels in Feed						
Feeds from 6 manufacturers used in 11 USF&WS National Fish Hatcheries	1.94*	11.33	nr	0.227	16.7	Maule et al., 2006
Mean of 8 commercial salmon aquaculture feeds from Scottish sources	105	12.1	5.1	nr	28.8	Jacobs et al., 2002
Mean of fish feed/fishmeal samples originating from Canada (n=14), U.S. (n=7), Iceland (n=1), Peru (n=1), and Russia (n=1)	Can.–30.7† U.S.–16.5† Ice.–12.1† Peru–0.6† Rus.–12.7†	Can.–21.1 U.S.–23.3 Ice.– <7 Peru–nr Rus.–nr	nr	Can.–1.0 U.S.–1.1 Ice.–0.23 Peru–ND Rus.–0.22	nr	CFIA, 2006
Feed from the 2 largest global suppliers for commercial salmon aquaculture; 9 samples from North and South America and 4 samples from Europe	15 (approx. Amer. median) 60 (approx. Eur. median)	nr	nr	1 (approx. Amer. median) 4 (approx. Eur. median)	nr	Hites et al., 2004
Confidential	nr	nr	nr	1.21	nr	Hermann et al., 2004
WDFW trout hatchery feed	*13.8*	*8.2*	*ND (<0.25)*	*0.75*	*16.8*	*present study, 2005*
Regulatory Levels in Feed						
U.S. Food and Drug Admin.	2,000	ne	ne	ne	ne	21 CFR 109.30(a)(6)
European Union	ne	ne	ne	2.25	ne	Directive 2001/102/EC 21

*sum of 14 dioxin-like congeners
†sum of 72 congeners
nr - not reported
ND – not detected
ne - not established

Maule et al. (2006) analyzed numerous batches of feeds used at eleven U.S. Fish and Wildlife Service (USF&WS) National Fish Hatcheries. They found a lower mean dioxin TEQ and ΣPCB than reported here, although the ΣPCB was derived from only 14 dioxin-like PCB congeners. Interestingly, the bulk of the dioxin TEQ was derived from 2,3,7,8-TCDD and 1,2,3,7,8-PeCDD similar to the feed samples analyzed for the present survey. ΣDDT and percent lipid contents were similar between the two studies.

Comparison of contaminants in feed and salmon fillet from commercial aquaculture operations showed ΣPCB, ΣDDT, and ΣPBDE higher in feed compared to fish tissue at similar proportions to those reported here (Jacobs et al., 2002). Although the mean ΣDDT was similar to results for the present study, ΣPCB was an order of magnitude higher. The feeds, which were from Scottish sources, had much higher lipids (mean of 28.8%) than found here (mean of 16.8%). Hites et al. (2004) reported ΣPCB and dioxin TEQ levels approximately four-fold higher in commercial salmon aquaculture feeds from Europe compared to North and South America. Like other results, the Hites et al. (2004) study found POP concentrations slightly higher in feed compared to fish, on average, but lipid concentrations were not given.

The Canadian Food Inspection Agency (CFIA) analyzed 24 samples originating primarily from Canada and the U.S., but also from Iceland, Peru, and Russia, although little additional information is provided about the samples (CFIA, 2006). ΣPCB for U.S., Icelandic, and Russian samples were similar to those reported here, while Canadian feed had about twice the concentration on average. DDT levels were about twice the levels reported in other samples, while the dioxin TEQs tended to be within ranges reported by other investigators.

The only reported PBDE analysis of feed was done in the Jacobs et al. (2002) study which found ΣPBDE averaging 20 times the analytical reporting limits for the present study. PBDE-47 was the predominant congener found and, although not found in WDFW hatchery feeds, it was the major congener in rainbow trout from hatcheries and lakes and is one of the most abundant congeners found in Washington freshwater fish (Johnson and Olson, 2001).

None of the average POP concentrations reported in these studies violated residue tolerances from the applicable regulations, with the exception of the European median dioxin TEQ reported by Hites et al. (2004). A study by Herrmann et al. (2004), designed specifically to look at compliance with EU standards, found a mean concentration of dioxin TEQ about one-half the EU residue tolerance. However, 95th percentile values for both fish feed (2.71 pg/g) and fish oil (6.30 pg/g) exceeded the dioxin TEQ standards.

Depuration/Uptake of Contaminants in Lakes

One of the underlying goals of this study was to determine if lakes act as purifying environments or if hatchery fish stocked in lakes accumulate additional contaminants. Cursory inspection of the data suggests depuration occurs for PCBs and chlorinated pesticides other than DDT compounds, and no differences are seen for ΣDDT (with the Fan Lake outlier removed) and ΣPBDE. Although pairwise comparisons show no statistical differences between ΣPCB in hatchery and lake rainbow trout, mean ΣPCB in hatchery fish are substantially higher, even when the outlier from Chelan Hatchery (67 ng/g) is removed.

Part of the decrease in ΣPCB may be the significant decrease in lipid content of the planted fish. It is a well-known fact that PCB concentrations are often positively correlated with lipid in tissue. Lipid metabolism, and in particular lipid catabolysis, to meet energy requirements may mobilize lipids from muscle to high lipid organs such as the liver, kidney, and brain, thus decreasing muscle PCB concentrations (Jørgensen et al., 2002). Gamete production is another mechanism for transfer of PCBs due to loss of muscle lipids, although it is unlikely that the fish examined had reached sexual maturity.

As mentioned previously, the absence of a strong correlation between lipids and ΣPCB suggests PCB concentrations are controlled by factors other than the percentage of lipid. One possible explanation for altered PCB concentrations is that some lakes allow PCBs to be shed from muscle tissue while others increase the PCB accumulated by fish. Molson, Warden, South Lewis County Park, and Lone lakes received fish with the highest ΣPCB, and all four lakes had the greatest depuration. Increases in ΣPCB only occurred where concentrations were low-to-moderate in hatchery fish (with the exception of the very small increase in Spokane Hatchery-Chapman Lake fish), suggesting net accumulation only occurs when PCB concentrations in tissue are initially low.

ΣDDT concentrations were low in muscle tissue of hatchery rainbow trout precluding an opportunity to observe substantial depuration. As many lakes had increased levels of ΣDDT as had decreases, all small changes with the exception of Fan Lake.

Fan Lake is a medium-sized (32 hectare) lake approximately 20 miles north of Spokane, situated in a small drainage basin (1,600 hectares). It is one of the lowermost in a chain of lakes along the West Branch of the Little Spokane River. Since the Fan Lake basin is relatively undeveloped and the lake is connected to other lakes only through its outlet, it appears unlikely that high ΣDDT accumulating in Fan Lake rainbow trout is a result of basin-wide or area-wide contamination.

Comparison of Tissue Concentrations to Applicable Criteria

Criteria to protect human health from harmful pollutants in ingested water and fish were issued to Washington State in EPA's 1992 National Toxics Rule (NTR, 40 CFR 131.36). The human health-based criteria, if met, will generally ensure that public health concerns do not arise, and that fish advisories are not needed. Sampling of either water or edible fish tissue may be conducted to assess compliance with the NTR criteria (Ecology, 1992), but tissue is generally

preferred because POPs are often found at concentrations in water below reasonably available laboratory detection limits.

It should be recognized that POPs exceeding the NTR criteria in fish tissue do not necessarily signal the need for a fish consumption advisory, nor does it imply the existence of a public health concern. Assessment of risks to the fishing public and consumption advice is carried out by the Washington State Department of Health (DOH), often based on data collected by Ecology and WDFW. While DOH supports Ecology's use of the NTR criteria for identifying potentially contaminated waters and for developing source controls to keep water quality at or below criteria, it does not use the NTR criteria to establish fish advisories. Instead, DOH evaluates contaminants in fish tissue using established risk assessment paradigms. These include tools for:

1. Analysis of risks – calculating allowable meal limits based on known contaminant concentrations, estimates of exposure in specific groups or populations.
2. Risk management – e.g., reduction in contaminants through preparation and cooking techniques, known health benefits from fish consumption, contaminant concentrations or health risks associated with replacement foods, and cultural importance of fish.
3. Risk communication – the outreach component of a fish advisory or a conclusion that an advisory is unnecessary.

Table 8 shows NTR criteria for chemicals analyzed in the present 2005 survey compared to concentrations in rainbow trout. NTR criteria have not been established either for PBDEs or for some of the chlorinated pesticides analyzed here.

There are 15 instances where contaminants in rainbow trout exceed NTR criteria. Most of the exceedances are for ΣPCB (three lakes and six hatcheries), followed by dieldrin (one lake and four hatcheries) and 4,4'-DDE (one lake). In all, seven of the eleven hatchery samples (including Troutlodge) exceeded NTR criteria for at least one chemical, and five of the ten planted fish samples exceeded the criteria.

When criteria are not met, the waterbody is considered impaired and placed on the federal Clean Water Act section 303(d) list and may require a Total Maximum Daily Load (TMDL) study to address the impairment. TMDLs identify the sources of a pollutant and allocate pollutant loads among sources in order to bring the waterbody in compliance with standards.

One of the questions this study sought to answer is whether contaminants accumulated by fish in hatcheries could contribute, in whole or in part, to the water quality impairment of a waterbody and its consequent addition to the 303(d) list. This question was not intended to be directed at the lakes analyzed in the present survey. Instead, at issue is whether hatchery-derived contamination is worthy of inquiry by investigators who are conducting surveys on individual lakes and streams where the source of contamination is uncertain and where waterbodies may be considered for 303(d) listing.

Table 8. National Toxics Rule Criteria Compared to Contaminant Residues in Hatchery and Planted Rainbow Trout Fillet (ng/g ww except pg/g ww for 2,3,7,8-TCDD).

Contaminant	Criterion	Concentrations in present study	No. of Criterion Exceedances	Location(s) of Criterion Exceedance
ΣPCB	5.3	4.85 - 67	9	Chapman Lake Molson Lake South Lewis Co. Park Pond Chelan Hatchery Columbia Basin Hatchery Mossyrock Hatchery Troutlodge Arlington Hatchery Spokane Hatchery
4,4'-DDT	32	0.14 - 1.9		
4,4'-DDE	32	1.9 - 45	1	Fan Lake
4,4'-DDD	45	0.29 - 9.6		
2,3,7,8-TCDD	0.07	0.011 - 0.053		
Aldrin	0.65	ND		
Dieldrin	0.65	0.32 - 0.88	5	Warden Lake Mossyrock Hatchery Spokane Hatchery Arlington Hatchery Eells Springs Hatchery
Endrin	3,216	ND		
Endrin Aldehyde	3,216	ND		
alpha-BHC	1.7	ND		
beta-BHC	1.6	ND		
gamma-BHC (Lindane)	8.2	ND		
Chlordane (total)	8.3	ND		
Endosulfan I	540	ND		
Endosulfan II	540	ND		
Endosulfan Sulfate	540	ND		
Heptachlor	2.4	ND		
Heptachlor Epoxide	1.2	ND		
Hexachlorobenzene	6.7	0.10 - 0.73		
Toxaphene	9.8	ND		

ND - not detected

To examine this question on a statewide basis, results of the present survey were compared to all of the data on contaminants in tissue in Washington. In order to provide a suitably comparable data set, data that met the following conditions were extracted from Ecology's Environmental Information Management (EIM) database; 1) samples analyzed since 1998, 2) results only for rainbow trout, brown trout, and cutthroat trout since these are the primary resident species raised to catchable size at WDFW hatcheries, and 3) fillet data only. Only results for ΣPCB, 4,4'-DDE, and dieldrin were selected since these were the contaminants found here that exceeded NTR criteria. Waterbodies known to have large sources of these chemicals (e.g., Spokane River for PCBs, Yakima River for 4,4'-DDE and dieldrin) were removed.

Figure 4 shows the cumulative distribution of all ΣPCB data for fish in Washington using the selection process previously described. Sixty percent of the 46 samples exceed the NTR criterion, and approximately 50% of samples statewide had concentrations that fall into the range of detectable ΣPCB concentrations in hatchery rainbow trout, excluding the Chelan Hatchery sample. One hundred percent of samples fall into the hatchery ΣPCB range when all of the hatchery rainbows are considered.

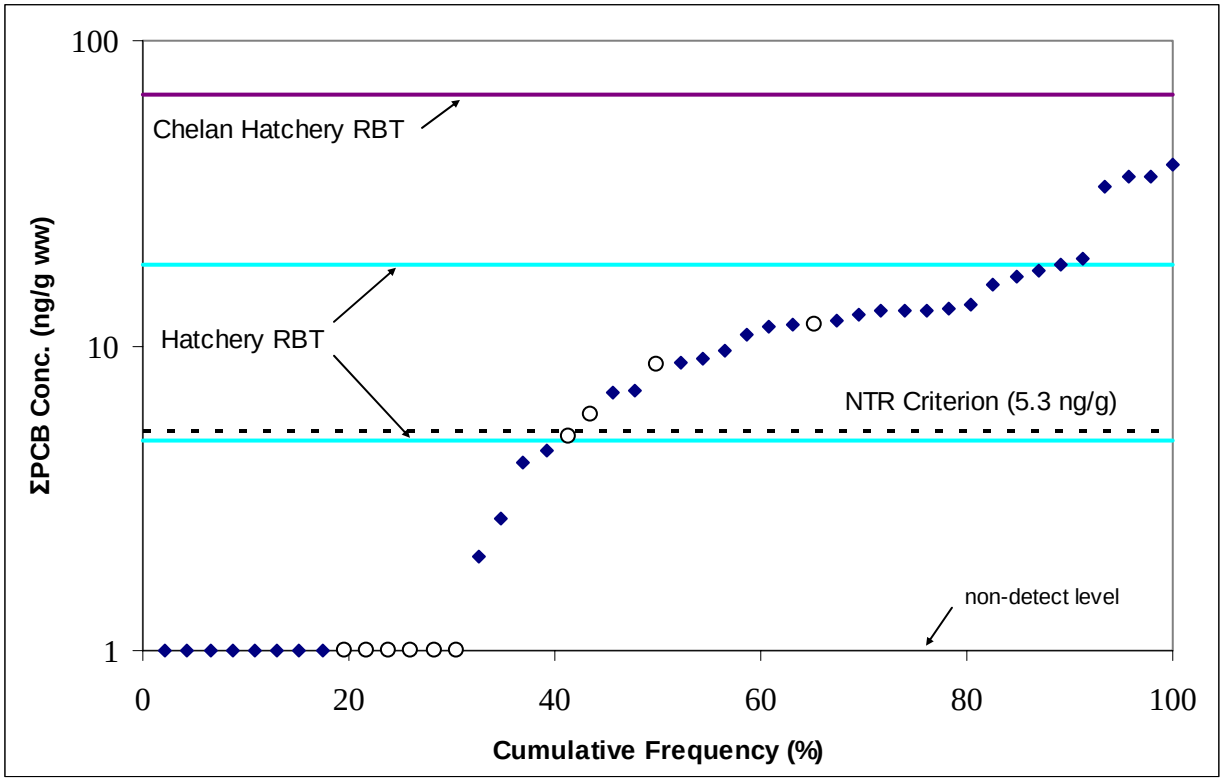

Figure 4. Cumulative Frequency Distribution of ΣPCB Concentrations in Fillet Tissue of Rainbow Trout (RBT), Cutthroat Trout, and Brown Trout from Washington Lakes and Streams. (Open circles represent data from lakes sampled in the present survey. Solid gray lines bound the range of detected ΣPCB concentrations in hatchery fish. Dashed line is the NTR criterion for ΣPCBs.)

Only two of the 38 trout samples (5%) exceed the NTR criterion for 4,4'-DDE, including the Fan Lake result found in the present survey (Figure 5). Approximately 40% of the samples are within the hatchery fish range, with more than 70% falling below the maximum 4,4'-DDE level in hatchery rainbows.

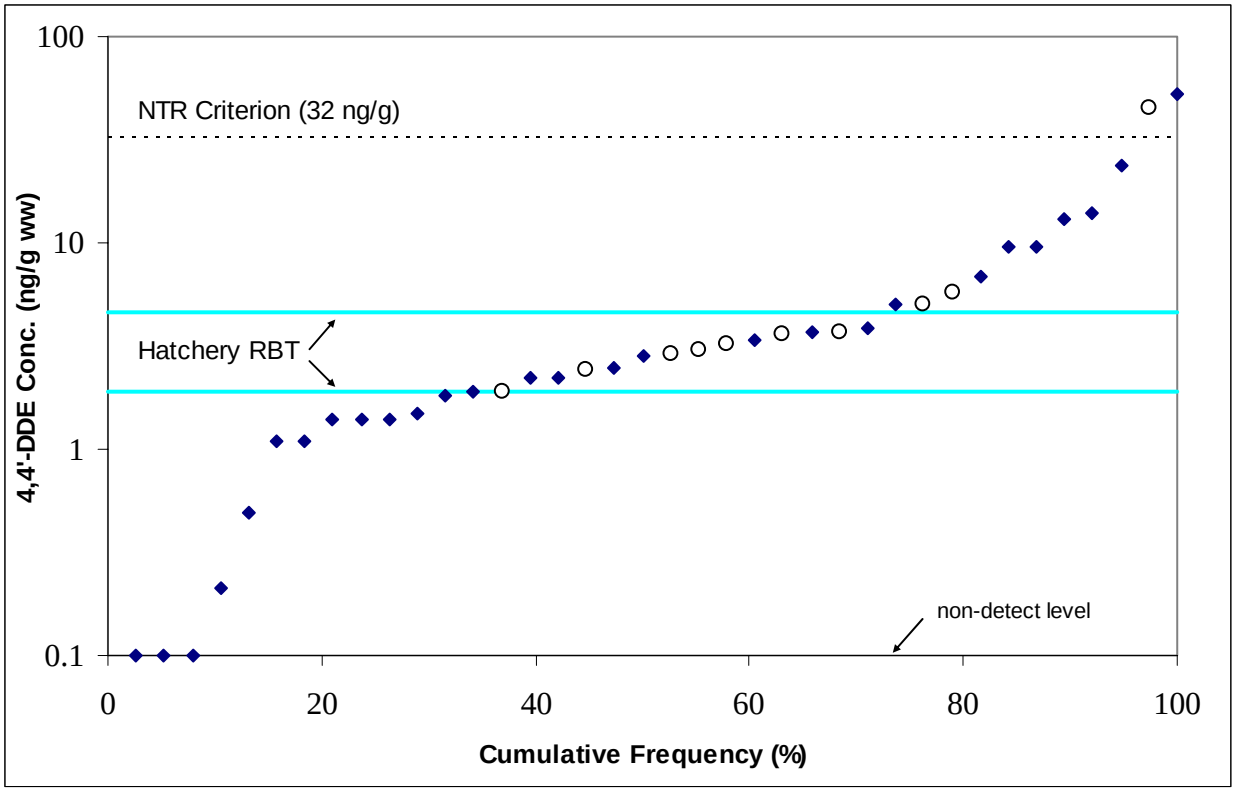

Figure 5. Cumulative Frequency Distribution of 4,4'-DDE Concentrations in Fillet Tissue of Rainbow Trout (RBT), Cutthroat Trout, and Brown Trout from Washington Lakes and Streams. (Open circles represent data from lakes sampled in the present survey. Solid gray lines bound the range of detected 4,4'-DDE concentrations in hatchery fish. Dashed line is the NTR criterion for 4,4'-DDE.)

None of the 38 samples screened from the EIM database had detectable levels of dieldrin; North Lake and Warden Lake are the only "unpolluted" sites where dieldrin has been detected in common trout species (Figure 6). Dieldrin in Warden Lake rainbow trout (0.76 ng/g) exceeds the NTR criterion (0.65 ng/g).

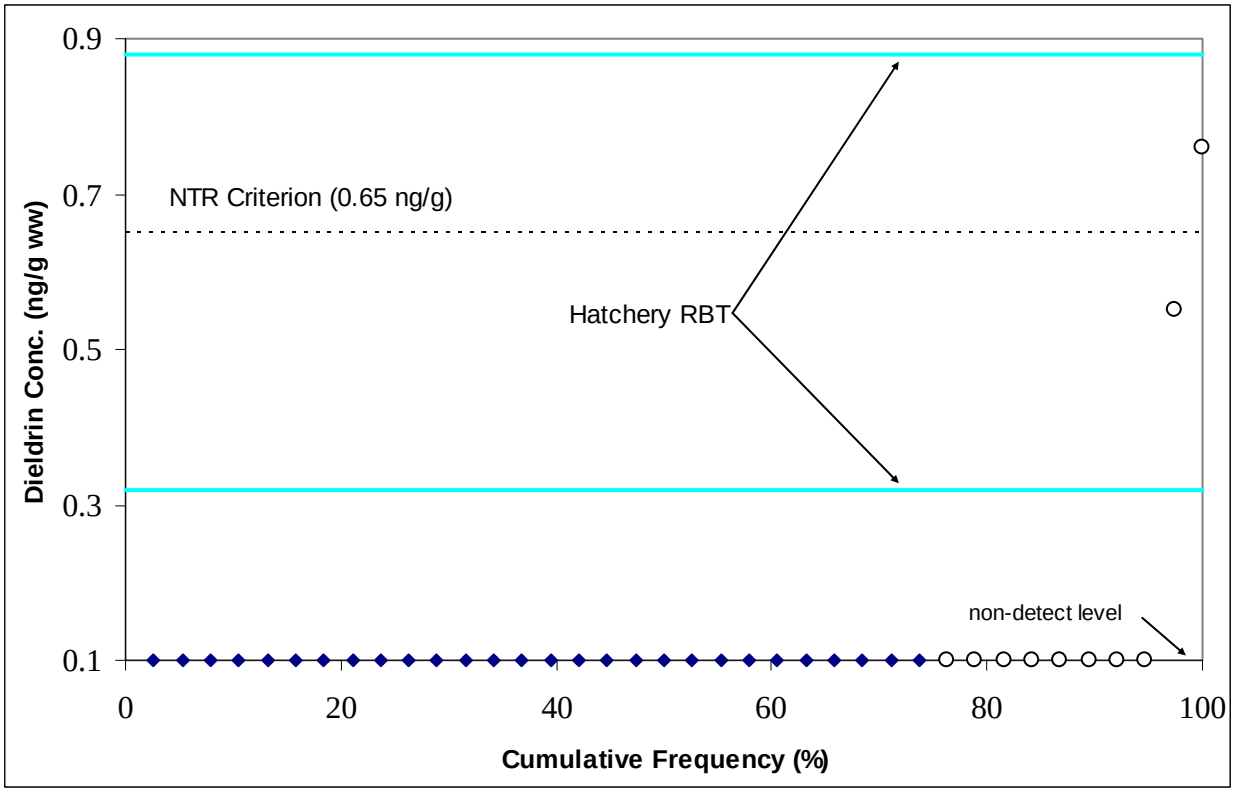

Figure 6. Cumulative Frequency Distribution of Dieldrin Concentrations in Fillet Tissue of Rainbow Trout (RBT), Cutthroat Trout, and Brown Trout from Washington Lakes and Streams. (Open circles represent data from lakes sampled in the present survey. Solid gray lines bound the range of detected dieldrin concentrations in hatchery fish. Dashed line is the NTR criterion for dieldrin.)

There are several inferences that may be drawn from these comparisons to screened data. One supposition is that some portion of POPs found in trout is derived from burdens obtained at a hatchery, although this presumes that the fish were planted. However, there is no concomitant information on the origin of the fish used for these comparisons.

The extent of hatchery-derived POPs remaining in planted fish is variable and appears to depend largely on the amount of time lapsed since stocking. Trout caught around the time of opening day – typically mid-to-late April – will have contaminant concentrations nearly identical to concentrations found in fish just prior to their removal from hatcheries. It appears that as the fishing season progresses, the concentrations in fillet tissue generally decrease, particularly for PCBs and some chlorinated pesticides, although this is a pattern which is less consistent for DDT compounds and PBDEs. Unfortunately, the planted cohorts that were sampled for each hatchery-planted pair could not be sampled further to track fillet concentrations during subsequent periods.

One of the implications of these results, particularly from the practical standpoint of a regulatory agency, is that waterbodies may be included on the 303(d) list due to contamination stemming from hatcheries. Taken further, 303(d) listed waters often require a TMDL to assess contaminant sources. Sources considered for TMDLs are typically point sources (e.g., piped effluent) and nonpoint sources (e.g., agricultural and urban runoff, atmospheric deposition) which normally occur in the vicinity of the impaired waterbody. However, no known TMDLs in Washington have included hatchery fish as a contaminant source. For PCBs, and to a lesser extent dieldrin, hatchery fish may contribute to impairment and, in some cases, may cause the bulk of impairment. Therefore, TMDL investigators may want to consider including hatchery fish as contaminant sources among other sources.

Conclusions

Rainbow trout acquire low-to moderate concentrations of persistent organic pollutants (POPs) while residing at Washington Department of Fish and Wildlife (WDFW) hatcheries. Feed used to raise rainbow trout to catchable size (≥6") contains concentrations of PCBs, DDT compounds, PCDD/Fs, and several additional chlorinated pesticides at higher wet-weight concentrations than in fillet tissue of the fish specimens analyzed, suggesting that the POP accumulation pathway is primarily through the feed. This is consistent with findings of other aquacultural studies, although other possible pathways of contaminant accumulation were not examined for this study. Low levels of PBDEs were also present in rainbow trout tissue, but were not detected in trout feed.

Fish feed is high in lipids and shows variable amounts of contaminants, but POP concentrations were not correlated with the percent lipid in feed samples. This suggests the source of lipids, largely derived from marine oil, is an important determinant in POP concentrations. The positive correlation between feed and fish ΣPCB and between feed and fish ΣDDT supports the conclusion that feed is the primary contaminant source to hatchery fish. WDFW trout hatchery feed has POP concentrations similar or lower than feeds analyzed in other studies, and appears to have much lower PCB concentrations than feeds used in commercial salmon aquaculture.

It appears that fillet tissue concentrations of some POPs, particularly PCBs, decrease following stocking in lakes, although this finding is inconclusive. ΣDDT concentrations may increase in the lake environment even as fillet lipid concentrations decrease significantly. Fish from only one location – Fan Lake in Pend Oreille County – showed a substantial increase in contaminants (DDT compounds) following residence in the wild.

In the 21 rainbow trout fillet samples analyzed (11 from hatcheries including Troutlodge and 10 from lakes), there are 15 instances where contaminants exceed (do not meet) regulatory criteria. Most of the exceedances are for ΣPCB (three lakes and six hatcheries), followed by dieldrin (one lake and four hatcheries) and 4,4'-DDE (one lake). Considering the POP levels in catchable rainbow trout just prior to planting, it appears likely that at least part of the contaminant burden is hatchery-derived, with the notable exception of DDT compounds in Fan Lake as described previously.

Based on comparisons between waterbodies in Washington State and POP data reported here, it is possible that trout caught in "unpolluted" lakes and streams contain contaminants originating from WDFW hatcheries. It is also possible that some listings for impaired waters, particularly listings for PCBs, may be due to hatchery-contaminated fish. Therefore, Total Maximum Daily Load (TMDL) project managers may want to consider hatchery fish as a source of contaminant loads.

Recommendations

Based on results of this 2005 study, it is recommended that fish feed and trout fillet tissue sampling be expanded to include all 26 WDFW hatcheries raising catchable trout. Samples should be analyzed for the persistent organic pollutants (POPs) in the present study, with PCDD/F analysis included for all samples. Water in hatcheries should also be sampled where contaminant levels in fish are exceptionally high. Any water sampling should be performed using semi-permeable membrane devices or other methods to achieve low detection limits for POPs.

More data are needed to assess depuration or accumulation of contaminants in catchable trout following planting in lakes. Ideally, fish could be sampled during several periods to better track trends in contaminant levels over time. Whole fish analysis should also be considered along with fillet sampling, to determine if contaminant burdens are conserved in fish following mobilization of lipids in muscle tissue.

A review of the current 303(d) list should be conducted to identify cases where tissue data used to assess impairment may have come from WDFW catchable trout plants. TMDL project managers should consider the implications of hatchery fish as a possible source of contaminants to waterbodies being assessed.

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Appendices

Appendix A – Glossary of Acronyms and Units

303(d) – Section 303(d) of the federal Clean Water Act
CFIA – Canadian Food Inspection Agency
DDD – 1,1-dichloro-2,2-bis(*p*-chlorophenyl)ethane
DDE – 1,1-dichloro-2,2-bis(*p*-chlorophenyl)ethylene
DDT – 1,1,1-trichloro-2,2-bis(*p*-chlorophenyl)ethane
DOH – Washington State Department of Health
ECD – electron capture detector
Ecology – Washington State Department of Ecology
EIM – Environmental Information Management
EPA – U.S. Environmental Protection Agency
EU – European Union
GC – gas chromatography
MEL – Manchester Environmental Laboratory
MS – mass spectrometry
NIST – National Institute of Standards and Technology
NTR – National Toxics Rule
PBDE – polybrominated diphenyl ether
PCB – polychlorinated biphenyl
PCDD – polychlorinated dibenzo-*p*-dioxin
PCDF – polychlorinated dibenzofuran
POP – persistent organic pollutant
SRM – standard reference material
TEF – toxic equivalency factor
TEQ – toxic equivalent
TMDL – total maximum daily load (water cleanup plan)
USF&WS – U.S. Fish and Wildlife Service
WDFW – Washington Department of Fish and Wildlife
ww – wet weight
Σ – sum of

Units of measurement

ng/g – nanograms per gram (parts per billion)
pg/g – picograms per gram (parts per trillion)

Appendix B – Target Analytes and Reporting Limits

Table B. Target Analytes and Reporting Limits

Analyte	Reporting Limit (ng/g ww)	Analyte	Reporting Limit (ng/g ww)	Analyte	Reporting Limit (pg/g ww)
PCBs		**Chlorinated Pesticides**		**PCDDs/PCDFs**	
Aroclor-1016	2.3 – 5.0	2,4'-DDE	0.47 – 1.0	2,3,7,8-TCDD	0.03 – 0.05
Aroclor-1221	2.3 – 5.0	2,4'-DDD	0.47 – 1.0	1,2,3,7,8-PeCDD	0.07 – 0.11
Aroclor-1232	2.3 – 5.0	2,4'-DDT	0.47 – 1.0	1,2,3,4,7,8-HxCDD	0.05 – 0.2
Aroclor-1242	2.3 – 5.0	4,4'-DDT	0.47 – 1.0	1,2,3,6,7,8-HxCDD	0.05 – 0.2
Aroclor-1248	2.3 – 5.0	4,4'-DDE	0.47 – 1.0	1,2,3,7,8,9-HxCDD	0.05 – 0.2
Aroclor-1254	2.3 – 5.0	4,4'-DDD	0.47 – 1.0	1,2,3,4,6,7,8-HpCDD	0.05 – 0.2
Aroclor-1260	2.3 – 5.0	DDMU	0.47 – 1.0	OCDD	0.36
PBDEs		Aldrin	0.47 – 1.0	2,3,7,8-TCDF	0.03 – 0.05
		Dieldrin	0.47 – 10	1,2,3,7,8-PeCDF	0.06 – 0.1
PBDE-47	0.23 – 0.65	Endrin	0.96 – 10	2,3,4,7,8-PeCDF	0.04 – 0.12
PBDE-66	0.25 – 0.50	Endrin Aldehyde	0.96 – 10	1,2,3,4,7,8-HxCDF	0.2
PBDE-71	0.25 – 0.50	Endrin Ketone	0.96 – 10	1,2,3,6,7,8-HxCDF	0.05 – 0.1
PBDE-99	0.25 – 0.50	alpha-BHC	0.47 – 1.0	2,3,4,6,7,8-HxCDF	0.05 – 0.2
PBDE-100	0.25 – 0.50	beta-BHC	0.47 – 1.0	1,2,3,7,8,9-HxCDF	0.05 – 0.2
PBDE-138	0.25 – 0.50	gamma-BHC (Lindane)	0.47 – 1.0	1,2,3,4,6,7,8-HpCDF	0.05 – 0.2
PBDE-153	0.25 – 0.50	delta-BHC	0.47 – 1.0	1,2,3,4,7,8,9-HpCDF	0.05 – 0.2
PBDE-154	0.25 – 0.50	cis-Chlordane (alpha)	0.47 – 1.0	OCDF	0.21 – 0.23
PBDE-183	0.25 – 0.50	trans-Chlordane (gamma)	0.47 – 1.0		
PBDE-190	0.25 – 0.50	Oxychlordane	0.47 – 1.0		
PBDE-209	1.2 – 6.2	Chlordane (technical)	0.98 – 5.0		
		Chlorpyriphos	0.96 – 10		
		Dacthal (DCPA)	0.96 – 10		
		Endosulfan I	0.96 – 10		
		Endosulfan II	0.96 – 10		
		Endosulfan Sulfate	0.96 – 10		
		Heptachlor	0.47 – 1.0		
		Heptachlor Epoxide	0.96 – 10		
		Hexachlorobenzene	0.47 – 1.0		
		Methoxychlor	0.47 – 1.0		
		Mirex	0.47 – 1.0		
		cis-Nonachlor	0.47 – 1.0		
		trans-Nonachlor	0.47 – 1.0		
		Pentachloroanisole	0.47 – 1.0		
		Toxaphene	0.98 – 10		

Appendix C – Complete Results of Lipid and Contaminant Analysis

Table C-1. Complete Results of Percent Lipids and PCB Aroclor Analysis of Feed and Fish Tissue Samples (ng/g ww)

	Sample Number	Lipids (%)	PCB-aroclor 1016		PCB-aroclor 1221		PCB-aroclor 1232		PCB-aroclor 1242		PCB-aroclor 1248		PCB-aroclor 1254		PCB-aroclor 1260		PCB-aroclor 1262		PCB-aroclor 1268	
Hatchery Feed																				
Arlington	5144102	18.19	2.5	UJ	2.5	UJ	2.5	UJ	2.7	J	5.5	UJ	8.1	J	3	J	2.5	UJ	2.5	UJ
Chelan	5144096	13.75	2.5	U	2.5	U	2.5	U	5	UJ	8	NJ	18		8.8		2.5	U	2.5	U
Columbia Basin	5144098	14.47	2.4	UJ	2.4	UJ	2.4	UJ	2.4	UJ	4.7	UJ	7.6	J	4	J	2.4	UJ	2.4	UJ
Eells Spring	5144103	12.7	2.4	UJ	2.4	UJ	2.4	UJ	2.6	J	4.6	UJ	6.9	J	3	J	2.4	UJ	2.4	UJ
Ford	5144099	25.75	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ
Ford-Dup	5144099-Dup	25.95	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U
Mossyrock	5144097	19.64	2.5	UJ	2.5	UJ	2.5	UJ	3.8	J	10	UJ	16	J	7.8	J	2.5	UJ	2.5	UJ
Puyallup	5144104	15.75	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ
Puyallup-Dup	5144104-Dup	16.52	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U
Spokane	5144100	15.79	2.5	UJ	2.5	UJ	2.5	UJ	2.8	J	6.1	UJ	9.3	J	4.3	J	2.5	UJ	2.5	UJ
Tucannon	5144101	15.01	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.8	J	5.4	NJ	2.5	UJ	2.5	UJ	2.5	UJ
Vancouver	5144095	16.08	2.5	UJ	2.5	UJ	2.5	UJ	2.6	J	5.5	UJ	7.7	NJ	3	J	2.5	UJ	2.5	UJ
Hatchery Rainbows																				
Arlington	5144087	3.97	2.5	U	2.5	U	2.5	U	2.1	J	3.5	UJ	6.5	NJ	3.5	J	2.5	U	2.5	U
Chelan	5144081	3.05	2.4	U	2.4	U	2.4	U	8.5	UJ	20	NJ	47		3.4	UJ	2.4	U	2.4	U
Columbia Basin	5144083	4.1	2.5	U	2.5	U	2.5	U	2.5	U	3.5	J	9		6		3.6	UJ	2.5	U
Eells Spring	5144088	2.7	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U
Eells Spring-Dup	5144088-Dup	2.13	2.4	U	2.4	U	2.4	U	2.4	U	2.4	U	2.4	U	2.4	U	2.4	U	2.4	U
Ford	5144084	2.35	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U
Mossyrock	5144082	2.69	2.4	U	2.4	U	2.4	U	2.4	U	4.8	UJ	9	J	6.8	J	2.4	U	2.4	U
Puyallup	5144089	3.07	2.3	U	2.3	U	2.3	U	2.3	U	2.3	U	2.3	U	2.3	U	2.3	U	2.3	U
Spokane	5144085	2.48	2.5	U	2.5	U	2.5	U	2.5	U	2.6	J	5.2	NJ	3.9	J	2.5	U	2.5	U
Troutlodge	5144090	5.39	2.4	U	2.4	U	2.4	U	2.6	J	5.6	UJ	7.9		3.9	J	2.4	U	2.4	U
Tucannon	5144086	3.69	2.4	U	2.4	U	2.4	U	2.4	U	2.4	U	2.4	U	2.4	U	2.4	U	2.4	U
Vancouver	5144080	4.86	2.5	U	2.5	U	2.5	U	2.5	U	3.2	NJ	3.6	NJ	2.9	NJ	2.5	U	2.5	U
Vancouver-Dup	5144080-Dup	3.14	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U

Table C-1 (cont'd). Complete Results of Percent Lipids and PCB Aroclor Analysis of Feed and Fish Tissue Samples (ng/g ww)

	Sample Number	Lipids (%)	PCB-aroclor 1016		PCB-aroclor 1221		PCB-aroclor 1232		PCB-aroclor 1242		PCB-aroclor 1248		PCB-aroclor 1254		PCB-aroclor 1260		PCB-aroclor 1262		PCB-aroclor 1268	
Planted Rainbows																				
Chapman Lake	5248102	0.44	5	U	5	U	5	U	5	U	5	U	6.7	J	5.1	J	5	U	5	U
Donnie Lake	5248103	1.29	5	U	5	U	5	U	5	U	5	U	5	U	5	U	5	U	5	U
Fan Lake	5248104	2.66	5	U	5	U	5	U	5	U	5	U	5	U	5	U	5	U	5	U
Lacamas Lake	5248100	1.5	5	U	5	U	5	U	5	U	5	U	5	U	5	U	5	U	5	U
Lone Lake	5248108	1.63	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U
Lone Lake-Dup	5248108-Dup	1.71	4.8	U	4.8	U	4.8	U	4.8	U	4.8	U	4.8	U	4.8	U	4.8	U	4.8	U
Molson Lake	5248101	2.05	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	8.6	J	4.9	U	4.9	U	4.9	U
North Lake	5248106	0.65	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U
South Lewis Co. Park Pond	5248105	0.77	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	5.9	J	4.9	U	4.9	U	4.9	U
Summit Lake	5248109	0.35	4.8	U	4.8	U	4.8	U	4.8	U	4.8	U	4.7	J	4.8	U	4.8	U	4.8	U
Summit Lake-Dup	5248109-Dup	0.45	5.0	U	5.0	U	5.0	U	5.0	U	5.0	U	**5.2**	J	5.0	U	5.0	U	5.0	U
Warden Lake	5248107	0.61	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U

U - Analyte was not detected at or above the reported result
UJ - Analyte was not detected at or above the reported estimated result
J - Analyte was positively identified. The associated numerical result is an estimate
NJ - There is evidence that the analyte is present. The associated numerical result is an estimate
Dup - Duplicate

Table C-2. Complete Results of DDT Analysis of Feed and Fish Tissue Samples (ng/g ww)

	Sample Number	2,4'-DDD		2,4'-DDE		2,4'-DDT		4,4'-DDD		4,4'-DDE		4,4'-DDT	
Hatchery Feed													
Arlington	5144102	0.37	J	0.49	U	0.49	U	2.3		2.8		0.78	NJ
Chelan	5144096	0.57	UJ	0.5	U	0.5	U	3.1		4.8		1.5	
Columbia Basin	5144098	0.28	J	0.49	U	0.49	U	1.4		4.4		0.21	J
Eells Spring	5144103	0.38	J	0.48	U	0.48	U	2		2.8		0.72	J
Ford	5144099	0.12	J	0.5	U	0.12	J	0.61		2.3		0.31	J
Ford- Dup	5144099-Dup	0.17	J	0.5	U	0.13	J	0.86		2.4		0.38	J
Mossyrock	5144097	0.43	J	0.5	U	0.5	U	3.4		6		1.2	NJ
Puyallup	5144104	0.32	J	0.5	U	0.15	J	1.2		4.3		0.49	J
Puyallup-Dup	5144104-Dup	0.3	J	0.49	U	0.15	J	1.4		4.3	NJ	0.55	
Spokane	5144100	0.2	J	0.5	U	0.5	U	0.77		4.2		0.7	NJ
Tucannon	5144101	0.85		0.5	U	0.16	J	4.4		15		0.7	J
Vancouver	5144095	0.43	J	0.5	U	0.5	U	2.1		2.6		0.69	J
Hatchery Rainbows													
Arlington	5144087	0.12	J	0.49	U	0.49	U	0.84		3.8		0.49	U
Chelan	5144081	0.49	U	0.49	U	0.49	U	0.7		2.8		0.6	NJ
Columbia Basin	5144083	0.5	U	0.61	UJ	0.5	U	1.8		3.9		0.77	NJ
Eells Spring	5144088	0.1	J	0.5	U	0.5	U	0.55		1.8		0.25	J
Eells Spring-Dup	5144088-Dup	0.49	U	0.49	U	0.49	U	0.5		1.9		0.24	J
Ford	5144084	0.49	U	0.49	U	0.49	U	0.33	J	2.2		0.49	U
Mossyrock	5144082	0.48	U	0.48	U	0.48	U	0.91		2.7		0.24	NJ
Puyallup	5144089	0.47	U	0.47	U	0.47	U	0.29	J	2		0.14	J
Spokane	5144085	0.49	U	0.49	U	0.49	U	0.52		2.2		0.2	J
Troutlodge	5144090	0.15	J	0.49	U	0.49	U	1.5		3.4		0.65	J
Tucannon	5144086	0.49	U	0.49	U	0.49	U	0.52		4.6		0.18	J
Vancouver	5144080	0.5	U	0.5	U	0.5	U	0.7		4		0.26	J
Vancouver-Dup	5144080-Dup	0.12	J	0.5	U	0.5	U	0.59		2.1		0.27	J
Planted Rainbows													
Chapman Lake	5248102	1	U	1	U	1	U	1	U	5.1		1	U
Donnie Lake	5248103	1	U	1	U	1	U	1	U	3.6		1	U
Fan Lake	5248104	1	U	1	U	1	U	9.6	J	45		1.9	J
Lacamas Lake	5248100	1	U	1	U	1	U	1	U	3.0		1	U
Lone Lake	5248108	0.98	U	0.98	U	0.98	U	0.98	U	1.9	J	0.98	U
Lone Lake-Dup	5248108-Dup	0.97	U	0.97	U	0.97	U	0.97	U	1.9	J	0.97	U
Molson Lake	5248101	0.98	U	0.98	U	0.98	U	0.98	U	5.8		0.98	U
North Lake	5248106	0.97	U	0.97	U	0.97	U	0.97	U	2.9		0.97	U
South Lewis Co. Park Pond	5248105	0.96	U	0.98	U	0.98	U	0.96	U	2.4		0.96	UJ
Summit Lake	5248109	0.96	U	0.96	U	0.96	U	0.96	U	3.0		0.96	U
Summit Lake-Dup	5248109-Dup	1.0	U	1.0	U	1.0	U	1.0	U	3.3		1.0	U
Warden Lake	5248107	0.97	U	0.97	U	0.97	U	0.97	U	3.7		0.97	U

U - Analyte was not detected at or above the reported result
UJ - Analyte was not detected at or above the reported estimated result
J - Analyte was positively identified. The associated numerical result is an estimate
NJ - There is evidence that the analyte is present. The associated numerical result is an estimate

Table C-3. Complete Results of PBDE Analysis of Feed and Fish Tissue Samples (ng/g ww)

	Sample Number	PBDE-047		PBDE-066		PBDE-071		PBDE-099		PBDE-100		PBDE-138	
Hatchery Feed													
Arlington	5144102	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ
Chelan	5144096	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ
Columbia Basin	5144098	0.24	UJ	0.24	UJ	0.24	UJ	0.24	UJ	0.24	UJ	0.24	UJ
Eells Spring	5144103	0.24	UJ	0.24	UJ	0.24	UJ	0.24	UJ	0.24	UJ	0.24	UJ
Ford	5144099	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ
Ford-Dup	5144099-Dup	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ
Mossyrock	5144097	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ
Puyallup	5144104	0.25	U	0.25	U	0.25	U	0.25	U	0.25	U	0.25	U
Puyallup-Dup	5144104-Dup	0.25	U	0.25	U	0.25	U	0.25	U	0.25	U	0.25	U
Spokane	5144100	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ
Tucannon	5144101	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ
Vancouver	5144095	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ
Hatchery Rainbows													
Arlington	5144087	0.64		0.24	U	0.24	U	0.24	U	0.24	U	0.24	U
Chelan	5144081	0.95		0.24	U	0.14	J	0.24	U	0.24	U	0.24	U
Columbia Basin	5144083	0.65	J	0.25	U	0.16	J	0.089	NJ	0.25	U	0.25	U
Eells Spring	5144088	0.55		0.25	U	0.25	U	0.25	U	0.25	U	0.25	U
Eells Spring-Dup	5144088-Dup	0.49		0.24	U	0.24	U	0.24	U	0.24	U	0.24	U
Ford	5144084	0.24	J	0.25	U	0.25	U	0.25	U	0.25	U	0.25	U
Mossyrock	5144082	0.73		0.24	U	0.16	J	0.24	U	0.24	U	0.24	U
Puyallup	5144089	0.24		0.23	U	0.23	U	0.23	U	0.23	U	0.23	U
Spokane	5144085	0.55		0.25	U	0.25	U	0.25	U	0.11	J	0.1	J
Troutlodge	5144090	0.62		0.24	U	0.15	J	0.07	J	0.24	U	0.24	U
Tucannon	5144086	0.27		0.24	U	0.24	U	0.24	U	0.24	U	0.24	U
Vancouver	5144080	0.72		0.25	U	0.13	J	0.25	U	0.25	U	0.25	U
Vancouver-Dup	5144080-Dup	0.56		0.25	U	0.25	U	0.25	U	0.25	U	0.25	U
Planted Rainbows													
Chapman Lake	5248102	0.91		0.5	U	0.5	U	0.1	NJ	0.5	U	0.5	U
Donnie Lake	5248103	0.25	J	0.5	U	0.5	U	0.5	U	0.5	U	0.5	U
Fan Lake	5248104	0.4	J	0.5	U	0.5	U	0.5	U	0.5	U	0.5	U
Lacamas Lake	5248100	0.29	J	0.5	U	0.5	U	0.5	U	0.13	J	0.5	U
Lone Lake	5248108	0.65		0.49	U	0.49	U	0.24	J	0.15	J	0.49	U
Lone Lake-Dup	5248108-Dup	0.54		0.48	U	0.48	U	0.13	J	0.20	J	0.48	U
Molson Lake	5248101	0.49	UJ	0.49	UJ	0.49	UJ	0.49	UJ	0.49	UJ	0.49	UJ
North Lake	5248106	0.78		0.48	U	0.48	U	0.29	J	0.16	J	0.48	U
South Lewis Co. Park Pond	5248105	1		0.49	U	0.49	U	0.33	J	0.49	U	0.49	U
Summit Lake	5248109	0.87		0.48	U	0.48	U	0.12	J	0.14	J	0.48	U
Summit Lake-Dup	5248109-Dup	0.65	U	0.50	U	0.50	U	0.50	U	0.50	U	0.50	U
Warden Lake	5248107	0.46	J	0.48	U	0.48	U	0.48	U	0.48	U	0.48	U

Table C-3 (cont'd). Complete Results of PBDE Analysis of Feed and Fish Tissue Samples (ng/g ww)

	Sample Number	PBDE-153		PBDE-154		PBDE-183		PBDE-190		PBDE-209	
Hatchery Feed											
Arlington	5144102	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	1.2	UJ
Chelan	5144096	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	1.2	UJ
Columbia Basin	5144098	0.24	UJ	0.24	UJ	0.24	UJ	0.24	UJ	1.2	UJ
Eells Spring	5144103	0.24	UJ	0.24	UJ	0.24	UJ	0.24	UJ	1.2	UJ
Ford	5144099	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	1.2	UJ
Ford-Dup	5144099-Dup	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	1.2	UJ
Mossyrock	5144097	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	1.2	UJ
Puyallup	5144104	0.25	U	0.25	U	0.25	U	0.25	U	1.2	U
Puyallup-Dup	5144104-Dup	0.25	U	0.25	U	0.25	U	0.25	U	1.2	U
Spokane	5144100	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	1.2	UJ
Tucannon	5144101	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	1.2	UJ
Vancouver	5144095	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	1.2	UJ
Hatchery Rainbows											
Arlington	5144087	0.24	U	0.24	U	0.24	U	0.24	U	3.1	U
Chelan	5144081	0.24	U	0.24	U	0.24	U	0.24	U	3	U
Columbia Basin	5144083	0.25	U	0.25	U	0.25	U	0.25	U	3.1	U
Eells Spring	5144088	0.25	U	0.25	U	0.25	U	0.25	U	3.1	U
Eells Spring-Dup	5144088-Dup	0.24	U	0.24	U	0.24	U	0.24	U	3.0	U
Ford	5144084	0.25	U	0.25	U	0.25	U	0.25	U	3.1	U
Mossyrock	5144082	0.24	U	0.24	U	0.24	U	0.24	U	3	U
Puyallup	5144089	0.23	U	0.23	U	0.23	U	0.23	U	2.9	U
Spokane	5144085	0.25	U	0.25	U	0.25	U	0.25	U	0.34	J
Troutlodge	5144090	0.24	U	0.24	U	0.24	U	0.24	U	3	U
Tucannon	5144086	0.24	U	0.24	U	0.24	U	0.24	U	3	U
Vancouver	5144080	0.25	U	0.25	U	0.25	U	0.25	U	3.1	U
Vancouver-Dup	5144080-Dup	0.25	U	0.25	U	0.25	U	0.25	U	3.1	U
Planted Rainbows											
Chapman Lake	5248102	0.5	U	0.5	U	0.5	U	0.5	U	6.2	U
Donnie Lake	5248103	0.5	U	0.5	U	0.5	U	0.5	U	6.2	U
Fan Lake	5248104	0.5	U	0.5	U	0.5	U	0.5	U	6.2	U
Lacamas Lake	5248100	0.5	U	0.5	U	0.5	U	0.5	U	6.2	U
Lone Lake	5248108	0.49	U	0.49	U	0.49	U	0.49	U	6.1	U
Lone Lake-Dup	5248108-Dup	0.48	U	0.48	U	0.48	U	0.48	U	6.0	U
Molson Lake	5248101	0.49	UJ	0.49	UJ	0.49	UJ	0.49	UJ	2.4	UJ
North Lake	5248106	0.48	U	0.48	U	0.48	U	0.48	U	6.1	U
South Lewis Co. Park Pond	5248105	0.49	U	0.49	U	0.49	U	0.49	U	6.1	U
Summit Lake	5248109	0.48	U	0.48	U	0.48	U	0.48	U	6	U
Summit Lake-Dup	5248109-Dup	0.50	U	0.50	U	0.50	U	0.50	U	6.2	U
Warden Lake	5248107	0.48	U	0.48	U	0.48	U	0.48	U	6.1	U

U - Analyte was not detected at or above the reported result

UJ - Analyte was not detected at or above the reported estimated result

J - Analyte was positively identified. The associated numerical result is an estimate

Table C-4. Complete Results of PCDD/F Analysis of Feed Samples (pg/g ww)

Sample Number	TEF	Vancouver Hatchery			Mossyrock Hatchery			Ford Hatchery			Ford Hatchery			Spokane Hatchery		
		Sample 05144105		TEQ	Sample 05144106		TEQ	Sample 05144107		TEQ	Sample 05144107-Dup		TEQ	Sample 05144108		TEQ
2,3,7,8-TCDF	0.1	0.525		0.0525	0.798		0.0798	0.182		0.0182	0.222		0.0222	1.4		0.14
1,2,3,7,8-PeCDF	0.05	0.251	J	0.01255	0.591		0.02955	0.1	UJ	0	0.115	J	0.00575	0.218	J	0.0109
2,3,4,7,8-PeCDF	0.5	0.126	J	0.063	0.226	J	0.113	0.12	UJ	0	0.12	UJ	0	0.246	J	0.123
1,2,3,4,7,8-HxCDF	0.1	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0
1,2,3,6,7,8-HxCDF	0.1	0.1	UJ	0	0.1	UJ	0	0.1	UJ	0	0.1	UJ	0	0.1	UJ	0
2,3,4,6,7,8-HxCDF	0.1	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0
1,2,3,7,8,9-HxCDF	0.1	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0
1,2,3,4,6,7,8-HpCDF	0.01	0.2	UJ	0	0.207	J	0.00207	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0
1,2,3,4,7,8,9-HpCDF	0.01	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0
OCDF	0.0001	0.404	J	0.00004	0.403	J	0.00004	0.32	J	0.00003	0.344	J	0.00003	0.377	J	0.00004
2,3,7,8-TCDD	1	0.186		0.186	0.367		0.367	0.05	UJ	0	0.05	UJ	0	0.192		0.192
1,2,3,7,8-PeCDD	1	0.215	J	0.215	0.534		0.534	0.11	UJ	0	0.11	UJ	0	0.637		0.637
1,2,3,4,7,8-HxCDD	0.1	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0
1,2,3,6,7,8-HxCDD	0.1	0.235	J	0.0235	0.494		0.0494	0.2	UJ	0	0.2	UJ	0	0.437		0.0437
1,2,3,7,8,9-HxCDD	0.1	0.2	UJ	0	0.333		0.0333	0.2	UJ	0	0.2	UJ	0	0.339		0.0339
1,2,3,4,6,7,8-HpCDD	0.01	0.857		0.00857	1.6		0.016	0.524		0.00524	0.474		0.00474	1.25		0.0125
OCDD	0.0001	8.14		0.00081	16.5		0.00165	3.38		0.00034	3.3		0.00033	11.5		0.00115
TEQ total				0.56197			1.22581			0.02381			0.03305			1.19419

TEF - Toxicity Equivalence Factor from Van den Berg et al., 1998
TEQ - Toxic Equivalent
UJ - The analyte was not detected at or above the reported estimated result
J - The analyte was positively identified. The associated numerical value is an estimate

Table C-5. Complete Results of PCDD/F Analysis of Rainbow Trout Tissue Samples (pg/g ww)

Sample Number	TEF	Vancouver Hatchery			Mossyrock Hatchery			Ford Hatchery			Ford Hatchery			Spokane Hatchery		
		Sample 05144092		TEQ	Sample 05144093		TEQ	Sample 05144091		TEQ	Sample 05144091-Dup		TEQ	Sample 05144094		TEQ
2,3,7,8-TCDF	0.1	0.187		0.0187	0.282		0.0282	0.037	J	0.0037	0.042	J	0.0042	0.163		0.0163
1,2,3,7,8-PeCDF	0.05	0.111	J	0.00555	0.143	J	0.00715	0.06	UJ	0	0.06	UJ	0	0.06	UJ	0
2,3,4,7,8-PeCDF	0.5	0.041	J	0.0205	0.04	UJ	0	0.04	UJ	0	0.04	UJ	0	0.04	UJ	0
1,2,3,4,7,8-HxCDF	0.1	0.07	J	0.007	0.051	J	0.0051	0.075	J	0.0075	0.053	J	0.0053	0.067	J	0.0067
1,2,3,6,7,8-HxCDF	0.1	0.05	UJ	0	0.05	UJ	0	0.05	UJ	0	0.05	UJ	0	0.05	UJ	0
2,3,4,6,7,8-HxCDF	0.1	0.06	UJ	0	0.06	UJ	0	0.06	UJ	0	0.06	UJ	0	0.06	UJ	0
1,2,3,7,8,9-HxCDF	0.1	0.06	UJ	0	0.06	UJ	0	0.06	UJ	0	0.06	UJ	0	0.06	UJ	0
1,2,3,4,6,7,8-HpCDF	0.01	0.095	J	0.00095	0.078	J	0.00078	0.094	J	0.00094	0.105	J	0.00105	0.082	J	0.00082
1,2,3,4,7,8,9-HpCDF	0.01	0.05	UJ	0	0.05	UJ	0	0.05	UJ	0	0.05	UJ	0	0.05	UJ	0
OCDF	0.0001	0.31	J	0.00003	0.225	J	0.00002	0.313	J	0.00003	0.289	J	0.00003	0.336	J	0.00003
2,3,7,8-TCDD	1	0.03	UJ	0	0.03	UJ	0	0.03	UJ	0	0.03	UJ	0	0.03	UJ	0
1,2,3,7,8-PeCDD	1	0.07	UJ	0	0.07	UJ	0	0.07	UJ	0	0.07	UJ	0	0.07	UJ	0
1,2,3,4,7,8-HxCDD	0.1	0.10000	UJ	0	0.10000	UJ	0	0.10000	UJ	0	0.10000	UJ	0	0.10000	UJ	0
1,2,3,6,7,8-HxCDD	0.1	0.08	UJ	0	0.08	UJ	0	0.08	UJ	0	0.08	UJ	0	0.08	UJ	0
1,2,3,7,8,9-HxCDD	0.1	0.05	UJ	0	0.05	UJ	0	0.05	UJ	0	0.05	UJ	0	0.05	UJ	0
1,2,3,4,6,7,8-HpCDD	0.01	0.08	UJ	0	0.08	UJ	0	0.08	UJ	0	0.08	UJ	0	0.08	UJ	0
OCDD	0.0001	0.36	UJ	0	0.36	UJ	0	0.36	UJ	0	0.36	UJ	0	0.36	UJ	0
TEQ total				0.05273			0.04125			0.01217			0.01058			0.02385

TEF - Toxicity Equivalence Factor from Van den Berg et al., 1998
TEQ - Toxic Equivalent
UJ - The analyte was not detected at or above the reported estimated result
J - The analyte was positively identified. The associated numerical value is an estimate

Table C-5. Complete Results of Chlorinated Pesticide (Excluding DDT Compounds) Analysis of Feed and Fish Tissue Samples (ng/g ww)

	Sample Number	Aldrin		Alpha-BHC		Beta-BHC		Chlordane		Chlor-pyriphos		cis-Chlordane	
Hatchery Feed													
Arlington	5144102	0.49	UJ	0.49	U	0.49	U	2.5	U	2.5	UJ	0.52	
Chelan	5144096	0.5	UJ	0.5	U	0.5	U	2.5	U	2.5	U	0.5	U
Columbia Basin	5144098	0.49	UJ	0.49	U	0.49	U	2.4	U	2.4	U	0.68	NJ
Eells Spring	5144103	0.48	UJ	0.48	U	0.48	U	2.4	U	2.4	UJ	0.47	NJ
Ford	5144099	0.5	UJ	0.5	U	0.5	U	2.5	U	2.5	U	0.19	J
Ford-Dup	5144099-Dup	0.50	UJ	0.50	U	0.50	U	2.5	U	2.5	UJ	0.25	J
Mossyrock	5144097	0.5	UJ	0.5	U	0.5	U	2.5	U	2.5	U	1.2	NJ
Puyallup	5144104	0.5	UJ	0.5	U	0.5	U	2.5	U	10	UJ	0.5	U
Puyallup-Dup	5144104-Dup	0.49	UJ	0.49	U	0.49	U	2.5	U	9.9	UJ	0.49	U
Spokane	5144100	0.5	UJ	0.5	U	0.5	U	2.5	U	2.5	UJ	0.39	NJ
Tucannon	5144101	0.5	UJ	0.5	U	0.5	U	2.5	U	2.5	UJ	0.5	U
Vancouver	5144095	0.5	UJ	0.5	U	0.5	U	2.5	U	2.5	U	0.52	
Hatchery Rainbows													
Arlington	5144087	0.49	UJ	0.49	U	0.49	U	2.5	U	2.5	U	0.24	NJ
Chelan	5144081	0.49	UJ	0.49	U	0.49	U	2.4	U	2.4	U	0.49	U
Columbia Basin	5144083	0.5	UJ	0.5	U	0.5	U	2.5	U	2.5	U	0.25	NJ
Eells Spring	5144088	0.5	UJ	0.5	U	0.5	U	2.5	U	2.5	U	0.5	U
Eells Spring-Dup	5144088-Dup	0.49	UJ	0.49	U	0.49	U	2.4	U	2.4	U	0.49	U
Ford	5144084	0.49	UJ	0.49	U	0.49	U	2.5	U	2.5	U	0.49	U
Mossyrock	5144082	0.48	UJ	0.48	U	0.48	U	2.4	U	2.4	U	0.31	NJ
Puyallup	5144089	0.47	UJ	0.47	U	0.47	U	2.3	U	2.3	U	0.47	U
Spokane	5144085	0.49	UJ	0.49	U	0.49	U	2.5	U	2.5	U	0.49	U
Troutlodge	5144090	0.49	UJ	0.49	U	0.49	U	2.4	U	1.2	J	0.44	NJ
Tucannon	5144086	0.49	UJ	0.49	U	0.49	U	2.4	U	2.4	U	0.17	J
Vancouver	5144080	0.5	UJ	0.5	U	0.5	U	2.5	U	2.5	U	0.2	NJ
Vancouver-Dup	5144080-Dup	0.50	UJ	0.50	U	0.50	U	2.5	U	2.5	U	0.17	NJ
Planted Rainbows													
Chapman Lake	5248102	1	UJ	1	UJ	1	U	5	U	4	U	1	U
Donnie Lake	5248103	1	UJ	1	UJ	1	U	5	U	4	U	1	U
Fan Lake	5248104	1	UJ	1	UJ	1	U	5	U	4	U	1	U
Lacamas Lake	5248100	1	UJ	1	UJ	1	U	5	U	4	U	1	U
Lone Lake	5248108	0.98	UJ	0.98	UJ	0.98	U	0.98	U	3.9	U	0.98	U
Lone Lake-Dup	5248108-Dup	0.97	UJ	0.97	UJ	0.97	U	4.8	U	3.9	U	0.97	U
Molson Lake	5248101	0.98	UJ	0.98	UJ	0.98	U	4.9	U	3.9	U	0.98	U
North Lake	5248106	0.97	UJ	0.97	UJ	0.97	U	4.9	U	3.9	U	0.97	U
South Lewis Co. Park Pond	5248105	0.98	UJ	0.96	UJ	0.96	U	2.4	U	0.96	U	0.96	UJ
Summit Lake	5248109	0.96	UJ	0.96	UJ	0.96	U	4.8	U	3.8	U	0.96	U
Summit Lake-Dup	5248109-Dup	1.0	UJ	1.0	UJ	1.0	U	5.0	U	4.0	U	1.0	U
Warden Lake	5248107	0.97	UJ	0.97	UJ	0.97	U	4.9	U	3.9	U	0.97	U

Table C-5 (cont'd). Complete Results of Chlorinated Pesticide (Excluding DDT Compounds) Analysis of Feed and Fish Tissue Samples (ng/g ww)

	Sample Number	cis-Nonachlor		Dacthal (DCPA)		DDMU		Delta-BHC		Dieldrin	
Hatchery Feed											
Arlington	5144102	0.49	U	2.5	UJ	0.69	NJ	0.49	UJ	2.5	UJ
Chelan	5144096	0.5	U	2.5	U	1.5	NJ	0.5	UJ	2.5	U
Columbia Basin	5144098	0.49	U	2.4	UJ	0.73	NJ	0.49	UJ	2.4	UJ
Eells Spring	5144103	0.48	U	2.4	UJ	0.58	NJ	0.48	UJ	2.4	UJ
Ford	5144099	0.5	U	2.5	UJ	0.4	NJ	0.5	UJ	2.5	UJ
Ford-Dup	5144099-Dup	0.50	U	2.5	UJ	0.50	U	0.50	UJ	2.5	UJ
Mossyrock	5144097	0.5	U	2.5	U	1.2	NJ	0.5	UJ	3.8	NJ
Puyallup	5144104	0.5	U	10	UJ	1.1	NJ	0.5	UJ	10	UJ
Puyallup-Dup	5144104-Dup	0.49	U	0.49	UJ	1.0	NJ	0.49	UJ	9.9	UJ
Spokane	5144100	0.5	U	2.5	UJ	0.85	NJ	0.5	UJ	2.5	UJ
Tucannon	5144101	0.5	U	2.5	UJ	3.9	NJ	0.5	UJ	2.5	UJ
Vancouver	5144095	0.5	U	2.5	U	0.6	NJ	0.5	UJ	2.5	U
Hatchery Rainbows											
Arlington	5144087	0.49	U	2.5	U	0.9	NJ	0.49	UJ	0.77	J
Chelan	5144081	0.49	U	2.4	U	0.52	NJ	0.49	UJ	0.58	NJ
Columbia Basin	5144083	0.5	U	2.5	U	0.63	NJ	0.5	UJ	0.5	J
Eells Spring	5144088	0.5	U	2.5	U	0.5	U	0.5	UJ	0.75	J
Eells Spring-Dup	5144088-Dup	0.49	U	2.4	U	0.49	U	0.49	UJ	0.63	NJ
Ford	5144084	0.49	U	2.5	U	0.49	U	0.49	UJ	0.49	UJ
Mossyrock	5144082	0.48	U	2.4	U	0.48	U	0.48	UJ	0.88	NJ
Puyallup	5144089	0.47	U	2.3	U	0.47	U	0.47	UJ	0.47	UJ
Spokane	5144085	0.49	U	2.5	U	0.49	U	0.49	UJ	0.78	J
Troutlodge	5144090	0.49	U	2.4	U	0.49	NJ	0.49	UJ	0.75	J
Tucannon	5144086	0.49	U	2.4	U	0.62		0.49	UJ	0.49	UJ
Vancouver	5144080	0.5	U	2.5	U	0.5	U	0.5	UJ	0.33	NJ
Vancouver-Dup	5144080-Dup	0.50	U	2.5	U	0.50	U	0.50	UJ	0.30	J
Planted Rainbows											
Chapman Lake	5248102	1.0	U	4.0	U	1.0	U	1.0	UJ	0.79	UJ
Donnie Lake	5248103	1.0	U	4.0	U	1.0	U	1.0	UJ	0.79	UJ
Fan Lake	5248104	1.0	U	4.0	U	4.4		1.0	UJ	0.80	UJ
Lacamas Lake	5248100	1.0	U	4.0	U	1.0	U	1.0	UJ	0.80	UJ
Lone Lake	5248108	0.98	U	3.9	U	0.98	U	0.98	UJ	0.78	UJ
Lone Lake-Dup	5248108-Dup	0.97	U	3.9	U	0.97	U	0.97	UJ	0.78	UJ
Molson Lake	5248101	0.98	U	3.9	U	0.98	U	0.98	UJ	0.78	UJ
North Lake	5248106	0.97	U	3.9	U	0.97	U	0.97	UJ	0.55	NJ
South Lewis Co. Park Pond	5248105	0.96	U	0.96	U	0.98	U	0.96	U	0.96	U
Summit Lake	5248109	0.96	U	3.8	U	0.96	U	0.96	UJ	0.77	UJ
Summit Lake-Dup	5248109-Dup	1.0	U	4.0	U	1.0	U	1.0	UJ	0.80	UJ
Warden Lake	5248107	0.97	U	3.9	U	0.97	U	0.97	UJ	0.76	NJ

Table C-5 (cont'd). Complete Results of Chlorinated Pesticide (Excluding DDT Compounds) Analysis of Feed and Fish Tissue Samples (ng/g ww)

	Sample Number	Endo-sulfan I		Endo-sulfan II		Endo-sulfan Sulfate		Endrin		Endrin Aldehyde		Endrin Ketone	
Hatchery Feed													
Arlington	5144102	2.5	UJ	2.5	UJ	REJ		2.5	UJ	REJ		REJ	
Chelan	5144096	2.5	U	2.5	UJ	REJ		2.5	U	REJ		REJ	
Columbia Basin	5144098	2.4	UJ	2.4	UJ	REJ		2.4	UJ	REJ		REJ	
Eells Spring	5144103	2.4	UJ	2.4	UJ	REJ		2.4	UJ	REJ		REJ	
Ford	5144099	2.5	UJ	2.5	UJ	REJ		2.5	UJ	REJ		REJ	
Ford-Dup	5144099-Dup	2.5	UJ	2.5	UJ	REJ		2.5	UJ	REJ		REJ	
Mossyrock	5144097	2.5	U	2.5	UJ	REJ		2.5	U	REJ		REJ	
Puyallup	5144104	10	UJ	10	UJ	10	UJ	10	UJ	10	UJ	10	UJ
Puyallup-Dup	5144104-Dup	9.9	UJ	9.9	UJ	9.9	UJ	9.9	UJ	9.9	UJ	9.9	UJ
Spokane	5144100	2.5	UJ	2.5	UJ	REJ		2.5	UJ	REJ		REJ	
Tucannon	5144101	2.5	UJ	2.5	UJ	REJ		2.5	UJ	REJ		REJ	
Vancouver	5144095	2.5	U	2.5	UJ	REJ		2.5	U	REJ		REJ	
Hatchery Rainbows													
Arlington	5144087	2.5	U	2.5	UJ	REJ		2.5	U	REJ		REJ	
Chelan	5144081	2.4	U	2.4	UJ	REJ		2.4	U	REJ		REJ	
Columbia Basin	5144083	2.5	U	2.5	UJ	REJ		2.5	U	REJ		REJ	
Eells Spring	5144088	2.5	U	2.5	UJ	REJ		2.5	U	REJ		REJ	
Eells Spring-Dup	5144088-Dup	2.4	U	2.4	UJ	REJ		2.4	U	REJ		REJ	
Ford	5144084	2.5	U	2.5	UJ	REJ		2.5	U	REJ		REJ	
Mossyrock	5144082	2.4	U	2.4	UJ	REJ		2.4	U	REJ		REJ	
Puyallup	5144089	2.3	U	2.3	UJ	REJ		2.3	U	REJ		REJ	
Spokane	5144085	2.5	U	2.5	UJ	REJ		2.5	U	REJ		REJ	
Troutlodge	5144090	2.4	U	2.4	UJ	REJ		2.4	U	REJ		REJ	
Tucannon	5144086	2.4	U	2.4	UJ	REJ		2.4	U	REJ		REJ	
Vancouver	5144080	2.5	U	2.5	UJ	REJ		2.5	U	REJ		REJ	
Vancouver-Dup	5144080-Dup	2.5	U	2.5	UJ	REJ		2.5	U	REJ		REJ	
Planted Rainbows													
Chapman Lake	5248102	4.0	U	4.0	U	4.0	UJ	4.0	U	4.0	UJ	4.0	U
Donnie Lake	5248103	4.0	U	4.0	U	4.0	UJ	4.0	U	4.0	UJ	4.0	U
Fan Lake	5248104	4.0	U	4.0	U	4.0	UJ	4.0	U	4.0	UJ	4.0	U
Lacamas Lake	5248100	4.0	U	4.0	U	4.0	UJ	4.0	U	4.0	UJ	4.0	U
Lone Lake	5248108	3.9	U	3.9	U	3.9	UJ	3.9	U	3.9	UJ	3.9	U
Lone Lake-Dup	5248108-Dup	3.9	U	3.9	U	3.9	UJ	3.9	U	3.9	UJ	3.9	U
Molson Lake	5248101	3.9	U	3.9	U	3.9	UJ	3.9	U	3.9	UJ	3.9	U
North Lake	5248106	3.9	U	3.9	U	3.9	UJ	3.9	U	3.9	UJ	3.9	U
South Lewis Co. Park Pond	5248105	0.96	U	0.96	U	0.96	UJ	0.96	U	0.96	UJ	0.96	U
Summit Lake	5248109	3.8	U	3.8	U	3.8	UJ	3.8	U	3.8	UJ	3.8	U
Summit Lake-Dup	5248109-Dup	4.0	U	4.0	U	4.0	UJ	4.0	U	4.0	UJ	4.0	U
Warden Lake	5248107	3.9	U	3.9	U	3.9	UJ	3.9	U	3.9	UJ	3.9	U

Table C-5 (cont'd). Complete Results of Chlorinated Pesticide (Excluding DDT Compounds) Analysis of Feed and Fish Tissue Samples (ng/g ww)

	Sample Number	Heptachlor		Heptachlor Epoxide		Hexachloro -benzene		Lindane		Methoxy-chlor		Mirex	
Hatchery Feed													
Arlington	5144102	0.49	U	2.5	UJ	0.16	J	0.49	U	2.5	UJ	0.49	UJ
Chelan	5144096	0.5	U	2.5	U	0.31	J	0.5	U	2.5	UJ	0.5	UJ
Columbia Basin	5144098	0.49	U	2.4	UJ	0.49	UJ	0.49	U	2.4	UJ	0.49	UJ
Eells Spring	5144103	0.48	U	2.4	UJ	0.15	J	0.48	U	2.4	UJ	0.48	UJ
Ford	5144099	0.5	U	2.5	UJ	0.16	J	0.5	U	2.5	UJ	0.5	UJ
Ford-Dup	5144099-Dup	0.50	U	2.5	UJ	0.19	J	0.50	U	2.5	UJ	0.50	UJ
Mossyrock	5144097	0.5	U	2.5	U	0.3	J	0.5	U	2.2	J	0.5	UJ
Puyallup	5144104	0.5	U	10	UJ	0.5	UJ	0.5	U	4.9	J	0.5	UJ
Puyallup-Dup	5144104-Dup	0.49	U	9.9	UJ	0.10	J	0.49	U	4.0	J	0.49	UJ
Spokane	5144100	0.5	U	2.5	UJ	0.12	J	0.5	U	2.5	UJ	0.5	UJ
Tucannon	5144101	0.5	U	2.5	UJ	0.16	J	0.5	U	2.5	UJ	0.5	UJ
Vancouver	5144095	0.5	U	2.5	U	0.17	J	0.5	U	2.5	UJ	0.5	UJ
Hatchery Rainbows													
Arlington	5144087	0.49	U	0.49	U	0.23	J	0.49	U	2.5	U	0.49	U
Chelan	5144081	0.49	U	0.49	U	0.49	U	0.49	U	2.4	U	0.49	U
Columbia Basin	5144083	0.5	U	0.5	U	0.5	U	0.5	U	2.5	U	0.5	U
Eells Spring	5144088	0.5	U	0.5	U	0.5	U	0.5	U	2.5	U	0.5	U
Eells Spring-Dup	5144088-Dup	0.49	U	0.49	U	0.49	U	0.49	U	2.4	U	0.49	U
Ford	5144084	0.49	U	0.49	U	0.49	U	0.49	U	2.5	U	0.49	U
Mossyrock	5144082	0.48	U	0.48	U	0.48	U	0.48	U	2.4	U	0.48	U
Puyallup	5144089	0.47	U	0.47	U	0.47	U	0.47	U	2.3	U	0.47	U
Spokane	5144085	0.49	U	0.49	U	0.1	J	0.49	U	2.5	U	0.49	U
Troutlodge	5144090	0.49	U	0.49	U	0.49	U	0.49	U	2.4	U	0.49	U
Tucannon	5144086	0.49	U	0.49	U	0.49	U	0.49	U	2.4	U	0.49	U
Vancouver	5144080	0.5	U	0.5	U	0.3	NJ	0.5	U	2.5	U	0.5	U
Vancouver-Dup	5144080-Dup	0.50	U	0.50	U	0.50	U	0.50	U	2.5	U	0.50	U
Planted Rainbows													
Chapman Lake	5248102	1	U	0.79	UJ	1	U	1	U	4	U	1	U
Donnie Lake	5248103	1	U	0.79	UJ	1	U	1	U	4	U	1	U
Fan Lake	5248104	1	U	0.8	UJ	1	U	1	U	4	U	1	U
Lacamas Lake	5248100	1	U	0.8	UJ	1	U	1	U	4	U	1	U
Lone Lake	5248108	0.98	U	0.78	UJ	0.98	U	0.98	U	3.9	U	0.98	U
Lone Lake-Dup	5248108-Dup	0.97	U	0.78	UJ	0.97	U	0.97	U	3.9	U	0.97	U
Molson Lake	5248101	0.98	U	0.78	UJ	0.98	U	0.98	U	3.9	U	0.98	U
North Lake	5248106	0.97	U	0.78	UJ	0.97	U	0.97	U	3.9	U	0.97	U
South Lewis Co. Park Pond	5248105	0.98	U	0.96	U	0.98	U	0.96	U	0.96	U	0.98	U
Summit Lake	5248109	0.96	U	0.77	UJ	0.69	J	0.96	U	3.8	U	0.96	U
Summit Lake-Dup	5248109-Dup	1.0	U	0.80	UJ	0.76	J	1.0	U	4.0	U	1.0	U
Warden Lake	5248107	0.97	U	0.78	UJ	0.97	U	0.97	U	3.9	U	0.97	U

Table C-5 (cont'd). Complete Results of Chlorinated Pesticide (Excluding DDT Compounds) Analysis of Feed and Fish Tissue Samples (ng/g ww)

	Sample Number	Oxychlordane		Pentachloro -anisole		Toxaphene		trans-Chlordane		trans-Nonachlor	
Hatchery Feed											
Arlington	5144102	0.49	U	0.49	UJ	4.9	U	0.49	U	0.68	
Chelan	5144096	0.5	U	0.5	UJ	5	U	1.1	NJ	1.1	NJ
Columbia Basin	5144098	0.49	U	0.49	UJ	3.9	NJ	0.2	J	0.7	
Eells Spring	5144103	0.48	U	0.48	UJ	4.8	U	0.19	J	0.44	J
Ford	5144099	0.5	U	0.5	UJ	5	U	0.5	U	0.15	J
Ford-Dup	5144099-Dup	0.50	U	0.50	UJ	5.0	U	0.50	U	0.16	J
Mossyrock	5144097	0.5	U	0.15	J	5	U	0.2	NJ	1.2	
Puyallup	5144104	0.5	U	0.5	UJ	3.6	J	0.5	U	0.5	U
Puyallup-Dup	5144104-Dup	0.49	U	0.49	UJ	4.9	U	0.49	U	0.49	U
Spokane	5144100	0.5	U	0.5	UJ	2.6	J	0.5	U	0.8	
Tucannon	5144101	0.5	U	0.11	J	5	U	0.5	U	0.3	J
Vancouver	5144095	0.5	U	0.5	UJ	5	U	0.5	U	0.77	J
Hatchery Rainbows											
Arlington	5144087	0.49	U	0.49	U	4.9	U	0.49	U	0.27	J
Chelan	5144081	0.49	U	0.49	U	4.9	U	0.49	U	0.42	J
Columbia Basin	5144083	0.5	U	0.5	U	5	U	0.5	U	0.45	J
Eells Spring	5144088	0.5	U	0.5	U	5	U	0.5	U	0.2	J
Eells Spring-Dup	5144088-Dup	0.49	U	0.49	U	4.9	U	0.49	U	0.17	J
Ford	5144084	0.49	U	0.49	U	4.9	U	0.49	U	0.49	U
Mossyrock	5144082	0.48	U	0.48	U	4.8	U	0.48	U	0.3	NJ
Puyallup	5144089	0.47	U	0.47	U	4.7	U	0.47	U	0.094	J
Spokane	5144085	0.49	U	0.49	U	4.9	U	0.49	U	0.49	U
Troutlodge	5144090	0.49	U	0.49	U	4.9	U	0.49	U	0.57	J
Tucannon	5144086	0.49	U	0.18	J	4.9	U	0.49	U	0.12	NJ
Vancouver	5144080	0.5	U	0.21	J	5	U	0.5	U	0.3	J
Vancouver-Dup	5144080-Dup	0.50	U	0.50	U	5.0	U	0.50	U	0.20	J
Planted Rainbows											
Chapman Lake	5248102	1	U	1	U	9.9	U	1	U	1	U
Donnie Lake	5248103	1	U	1	U	9.9	U	1	U	1	U
Fan Lake	5248104	1	U	1	U	10	U	1	U	1	U
Lacamas Lake	5248100	1	U	0.47	J	10	U	1	U	1	U
Lone Lake	5248108	0.98	U	0.98	U	0.98	U	0.98	U	0.98	U
Lone Lake-Dup	5248108-Dup	0.97	U	0.97	U	9.7	U	0.97	U	0.97	U
Molson Lake	5248101	0.98	U	0.98	U	9.8	U	0.98	U	0.98	U
North Lake	5248106	0.97	U	0.97	U	9.7	U	0.97	U	0.97	U
South Lewis Co. Park Pond	5248105	0.96	U	0.96	U	9.6	U	0.96	U	0.98	U
Summit Lake	5248109	0.96	U	0.96	U	9.6	U	0.96	U	0.96	U
Summit Lake-Dup	5248109-Dup	1.0	U	1.0	U	10	U	1.0	U	1.0	U
Warden Lake	5248107	0.97	U	0.97	U	9.7	U	0.97	U	0.97	U

Dup – Duplicate
U - Analyte was not detected at or above the reported result
UJ - Analyte was not detected at or above the reported estimated result
J - Analyte was positively identified. The associated numerical result is an estimate
NJ - There is evidence that the analyte is present. The associated numerical result is an estimate
REJ - Data are unusable for all purposes

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ORIGINAL ARTICLE

Contaminant exposure in outmigrant juvenile salmon from Pacific Northwest estuaries of the United States[1]

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Abstract To better understand the dynamics of contaminant uptake in outmigrant juvenile salmon in the Pacific Northwest, concentrations of polychlorinated biphenyls (PCBs), DDTs, polycylic aromatic hydrocarbons (PAHs) and organochlorine pesticides were measured in tissues and prey of juvenile chinook and coho salmon from several estuaries and hatcheries in the US Pacific Northwest. PCBs, DDTs, and PAHs were found in tissues (whole bodies or bile) and stomach contents of chinook and coho salmon sampled from all estuaries, as well as in chinook salmon from hatcheries. Organochlorine pesticides were detected less frequently. Of the two species sampled, chinook salmon had the highest whole body contaminant concentrations, typically 2–5 times higher than coho salmon from the same sites. In comparison to estuarine chinook salmon, body burdens of PCBs and DDTs in hatchery chinook were relatively high, in part because of the high lipid content of the hatchery fish. Concentrations of PCBs were highest in chinook salmon from the Duwamish Estuary, the Columbia River and Yaquina Bay, exceeding the NOAA Fisheries' estimated threshold for adverse health effects of 2400 ng/g lipid. Concentrations of DDTs were especially high in juvenile chinook salmon from the Columbia River and Nisqually Estuary; concentrations of PAH metabolites in bile were highest in chinook salmon from the Duwamish Estuary and Grays Harbor. Juvenile chinook salmon are likely absorbing some contaminants during estuarine residence through their prey, as PCBs, PAHs, and DDTs were consistently present in stomach contents, at concentrations significantly correlated with contaminant body burdens in fish from the same sites.

Keywords Chinook salmon · Coho salmon · Contaminants · PAHs · PCBs · DDTs · Pesticides · Washington · Oregon · Estuary

1 Introduction

Estuaries are important habitats for salmon during the juvenile stage of their life cycle, when they make the transition from freshwater to the ocean (Healey, 1982). Estuaries provide outmigrating juvenile salmon with a refuge from predators, a rich food supply that supports rapid growth, and appropriate conditions for the physiological adaptation to saltwater (Dorcey *et al.*, 1978; Simenstad *et al.*, 1982). However, urban and industrial development may impair the quality of estuarine habitats. Estuaries located near urban centers often receive inputs of toxic contaminants from municipal and industrial activities (Brown *et al.*, 1998; USEPA,

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1997), which may be taken up by juvenile salmon and their prey. Because juvenile salmon are in a period of rapid development, and undergoing many physiological changes during their residence in estuarine environments, they may be especially vulnerable to the deleterious effects of toxic chemicals.

The well-documented presence of chemically contaminated sediments in Puget Sound urban estuaries (e.g., Malins *et al.*, 1982) prompted a series of studies to examine the degree to which juvenile salmon were exposed to toxic chemicals during estuarine residence (McCain *et al.*, 1990; Varanasi *et al.*, 1993; Stein *et al.*, 1995; Stehr *et al.*, 2000). Juvenile salmon (primarily chinook and coho, *Onchorhynchus tshawytscha* and *O. kisutch)* were sampled from several urban and non-urban estuaries in Puget Sound including the Green River/Duwamish Estuary system in Seattle, the Puyallup River/Hylebos Waterway system in Tacoma, and the more rural Snohomish River and Nisqually River Estuaries. Juvenile chinook salmon from hatcheries associated with sampled estuaries were also collected and whole bodies and stomach contents were analyzed for chemical concentrations. Results of these surveys showed that outmigrating juvenile chinook salmon from the Duwamish and Hylebos Waterways exhibited consistent evidence of exposure to contaminants. Juvenile chinook salmon from the Snohomish Estuary, which has some urban development, also appeared to be exposed to contaminants, but to a much lesser degree than salmon from the Duwamish and Hylebos Waterways. In addition, when held in tanks with flow-through seawater for a period of several months, juvenile salmon from the Duwamish Estuary exhibited reduced growth and reduced disease resistance when compared to salmon from either the Green River Hatchery (the primary source of salmon for the Duwamish Estuary) or to salmon from the nonurban Nisqually system (Arkoosh *et al.*, 1998; Casillas *et al.*, 1995). Similar effects were observed for juvenile salmon from the Hylebos Waterway (Arkoosh *et al.*, 2001; Casillas *et al.*, 1998). Chemical contaminant exposure in the estuary appeared to place additional stresses on juvenile chinook salmon that could affect their long-term health and survival as they enter the marine environment.

To increase our knowledge of concentrations of chemical contaminants in outmigrant salmon in the Pacific Northwest, we carried out an expanded study from 1996–2001 in which juvenile coho and chinook salmon were collected for contaminant analyses from a number estuaries in Washington and Oregon. Classified by the overall level of development and channel alteration in each estuary (Cortright *et al.*, 1987), the sampling areas included: five deep draft estuaries, with the maximum level channel alteration and urban development (Duwamish Estuary, Columbia River, Grays Harbor, Yaquina Bay, and Coos Bay); two shallow draft estuaries with less extensive channel alteration and some urban and industrial development (Tillamook Bay and Coquille River), four conservation estuaries, where channel alteration is minimal and development is limited (Skokomish Estuary, Nisqually Estuary, Willapa Bay and Alsea Bay); and two natural estuaries, which are largely undeveloped for residential, commercial or industrial uses (Elk River and Salmon River). Predominantly wild fish were collected in the estuaries, although some fish of hatchery origin may have been sampled due to incomplete marking of hatchery fish. Juvenile chinook salmon were also sampled from regional hatcheries to evaluate contaminant uptake during rearing but prior to release. Our results indicate that exposure to chemical contaminants is widespread in outmigrant juvenile chinook and coho salmon, and concentrations in tissues of chinook salmon from several estuaries are high enough to pose a potential threat to their health and survival.

2 Materials and methods

2.1 Collecting juvenile salmon

Juvenile, subyearling chinook salmon were collected from a number of Washington and Oregon estuaries over a 6-year period (1996–2001; Fig. 1; Table 1). The Washington estuaries included: Skokomish and Nisqually Estuaries; Duwamish Estuary, and Grays Harbor and Willapa Bay. The Oregon estuaries included the Columbia, Salmon, Coquille, and Elk Rivers; and Yaquina, Alsea, and Coos Bays. Juvenile coho were also collected from Grays Harbor and Willapa, Yaquina, Alsea, and Coos Bays during 1998 (Fig. 1; Table 1). Due to the pattern of salmon movement in the estuaries, we generally sampled on early morning outgoing tides. Salmon were caught with a beach seine net 36.6 meters in length. The wings of

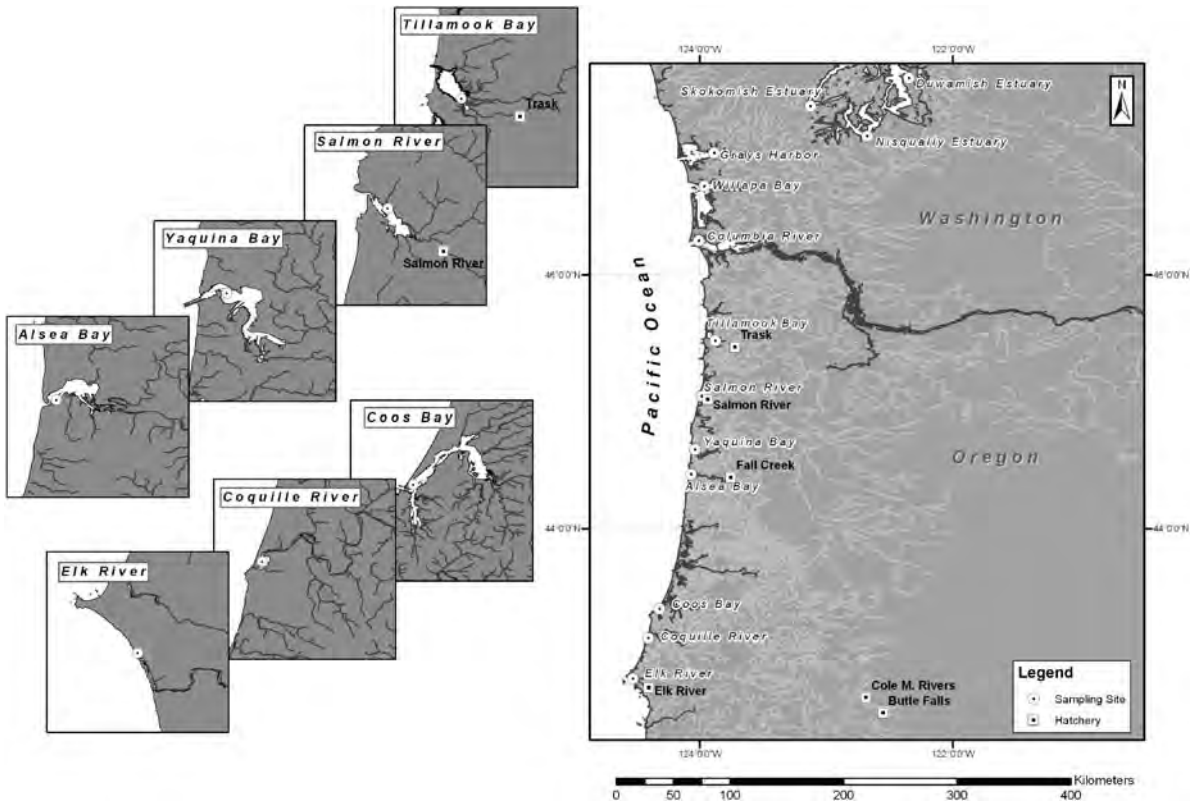

Fig. 1 Locations of hatcheries and estuaries where juvenile coho and chinook salmon were collected

the net were 18 meters long by 2.3 meters deep with 0.6 cm mesh.

Appropriate sampling permits were obtained from the National Marine Fisheries Service (NMFS), and the Oregon and Washington Departments of Fish & Wildlife prior to sampling. To ensure sampling of wild fish instead of hatchery-reared fish we attempted to collect fish from field sites prior to releases from hatcheries or other programs (such as the Salmon and Trout Enhancement Program or STEP). Although a few finclipped hatchery fish were collected and sampled, we did not include these fish in our analyses. Once target salmonids were removed from the net they were placed in insulated aerated tanks and transported live to the nearest laboratory, either the Hatfield Marine Science Center in Newport, Oregon; the University of Oregon's Oregon Institute of Marine Biology in Charleston, Oregon; the U.S. Fish and Wildlife's Olympia Fish Health Center in Olympia, Washington, the Point Adams Field Station in Hammond, Oregon or the Northwest Fisheries Science Center in Seattle, Washington, where they were necropsied within a few hours of collection. Juvenile chinook salmon were also obtained directly from several hatcheries (Fall Creek, Butte Falls, Cole M. Rivers, Elk River, Salmon River, and Trask; see Fig. 1 for locations) to evaluate contaminant uptake during hatchery rearing. Juvenile hatchery coho salmon were not available for sampling at the time of the survey.

Fish to be necropsied were measured (to the nearest mm) and weighed (to the nearest 0.1 g), then sacrificed by a blow to the head. Bile and stomach contents were removed, and composites of 10–15 fish each were generated. Whole gutted bodies from 10 fish were also collected and composited. Bile and stomach contents samples were frozen and stored at $-80\,^{\circ}C$ and whole body samples were frozen and stored at $-20\,^{\circ}C$ until chemical analyses were performed. Sampling sites, dates, and sample types collected are listed in Table 1. Because of limitations associated with fish availability and tissue requirements for analysis, not all samples types could be collected each year from all sites.

Table 1 Sites sampled in Washington and Oregon for juvenile salmonids. Sites were classified by estuary type according to Cortright *et al.* (1987). N = natural estuary; C = conservation estuary; S = shallow draft estuary; D = deep draft estuary NS = not sampled; CH = chinook sampled; CO = coho sampled. wb = whole body sampled; b = bile sampled; s = stomach contents sampled

	1996	1997	1998	1999	2000	2001
WA						
Skokomish Estuary (C)	NS	NS	CH (wb,b)	CH (wb,b)	CH (b)	NS
Duwamish Estuary (D)	NS	NS	CH (wb,b)	CH (wb,b,s)	NS	NS
Nisqually Estuary (C)	NS	NS	CH (wb,b,s)	CH (wb,b,s)	NS	NS
Grays Harbor (D)	NS	NS	CH (wb,b.s) CO (wb,b,s)	CH (wb,b,s)	NS	NS
Willapa Bay (C)	NS	NS	CH (wb,b,s) CO (wb,b,s)	CH (wb,b,s)	NS	NS
Columbia River (D)	NS	NS	CH (wb,s)	CH (wb,b,s)	CH(b,s)	CH (b)
OR						
Salmon River (N)	CH (wb)	NS	CH (b)	CH (wb,s)	CH (wb,s)	CH (wb,s)
Yaquina Bay (D)	NS	NS	CH (wb,b,s) CO (wb,b,s)	CH (wb,b,s)	CH (wb,s)	CH (b) CO (wb,s)
Alsea Bay (C)	CH (wb,b)	NS	CH (wb,s) CO (wb,b,s)	CH (wb,b,s)	CH (wb,b,s)	CH (wb,b,s) CO (wb,s)
Coos Bay (D)	CH (wb)	NS	CH (wb,b,s) CO (wb,b,s)	CH (wb,b,s)	CH (wb,s)	NS
Coquille River (S)	CH (wb)	NS	NS	NS	NS	NS
Elk River (N)	CH (wb)	NS	CH (wb,b.s)	NS	CH (wb,s)	CH (wb,b,s)
Salmon River Hatchery	CH (wb)	NS	NS	NS	NS	NS
Fall Creek Hatchery	CH (wb)	NS	NS	NS	NS	NS
Trask Hatchery	CH (wb)	NS	NS	NS	NS	NS
Butte Falls Hatchery	CH (wb)	NS	NS	NS	NS	NS
Cole M. Rivers Hatchery	CH (wb)	NS	NS	NS	NS	NS
Elk River Hatchery	CH (wb)	NS	CH (wb,s)	NS	NS	NS

2.2 Sample analyses

2.2.1 Organochlorine and aromatic hydrocarbon analyses of composite whole body and stomach content samples

Samples in this study were analyzed using a performance-based measurement system (Telliard, 1999), described in detail by Sloan *et al.* (1993) and updated in Sloan *et al.* (2005). Briefly, after the addition of surrogate standards, samples of up to 3 g were extracted with dichloromethane either by homogenizing in the presence of sodium sulfate (Sloan *et al.*, 1993) or utilizing accelerated solvent extraction (Sloan *et al.*, 2005). For composite whole body samples, a portion of the extract was taken for gravimetric lipid determination. The portion of the extract to be analyzed underwent initial cleanup by filtering through silica gel and neutral alumina, followed by the addition of a recovery standard to determine the fraction of the total extract analyzed. After further sample cleanup using high-performance liquid chromatography with size-exclusion chromatography, the sample fraction containing organochlorines (OCs) and 2–6 ring aromatic hydrocarbons was collected. The fraction was reduced in volume, a GC standard was added, and the sample was analyzed using high-resolution gas chromatography coupled with electron capture detection (samples analyzed for OCs 1996–1998; Sloan *et al.*, 1993) or mass spectrometry with selected-ion monitoring (samples analyzed for OCs 1999–2001; Sloan *et al.*, 2005) with 5–10 levels of calibration standards. Concentrations of aromatic hydrocarbons (stomach contents samples only) were analyzed in all sampling years by high-resolution gas chromatography with mass spectrometry using selected ion monitoring and 5–6 levels of calibration standards. Quality assurance measures included analysis of a certified reference material and a

laboratory blank with each batch of samples. Performance criteria were met for all samples and sample batches.

Analyses for OCs included individual PCB (polychlorinated biphenyl) congeners, DDTs, chlordanes, lindane, aldrin, dieldrin and mirex. PCBs measured over all years included a standard list of 17 congeners (IUPAC numbers 18, 28, 44, 52, 95, 101, 105, 118, 128, 138, 153, 170, 180, 187, 195, 206, and 209). Total PCBs was calculated by summing the concentrations of these individual congeners and multiplying the result by two. This formula provides a good estimate of the total PCBs in a typical environmental sample of sediments or animals feeding on lower trophic levels, where a mixture of Aroclors 1254 and 1260 is the predominant pattern (Lauenstein *et al.*, 1993). Summed DDTs (ΣDDTs) levels were calculated by summing the concentrations of *o,p′*- and *p,p′*-DDD, *o,p′*- and *p,p′*-DDE, and *o,p′*- and *p,p′*-DDT. Summed chlordanes (ΣCHLDs) were calculated by summing the concentrations of heptachlor, heptachlor epoxide, γ-chlordane, α-chlordane, oxychlordane, *cis*-nonachlor, *trans*-nonachlor and nonachlor III. Summed low molecular weight aromatic hydrocarbons (ΣLAHs) were determined by adding the concentrations of biphenyl, naphthalene, 1-methylnaphthalene, 2-methylnaphthalene, 2,6-dimethylnapthalene, acenaphthene, fluorene, phenanthrene; 1-methylphenanthrene, and anthracene. Summed high molecular weight aromatic hydrocarbons (ΣHAHs) were calculated by adding the concentrations of fluoranthene, pyrene, benz[a]anthracene, chrysene, benzo[a]pyrene, benzo[e]pyrene, perylene, dibenz[a,h]anthracene, benzo[b]fluoranthene, benzo[k]fluoranthene, indenopyrene, and benzo[ghi]perylene. Summed total aromatic hydrocarbons (ΣAHs) were calculated by adding ΣHAHs and ΣLAHs.

2.2.2 PAH metabolites in bile

Composite samples of bile were analyzed by high-performance liquid chromatography with fluorescence detection (HPLC/uvf) for aromatic hydrocarbon (AH) metabolites as described in Krahn *et al.* (1986). In brief, bile was injected directly onto a C18 reverse-phase column (Phenomenex Synergi Hydro) and eluted with a linear gradient from 100% water (containing a trace amount of acetic acid) to 100% methanol at a flow of 1.0 mL/min. Chromatograms were recorded at the following wavelength pairs: 1) 260/380 nm where several 3–4 ring compounds (e.g., phenanthrene) fluoresce and 2) 380/430 nm where 4–5 ring compounds (e.g., benzo[a]pyrene) fluoresce. Peaks eluting after 5 minutes were integrated and the areas of these peaks were summed. The concentrations of fluorescent AHs in bile were determined using phenanthrene (PHN) and benzo[a]pyrene (BaP) as external standards and converting the fluorescence response of bile to phenanthrene (ng PHN equivalents/g bile), and benzo[a]pyrene (ng BaP equivalents/g bile) equivalents. Bile metabolites fluorescing at phenanthrene wavelengths were considered an indicator of exposure to low molecular weight PAHs, while metabolites fluorescing at benzo[a]pyrene (BaP) wavelengths were considered as an indicator of exposure to high molecular weight PAHs.

2.2.3 Statistical methods

Statistical analyses were conducted with the Statview©statistical software package (SAS Institute, Inc., Cary, NC, USA). Temporal and intersite differences in tissue, stomach contents, and bile contaminant concentrations were determined by ANOVA. Data were log-transformed as necessary to achieve a normal distribution. The significance level for all analyses was set at $\alpha = 0.05$.

3 Results

3.1 Lipid content in whole bodies

Lipid content (as total extractable organics) in bodies of chinook salmon collected from the estuaries varied from 0.8% in fish from Tillamook Bay to 3.5% in fish from Coquille River, with an average concentration of 2.4% (Fig. 2; Table 2). Lipid levels in juvenile coho salmon were slightly lower, with an average concentration of 1.2% (Fig. 2; Table 2), but not significantly different than levels in estuarine chinook salmon (ANOVA, $p = 0.08$). Lipid concentrations in hatchery chinook salmon were significantly higher than in estuary chinook (ANOVA, $p = 0.001$), with an average concentration of 7.9% (Fig. 2; Table 2). The number of samples collected (typically one composite per site or hatchery) was too small for intersite or interhatchery differences to be meaningfully evaluated,

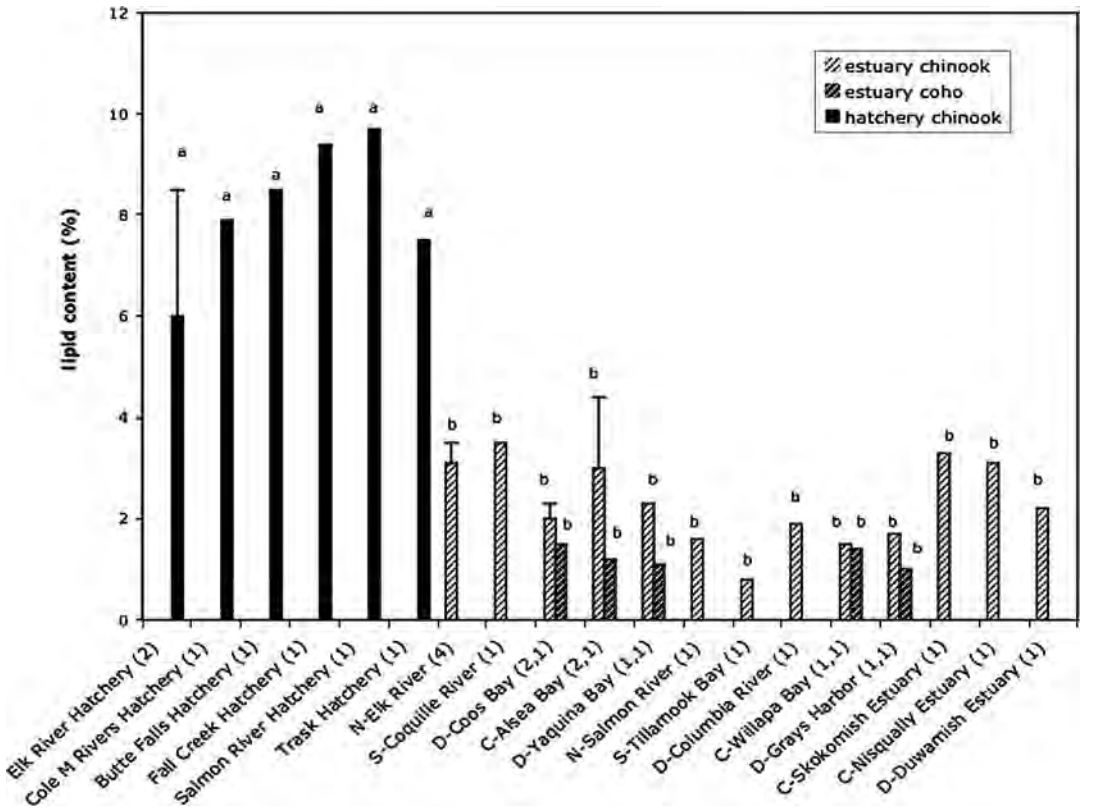

Fig. 2 Mean lipid content (%, as total extractable organics, ± SE) in whole bodies of chinook and coho salmon from Pacific Northwest estuaries and juvenile chinook salmon from associated hatcheries. N = natural estuary; C = conservation estuary; S = shallow draft estuary; D = deep draft estuary. Numbers in parentheses indicate number of composite samples (10–15 fish each) analyzed per site or group. Measurements with different letters are significantly different (ANOVA, $p < 0.05$)

but concentrations tended to be fairly uniform within the sampling groups (i.e, estuarine chinook, estuarine coho, and hatchery chinook).

3.2 Organochlorine contaminants in whole bodies

Concentrations of PCBs in whole bodies of estuarine chinook salmon (Fig. 3, Tables 2 and 3) were quite variable, ranging from ∼500 ng/g lipid weight (lw) in salmon from Elk River and Coquille Estuaries to 3100 ng/g lw in salmon from the Duwamish Estuary in Seattle (or from 3.6 ng/g wet weight (ww) at Salmon River to 103 ng/g ww at Duwamish). The lowest concentrations of PCBs were found in chinook salmon from Elk River Estuary, Coquille River, Alsea Bay Estuary, Salmon River, and Tillamook Bay; wet weight PCB concentrations were less than 20 ng/g ww at all these sites, and lipid weight PCB concentrations were below 600 ng/g lw in chinook from Elk River Estuary, Coquille River, and Tillamook. The highest PCB concentrations (2500–3100 ng/g lw or 45–103 ng/g ww) were found in salmon from Yaquina Bay, the Columbia River, and the Duwamish Estuary.

Concentrations of PCBs in juvenile coho salmon (Fig. 3, Tables 2 and 3) tended to be lower than those in chinook salmon. At sites where both species were collected, the mean PCB concentration overall was significantly lower in coho than in chinook on both a lipid weight and wet weight basis (1030 vs. 1650 ng/g lw, $p = 0.018$; 10 vs. 30 ng/g ww; $p = 0.0026$). No significant differences were observed in PCB concentrations in coho salmon from different sampling sites, but the number of samples was very small.

The mean concentration of PCBs in juvenile chinook salmon from hatcheries (Fig. 3, Tables 2 and 3) was relatively low on a lipid weight basis (620 ng/g lw),

Table 2 Contaminant concentration mean values (± SE), ranges, and sites where high and low values were observed in juvenile chinook and coho salmon from Pacific Northwest estuaries and juvenile chinook salmon from Pacific Northwest hatcheries. Values with different superscripts are significantly different (ANOVA, $p = 0.05$) in estuarine chinook, estuarine coho, and hatchery chinook

	Estuaries		Hatcheries
	Chinook	Coho	Chinook
% lipid	2.4 ± 0.2 (n = 19)[a]	1.2 ± 0.1 (n = 5)[a]	7.9 ± 0.8 (n = 7)[b]
	0.8–3.5.%	1.1–1.5%	6–9.7%[1]
	Tillamook–Coquille	Grays Hbr.-Coos	Elk–Salmon
Body PCBs	27 ± 4 (n = 65)[a]	9.7 ± 1.6 (n = 9)[b]	46 ± 3 (n = 7)[c]
(ng/g wet wt)	3.6–103	6–16	39–59
	Salmon–Duwamish	Alsea–Grays Hbr.	Trask–Salmon
Body PCBs	1650 ± 190 (n = 19)[a]	1030 ± 230 (n = 5)[a]	620 ± 50 (n = 7)[b]
(ng/g lipid)	516–3099	470–1564	521–760
	Elk R.–Duwamish	Willapa-Grays Hbr.	Fall Cr.–Elk
Body DDTs	13 ± 2 (n = 65)[a]	1.7 ± 0.3 (9)[b]	34 ± 3 (7)[c]
(ng/g wet wt)	0.5–41	0.9–3.4	27–45
	Tillamook–Columbia.	Willapa-Grays Hbr.	Trask–Salmon
Body DDTs	550 + 120 (n = 19)	140 + 50 (n = 5)	436 + 234 (n = 7)
(ng/g lipid)	62–2280	66–333	354–507
	Tillamook–Columbia	Willapa-Grays Hbr.	Trask–Elk
Whole body	0.63 ± 0.06 (n = 65)[a]	0.21 ± 0.03 (n = 9)[b]	0.72 ± 0.03 (n = 7)[a]
DDT/PCB ratio	0.10–1.1	0.13–0.26	0.68–0.75
	Tillamook–Salmon	Coos-Alsea	Elk/Trask–Salmon
FACs-BaP	364 ± 96 (n = 47)	218 ± 26 (n = 10)	ND
(ng/g bile)	108–1925	136–298	
	Alsea–Duwamish	Yaquina–Grays Hbr.	
FACs-PHN	44600 ± 15900 (n = 47)	17600 ± 2040 (n = 10)	
(ng/g bile)	9270–359000	12900–25400	ND
	Nisqually-Duwamish	Yaquina–Coos Bay	
Stomach contents	18.6 ± 5.7 (n = 35)	11.6 ± 2.5 (n = 9)	13 (n = 1)
PCBs	4.5–200	5.4–22	
(ng/g wet wt)	Salmon–Duwamish	Alsea–Grays Hbr.	Elk
Stomach contents	8.3 ± 2.9 (n = 35)	1.5 ± 0.4 (n = 9)	4.5 (n = 1)
DDTs	0.6–45	0.9–2.3	
(ng/g wet wt)	Elk.–Grays Hbr.	Alsea–Grays Hbr.	Elk
Stomach contents	415 ± 235 (n = 35)[a]	40 ± 19 (n = 9)[b]	28 (n = 1)[b]
ΣLAHs	12–8000	10–69	
(ng/g wet wt)	Elk-Duwamish	Coos Bay-Alsea Bay	Elk
Stomach contents	594 ± 353 (n = 35)[a]	5.4 ± 1.7 (n = 35)[b]	5 (n = 1)[b]
ΣHAHs	1.3–6300	1.3–10	
(ng/g wet wt)	Elk/Salmon-Willapa	Coos Bay–Grays Hbr.	Elk

comparable to concentrations observed in estuary chinook and coho salmon from rural estuaries (e.g., Elk River, Coquille River, Alsea Bay). On a wet weight basis, however, the mean PCB concentration in hatchery chinook was quite high (47 ng/g ww), comparable to concentrations in moderately to heavily urbanized estuaries (Table 3).

Concentrations of ΣDDTs in estuarine chinook salmon bodies ranged from 62 ng/g lw at Tillamook Bay to 2280 ng/g lw in the Columbia River (or from below 0.5 ng/g ww in fish from Tillamook Bay to 41 ng/g ww in fish from the Columbia River) (Fig. 4, Tables 2 and 3), with a mean concentration of 550 ng/g lw or 13 n/g ww (Fig. 4; Tables 2 and 3). Concentrations

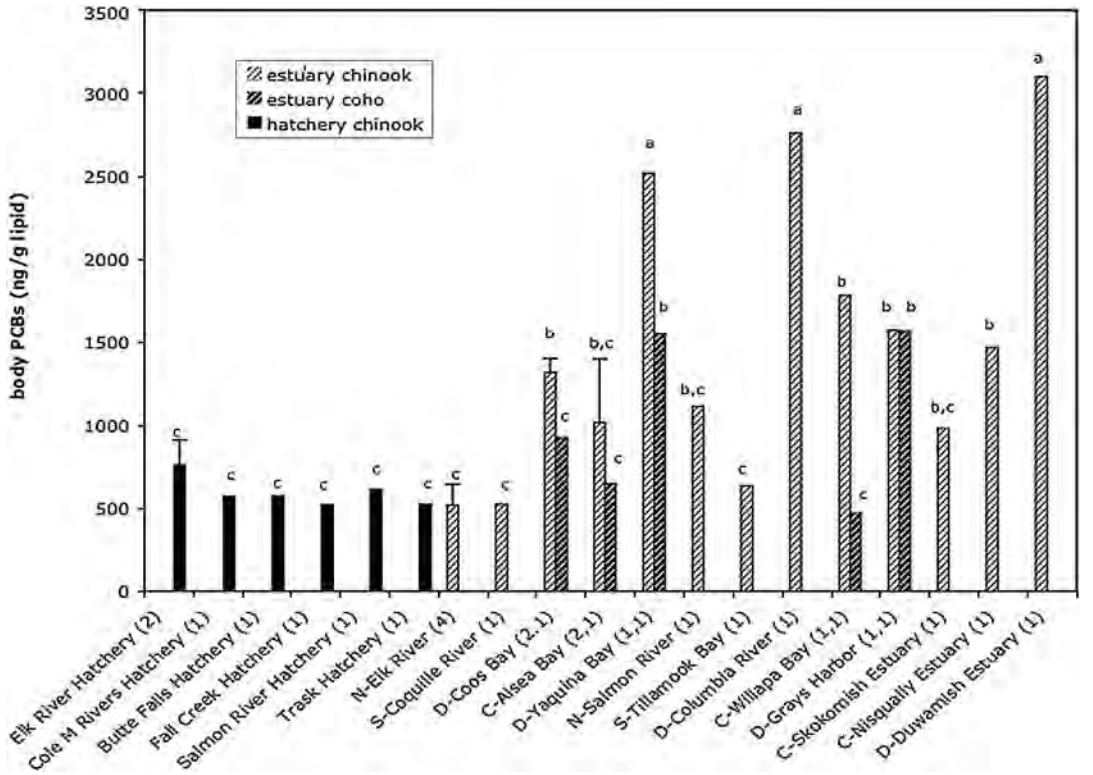

Fig. 3 Mean concentrations of ΣPCBs (ng/g lipid, ± SE) in whole bodies of juvenile chinook and coho salmon from Pacific Northwest Estuaries and juvenile chinook salmon from associated hatcheries. N = natural estuary; C = conservation estuary; S = shallow draft estuary; D = deep draft estuary. Numbers in parentheses indicate number of composite samples (10–15 fish each) analyzed per site or group. Measurements with different letters are significantly different (ANOVA, $p < 0.05$)

of ΣDDTs were low in fish from Tillamook Bay, Alsea Bay, and Elk River on both a wet wt and lipid wt basis (below 250 ng/g lw and 5 ng/g ww); at Coquille River lipid wt DDT concentrations were comparable but wet wt concentrations were higher, while the reverse was true for chinook from Salmon River. Concentrations of ΣDDTs were relatively high (over 1000 ng/g lw or 25 ng/g ww) in fish from the Nisqually, Duwamish, and Columbia River Estuaries. Fish with the highest ΣDDT concentrations were from the Columbia River, where levels were over 2200 ng/g lw or 40 ng/g ww.

In juvenile coho salmon, the maximum ΣDDT concentration was 333 ng/g lw or 3.4 n/g ww in fish from Grays Harbor (Fig. 4; Tables 2 and 3), while the mean concentration was 140 ng/g lw or 1.7 ng/g ww. When coho and chinook salmon collected from the same sites were compared, ΣDDT concentrations were much lower in coho salmon (1.7 ± 0.3 ng/g ww vs. 8.8 ng/g ww, $p = 0.0026$; or 137 ng/g lw vs. 551 ± 95 ng/g lw, $p \leq 0.001$).

On a wet weight basis, concentrations of ΣDDTs in whole bodies of juvenile Chinook collected from the hatcheries were fairly high, with the mean concentrations for all hatcheries significantly above the mean concentrations measured in estuarine chinook and coho (Tables 2 and 3). However, because of the high lipid content of the hatchery fish, their whole body ΣDDT concentrations on a lipid weight basis were more moderate (400–500 ng/g lw), and did not differ significantly from mean concentrations in estuarine salmon (Fig. 4; Tables 2 and 3).

Of the six DDTs measured in salmon whole bodies, *p*,*p*′-DDE predominated in whole bodies of both coho and chinook salmon from all estuaries and hatcheries sampled, accounting for 75–100% of DDTs measured (Fig. 5; Table 3). The second most prominent DDT was *p*,*p*′-DDD; it accounted for 10–20% of DDTs measured in chinook and coho salmon from most sites. Additionally, *p*,*p*′-DDT was present at several sites, accounting for 3–6% of total DDTs in chinook salmon

Table 3 Mean concentrations (± SE) in ng/g, wet wt of ΣPCBs, ΣDDTs, and DDT isomers in whole bodies of juvenile chinook and coho salmon collected from Pacific Northwest estuaries and juvenile chinook salmon from Pacific Northwest hatcheries. Compounds were measured by GC/ECD in samples collected from 1996–1998 and by GC/MS in samples collected from 1999–2001. Values with different letter superscripts are significantly different (ANOVA, $p \leq 0.05$)

Site	ΣPCBs	ΣDDTs	*o,p′*-DDD	*o,p′*-DDE	*o,p′*-DDT	*p,p′*DDD	*p,p′*-DDE	*p,p′*-DDT
Estuary chinook								
Columbia River (6)	50 ± 14^{b}	41 ± 3^{a}	0.6 ± 0.1^{a}	0.27 ± 0.0^{a}	0.71 ± 0.15^{a}	6.2 ± 0.64^{a}	31 ± 2.3^{a}	2.4 ± 0.6^{a}
Alsea Bay (8)	11 ± 3^{c}	2.4 ± 0.5^{d}	$<$DL^{b}	0.05 ± 0.05^{b}	$<$DL^{c}	0.32 ± 0.25^{b}	2.8 ± 0.8^{c}	0.11 ± 0.09^{b}
Elk River (2)	9.9 ± 3.9^{c}	4.7 ± 2.6^{d}	0.04 ± 0.03^{b}	$<$DL^{b}	0.02 ± 0.03^{c}	0.5 ± 0.4^{b}	4.1 ± 2.1^{c}	0.21 ± 0.15^{b}
Grays Harbor (3)	$27 \pm 8^{b,c}$	11.3 ± 4^{c}	0.07 ± 0.07^{b}	$<$DL^{b}	$<$DL^{c}	1.1 ± 0.6^{b}	9.9 ± 3.3^{b}	0.1 ± 0.1^{b}
Salmon River (11)	3.6 ± 1.6^{c}	1.9 ± 0.5^{d}	$<$DL^{b}	$<$DL^{b}	$<$DL^{c}	0.16 ± 0.09^{b}	1.7 ± 0.4^{c}	0.11 ± 0.06^{b}
Skokomish Estuary (3)	$29 \pm 2^{b,c}$	19.9 ± 1.5^{b}	0.08 ± 0.08^{b}	$<$DL^{b}	0.05 ± 0.05^{c}	1.9 ± 0.15^{b}	17.3 ± 1.2^{b}	0.27 ± 0.18^{b}
Willapa Bay (3)	$24^{b,c}$	12.3 ± 0.4^{c}	<DLb	$<$DL^{b}	$<$DL^{c}	0.62 ± 0.14^{b}	11.2 ± 0.7^{b}	0.14 ± 0.14^{b}
Yaquina Bay (7)	46 ± 1^{b}	7.8 ± 2.2^{d}	$<$DL^{b}	$<$DL^{b}	0.07 ± 0.07^{b}	0.48 ± 0.11^{b}	6.8 ± 1.8^{b}	0.41 ± 0.14^{b}
Coos Bay (3)	$22 \pm 3^{b,c}$	10.8 ± 1.3^{c}	$<$DL^{b}	$<$DL^{b}	0.02 ± 0.02^{c}	0.59 ± 0.09^{b}	9.8 ± 1.1^{b}	0.45 ± 0.12^{b}
Duwamish Estuary (3)	103 ± 29^{a}	27 ± 1^{b}	0.36 ± 0.03	0.18 ± 0.09^{a}	$0.09 \pm .09^{b}$	3.5 ± 0.4^{a}	22 ± 0.6^{a}	0.61 ± 0.14^{b}
Nisqually Esuary (3)	40 ± 4^{b}	30 ± 4^{b}	0.26 ± 0.03	0.09 ± 0.09^{b}	0.04 ± 0.04^{c}	3.4 ± 0.5^{a}	26 ± 3.5^{a}	0.34 ± 0.09^{b}
Coquille River (1)	$18^{b,c}$	$9.2^{c,d}$	$<$DL^{b}	$<$DL^{b}	$<$DL^{c}	1.3^{b}	7.3^{b}	0.58^{b}
Tillamook Bay (1)	5.1^{c}	0.5^{d}	$<$DL^{b}	$<$DL^{b}	$<$DL^{c}	$<$DL^{b}	0.47^{c}	$<$DL
Hatchery chinook								
Fall Creek (1)	49^{b}	39^{a}	0.51^{a}	$<$DL^{b}	0.03^{c}	5.4^{a}	32^{a}	1.3^{a}
Butte Falls (1)	49^{b}	35^{a}	0.56^{a}	$<$DL^{b}	$<$DL^{c}	4.9^{a}	28^{a}	1.5^{a}
Cole M. Rivers (1)	45^{b}	31^{a}	0.8^{a}	$<$DL^{b}	0.09^{b}	6.1^{a}	22^{a}	2.0^{a}
Elk River (2)	42^{b}	30 ± 10^{b}	0.04^{b}	$<$DL^{b}	0.21^{a}	4.2^{a}	23^{a}	1.7^{a}
Salmon River (1)	59^{b}	45^{a}	0.9^{a}	$<$DL^{b}	0.26^{a}	8.3^{a}	32^{a}	3.0^{a}
Trask (1)	39^{b}	27^{b}	0.67^{a}	$<$DL^{b}	$<$DL^{c}	4.5^{a}	20^{a}	1.3^{a}
Estuary Coho								
Alsea Bay (3)	5.9 ± 1^{c}	1.4 ± 0.2^{d}	$<$DL^{b}	$<$DL^{b}	$<$DL^{c}	0.08 ± 0.04^{b}	1.3 ± 0.2^{c}	$<$DL^{b}
Coos Bay (1)	14^{c}	1.8^{d}	$<$DL^{b}	$<$DL^{b}	$<$DL^{c}	$<$DL^{b}	1.8^{c}	$<$DL^{b}
Grays Harbor (1)	$27^{b,c}$	3.4^{d}	$<$DL^{b}	$<$DL^{b}	$<$DL^{c}	0.26^{b}	3.0^{c}	0.13^{b}
Willapa Bay (1)	6.4^{c}	0.9^{d}	$<$DL^{b}	$<$DL^{b}	$<$DL^{c}	0.13^{b}	0.63^{c}	0.12^{b}
Yaquina Bay (3)	11^{c}	1.7 ± 0.4^{d}	$<$DL^{b}	$<$DL^{b}	$<$DL^{c}	0.13 ± 0.07^{b}	1.6 ± 0.4^{c}	0.4 ± 0.02^{b}

from the Columbia River, Yaquina Bay, Grays Harbor, and Salmon River, 4% of total DDTs in juvenile coho from Grays Harbor, and 13% of total DDTs in coho from Willapa Bay. In hatchery chinook salmon, *p,p′*-DDT accounted for an average of 5% of total DDTs. Concentrations of estrogenic *o,p′*-DDT, *o,p′*-DDD, and *o,p′*-DDE (Fig. 6) were below detection limits in all coho and many chinook salmon sampled, but were present at concentrations above 0.1 ng/g ww or 10 n/g lw in chinook salmon from the Columbia, Nisqually, Duwamish and Yaquina Bay Estuaries. As with ΣDDTs, concentrations of the *o,p′* isomers were highest in chinook from the Columbia River. In hatchery chinook salmon, they averaged 8 ng/g lw.

We calculated the ΣDDTs/ΣPCBs ratios in whole body samples of chinook and coho salmon to identify groups of fish with distinct contaminant profiles (Fig. 7). In coho salmon, the mean ΣDDTs/ΣPCBs ratio was 0.2, and in estuarine chinook salmon, the mean ratio was 0.4. In both coho and chinook salmon from most of the sites we sampled (Nisqually, Skokomish, Coos Bay, Alsea Bay Estuary, Salmon River Estuary, Willapa Bay, Elk River Estuary, Duwamish Estuary, Tillamook Bay, Yaquina Bay), ΣDDT/ΣPCB ratios were 0.5 or lower. This was not true, however, of chinook salmon from the Columbia River, whose ΣDDTs/ΣPCBs ratios were 1.0–1.1. In hatchery chinook, the mean ΣDDTs/ΣPCBs ratio was ∼0.7.

In addition to PCBs and DDTs, chlordanes, hexachlorobenzene, and dieldrin were detected in whole bodies of estuarine chinook and coho salmon from one or more sampling sites, but at much lower concentrations than PCBs or DDTs (mean concentrations ranging from <1 ng/g ww to 4 ng/g ww; Table 4). Of the

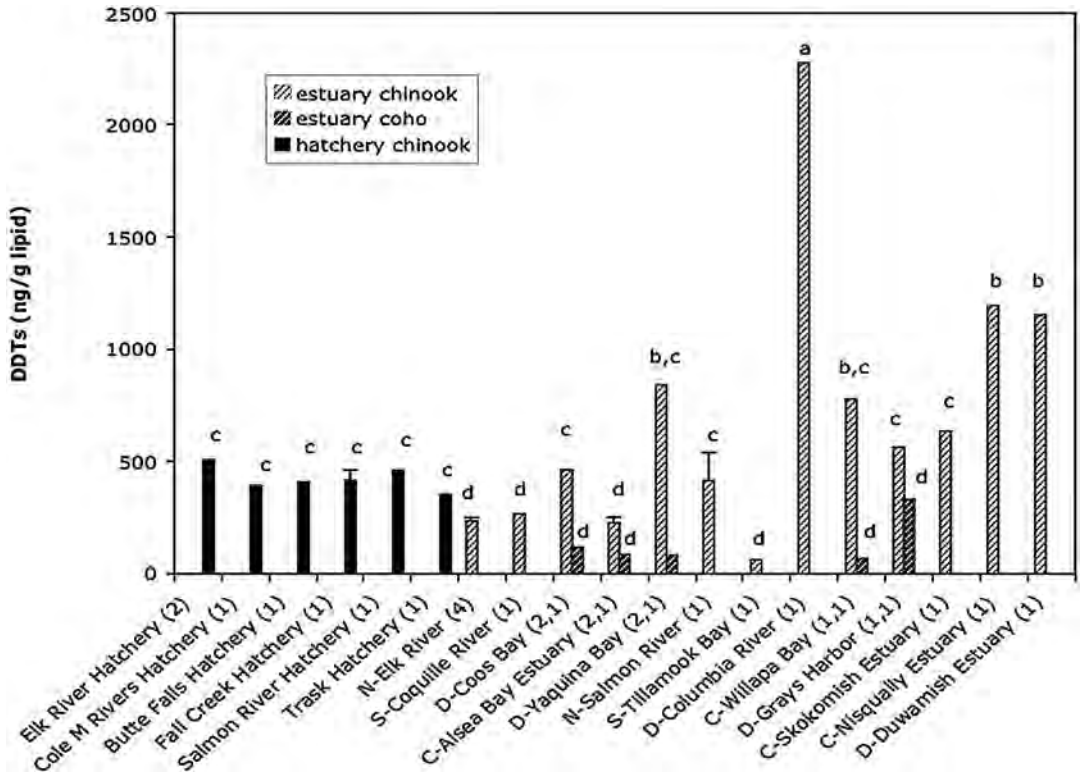

Fig. 4 Mean concentrations of ΣDDTs (ng/g lipid, ± SE) in whole bodies of juvenile chinook and coho salmon from Pacific Northwest estuaries and juvenile chinook salmon from associated hatcheries. N = natural estuary; C = conservation estuary; S = shallow draft estuary; D = deep draft estuary. Numbers in parentheses indicate number of composite samples (10–15 fish each) analyzed per site or group. Measurements with different letters are significantly different (ANOVA, $p < 0.05$)

pesticides detected, chlordanes were generally found at the highest concentrations. Other OC pesticides (i.e., lindane, mirex and aldrin) were below the limits of detection (generally <0.5 ng/g ww) in all samples. Dieldrin, chlordanes, and HCB were detected in whole bodies of juvenile chinook from all sampled hatcheries, typically at concentrations in the 1–5 ng/g ww range. Concentrations were comparable to the highest levels reported in estuarine chinook and coho (Table 4).

3.3 Bile metabolites

Levels of high molecular weight AH metabolites in bile (FACs-BaP) were low to moderate (100–400 ng/g bile) in juvenile fall chinook and coho salmon collected from most of the estuaries sampled along the Washington and Oregon Coast (Fig. 8). Concentrations in chinook salmon from the Duwamish Estuary (∼1930 ng BaP equiv/g bile) were significantly higher than in fish from any other sites. FAC-BaP levels were also somewhat elevated (350–500 ng/g bile) in chinook salmon from the Columbia River, Skokomish Estuary, Grays Harbor, and Willapa Bay, and in coho salmon from Grays Harbor. Lowest concentrations were observed in chinook and coho salmon from Elk River Estuary, Yaquina Bay Estuary, and Alsea Bay Estuary. At 100–200 ng BaP equiv/g bile, concentrations of FACs-BaP in fish at these sites were significantly lower than in chinook salmon from the Columbia, Skokomish, Willapa Bay, and Duwamish sites, and in chinook and coho salmon from Grays Harbor.

Concentrations of metabolites of low molecular weight PAHs (FAC-PHN; Fig. 8) were also significantly higher in chinook salmon from the Duwamish Estuary (359,000 ng PHN equiv/g bile) than in fish from any other sites. Concentrations in chinook salmon from Grays Harbor, Coos Bay, and the Columbia River (60,000–70,000 ng PHN equiv/g bile) were much lower than in the Duwamish chinook, but significantly above levels in either coho or

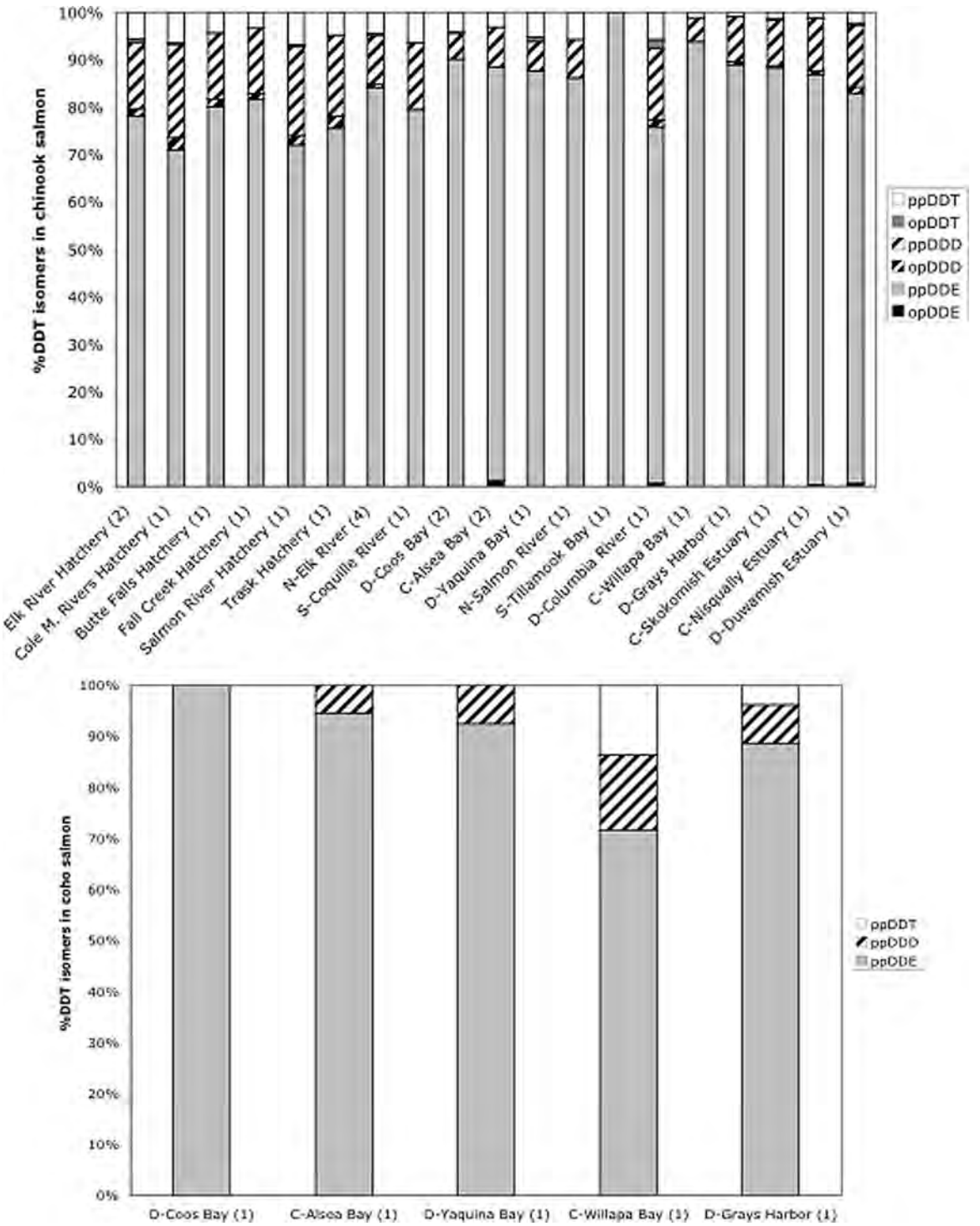

Fig. 5 Proportions of various DDTs in composite whole body samples of juvenile chinook and coho salmon collected from Pacific Northwest estuaries and hatcheries. N = natural estuary; C = conservation estuary; S = shallow draft estuary; D = deep draft estuary. Numbers in parentheses indicate number of composite samples (10–15 fish each) analyzed per site or group

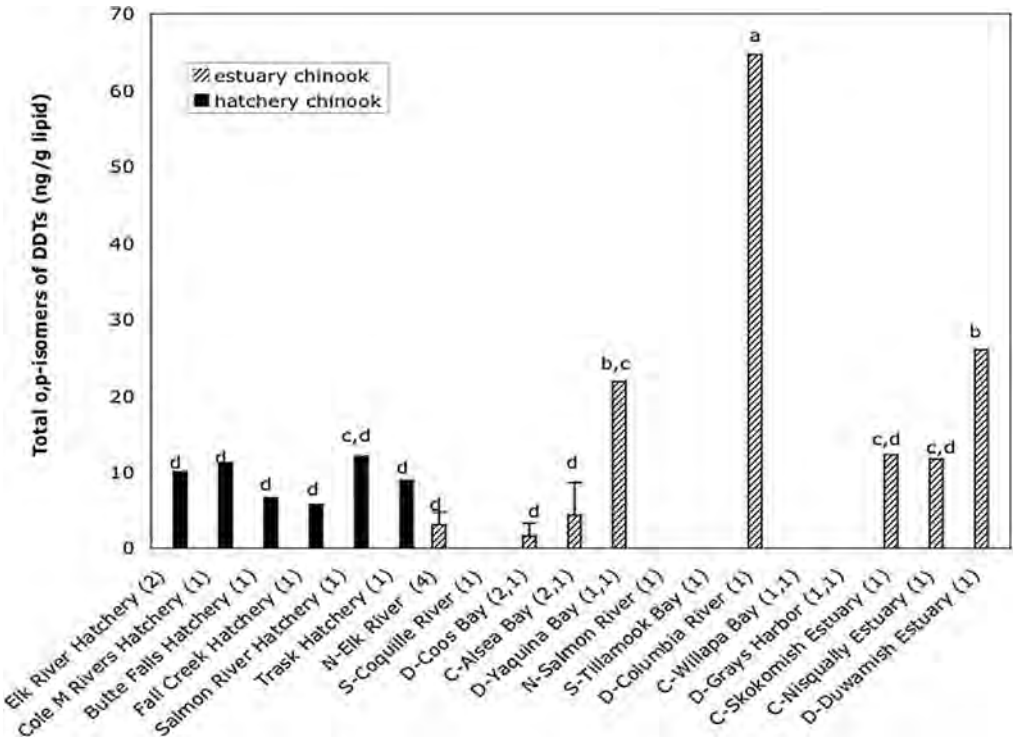

Fig. 6 Mean concentrations of Σ*o*, *p*′-isomers of DDTs (ng/g lipid, ± SE) in whole bodies of juvenile chinook and coho salmon from Pacific Northwest estuaries and juvenile chinook salmon from associated hatcheries. N = natural estuary; C = conservation estuary; S = shallow draft estuary; D = deep draft estuary. Numbers in parentheses indicate number of composite samples (10–15 fish each) analyzed per site or group. Measurements with different letters are significantly different (ANOVA, $p < 0.05$). Values were below detection limits for coho from all sites where they were sampled, and for chinook from Coquille River

chinook salmon from the other sampling sites, whose biliary FACs-PHN concentrations were 30,000 ng PHN equiv/g bile or less. Bile sample could not be collected from chinook salmon at the hatcheries.

3.4 Contaminants in stomach contents

Several classes of contaminants, including PCBs, DDTs, and low and high molecular weight PAHs, were present at detectable concentrations in stomach contents of outmigrant juvenile chinook and coho salmon. Concentrations of ΣLAHs in stomach contents of estuarine chinook salmon (Fig. 9; Table 2) ranged from 12 ng/g ww at the Elk River Estuary to 8000 ng/g ww at the Duwamish Estuary. Concentrations of ΣLAHs were also fairly high in fish from Willapa Bay, Yaquina Bay, and Grays Harbor in comparison to other sites, ranging from 350 to 1400 ng/g ww. Concentrations of ΣLAHs in stomach contents of chinook and coho salmon from all other sites were <100 ng/g ww (Fig. 9; Table 2). At sites where both species were collected, average ΣLAH concentrations in stomach contents of chinook salmon were higher than in coho salmon (920 ng/g ww vs. 5 ng/g ww). In chinook salmon from Elk River Hatchery, the concentration of ΣLAHs in stomach contents was 28 ng/g ww (Fig. 9; Table 2).

Concentrations of ΣHAHs in stomach contents of juvenile chinook salmon (Fig. 9, Table 2) were highest in fish from the Duwamish Estuary and Willapa Bay (6000–6300 ng/g ww). Concentrations of ΣHAHs at Grays Harbor and Yaquina Bay (330–340 ng/g ww) were also relatively high in comparison to other sites, where concentrations were ∼20 ng/g ww and below. The lowest levels ΣHAHs (1–2 ng/g ww) were observed in chinook from Salmon River and Elk River Estuary sites. In coho salmon (Fig. 9; Table 2) concentrations of ΣHAHs in stomach contents were ∼10 ng/g ww or below in fish from all sites; at sites where both species were collected, ΣHAH concentrations were higher in chinook salmon than in coho salmon (323 ng/g ww vs. 40 ng/g ww). In chinook and coho salmon from most sampling sites, HAHs accounted for

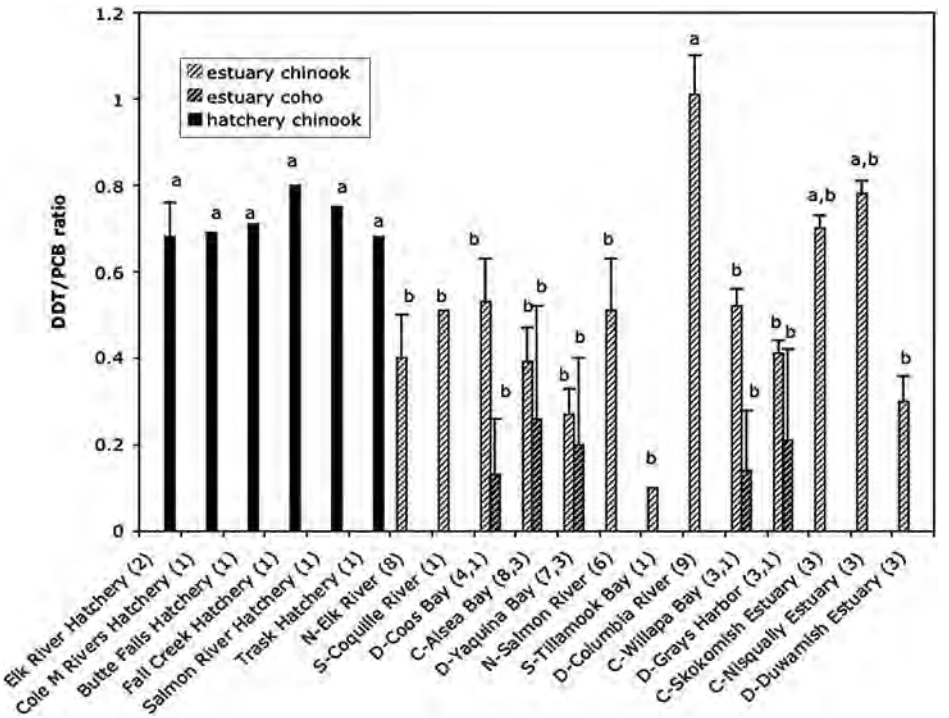

Fig. 7 Mean ΣDDT/ΣPCB ratios (± SE) in whole bodies of juvenile chinook and coho salmon from Pacific Northwest estuaries and juvenile chinook salmon from associated hatcheries. N = natural estuary; C = conservation estuary; S = shallow draft estuary; D = deep draft estuary. Numbers in parentheses indicate number of composite samples (10–15 fish each) analyzed per site or group. Measurements with different letters are significantly different (ANOVA, $p < 0.05$)

10–20% of total AHs. However, in chinook salmon from the Duwamish, Grays Harbor, Yaquina Bay, and Willapa Bay, HAHs were more predominant, accounting for 30–70% of total AHs. In chinook salmon from the Elk River Hatchery (Fig. 9), ΣHAH concentrations were relatively low (5 ng/g ww) and accounted for about 15% of total AHs.

Concentrations of ΣPCBs in stomach contents of estuarine chinook salmon (Fig. 10; Table 2) ranged from 5 ng/g ww in fish from the Salmon River Estuary to 200 ng/g ww in fish from the Duwamish Estuary. Concentrations of PCBs in salmon from the Columbia River and Grays Harbor were about 40 ng/g ww, and concentrations were about 20 ng/g ww or less at all other sampling sites. Lowest levels (5–10 ng/g ww) were observed at Yaquina Bay, Alsea Bay, Coos Bay, Elk River, and Salmon River Estuaries. In coho salmon (Fig. 10, Table 2), PCB concentrations in stomach contents ranged from 5 ng/g ww in fish from Alsea Bay Estuary to 22 ng/g ww in fish from Willapa Bay. At sites where both species were collected, PCB concentrations were similar in stomach contents of chinook salmon and coho salmon, 14 ng/g ww vs. 12 ng/g ww. At the Elk River Hatchery, PCB concentrations in stomach contents were 13 ng/g ww, comparable to levels in estuarine chinook salmon from non-urban sites (Fig. 10; Table 2).

Concentrations of ΣDDTs in stomach contents of estuarine chinook salmon (Fig. 11; Table 2) were highest in fish from Grays Harbor (45 ng/g ww) and the Columbia River (39 ng/g ww), significantly higher than in fish from all other sites. In stomach contents of chinook from all sampling sites except for the Columbia River and Grays Harbor, ΣDDT concentrations were <10 ng/g ww. Concentrations of ΣDDTs in stomach contents of coho salmon (Fig. 11, Table 2) were low (3 ng/g ww) in fish from all sites. At sites where both species were collected, ΣDDT concentrations were higher in chinook salmon than in coho salmon (9 ng/g ww vs. 1.5 ng/g ww). In chinook salmon from the Elk River Hatchery (Fig. 11, Table 2), concentrations of DDTs were also relatively low, 4.5 ng/g ww.

In stomach contents, as in tissues, p,p'-DDE was the predominant isomer detected, accounting for about

Table 4 Mean concentrations (± SE) in ng/g, wet wt of selected organochlorine pesticides in bodies of juvenile chinook and coho salmon collected from Pacific Northwest estuaries and hatcheries. Σchlordanes = summed concentrations of heptachlor, heptachlor epoxide, γ-chlordane, α-chlordane, *cis*-nonachlor, *trans*-nonachlor and nonachlor III. DL = detection limit. Pesticides were measured by GC/ECD in samples collected from 1996–1998 and by GC/MS in samples collected from 1999–2001. Values with different letter superscripts are significantly different (ANOVA, $p < 0.05$). Lindane was also measured, but was below DL (generally < 0.5 ng/g ww) in all samples

Site	dieldrin	aldrin	Σchlordanes	HCB	Mirex
Estuary Chinook					
Columbia River (6)	1.9 ± 0.88^{a}	$<DL^{b}$	3.1 ± 0.26^{b}	0.63 ± 0.05^{b}	$<DL^{a}$
Coquille River (1)	0.56^{b}	0.29^{a}	1.5^{c}	$0.65^{a,b}$	0.35^{c}
Alsea Bay (8)	0.69 ± 0.39^{b}	$<DL^{b}$	0.47 ± 0.30^{c}	0.21 ± 0.11^{b}	$<DL^{a}$
Coos Bay (4)	$0.83 \pm 0.83^{a,b}$	$<DL^{b}$	0.73 ± 0.12^{c}	0.33 ± 0.09^{b}	$<DL^{a}$
Duwamish Estuary (3)	$0.97 \pm 0.08^{a,b}$	$<DL^{b}$	4.3 ± 0.18^{a}	0.74 ± 0.09^{b}	$<DL^{a}$
Elk River (2)	0.14 ± 0.11^{b}	$<DL^{b}$	0.64 ± 0.33^{c}	0.21 ± 0.09^{b}	0.06 ± 0.06^{a}
Grays Harbor (3)	0.04 ± 0.04^{b}	$<DL^{b}$	1.53 ± 0.67^{c}	0.26 ± 0.06^{b}	$<DL^{a}$
Nisqually Estuary (3)	$0.71 \pm 0.14^{a,b}$	$<DL^{b}$	3.2 ± 0.46^{b}	0.59 ± 0.12^{b}	0.05 ± 0.05^{a}
Salmon River (11)	$0.78 \pm 0.38^{a,b}$	$<DL^{b}$	0.15 ± 0.09^{c}	0.08 ± 0.04^{c}	$<DL^{a}$
Skokomish Estuary (3)	0.28 ± 0.09^{b}	$<DL^{b}$	2.45 ± 0.51^{b}	0.46 ± 0.15^{b}	0.04 ± 0.04^{a}
Tillamook Bay (1)	$<DL^{b}$	$<DL^{b}$	$<DL^{c}$	$<DL^{c}$	$<DL^{a}$
Yaquina Bay (7)	0.06 ± 0.06^{b}	$<DL^{b}$	1.1 ± 0.6^{c}	0.18 ± 0.08^{b}	$<DL^{a}$
Willapa Bay (3)	$<DL^{b}$	$<DL^{b}$	0.32 ± 0.04^{c}	0.13 ± 0.07^{b}	$<DL^{a}$
Hatchery chinook					
Fall Creek (1)	2.1^{a}	0.22^{a}	4.5^{a}	1.2^{a}	$<DL^{a}$
Butte Falls (1)	1.9^{a}	0.25^{a}	4.7^{a}	1.1^{a}	$<DL^{a}$
Cole M. Rivers (1)	2.3^{a}	$<DL^{b}$	4.2^{a}	$0.88^{a,b}$	$<DL^{a}$
Elk River (2)	1.4 ± 0.9^{a}	$<DL^{b}$	3.7^{a}	$0.65^{a,b}$	0.13 ± 0.13^{b}
Trask (1)	1.7^{a}	$<DL^{b}$	3.6^{a}	$0.87^{a,b}$	$<DL^{a}$
Salmon River (1)	3.7^{a}	$<DL^{b}$	4.4^{a}	1.1^{a}	$<DL^{a}$
Estuary coho					
Alsea Bay (3)	2.5 ± 0.3^{a}	$<DL^{b}$	0.17 ± 0.04^{c}	0.2 ± 0.03b	$<DL^{a}$
Coos Bay (1)	3.3 ± 0.3^{a}	$<DL^{b}$	0.2^{c}	0.16^{b}	0.64^{d}
Grays Harbor (1)	$<DL^{b}$	$<DL^{b}$	0.35^{c}	0.13^{b}	$<DL^{a}$
Willapa Bay (1)	$<DL^{b}$	$<DL^{b}$	0.44 ± 0.26^{c}	0.13 ± 0.0^{b}	$<DL^{a}$
Yaquina Bay (3)	$<DL^{b}$	$<DL^{b}$	0.10^{c}	0.09^{b}	$<DL^{a}$

60–100% of ΣDDTs in stomach contents of both coho and chinook salmon from all sites (Fig. 12; Table 5). Additionally, *p,p′*-DDD and *p,p′*-DDT were found in both chinook and coho salmon stomach contents from several sites, with highest concentrations in juvenile chinook from the Columbia River (5.9 and 2.5 ng/g ww for *p,p′*-DDD and *p,p′*-DDT, respectively). These isomers accounted for 5–25% of total DDTs. In comparison with salmon whole bodies, *p,p′*-DDT was found at higher concentrations in stomach contents. The *o,p'*-DDTs were found only in stomach contents of chinook salmon from the Columbia River, which had measurable concentrations (0.6–1.1 ng/g ww) of both *o,p′*-DDT and *o,p′*-DDD. In stomach contents of juvenile chinook from the Elk River Hatchery, the only DDT isomer found was *p,p′*-DDE, which was present at a concentration of 4.5 ng/g ww.

In addition to PCBs, DDTs, and PAHs, chlordanes HCBs, HCHs, dieldrin, and mirex were detected in stomach contents of estuarine chinook or coho from one or more sampling sites (Table 6). In stomach contents of chinook from the Elk River Hatchery, chlordanes, HCB, and mirex were detected, all at relatively low levels (0.7–1.4 ng/g ww). Aldrin was below the limits of detection in all samples.

3.5 Relationship between contaminants in stomach contents and in salmon bodies

In chinook salmon, concentrations of PCBs and DDTs in stomach contents were significantly and positively

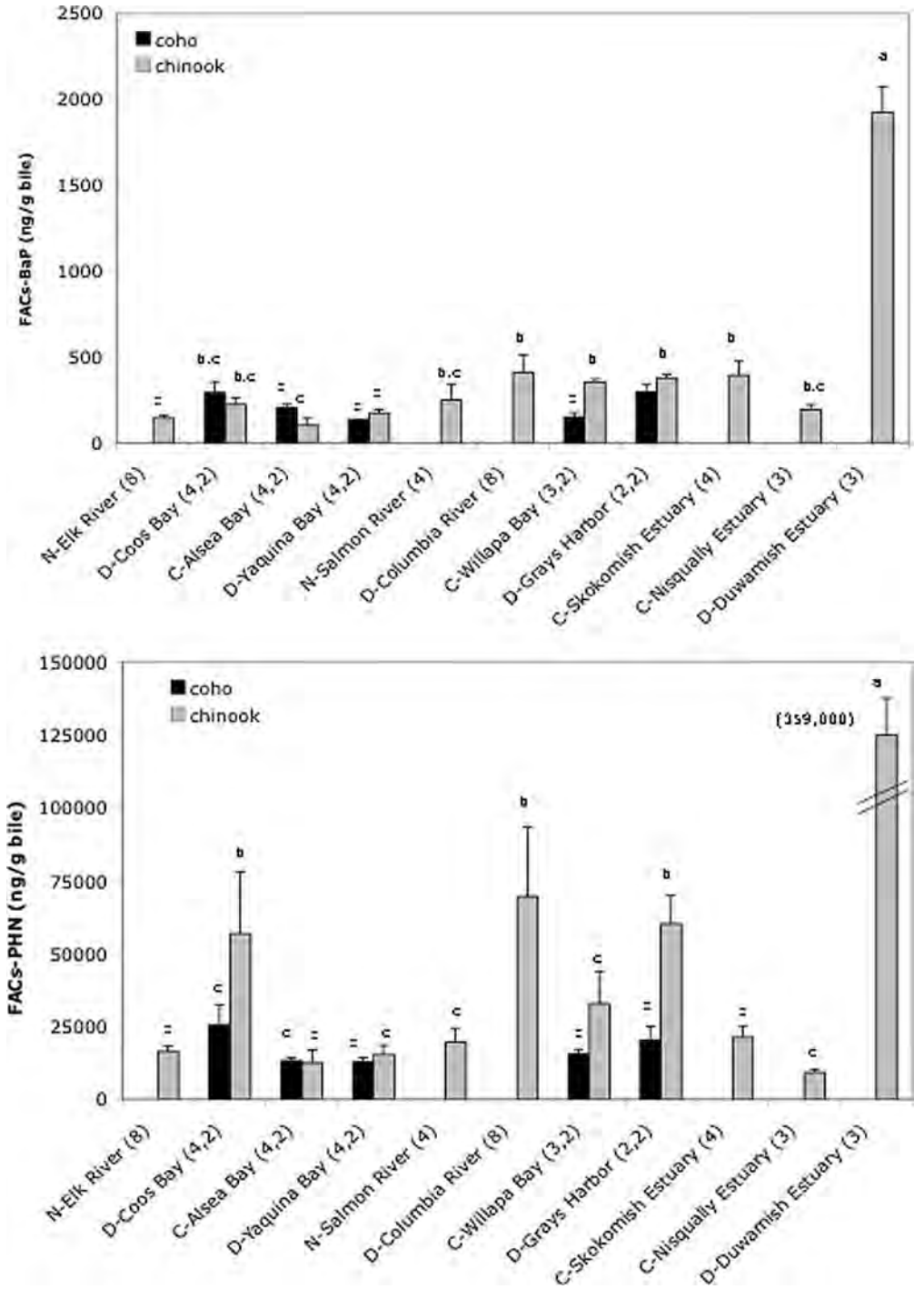

Fig. 8 Mean concentrations of fluorescent aromatic compounds (± SE) measured at phenanthrene wavelengths (FACs-PHN) and benzo[a]pyrene wavelengths (BaP-FACs) in bile of juvenile chinook and coho almon from Pacific Northwest estuaries. N = natural estuary; C = conservation estuary; S = shallow draft estuary; D = deep draft estuary. Bile metabolites measured at PHN and BaP wavelengths are representative of metabolites of low and high molecular weight PAHs, respectively. Numbers in parentheses indicate number of composite samples (10–15 fish each) analyzed per site or group. Measurements with different letters are significantly different (ANOVA, $p < 0.05$)

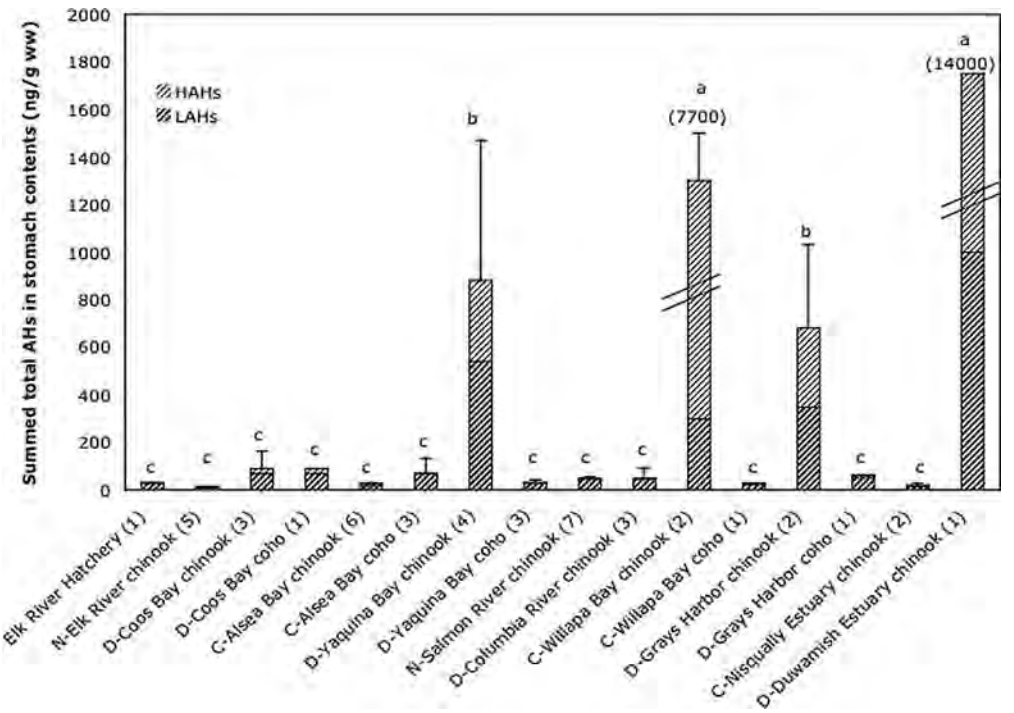

Fig. 9 Mean concentrations of total aromatic hydrocarbons (ΣAHs) (ng/g wet wt, ± SE) in stomach contents of juvenile chinook and coho salmon from Pacific Northwest estuaries and juvenile chinook salmon from Elk River hatchery. N = natural estuary; C = conservation estuary; S = shallow draft estuary; D = deep draft estuary. Contributions of low molecular weight and high molecular weight AHs (LAHs and HAHs) to totals are indicated. Numbers in parentheses indicate number of composite samples (10–15 fish each) analyzed per site or group. Measurements with different letters are significantly different (ANOVA, $p < 0.05$)

correlated with body burdens of the same contaminants. For PCBs ($n = 46$), $r^2 = 0.32$, $p = 0.0001$; while for DDTs ($n = 40$), $r^2 = 0.38$, $p = 0.0001$. In coho salmon, concentrations of contaminant in bodies and stomach contents were also positively correlated, but relationships were marginally significant ($0.06 \leq p \leq 0.08$), in part because of smaller sample size. For body DDTs vs. stomach DDTs ($n = 9$), $r^2 = 0.34$, $p = 0.06$. For body PCBs vs. stomach PCBs ($n = 9$), $r^2 = 0.29$, $p = 0.08$.

In estuarine chinook salmon, concentrations of PCBs and DDTs (ng/g ww) in whole bodies were 3–4 times as high as in stomach contents on average, while in coho salmon, concentrations of PCBs and DDTs in whole bodies and stomach contents were about the same or only slightly higher (1–1.3 times). For chinook salmon from the Elk River Hatchery (the only hatchery where stomach contents data were available), concentrations of PCBs (ng/g ww) were 4.7 times as high in bodies as in stomach contents, while concentrations of DDTs (ng/g ww) were 25 times as high in bodies as in stomach contents.

In chinook salmon, concentrations of PAH metabolites in bile and PAHs in stomach contents were significantly, positively correlated. For ΣLAHs vs. FACs-PHN, $n = 35$, $p = 0.0001$, $r^2 = 0.56$, and for ΣHAHs vs. FACs-BaP, $n = 35$, $p = 0.0006$, $r^2 = 0.28$. In coho salmon, on the other hand, there was no significant correlation between concentrations of either ΣHAHs or ΣLAHs in stomach contents and concentrations of PAH metabolites in bile. For ΣHAHs, $n = 5$, $r^2 = 0.07$, $p = 0.33$. For ΣLAHs, $n = 5$, $r^2 = 0.18$, $p = 0.26$.

4 Discussion

Estuarine and nearshore ecosystems provide a vital role as juvenile rearing habitat for salmonid species (Levy and Northcote, 1982; Gray *et al.*, 2002; Rice *et al.*, 2005), and can be particularly important in the recovery of species at risk (Feist *et al.*, 2003; Fresh *et al.*, 2005). Unfortunately, estuarine and coastal ecosystems are also among the environments that are most heavily

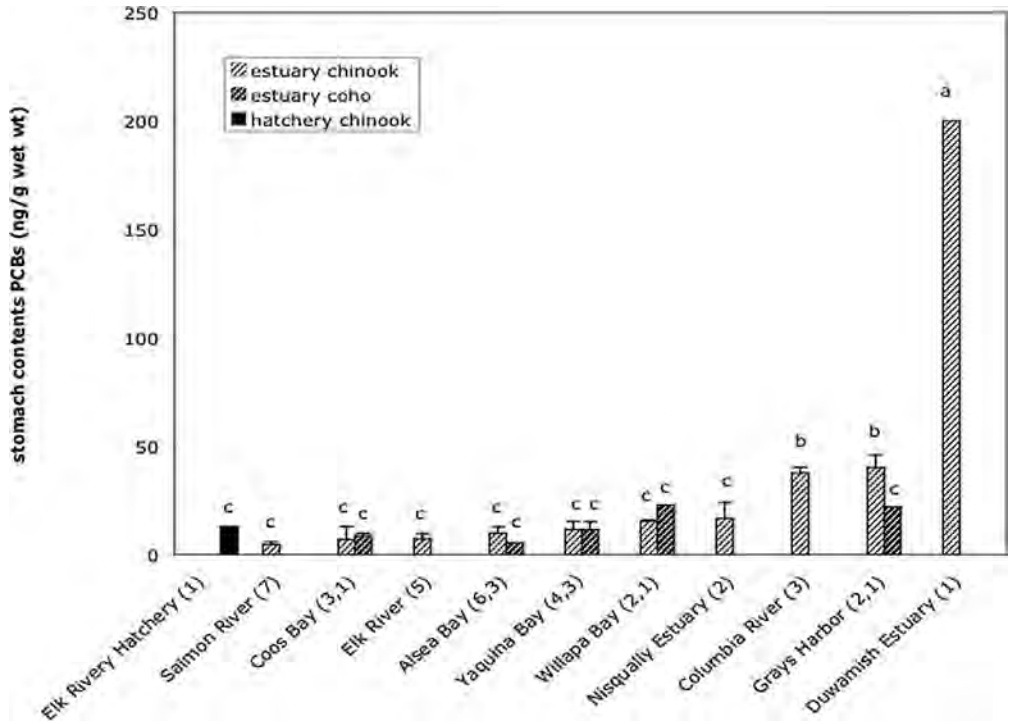

Fig. 10 Mean concentrations of ΣPCBs (ng/g wet wt. ± SE) in stomach contents of juvenile chinook and coho salmon from Pacific Northwest estuaries and juvenile chinook salmon from Elk River hatchery. N = natural estuary; C = conservation estuary; S = shallow draft estuary; D = deep draft estuary. Numbers in parentheses indicate number of composite samples (10–15 fish each) analyzed per site or group. Measurements with different letters are significantly different (ANOVA, $p < 0.05$)

impacted by anthropogenic activities (Shreffler *et al.*, 1990; Beck *et al.*, 2001; Rice *et al.*, 2005). Analyses of risks to salmon populations in estuarine environments have focused largely on alterations to or loss of physical habitat attributes (Bottom *et al.*, 2005; Gray *et al.*, 2002; Fresh *et al.*, 2005), but it is increasingly recognized that habitat degradation associated with chemical contaminants may also pose a significant risk to salmon populations (Spromberg and Meador, 2005; Fresh *et al.*, 2005; Loge *et al.*, 2005).

The importance of estuarine contamination in terms of the health of salmonid species depends in part on the life history strategy of the species in question. In general, ocean-type stocks, such as fall chinook, which spend an extended period during their first year of life in the estuary, are more vulnerable to the impacts of contaminants in this environment than stream-type stocks, such as coho salmon, which pass through the estuary relatively quickly (Fresh *et al.*, 2005). The same may be true of chum salmon, which have a long estuarine residence time (Dorcey *et al.*, 1978; Healey, 1982). Juvenile chum have shown relatively high contaminant body burdens at urban sites in previous surveys in Puget Sound, WA (Stehr *et al.*, 2000).

The results of the current study confirm that chemical contaminants are present in the prey and tissues of outmigrant juvenile salmon from a number of estuaries in the Pacific Northwest. The most widespread contaminants were PCBs, DDTs, and PAHs, which were observed in both tissues and stomach contents of chinook and coho salmon from all estuarine sampling sites, as well as in chinook salmon from local hatcheries. Although additional organochlorine pesticides (chlordanes, lindane, hexachlorobenzene, dieldrin, aldrin and mirex) were also detected in salmon tissues or stomach contents, the measured concentrations were relatively low. Like earlier studies in Puget Sound, the present study highlights the importance of the estuary as a source of exposure to chemical contaminants, especially for juvenile chinook salmon. The observation of elevated contaminant concentrations in stomach contents of salmon from sites in several estuaries indicates that fish are being exposed to these contaminants during estuarine residence through their

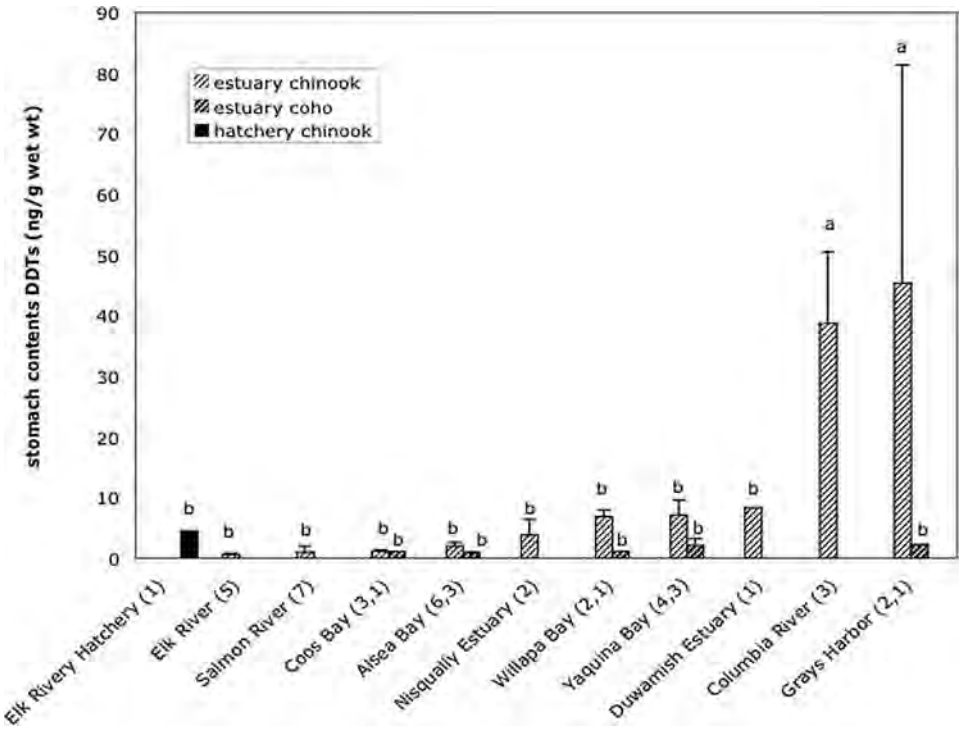

Fig. 11 Mean concentrations of ΣDDTs (ng/g ww, ± SE) in stomach contents of juvenile chinook and coho salmon from Pacific Northwest estuaries and juvenile chinook salmon from Elk River hatchery. N = natural estuary; C = conservation estuary; S = shallow draft estuary; D = deep draft estuary. Numbers in parentheses indicate number of composite samples (10–15 fish each) analyzed per site or group. Measurements with different letters are significantly different (ANOVA, $p < 0.05$)

prey. The hypothesis that this could be an important source of uptake is further supported by the significant correlations between concentrations of PCBs and DDTs in stomach contents and whole bodies of juvenile chinook salmon, and between PAHs in stomach contents and PAH metabolites in bile. Contaminants in the water column, and in suspended particulate material, are also potential sources of exposure, although they were not measured in this study. Depending on their origin, chinook and coho salmon from some populations could also be taking up certain contaminants through the water column or the diet in freshwater before entering the estuary. This is especially true if they are passing through urbanized watersheds. However, the potential contribution of contaminants in freshwater habitats to juvenile salmon body burdens cannot be evaluated based on the samples collected in the present study.

4.1 Species differences in contaminant uptake

Of the two species we examined, chinook salmon exhibited the highest degree of uptake and accumulation of contaminants. On both a lipid weight and a wet weight basis, contaminant concentrations in whole bodies of chinook salmon were significantly higher than in coho salmon sampled from the same sites, with levels typically 2–5 times as great in chinook than in coho salmon collected at the same sites. Concentrations of contaminants in chinook salmon stomach contents tended to be higher as well, although the difference was less marked. Additionally, correlations between contaminant body burdens and contaminant concentrations in stomach contents were stronger in chinook than in coho salmon.

These findings are consistent with results of other studies on chinook and coho salmon in the Great Lakes (Manchester-Neesvig *et al.*, 2001; Jackson *et al.*, 2001; Rohrer *et al.*, 1982), and are likely related to differences in life history and habitat use, as well as diet and metabolism. Assuming that the estuary is an important source of contaminants for outmigrant salmonids, these differences are consistent with the more prolonged period of estuarine residence in chinook salmon. Of the five species of Pacific salmon, chinook salmon

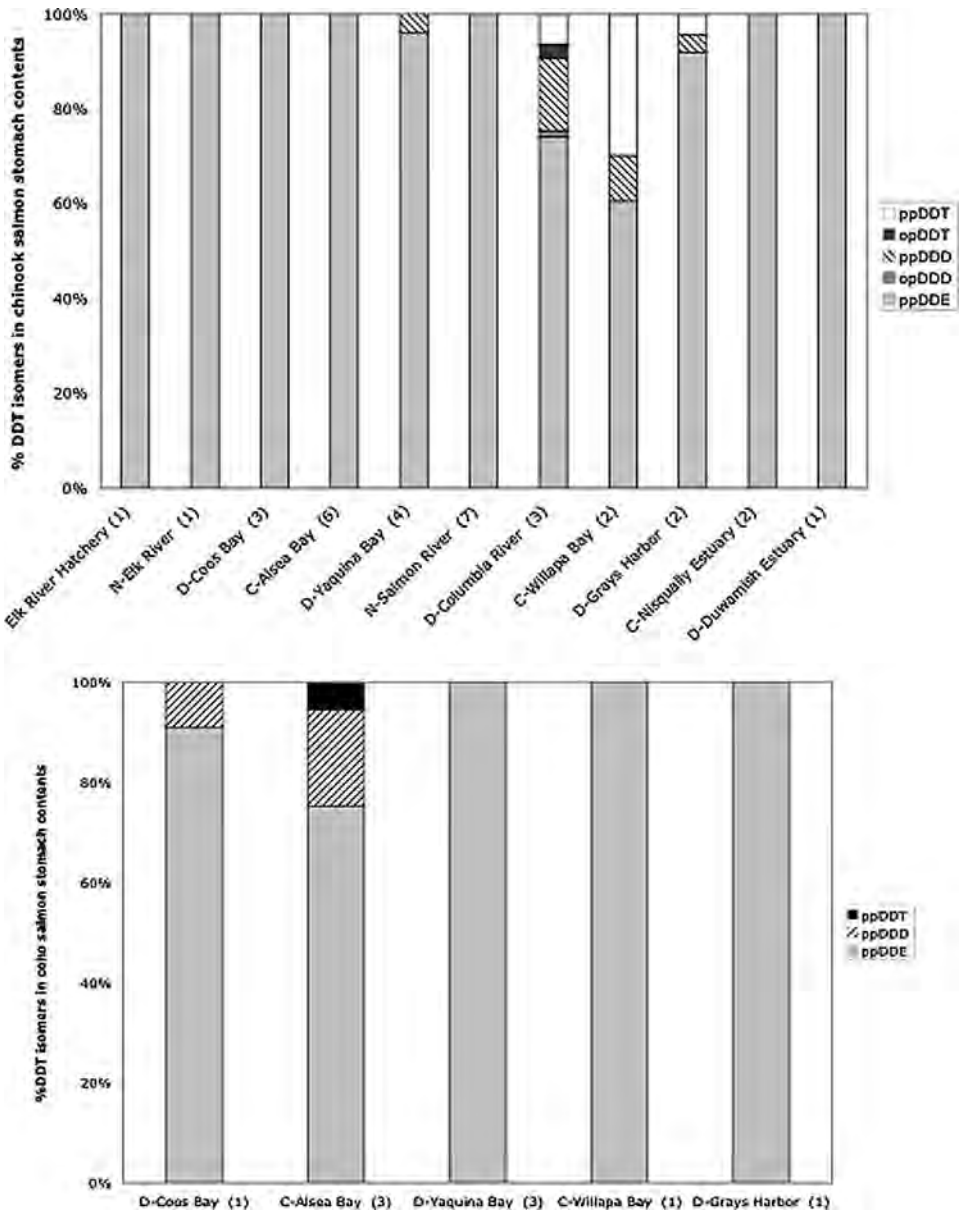

Fig. 12 Proportions of different DDTs in composite stomach contents samples of juvenile chinook and coho salmon collected from Pacific Northwest Estuaries. N = natural estuary; C = conservation estuary; S = shallow draft estuary; D = deep draft estuary. Numbers in parentheses indicate number of composite samples (10–15 fish each) analyzed per site or group

are most dependent upon estuaries during the early stages of their life cycle (Healey, 1982; 1991; Healey and Prince, 1995), typically residing in estuaries for one to two months (Simenstead *et al.*, 1982), but in some cases for up to 6 months (Healey, 1982; Reimers, 1973; Levy and Northcote, 1982; Simenstad *et al.*, 1982). Outmigrant juvenile coho, on the other hand, are much less estuarine-dependent, typically passing through the estuary within a few days (Moser *et al.*, 1991; McMahon and Holtby, 1992; Magnusson, 2003; Duffy *et al.*, 2005). Increased bioaccumulation in chinook salmon may also indicate that they are feeding at a higher trophic level than coho salmon, which would be supported by the generally higher concentrations of PCBs and DDTs in stomach contents of chinook salmon in comparison with levels in stomach contents of coho salmon collected from the same sites. This is consistent with dietary studies showing that,

Table 5 Mean concentrations (± SE) in ng/g wet wt of DDT isomers in stomach contents composites of juvenile chinook and coho salmon from Pacific Northwest estuaries, and juvenile chinook salmon from Elk River Hatchery. DDTs were measured by GC/ECD in samples collected from 1996–1998 and by GC/MS in samples collected from 1999–2001. Composites contain stomach contents from 10–15 fish. Values with different letter superscripts are significantly different (ANOVA, $p \leq 0.05$)

Site	*o,p′*-DDD	*o,p′*-DDT	*p,p′*-DDE	*p,p′*-DDD	*p,p′*-DDT
Hatchery chinook					
Elk River (1)	<DL^b	<DL^b	4.5^b	<DL^b	<DL^b
Estuary chinook					
Alsea Bay (6)	<DL^b	<DL^b	2.0 ± 0.6^b	<DL^b	<DL^b
Columbia River (3)	0.6 ± 0.6^a	1.1 ± 0.6^a	28.7 ± 9.1^a	5.9 ± 0.7^a	2.5 ± 1.4^a
Coos Bay (3)	<DL^b	<DL^b	1.1 ± 0.3^b	<DL^b	<DL^b
Duwamish Estuary (1)	<DL^b	<DL^b	5.8^b	<DL^b	2.5^a
Elk River (5)	<DL^b	<DL^b	0.6 ± 0.2^b	<DL^b	<DL^b
Grays Harbor (2)	<DL^b	<DL^b	41.7 ± 32.3^a	1.6 ± 1.6^b	2.1 ± 2.1^a
Nisqually Estuary (2)	<DL^b	<DL^b	3.5 ± 2.3^b	0.3 ± 0.3^b	<DL^b
Salmon River (7)	<DL^b	<DL^b	1.0 ± 1.0^b	<DL^b	<DL^b
Willapa Bay (2)	<DL^b	<DL^b	4.2 ± 0.4^b	0.7 ± 0.7^b	2.1 ± 2.1^a
Yaquina Bay (3)	<DL^b	<DL^b	6.9 ± 2.2^b	0.3 ± 0.3^b	<DL^b
Estuary coho					
Alsea Bay (3)	<DL^b	<DL^b	0.8 ± 0.1^b	0.11 ± 0.1^b	<DL^b
Coos Bay (1)	<DL^b	<DL^b	1.1^b	<DL^b	<DL^b
Grays Harbor (1)	<DL^b	<DL^b	2.3^b	<DL^b	<DL^b
Willapa Bay (1)	<DL^b	<DL^b	1.2^b	<DL^b	2.5^a
Yaquina Bay (3)	<DL^b	<DL^b	1.9 ± 0.9^b	0.2 ± 0.1^b	0.1 ± 0.1^b

Table 6 Mean concentrations (± SE) in ng/g, wet wt of selected organochlorine pesticides measured in stomach contents of juvenile chinook and coho salmon collected from the Pacific Northwest estuaries and hatcheries. Σchlordanes = summed concentrations of heptachlor, heptachlor epoxide, γ-chlordane, α-chlordane, *cis*-nonachlor, *trans*-nonachlor and nonachlor III. DL = detection limit. Pesticides were measured by GC/ECD in samples collected from 1996–1998 and by GC/MS in samples collected from 1999–2001. Values with different letter superscripts are significantly different (ANOVA, $p \leq 0.05$)

Site	lindane	dieldrin	Σchlordanes	HCB	mirex
Hatchery chinook					
Elk River (1)	<DL^b	<DL^b	1.4^c	0.7^b	0.7^b
Estuary chinook					
Alsea Bay (6)	<DL^b	<DL^b	<DL^c	0.6 ± 0.3^{b,c}	0.2 ± 0.2^b
Columbia River (3)	<DL^b	6.0 ± 6.0^a	0.8 ± 0.5^c	1.5 ± 0.8^{a,b}	0.3 ± 0.3^b
Coos Bay (3)	<DL^b	<DL^b	<DL^c	0.3 ± 0.2^c	0.6 ± 0.6^b
Duwamish Estuary (1)	<DL^b	<DL^b	12^a	<DL^c	2.5^b
Elk River (5)	<DL^b	<DL^b	1.4^c	0.3 ± 0.2^c	0.24 ± 0.25^b
Grays Harbor (2)	1.8 + 1.8^a	1.5 ± 1.5^{a,b}	6.1 ± 0.6^b	1.9 ± 1.9^a	2.7 ± 2.7^b
Nisqually Estuary (2)	<DL^b	0.9^b	0.5 ± 0.5^c	0.17 ± 0.17^c	<DL^b
Salmon River (7)	<DL^b	<DL^b	<DL^c	<DL^c	<DL^b
Willapa Bay (2)	<DL^b	6.5 ± 6.5^a	<DL^c	<DL^c	6 ± 6^a
Yaquina Bay (3)	0.6 + 0.6^a	<DL^b	1.8 ± 1.8^c	0.24 ± 0.24^c	0.4 ± 0.4^b
Estuary coho					
Alsea Bay (3)	<DL^b	<DL^b	0.17 ± 0.06^c	0.72 ± 0.22^b	<DL^b
Coos Bay (1)	<DL^b	4.0 ± 4.0^b	0.31^c	0.25^c	<DL^b
Grays Harbor (1)	<DL^b	<DL^b	<DL^c	<DL^c	<DL^b
Willapa Bay (1)	<DL^b	<DL^b	0.65^c	0.65^b	<DL^b
Yaquina Bay (3)	<DL^b	<DL^b	0.69 ± 0.36^c	0.12 ± 0.07^c	<DL^b

while there is considerable overlap in the diet of juvenile coho and chinook salmon, coho tend to consume a lower proportion of juvenile and larval fish and a higher proportion of invertebrates than chinook (Schabetsberger *et al.*, 2003; Brodeur and Pearcy, 1990).

4.2 Site-related differences in contaminant body burdens

Although contaminant concentrations in coho salmon showed no strong spatial trends, in chinook salmon there were marked intersite differences in contaminant concentrations in tissues and stomach contents, with highest exposure levels in the industrial and urbanized estuaries. Concentrations of PCBs were highest in samples from the Duwamish Estuary, and were similar to or somewhat lower than concentrations reported in earlier Puget Sound studies at this location (Stein *et al.*, 1995; Varanasi *et al.*, 1993; Meador *et al.*, 2002). Total PCB concentrations 2 to 3 times higher than those reported in this study have been measured in juvenile chinook collected from heavily contaminated Duwamish Estuary sites (Varanasi *et al.*, 1993; Meador *et al.*, 2002). The somewhat lower concentrations of PCBs observed in juvenile salmon sampled in the present study may be due to differences in sampling location, or because sampling occurred early in the season, when juvenile salmon may have only recently entered the estuary (Bottom *et al.*, 2005). The lower concentrations may also be reflective of a low proportion of hatchery fish in this sample. Such differences in contaminant concentrations between wild and hatchery-released fish have been noted in other studies (Meador *et al.*, 2002). In addition to Duwamish chinook, concentrations of PCBs were also relatively high in chinook salmon from the Columbia River and Yaquina Bay.

Interestingly, PCB concentrations in the juvenile chinook salmon we sampled were quite similar to concentrations reported in returning adult chinook salmon from Washington State (Missildine *et al.*, 2005). Mean concentrations of PCBs in adult chinook ranged from 48–50 ng/g ww in salmon returning to Puget Sound hatcheries (Deschutes and Issaquah), and from 15–29 ng/g ww in salmon returning to coastal hatcheries (Makah and Quinault). Although it is unlikely that exposures occurring in the juvenile stage make a major contribution to adult contaminant body burdens (O'Neill *et al.*, 1998), these data do suggest consistent exposure at multiple life stages for salmon from urban estuaries.

Concentrations of DDTs were especially high in juvenile chinook salmon from the Lower Columbia River and in the Nisqually Estuary in Puget Sound. The high DDT concentrations in Columbia River chinook are consistent with elevated DDT concentrations observed in other resident marine and freshwater fish from the Columbia River in earlier studies by EPA, NOAA, and USGS, and the States of Washington and Oregon (USEPA, 2000; Tetra-Tech Inc., 1993, 1994, 1996; LCREP, 1999; Brown *et al.*, 1998; Foster *et al.*, 2001a,b). As in most environmental samples, DDT breakdown products, especially *p,p′*-DDE, predominated in coho and chinook salmon body and stomach contents samples. However, *p,p′*-DDT and *o,p′*-DDT were also detected in samples from some sites, particularly chinook salmon from the Columbia River and Yaquina Bay, and coho salmon from Willapa Bay. The presence of these parent compounds suggests that there may be fresher sources of DDT in these areas, although the half-lives of *p,p′*- and *o,p′*-DDT in soils can be quite variable (ATSDR, 2002).

Concentrations of PAHs were especially high in stomach contents of fish from the Duwamish Estuary, Willapa Bay, Grays Harbor and Yaquina Bay, although very high concentrations of PAH metabolites in bile (i.e., $>$1000 ng/g bile for FACs-BaP and $>$200,000 ng/g bile for FACs-PHN) were observed only in fish from the Duwamish Estuary. In fish from more pristine estuaries such as Alsea Bay, Salmon River, Elk River, and Tillamook, PAH concentrations were lower than any of those previously reported in Puget Sound (Stein *et al.*, 1995; Varanasi *et al.*, 1993; McCain *et al.*, 1990). High molecular weight AHs, which originate primarily from combustion products (Varanasi *et al.*, 1992; MacDonald and Crecelius, 1994), accounted for a higher proportion of total AHs in stomach contents of fish from the Duwamish Estuary, Willapa Bay, Grays Harbor and Yaquina Bay, than in fish from other estuaries. This suggests that atmospheric emissions from incineration and automobile emissions may be major contamination sources in these areas, as well as releases from industries that generate high molecular weight PAHs (e.g., aluminum smelters, oil refineries, creosote plants; Varanasi *et al.*, 1992; MacDonald and Crecelius, 1994). The predominance of LAHs, which are primarily associated with petroleum products (Varanasi *et al.*, 1992; MacDonald and Crecelius,

1994), in stomach contents of salmon from Alsea Bay, Coos Bay, Nisqually, Salmon River, the Columbia River, and Elk River, suggests that PAHs in these areas come mainly from releases of fuel oil, crude oil, and related materials into the environment.

Ratios of ΣDDT/ΣPCB varied from site to site, indicating differences in contaminant profiles among different groups of fish. For example, the ΣDDT/ΣPCB ratio in bodies of salmon from the Columbia Estuary site (∼1.1) was higher than in juvenile chinook salmon the other estuarine sites, suggesting particularly high uptake of DDTs from the environment at this site. Fish from the Duwamish Estuary, the other hand, had one of the lowest DDT/PCB ratios, reflecting the very high concentrations of PCBs in fish from this site.

4.3 Contaminants in hatchery salmon

Measurable concentrations of PCBs and DDTs were also present in bodies of juvenile chinook salmon sampled directly from Pacific Northwest hatcheries. On a wet weight basis, concentrations of both PCBs and DDTs in hatchery chinook were relatively high, comparable to those in juvenile chinook from the more contaminated estuarine sites. However, as the lipid content of hatchery fish was also quite high (8% as compared to 1–3% in estuarine fish), when PCB and DDT body burdens were calculated on a lipid weight basis, concentrations in hatchery chinook were relatively low in comparison to levels in chinook from urban and industrialized estuaries. In stomach contents of juvenile hatchery chinook, levels of PAHs, PCBs, DDTs, were also relatively low, similar to concentrations in rural estuaries such as Elk River and Alsea Bay. This suggests that elevated contaminant concentrations in the hatchery fish we sampled are due not so much to high concentrations of contaminants in feed, but to the high body fat levels in hatchery reared juveniles that facilitate the uptake of lipid soluble contaminants. It is uncertain, though, whether the Elk River Hatchery sample is representative of feed from other sampled hatcheries, or of feeds in current use.

Chemical contaminants, especially PCBs, have been detected in hatchery fish and feed and in farmed fish in several other studies (Easton *et al.*, 2002; Parkins, 2003; Karl *et al.*, 2003; Hites *et al.*, 2004). Available data suggest that the problem is widespread, and also that contaminant concentrations in different lots of feed and in fish from different hatcheries are highly variable. Concentrations of PCBs in juvenile salmon from the Pacific Northwest hatcheries sampled in this study were similar to mean levels (∼50 ng/g ww) reported by Easton *et al.* (2002) and Hites *et al.* (2004) in farmed salmon. However, PCB concentrations in commercial feed analyzed by Easton *et al.* (2002) and Hites *et al.* (2004) were generally higher than PCB concentrations in stomach contents of Elk River Hatchery salmon, with a number of samples in the 30–90 ng/g ww range.

In the hatchery chinook we analyzed, the DDT isomers *p,p′*-DDT and *o,p′*-DDT made up a substantial proportion of DDTs present. This appears to be common in farmed and hatchery fish, and may indicate use of oils or fish meals from sources where there was relatively recent usage of DDTs (Jacobs *et al.*, 2002).

The observation of chemical contaminants in prerelease hatchery fish is likely to be a concern for the management of these animals. If contaminant body burdens are already moderate to high when fish leave the hatchery, they have an increased risk of reaching exposure concentrations during estuarine residence that could significantly reduce their likelihood of survival. Moreover, contaminated salmon may be a significant source of toxicants in the environment and in the food chain (Kreummel *et al.*, 2003). This represents a hazard for birds and other piscivorous wildlife. More comprehensive sampling of fish and feed from hatcheries is needed to determine the extent of this problem in the Pacific Northwest.

4.4 Potential health effects of contaminants on salmon

For some contaminants, exposure levels in juvenile salmon from selected sites are approaching concentrations that could affect their health and survival. Indeed, adverse health effects have been observed in juvenile salmon from the Duwamish Estuary, which is contaminated with PAHs and PCBs. Fish from this area showed immunosuppression, reduced disease resistance and decreased growth rates (Arkoosh *et al.*, 1991, 1994, 1998, 2001; Varanasi *et al.*, 1993; Casillas *et al.*, 1995, 1998), as well as biochemical alterations such as DNA damage (i.e., PAH-DNA adducts in liver) and induction of cytochrome P4501A (CYP1A), an enzyme that metabolizes selected contaminants including PAHs, dioxins and furans, and dioxin-like PCB congeners (Stein *et al.*, 1995; McCain *et al.*, 1990; Varanasi *et al.*, 1993; Collier *et al.*, 1998; Stehr *et al.*,

2000). These biochemical alterations are not necessarily indicative of adverse health effects in themselves, but are associated with disease conditions including reproductive and developmental abnormalities and liver disease (Williams *et al.*, 1998; Whyte *et al.*, 2000; Myers *et al.*, 2003). Fish from several sites sampled in the present study (Grays Harbor, Yaquina Bay, the Columbia River) had concentrations of PCBs, PAHs or both in tissues or stomach contents that were comparable to those found in Duwamish Estuary fish, suggesting that they may also be at risk for the types of adverse health effects documented in fish from that Puget Sound site. The possibility of increased disease-induced mortality is increased by recent finding of widespread occurrence of potentially lethal parasites and pathogens in juvenile chinook and coho salmon from the estuaries sampled in this study (Arkoosh *et al.*, 2004).

The potential for health risks in Pacific Northwest salmon can also be evaluated by comparing measured tissue contaminant concentrations against established effects thresholds. For PCBs, Meador *et al.* (2002) estimated a critical body residue of 2400 ng/g lipid for protection against 95% of effects ranging from enzyme induction to mortality, based on a range of sublethal effects observed in salmonids in peer-reviewed studies conducted by NMFS and other researchers. Mean PCB body burdens in juvenile salmon analyzed in this study were near or above 2400 ng/g lw in fish from three sampling sites, the Columbia River, the Duwamish Estuary, and Willapa Bay. These findings suggest that a significant portion of outmigrant juvenile chinook salmon from these sites may be at risk of some type of health impairment due to PCB exposure.

A threshold concentration for the impact of DDTs on listed salmon has not been systematically determined, unlike the PCBs (Meador *et al.*, 2002). Most reported effects in salmonids are associated with whole body tissue total DDT concentrations at or above 500 ng/g ww (Allison *et al.*, 1963; Burdick *et al.*, 1964; Buhler *et al.*, 1969; Johnson and Pecor, 1969; Peterson, 1976; Poels *et al.*, 1980), or about 5000 ng/g lipid, assuming that the test fish had a lipid content of around 10%, which is typical of laboratory-reared salmonids (Meador *et al.*, 2002). A number of recent studies suggest that certain DDT isomers, such as *o,p′*-DDT and *o,p′*-DDE, have estrogenic activity, and may have endocrine-disrupting or immunotoxic effects (Donohoe and Curtis, 1996; Arukwe *et al.*, 1998; Celius and Walther, 1998; Khan and Thomas, 1998; Christiansen *et al.*, 2000; Zaroogian *et al.*, 2001; Milston *et al.*, 2003; Papoulias *et al.*, 2003). However, measured or estimated body burdens associated with these effects are typically in the 10–20 ng/g ww or 100–200 ng/g lipid range or above. Lipid-adjusted concentrations of total DDTs and *o,p′*-isomers of DDTs approached these concentrations in some fish from the Columbia River, but DDT body burdens typically found in estuarine chinook and coho salmon were substantially lower. This suggests that, by themselves, body burdens of DDTs would be unlikely to cause adverse health effects in most Pacific Northwest juvenile salmon. However, DDTs do not occur in isolation in Pacific Northwest estuaries, but are present with a variety of other contaminants. Estrogenic DDT metabolites, for example, even at low concentrations, could act in concert with other estrogenic contaminants (e.g., plasticizers, pharmaceuticals, and surfactants) to alter reproductive processes or other physiological functions. In fact, some field studies have reported effect thresholds for DDTs lower than those observed in laboratory exposure studies [e.g., maternal muscle concentrations of 25–30 ng/g ww for increased yolk sac fry mortality in Baltic salmon; Vuorinen *et al.* (1997)], possibly because of the presence of other contaminants, as well as lower lipid concentrations in wild fish. More work is needed to understand the potential cumulative effects of DDTs and other contaminants present in salmon habitats.

Exposure to PAHs may also contribute to health risks in juvenile chinook salmon from some of the sampling sites. In juvenile chinook salmon from Puget Sound sites where immunosuppression and other health effects have been observed (Arkoosh *et al.*, 1991, 1994, 1998, 2001; Varanasi *et al.*, 1993; Stein *et al.*, 1995; Casillas *et al.*, 1995, 1998; Stehr *et al.*, 2000), concentrations of total PAHs in stomach contents of these fish were in the 1,200 to 8,000 ng/g ww range for ΣLAHs and in the 2,000 to 6,000 ng/g ww range for ΣHAHs, or 4,000 to 15,000 ng/g ww for total PAHs (Stein *et al.*, 1995; Varanasi *et al.*, 1993; Stehr *et al.*, 2000). In the present study, PAH concentrations in this range were detected once again in chinook salmon from the Duwamish Estuary, suggesting a potential for health risks to fish from this site. Concentrations of ΣHAHs were also surprisingly high in stomach contents of chinook salmon from Willapa Bay, but this was not reflected in bile metabolite levels of fish from this site. Additional sampling may be needed to determine if

there is consistent exposure to PAHs in Willapa Bay salmon.

In laboratory feeding studies where fish were exposed to PAHs alone, reported effect concentrations are somewhat higher than levels of PAHs measured in stomach contents of salmon from sites in where biological effects have been reported in the field, or PAH levels measured in the present study. Meador *et al*. (2005) found physiological changes in juvenile chinook exposed to 120 ppm total PAHs dry wt, or about 25,000 ng/g ww, while Bravo *et al*. (2005) observed immunosuppression, CYP1A induction and DNA damage in rainbow trout exposed to concentrations of 40,000 ng/g ww PAH in diet. Reported no effect doses for immunosuppressive and other physiological effects are in the 8,000–16,000 ng/g ww range (Palm *et al*., 2004; Meador *et al*., 2005). Total PAH concentrations in stomach contents of juvenile chinook collected from the Duwamish Estuary and Willapa Bay as part of this study are similar, and thus might be considered as being close to a threshold effect level. Moreover, PAHs may contribute to immunosuppressive or growth-altering impacts of other contaminants in environmental mixtures, even if they are below toxicity thresholds when considered alone (e.g., see Loge *et al*. (2005).

4.5 Trophic transfer and health effects on wildlife

Even if levels of bioaccumulative compounds such as DDTs and PCBs are not sufficient to cause direct effects on juvenile salmonids, they may represent a hazard to fish-eating predators through bioaccumulation and bioconcentration. The U.S. Fish and Wildlife Service (2004) estimated a no-observable adverse effects level (NOAEL) for impacts of fish prey on bald eagles of 60 ng/g ww for PCBs and 40 ng/g ww for DDTs, while Nendza *et al.* (1997) estimated a ΣDDTs NOAEL of 22–50 ng/g ww in fish tissue for impacts of related to bioaccumulation and bioconcentration of DDTs in estuarine systems. Juvenile chinook salmon sampled in this study from the Columbia River, the Duwamish Estuary, and the Nisqually Estuary had whole body DDT concentrations in the 20–50 ng/g ww range, and chinook salmon from the Duwamish Estuary had PCB concentrations above 60 ng/g ww, suggesting these fish may pose a hazard to fish-eating wildlife. Indeed, there is considerable evidence of bioconcentration of DDTs in birds and other wildlife that use the Columbia River, resulting in body burdens high enough to cause reproductive problems (Anthony *et al.*, 1993; USFWS, 1999, 2004; Thomas and Anthony, 2003; Henny *et al.*, 2003; Buck *et al.*, 2005).

4.6 Summary

Overall, the results of this study indicate significant exposure to PCBs, DDTs, and PAHs in outmigrant juvenile chinook salmon from several Pacific Northwest estuaries. Contaminant concentrations were generally highest in stomach contents and tissues of salmon from the deep draft estuaries, with the highest levels of urban and industrial development (i.e., the Duwamish Estuary, the Columbia River, Yaquina Bay, Coos Bay and Grays Harbor), and lowest in the natural estuaries (Elk River and Salmon River), which are largely undeveloped. However, relatively high concentrations of contaminants were detected in juvenile chinook from some of the conservation estuaries (Nisqually Estuary, Skokomish Estuary, Willapa Bay, and Alsea Bay), where land use is primarily agricultural. For example, concentrations of DDTs in salmon from the Nisqually Estuary were among the highest observed in this survey. For juvenile chinook salmon from the Duwamish Estuary, the Columbia River, and Yaquina Bay, whole body PCBs were within the range where they could potentially affect fish health and survival. In juvenile coho salmon, on the other hand, contaminant concentrations were relatively low, below estimated biological effects thresholds, and showed minimal variation from site to site. Juvenile chinook salmon are likely absorbing some contamination during estuarine residence through their prey, as PCBs, PAHs, and DDTs were consistently present in stomach contents, and PCBs and DDTs were significantly correlated with contaminant body burdens in fish from the same sites. Hatchery chinook also showed evidence of contaminant uptake. Although contaminant concentrations were not especially high in stomach contents of fish from the hatchery we tested, body burdens were elevated, in part because of the high lipid content of the fish. More research is needed to document exposure and associated effects of chemical contaminants on endangered Pacific Northwest salmon, but the available data show clearly that tissue burdens of some classes of contaminants are within the range where they could potentially affect survival and productivity of listed stocks or have adverse effects on the ecosystem of which salmon are a part.

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Persistent organic pollutants in aquafeed and Pacific salmon smolts from fish hatcheries in British Columbia, Canada

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ABSTRACT

Recent studies have reported the occurrence of persistent organic pollutants (POPs) in commercial feeds used by fish hatcheries and aquaculture facilities. We measured levels of polychlorinated biphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzo furans (PCDFs), as well as several organochlorine pesticides (OCPs) in several aquafeeds as well as fry and smolts collected from four federal salmonid hatcheries from coastal British Columbia. ΣPCBs in aquafeed samples ($n=22$) averaged 270±136 ng/g lipid. ΣDDTs, ΣToxaphene, ΣChlordanes, ΣChlorobenzenes, ΣHexachlorocyclohexanes, dieldrin, endrin and pentachloroanisole in feeds ranged between 0.6 and 200 ng/g lipid. ΣPCDD/F levels in feed samples were relatively low (77±50 pg/g lipid). In some cases, contaminant levels and patterns varied between different feed brands (i.e., Moore–Clarke, EWOS and OMP). ∑PCB concentrations (ng/g lipid) in Moore–Clarke (154.3±88.9) were significantly lower ($p<0.05$) than ∑PCBs in OMP (319.8±101) and EWOS (376.3±88.2) feeds, which is likely due to different constituent composition in the Moore–Clarke feed. As young hatchery salmon smolts are sequentially introduced to increasingly lipid-rich starter feeds (which contain higher contaminant burdens), dietary exposure of POPs generally increases during the rearing process. However, tissue residue concentrations of POPs in smolts remained relatively low compared to feed, as denoted by the relatively low biomagnification factors of the various POPs (BMFs between 0.2 and 3). 2,3,7,8 TCCD toxic equivalent concentrations (TEQs) in smolts (0.21±0.19 pg/g wet wt.) were generally below levels associated with dioxin-like toxic effects. The highest TEQ was observed in Robertson Creek smolts (0.38±0.11 pg/g wet wt). Biochemical response measurements (EROD activity and CYPIA levels) in liver tissue of those smolts did not show any indication of negative impacts associated with aryl hydrocarbon (Ah) receptor inducers such as dioxins/furans and dioxin-like PCBs.

1. Introduction

Approximately 5 to 6 million hatchery-raised juvenile Pacific salmon are released annually from Canadian rivers into the Pacific. Spawning wild Pacific salmon from coastal British Columbia (BC) may contribute an additional 4 to 5 times this number of juveniles (Heard, 1998). Fish hatcheries located on rivers and streams in BC fertilise, incubate and rear chum, coho, chinook, sockeye and/or pink salmon, as well as steelhead and cutthroat trout. Shortly after hatching, alevins become fry (i.e. lose their yolk sac) and further develop into smolts in rearing ponds that are normally supplied with water from a nearby stream or river. After a 3–12 month rearing period, depending on species, salmon smolts are released from the hatchery and commence downstream-saltwater migration.

Sources of protein, energy, essential fatty acids (EFA), and minerals for fry and developing smolts are generally provided in the form of aquafeeds produced mainly from fishmeal and other fishery products (i.e. fish oil, fish protein hydrolysates), (Tacon, 1994; National Research Council, 1993). Commercial aqauafeeds generally consist of whole bodies and/or parts of small fishes (e.g. anchovy, jack mackerel, pilchard, menhaden, capelin, tuna, herring, sardine, etc.) from the South Pacific and/or the North Atlantic regions (National Research Council, 1993). In some cases, other protein sources such as meals from meat and bone, poultry by-products, flash or spray-dried blood, feather, soybean, cottonseed and peanut are used in commercial fish feeds. The actual composition of these feeds depends on market availability and thus may vary accordingly.

Several studies have documented the occurrence of persistent organic pollutants (POPs) such as polychlorinated biphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzo furans (PCDFs) and organochlorine pesticides (OCPs) in commercial aquafeeds and farmed fish (Jacobs et al., 2002; Easton et al., 2002; Hites et al., 2004; Hamilton et al., 2005; Bell et al., 2005; Ikonomou et al., 2007;

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Maule et al., 2007). Recent studies show that replacement of fish oil/meal with vegetable oils and proteins, which is increasingly more common in commercial aquafeed production, can effectively reduce contaminant levels in feed and subsequently lower contaminant burdens in farm raised fish (Drew et al., 2007; Friesen et al., 2008).

POPs are long-lived organic chemicals that are generally resistant to chemical and biological degradation processes. Although many POPs have been restricted from use in North America and Europe since the 1970s, these compounds are still present in food chains worldwide, in some cases surpassing adverse effect levels or human consumption guidelines (Giesy et al., 1999; deBruyn et al., 2004; Hites et al., 2004). Certain PCB, PCDD and PCDF congeners may significantly contribute to "dioxin-like" toxicity (Safe et al., 1985) and related endocrine disruption effects (Krishnan and Safe, 1993). PCB exposure in developing Atlantic salmon smolts has been shown to negatively impact smolt behaviour and physiology, which may ultimately effect survival during saltwater migration (Lerner et al., 2007). Also, the environmental distribution of various endocrine disrupting chemicals (EDCs), including natural/synthetic estrogens as well as various commercial chemicals, may potentially impact development, growth and reproduction in wild fish (Jobling et al., 1998). A recent whole-ecosystem experiment involving the addition of 17α-ethynylestradiol to a freshwater Canadian lake indicates chronic exposure to estrogens and/or their mimics can impact fish reproduction and ultimately lead to population collapse (Kidd et al., 2007).

Chemical bioaccumulation in fish occurs through a combination of equilibrium partitioning between ambient water and fish lipids (bioconcentration) and dietary exposure and gastrointestinal uptake (biomagnification), (Gobas et al., 1993b, 1999; Fisk et al., 1998). For hydrophobic chemicals (with octanol–water partition coefficients or K_{OW}s $>10^5$) such as PCBs, PCDD/Fs and many OCPs, dietary uptake is the primary exposure route (Gobas et al., 1999). Biomagnification of POPs can occur due to efficient dietary absorption and very slow elimination rates of those compounds. Chemical biomagnification factors (BMFs), which are the ratio of a chemical's concentration (on a lipid weight basis) in an organism (C_B) and consumed food (C_D), (i.e., BMF=C_B/C_D, lipid wt.) typically range between 5 and 10 in fish (Gobas, 1993; Fisk et al., 1998; Morrison et al., 1997; Campfens and Mackay, 1997). It is important to assess the extent of dietary exposure and biomagnification of POPs in hatchery-raised smolts due to potential developmental effects associated with elevated burdens of these contaminants in fish (Safe et al., 1985; Walker and Peterson, 1991; Krishnan and Safe, 1993; Giesy et al., 2002).

In this paper, we report measured concentrations of organochlorine pesticides (OCPs), PCBs, PCDDs, PCDFs and corresponding 2,3,7,8 TCDD toxic equivalents (TEQs) in samples of commercial aquafeeds and in tissue of Pacific salmon fry and reared smolts collected from four federal fish hatcheries in BC. To assess potential toxicological impacts we also measured biochemical responses, including hepatic ethoxyresorufin *O*-deethylase (EROD) activity and cytochrome P450 (CYP1A) levels in hatchery smolts that exhibited the highest TEQs. The study provides a comprehensive assessment of (i) levels and patterns of a wide range of organic contaminants in several brands of commercial aquafeeds and (ii) the extent of dietary accumulation and toxicological implications of chemical residue levels in young developing salmon smolts from British Columbia hatcheries.

2. Materials and methods

2.1. Sites and sampling

Between February 1999 and May of 2000, samples of commercial aquafeed and Pacific salmon fry and smolts were obtained from four federal hatcheries from coastal BC, including Chilliwack (CW), Kitimat (KM) and Robertson Creek (RC) and Big Qualicum (BQ) fish hatcheries. Sample collection and hatchery information is provided in Table 1. A map showing locations of the four federal hatcheries is given in Fig. S1 (Supporting Information). Salmon species studied included chinook (*Oncorhynchus tshawytscha*) at Chilliwack, Kitimat and Robertson Creek hatcheries and coho (*Oncorhynchus kisutch*) at Big Qualicum. In total, we collected 22 samples of commercial aquafeed, including various formulations of Moore–Clarke (n=9), OMP (n=4) and EWOS (n=9). From these hatcheries we first collected samples of unfed salmon fry prior to feeding as well as samples of reared smolts (3–12 months later). Throughout the rearing process hatchery fry and smolts are fed a series of four starter feeds. At the time of the present study, Chilliwack used Moore–Clarke starter feeds (n=4), Kitimat used OMP starter feeds (n=4), Robertson Creek EWOS starter feeds (n=4) and Big Qualicum used a

Table 1
Sampling information for commercial aquafeed, fry and smolts collected at four federal fish hatcheries from coastal British Columbia

	Chilliwack	Kitimat	Robertson Creek	Big Qualicum
Species	Chinook	Chinook	Chinook	Coho
Rearing period	3 months	3 months	3 months	12 months
Sampling date (Fry)	February 1999	February 1999	February 1999	April 1999
Sampling date (Smolts)	April 1999	April 1999	April 1999	May 2000
Sampling date (Feeds)	February–April 1999	February–April 1999	February–April 1999	April 1999–May 2000
Feed brand	Moore–Clarke[a]	OMP[b]	EWOS vextra[c]	EWOS vextra+Moore–Clarke
Feed samples	n=9	n=4	n=9	n=9
Fry samples[d]	n=1	n=1	n=1	n=1
Smolt livers	n=3[e]	n=4[f]	n=3[g,h]	n=3[i]
Smolt carcass	NA	n=3[j]	n=3	n=6
PCBs, PCDD/F s	Aquafeed	Aquafeed	Aquafeed	Aquafeed
	Fry	Fry	Fry	Fry
	Smolt livers	Smolt livers	Smolt livers	Smolt livers
		Smolt carcass	Smolt carcass	Smolt carcass
Pesticides	Aquafeed only	Aquafeed only	Aquafeed only	Aquafeed only
EROD, Cytochrome P450 1A	NA	NA	n=6	NA

[a] Moore–Clarke feed consisted of Starter # 0; Starter # 1; Starter # 2; Starter # 3 feeds.
[b] OMP feed samples consisted of feeds with pellet sizes 1/32", 3/64", 1/16", 3/32".
[c] EWOS vextra samples consisted of Starter # 1; Starter # 2; Starter # 3 feeds.
[d] Fry samples consisted of one pooled composite sample (pool of >50 individual fry carcasses).
[e] Note these are pooled samples. Each sample consisted of 178, 185 and 164 individual smolt livers.
[f] Note these are pooled samples. Each sample consisted of 44, 91, 106 and 105 individual smolt livers.
[g] Note these are pooled samples. Each sample consisted of 237, 120 and 232 individual smolt livers.
[h] 6 smolt liver samples were taken for cytochrome P450 1A (CYP1A) analysis.
[i] Note these are pooled samples. Each independent sample consisted of >100 individual smolt livers.
[j] Note: smolt carcass samples represent whole fish except those sampled at the Kitimat hatchery, which had livers removed prior to chemical analysis.

combination of EWOS and Moore–Clarke starter feeds ($n=4$). Individual aquafeed samples were different formulations used at different times during the smolt growth period. For example, Moore–Clarke and EWOS vextra feeds were supplied first using starter # 0 (t_0), followed by starter # 1(t_1), starter # 2 (t_2) and starter # 3 (t_3). Similarly, OMP feed is categorized by pellet size (i.e., initial feed=1/32", followed by subsequent feeds with pellet sizes of 2/64", 1/16" and 3/32", which can be classified as starter #s 0–3 (t_0–t_3). All aquafeed formulations (i.e., Moore–Clarke, OMP, and EWOS) were analyzed for contaminant residue concentrations. At each hatchery we collected numerous unfed fry and post-feed smolts in order to obtain pooled samples of excised liver tissue and/or whole body (i.e., carcass). At one hatchery (Robertson Creek), composite smolt liver samples ($n=6$, pooled of >100 fish) were obtained for EROD and CYP1A determinations. Those livers used for EROD and CYP1A analysis were removed with scalpel and forceps immediately after killing the fish then snap frozen in liquid nitrogen then stored at −80 °C at all times prior to analysis. Pooled tissue samples (liver and/or carcass) used for contaminant residue analysis were obtained from the killed fish at Chilliwack ($n=3$, pool of >100 fish), Kitimat ($n=3$–4, pool of >100 fish), Robertson Creek ($n=3$, pool of >100 fish) and Big Qualicum ($n=3$–6, pool of >100 fish) using solvent rinsed utensils and stored at −20 °C prior to analyses.

2.2. Contaminant analysis

The methods used for simultaneous extraction, cleanup and quantification of PCBs, PCDDs, PCDFs and pesticides are reported in detail elsewhere (MacDonald et al., 1997; Ikonomou et al., 2001). Extraction and cleanup of all aquafeed, fry and smolt samples were performed at the Institute of Ocean Sciences, Sidney, BC, Canada. Briefly, samples (5–10 g) were homogenized using Na_2SO_4 in a mortar and spiked with a mixture of surrogate internal standards which contained all $^{13}C_{12}$-labeled PCDDs and PCDFs, $^{13}C_{12}$-labeled PCBs and $^{13}C_{12}$-labeled OCPs. Homogenates were transferred quantitatively into a 30 cm glass extraction column (3 cm outer diameter) and extracted with DCM/hexane (1:1 v/v). The extract was reduced to a few millilitres by rotary evaporation and recollected in 5 mL of 1:1 DCM:hexane. Extracts were first cleaned up by gel permeation chromatography (GPC) to remove bulk lipids. Because many pesticides are acid labile, the extracts were split by volume into two aliquots, 70% to be used for PCB and PCDD/F cleanup and 30% for pesticides. The pesticide extracts were cleaned up using florisil chromatography. For PCBs and PCDD/Fs the extracts were cleaned up using (i) silica gel chromatography (with layers of basic, neutral, acidic, neutral silica); (iii) activated alumina chromatography; and (iv) carbon fibre chromatography. Neutral silica was activated overnight, at 200 °C and then cooled to room temperature in a desiccating chamber over anhydrous calcium sulphate. Acidic silica was prepared by adding 25 g concentrated sulfuric acid to 50 g of neutral silica and shaken to a free-flowing powder. Basic silica was prepared by adding 14 g of 1 N sodium hydroxide (made up with toluene-washed water) to 40 g of neutral silica and shaken to a free-flowing powder. The amounts used to prepare the silica columns were: 2 g basic, 1 g neutral, 4 g acidic, 1 g neutral. Alumina was activated at 200 °C overnight and cooled to room temperature in a desiccating chamber until usage. The column was dry-packed with 10 g activated alumina followed by 1 g sodium sulphate. Granular anhydrous sodium sulphate was baked at 450 °C overnight and cooled to room temperature in a desiccating chamber.

Fractionation of the eluent containing PCBs, PCDDs and PCDFs was accomplished with an automated high performance liquid chromatography (HPLC) system utilising a carbon fibre (CF) packed with a 1:12 mixture of activated carbon/filter paper homogenate. Four separate fractions, including (I) di-*ortho* PCBs using 3% DCM in hexane at 2 mL·min^{-1} for 10 min, (II) mono-*ortho* PCBs using 50:50 DCM/cyclohexane at 2 mL·min^{-1} for 11 min, (III) non-*ortho* PCBs using 50:50 ethyl acetate:benzene at 2 mL·min^{-1} for 25 min and (IV) PCDDs and PCDFs in toluene backflush of the CF column at 4 mL·min^{-1} for 15 min. Each fraction was concentrated to less than 10 μL and spiked with the corresponding ^{13}C-labeled method performance standards $^{13}C_{12}$-1,2,3,4-TeCDD, $^{13}C_{12}$-1,2,3,7,8,9-HxCDD and $^{13}C_{12}$-PeCB-111) prior to instrumental analysis.

Extracts were analyzed for full congener PCDD/Fs and PCBs at the Institute of Ocean Sciences (IOS) laboratory using gas chromatography/high resolution mass spectrometry (GC/HRMS). Specifically, PCDD/F analyses included quantification of 17 congeners and PCB analyses included quantification of 167 individual congeners plus 34 coeluting bands (See Table S1, Supporting Information). The high resolution mass spectrometer was a Micromass Ultima (Micromass, UK) instrument equipped with an HP-6890 gas chromatograph and a CTC autosampler. We used a DB-5 fused silica capillary column (60 m×0.25 mm i.d., 0.1 μm film thickness, J&W Scientific, Folsom CA). Analyte solution (1 μl) was injected in splitless mode, at an injector temperature of 282 °C. For all analyses the HRMS was operated at 10,000 resolution under positive EI conditions and data were acquired in the Single Ion Resolving Mode (SIR). The GC/HRMS conditions, the criteria used for congener identification and quantification and the quality assurance—quality control procedures used for the quantification of PCDD/Fs and PCBs are described in detail elsewhere (MacDonald et al., 1997; Ikonomou et al., 2001). Samples were run in batches of 12, which included a procedural blank (consisting of Na_2SO_4), a performance evaluation standard (i.e., Certified Reference Material: namely a fortified salmon homogenate-CRM-EDF-2526-purchased from Cambridge Isotope Laboratories), and 10 samples (one of which was a replicate). Analyte concentrations were calculated by the internal standard isotope-dilution method using mean relative response factors (RRFs) determined from calibration standard runs made before and after each batch of samples was analyzed. Recoveries of all surrogate internal standards were between 60 and 110%. The limits of detection (LODs) for individual PCDD/Fs were between 0.15 and 0.40 pg/g%. LODs for individual PCBs were between 0.25 and 2.50 pg/g.

Pesticide quantification was conducted at the Freshwater Institute in Winnipeg, Canada. Analytes included toxaphene (full congener), chlordanes (heptachlor, heptachlor epoxide, C1A, C2/U5, C1b/U6, U3, U1, C3, C5, oxychlordane, *cis*- and *trans*-chlordanes and *cis*- and *trans*-nonachlor), DDTs (*p,p′*-DDT, *p,p′*-DDE and *p,p′*-DDD), hexachlorocyclohexane (α-HCH and β-HCH), chlorobenzenes (Penta-CBz and Hexa-CBz), deildrin, mirex, photo-mirex, pentachloroanisole (PCA), α-endosulphan (I), methoxychlor, triveratrol, tetraveratrol, and endrin, Toxaphene was performed by full congener gas chromatography/electron-capture negative ion high resolution mass spectrometry (GC/ECNI-HRMS) fitted with a 60 m DB-5MS capillary column (0.25 mm i.d., 0.25 μm film thickness, J&W Scientific, Folsom CA) as described in (Braekevelt et al., 2001). All other pesticides were determined by high resolution gas chromatography with an electron-capture detector (GC/ECD) using a 60 m DB-5 fused silica capillary column (0.25 mm i.d., 0.25 μm film thickness, J&W Scientific), as previously described (Stern et al., 2005). LODs for individual pesticides were between 0.01 and 0.03 ng/g.

Lipid contents of all aquafeed and salmon tissue samples were determined gravimetrically from a parallel extraction of a 5 g sub-sample (wet weight). Samples were homogenized and extracted with 100 mL of 1:1 DCM:hexane (as above), reduced by turboevaporation to a few mL, transferred into a preweighed aluminum dish, dried at 40 °C overnight and subsequently weighed. The measured weight of remaining lipid was used to calculate the percentage of lipid (% lipid) of original wet sample weight. Moisture content was determined by oven drying the sample at 105 °C for 48 h and weighing the sample before and after drying.

2.3. Biochemical response measurements

Liver tissue from Robsertson Creek smolts were stored at −80 °C were assessed for EROD activity, and CYP1A protein levels. EROD activity (in units of pmol/mg/min) was measured using a spectrofluorometric assay (Burke et al., 1985). Immunoblotting and densitometric quantitation of CYP1A was done using polyacrylamide gel electrophoresis (PAGE), (Laemmli, 1970). Samples were applied to the gels at 20 to 40 μg protein per lane. Microsomal proteins resolved by SDS–PAGE were transferred

electrophoretically onto nitrocellulose and probed with antibody (Towbin et al., 1979). Membranes were incubated with rabbit anti-trout CYP1A peptide IgG at a concentration of 10 $\mu g \cdot mL^{-1}$ (Lin et al., 1998). Bound primary antibody was located using alkaline phosphatase-conjugated goat anti-rabbit IgG. Immunoreactive proteins were detected by reaction with a substrate solution containing 0.01% nitro blue tetrazolium, 0.05% 5-bromo-4-chloro-3-indolyl phosphate, and 5 mM $MgCl_2$ in 0.1 M Tris–HCl buffer, pH 9.5. Assay conditions were optimized to ensure that colour development did not proceed beyond the linear range of the phosphatase reaction. Staining intensities of the immunoreactive bands were quantified with a PDI 420 oe scanning densitometer integrated with Quantity One Version 3 software (PDI Inc., Huntington Station, NY). The amount of immunoreactive protein was determined from the integral of the optical density of the stained band. Staining intensities of bands on each blot were normalized with a purified rat hepatic CYP1A1. A purified rat CYP1A1 sample was included on every gel as an internal standard to correct for blot-to-blot and day-to-day variability. The CYP1A protein bands were expressed as relative optical density per mg protein, i.e., ROD/mg protein rather than in terms of microsomal content (e.g., pmol/mg) because purified salmon CYP1A enzyme is not available for use as a calibration standard for quantification.

2.4. Data analysis and statistics

Concentration data are reported as ng/g lipid for PCBs and pesticides and pg/g lipid for PCDDs, PCDFs and 2,3,7,8 TCDD toxic equivalents (TEQs). TEQs were determined for individual PCDD/Fs and dioxin-like PCBs (DL-PCBs) using World Health Organization toxic equivalency factors (WHO-TEFs) developed for assessing 2,3,7,8 TCDD toxicity in fish (Van den Berg et al., 1998). Specifically, TEQs for individual congeners were determined by multiplying their measured concentrations by the corresponding TEF (Table S2, Supporting Information). ∑PCDD-TEQs, ∑PCDF-TEQs, ∑PCB-TEQs and a total PCDD/F+PCB TEQs (∑TEQs) were determined from the sums of the various toxic equivalent concentrations. For reared smolts, we determined lipid corrected biomagnification factors (BMFs) for individual PCBs and PCDD/Fs, which were calculated as the ratio of mean tissue residue concentration (on a lipid weight basis) in the fish (C_B) and their last consumed feed (C_D), (i.e., BMF=C_B/C_D, lipid wt.). BMFs were calculated to serve as a future guide for assessing contaminant concentrations in smolts from known concentrations in aquafeed. Physical–chemical properties including molecular weights (MW, $g \cdot mol^{-1}$) and octanol–water partition coefficient (K_{OW}s) of the various POPs were compiled from the literature (Mackay et al., 1992) to assess the influence of chemical properties on chemical bioaccumulation behaviour in smolts. One-Way Analyses of Variance (ANOVA) and Tukey's HSD comparison tests were performed to evaluate differences between chemical concentrations observed in aquafeeds and/or fish. Also, linear regression was used to evaluate the relationship between contaminant concentrations and lipid contents in aquafeed.

3. Results

3.1. Levels and patterns of POPs in commercial aquafeeds from BC fish hatcheries

Measured POP concentrations in commercial aquafeeds and Pacific salmon fry/smolts from four BC fish hatcheries are summarized in Tables 2 and 3, respectively. Mean ∑PCB concentrations observed in

Table 2
Mean concentrations of PCBs (ng/g lipid), PCDD/Fs (pg/g lipid), 2,3,7,8 TCDD Toxic equivalents (TEQs, pg/g lipid) and several pesticides in three commercial aquafeeds used at hatcheries from coastal British Columbia

	Commercial aquafeed		
	Moore–Clarke (n=9)	OMP (n=4)	EWOS (n=9)
% Lipid	19.6±2.63	15.4±2.15	14.9±2.14
DL-PCBs (ng/g lipid)			
PCB77	0.13±0.06	0.19±0.10	0.36±0.12
PCB81	0.005±0.002	0.009±0.005	0.013±0.004
PCB105	2.53±1.4	5.61±1.67	4.70±1.31
PCB114	0.14±0.08	0.37±0.09	0.27±0.06
PCB118	6.92±3.77	14.3±4.06	13.7±3.30
PCB123	0.11±0.05	0.29 0.09	0.43±0.31
PCB126	0.04±0.02	0.06±0.01	0.07±0.02
PCB156	0.66±0.34	1.54±0.44	1.52±0.41
PCB157	0.18±0.09	0.40±0.1	0.46±0.12
PCB167	0.37±0.21	0.87±0.19	0.96±0.28
PCB169	0.01±0.01	0.01±0.002	0.02±0.01
PCB189	0.05±0.03	0.15±0.05	0.17±0.06
∑PCBs[a]	154±89	319±101	376±88
Dioxins/Furans (pg/g lipid)			
2,3,7,8-TCDD	0.73±0.55	0.88±0.06	1.52±0.55
1,2,3,7,8-PeCDD	1.93±1.71	1.74±0.41	2.54±0.91
1,2,3,4,7,8-HxCDD	1.26±1.36	0.78±0.23	1.10±0.36
1,2,3,6,7,8-HxCDD	3.04±2.28	5.19±0.54	4.87±1.31
1,2,3,7,8,9-HxCDD	1.81±2.52	1.58±0.45	2.22±0.95
1,2,3,4,6,7,8-HpCDD	4.53±6.55	10.1±3.71	7.71±3.04
OCDD	30.3±45.5	82.7±36.3	47.9±22.3
2,3,7,8-TCDF	6.68±3.10	8.91±0.88	9.43±2.56
1,2,3,7,8-PeCDF	1.30±0.36	0.85±0.03	1.59±0.54
2,3,4,7,8-PeCDF	3.81±1.79	1.50±0.41	2.91±0.98
1,2,3,4,7,8-HxCDF	0.66±0.10	0.60±0.01	0.89±0.38
1,2,3,6,7,8-HxCDF	0.60±0.09	n.d.	0.71±0.23
2,3,4,6,7,8-HxCDF	0.65±0.11	n.d.	0.87±0.48
1,2,3,7,8,9-HxCDF	n.d.	n.d.	0.75
1,2,3,4,6,7,8-HpCDF	0.82±0.47	1.51±0.76	1.07±0.33
1,2,3,4,7,8,9-HpCDF	0.90±0.40	n.d.	n.d.
OCDF	0.98±0.27	3.18±1.96	1.20±0.32
∑PCDDs[b]	40.6±59.0	103±38.8	67.6±28.4
∑PCDFs[b]	13.1±5.80	14.7±3.0	16.3±5.24
2,3,7,8 TCCD Toxic Equivalents (TEQs) (pg/g lipid)[c]			
∑PCDD TEQ	1.61±2.46	2.66±0.79	4.46±1.69
∑PCDF TEQ	2.03±1.33	1.26±0.20	2.15±0.72
∑PCB TEQ	0.36±0.19	0.53±0.13	0.69±0.21
∑TEQs	3.99±2.94	4.45±1.08	7.31±2.36
Pesticides (ng/g lipid)[d]			
Heptachlor epoxide	2.76	2.66	4.63
Dieldrin	15.1	7.62	20.3
Mirex	n.d.	n.d.	n.d.
Photomirex	n.d.	n.d.	n.d.
Pentachloro-anisole	0.63	0.80	1.65
α-Endosulfan	n.d.	n.d.	n.d.
Methoxychlor	n.d.	n.d.	n.d.
Triveratrol	1.52	n.d.	n.d.
Tetraveratrol	0.27	n.d.	n.d.
Endrin	3.08	1.46	2.47
∑CBz	10.1	5.81	16.3
∑HCHs	2.10	7.76	10.4
∑Chlordanes	23.9	53.7	63.5
∑DDTs	39.1	380	140
∑Toxaphene	204	174	227
∑PESTs	302	634	486

Notes to Table 2:
Bold text represents the sum of a given compound class.
n.d.=non-detectable concentration.
[a] Full congener PCBs: 167 individual congeners and 34 coeluting bands of di-ortho, mono-ortho and non-ortho PCBs.
[b] PCDDs (7 congeners), PCDFs (10 congeners).
[c] TEQs were calculated using WHO-TEFs for fish from Van den Berg et al. (1998). ∑PCDD TEQs were calculated from concentrations and WHO-TEFs of 2,3,7,8-TCDD, 1,2,3,7,8-PeCDD, 1,2,3,4,7,8-HxCDD, 1,2,3,6,7,8-HxCDD, 1,2,3,7,8,9-HxCDD, 1,2,3,4,6,7,8-HpCDD, OCDD. ∑PCDF TEQs were calculated from concentrations and WHO-TEFs of 2,3,7,8-TCDF, 1,2,3,7,8-PeDF, 2,3,4,7,8-PeDF, 1,2,3,4,7,8-HxCDF, 1,2,3,6,7,8-HxCDF, 2,3,4,6,7,8-HxCDF, 1,2,3,7,8,9-HxCDF, 1,2,3,4,6,7,8-HpCDF, 1,2,3,4,7,8,9-HpCDF, OCDF. ∑PCB TEQTEQs were calculated from concentrations and WHO-TEFs of PCB77, PCB81, PCB105, PCB114, PCB118, PCB123, PCB126, PCB156, PCB157, PCB167, PCB169, PCB189. ∑TEQ=∑PCDD TEQ+∑PCDF TEQ+∑PCB TEQ.
[d] ∑CBz=sum of pentachlorobenze (PeCBz) and hexachlorobenzene (HCBz). ∑HCHs=sum of α-, β- and γ- HCH. ∑Chlordanes=sum of *cis*- and *trans*-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, heptachlor and heptachlor epoxide. ∑DDTs=sum of *p,p′*-DDT, *o,p′*-DDT, *p,p′*-DDE, *o,p′*-DDE, *p,p′*-DDD, *o,p′*-DDD. ∑Toxaphene=sum of 27 chloroborane (CHB) peaks in technical mixture.

Table 3
Mean concentrations of ∑PCB (ng/g lipid), and ∑PCDD, ∑PCDF and ∑TEQs (pg/g lipid) in unfed salmon fry carcasses (t_0) and post-feed salmon smolt liver and carcasses (t_3=6 months) collected at Chilliwack (CW), Kitimat (KM) and Robertson Creek (RC) and Big Qualicum (BQ) hatcheries from coastal British Columbia

	Chilliwack (Species: Chinook) (Feed: Moore–Clarke)			Kitimat (Species: Chinook) (Feed: OMP)			Robertson Creek (Species: Chinook) (Feed: EWOS)			Big Qualicum (Species: Coho) (Feed: EWOS+Moore–Clarke)		
	Unfed fry (t_0) (n=1)	Smolt liver (t_3) (n=3)	Smolts carcass[c] (t_3) N/A	Unfed fry (t_0) (n=1)	Smolt liver (t_3) (n=4)	Smolts carcass (t_3) (n=3)	Unfed fry (t_0) (n=1)	Smolt liver (t_3) (n=3)	Smolt carcass[c] (t_3) (n=3)	Unfed fry (t_0) (n=1)	Smolt liver (t_3) (n=3)	Smolt carcass (t_3) (n=6)
% Lipid	3.93 (–)	4.32 (0.09)	–	4.79 (–)	4.30 (0.17)	6.43 (0.08)	2.86 (–)	5.47 (0.52)	16.0 (4.2)	2.19 (–)	3.56 (0.32)	6.20 (1.6)
∑PCBs[a] (ng/g lipid)	691 (–)	285 (24)	–	453 (–)	272 (62)	397 (15)	244 (–)	756 (47)	303 (161)	474 (–)	310 (47)	563 (137)
∑PCDDs[b] (pg/g lipid)	25.4 (–)	22.2 (3.7)	–	72.1 (–)	53.3 (27.3)	11.0 (4.48)	25.3 (–)	32.8 (8.14)	2.01 (0.91)	41.6 (–)	21.6 (10.4)	15.5 (11.8)
∑PCDFs[b] (pg/g lipid)	43.9 (–)	13.7 (2.3)	–	10.0 (–)	7.94 (2.0)	8.08 (0.32)	8.04 (–)	21.3 (3.3)	7.04 (2.54)	45.2 (–)	9.76 (1.55)	17.7 (10.4)
∑PCB TEQ[c] (pg/g lipid)	1.27 (–)	0.58 (0.05)	–	0.61 (–)	0.45 (0.11)	0.57 (0.01)	0.26 (–)	1.29 (0.08)	0.56 (0.16)	0.70 (–)	0.57 (0.12)	0.94 (0.18)
∑PCDD TEQ[c] (pg/g lipid)	4.93 (–)	0.71 (1.1)	–	0.07 (–)	0.09 (0.07)	0.73 (1.04)	0.09 (–)	3.34 (1.8)	0.37 (0.32)	5.23 (–)	1.64 (2.49)	4.02 (2.49)
∑PCDF TEQ[c] (pg/g lipid)	4.61 (–)	1.96 (1.3)	–	0.50 (–)	0.36 (0.10)	0.40 (0.02)	0.40 (–)	2.3 (0.25)	0.55 (0.33)	4.32 (–)	0.49 (0.08)	1.77 (0.94)
∑TEQ[c] (pg/g lipid)	10.8 (–)	3.25 (2.3)	–	1.18 (–)	0.91 (0.27)	1.70 (1.1)	0.75 (–)	6.92 (2.1)	1.49 (0.71)	10.2 (–)	2.70 (2.5)	6.73 (3.6)

Numbers in brackets are standard deviations. n.d.=non-detectable concentration. NA=not analyzed.
[a] Full congener PCBs: 167 individual congeners and 34 coeluting bands of di-ortho, mono-ortho and non-ortho PCBs.
[b] PCDDs (7 congeners), PCDFs (10 congeners).
[c] TEQs were calculated using WHO-TEFs for fish from Van den Berg et al. (1998). ΣPCDD TEQs were calculated from concentrations and WHO-TEFs of 2,3,7,8-TCDD, 1,2,3,7,8-PeCDD, 1,2,3,4,7,8-HxCDD, 1,2,3,6,7,8-HxCDD, 1,2,3,7,8,9-HxCDD, 1,2,3,4,6,7,8-HpCDD, OCDD. ΣPCDF TEQs were calculated from concentrations and WHO-TEFs of 2,3,7,8-TCDF, 1,2,3,7,8-PeDF, 2,3,4,7,8-PeDF, 1,2,3,4,7,8-HxCDF, 1,2,3,6,7,8-HxCDF, 2,3,4,6,7,8-HxCDF, 1,2,3,7,8,9-HxCDF, 1,2,3,4,6,7,8-HpCDF, 1,2,3,4,7,8,9-HpCDF, OCDF. ΣPCB TEQs were calculated from concentrations and WHO-TEFs of PCB77, PCB81, PCB105, PCB114, PCB118, PCB123, PCB126, PCB156, PCB157, PCB167, PCB169, PCB189. ∑TEQ=ΣPCDD TEQ+ΣPCDF TEQ+ΣPCB TEQ.

Moore–Clarke, OMP and EWOS aquafeeds were 154.3±88.0, 319.8±101 and 376.3±88.2 ng/g lipid, respectively (Table 2). PCB concentrations in the Moore–Clarke feed were significantly lower ($p<0.05$) than those concentrations in the EWOS and OMP feeds. ∑PCDD and ∑PCDF concentrations in all aquafeed samples (13–103 pg·g^{-1} lipid), were several orders of magnitude below observed ∑PCB levels.

Mean 2,3,7,8 TCDD toxic equivalents (TEQs) in the Moore–Clarke, OMP and EWOS aquafeeds were comparable at concentrations of 3.99±2.94, 4.50±1.08, 7.30±2.36 pg·g^{-1} lipid, respectively (Table 2). PCDD/Fs generally exhibited 85–90% of ∑TEQs in aquafeeds. The major contributing PCDD/F congeners included 2,3,7,8 TCDD, 1,2,3,7,8 PeCDD, 2,3,7,8 TeCDF and 2,3,4,7,8 PeCDF. DL-PCBs (mainly PCB-126 and PCB-169) provided the balance (10–15%) of ∑TEQ.

Organochlorine pesticide concentrations in aquafeeds ranged from 0.6 ng/g lipid for pentachloroanisole (PCA) to over 200 ng/g lipid for ∑DDTs and ∑Toxaphene (Table 2). While DDTs, toxaphenes and chlordanes generally exhibited the highest concentrations, several pesticides were below detection limits (e.g., mirex, photmirex, α-endosulfan, methoxychlor). Other common pesticides such as dieldrin, endrin, CBz and HCHs were routinely detected in the feeds at concentrations between 1 and 20 ng/g lipid. The Moore–Clarke feed exhibited the lowest pesticide concentrations (compared to EWOS and OMP feeds), but this could not be confirmed statistically due to low sample size.

PCB and PCDD/F congener profiles were similar among the three aquafeeds and smolt tissue samples (Figs. 1 and 2). Tetrachloro (Cl_4)-heptachloro (Cl_7) PCBs (e.g., Cl_6-PCB-153, Cl_6-PCB-138, Cl_6-PCB-149, Cl_5-PCB-101, Cl_5-PCB-118, Cl_5-PCB-99, Cl_5-PCB-110 and Cl_7-PCB-180) were dominant, typically comprising >90% of ∑PCBs. OCDD, 1,2,3,6,7,8 HxCDD, 1,2,3,4,6,7,8 HpCDD and 2,3,7,8 TCDF were the dominant PCDD/F congeners. OCDD typically comprised >60% of the total PCDD/F concentrations. In contrast to the similar PCB and PCDD/F profile patterns observed in aquafeeds, the pesticide profiles were slightly different among the three feeds. In particular, OMP feeds exhibited a higher composition of DDTs compared to EWOS and Moore–Clarke feeds (Fig. 3).

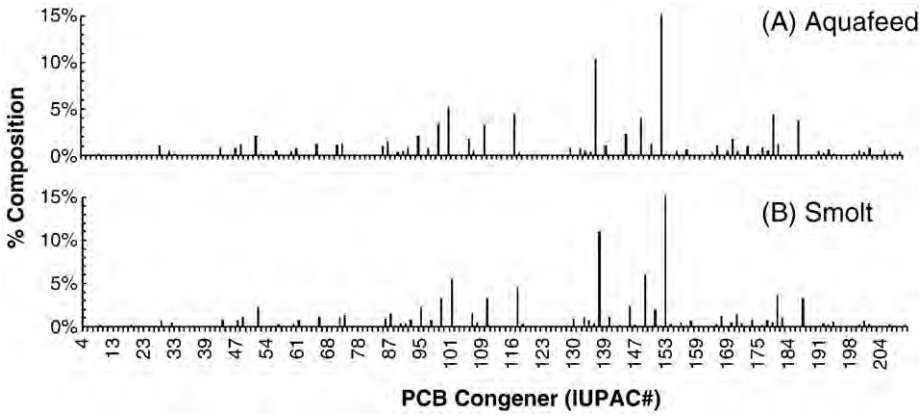

Fig. 1. Percent composition (% of ΣPCBs) for individual PCB congeners in (A) aquafeed and (B) Pacific salmon smolt liver from BC salmonid hatcheries. Congeners are numbered using IUPAC numbering system. For the following coeluting congeners, the congener in bold was the assigned number **4**/10, **7**/9, **8**/5, **16**/32, **33**/20, 42/**59**, **47**/48/75, **52**/73, **56**/60, 64/**71**/41, **70**/76, 74/**61**, 83/**109**, 84/**92**, **87**/115, **97**/86, **101**/90, **102**/93, 107/**108**, 117/125/**116**, **118**/106, 131/**142**, 134/**143**, **135**/144, **138**/163/164/160, **139**/140, **146**/161, **153**/132, **170**/190, 172/**192**, **174**/181, **187**/182, **203**/196.

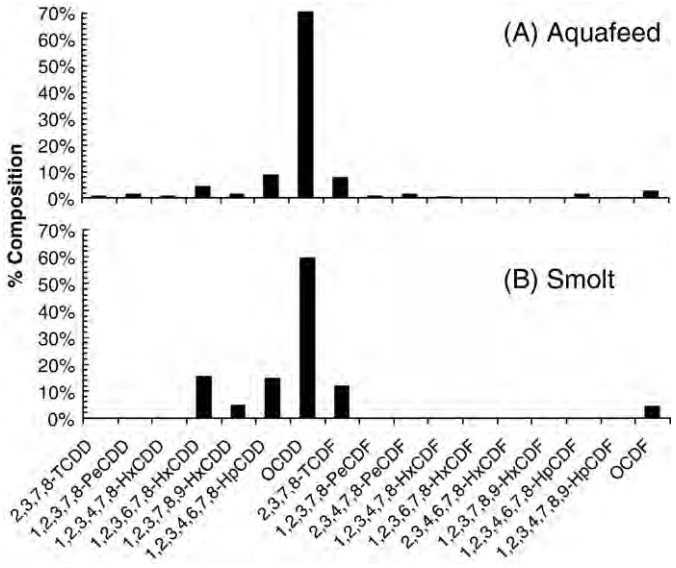

Fig. 2. Percent composition (% of ΣPCDD/Fs) for various PCDD and PCDF congeners in (A) aquafeed and (B) salmon smolt liver samples from BC salmonid hatcheries.

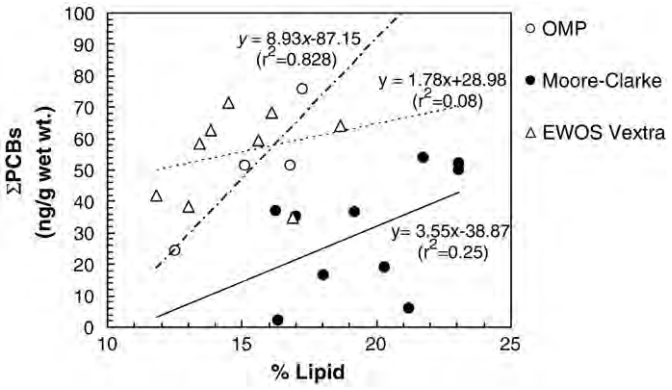

Fig. 4. Relationship between lipid content (% lipid) and ΣPCB concentration (ng/g wet weight) for three commercial aquafeeds, including OMP, EWOS and Moore–Clarke feeds.

3.2. Relationship between lipid content and contaminant concentrations in aquafeed

Lipid contents of the various aquafeeds analyzed in this study ranged between 11.8 and 23.3%. Mean lipid contents were highest in Moore–Clarke (19.6±2.6%) compared to OMP (15.4±2.1%) and EWOS (14.9±2.1%) feeds. For each of the feeds, contaminant concentrations (on a wet wt. basis) were positively correlated with the feed lipid content (Fig. 4). While Moore–Clarke feeds exhibited the highest lipid content, those feeds generally had the lowest contaminant concentrations.

3.3. Bioaccumulation of POPs in hatchery-fed salmon smolts

Mean ∑PCB concentrations in unfed fry samples were highest in chinook at Chilliwack (691 ng/g lipid) and lowest in chinook at Robertson Creek (244 ng/g lipid), (Table 3). Conversely, mean ∑PCB concentrations in chinook smolts were lowest at Chilliwack (272±25 ng/g lipid in liver tissue). Smolts at Robertson Creek exhibited the highest ∑PCBs (756±47 ng/g lipid in liver tissue). Robertson Creek chinook smolts also demonstrated relatively high ∑PCDD/F and ∑TEQ concentrations (∑PCDD/F=54.0±11.5 pg/g lipid, ∑TEQ=6.53±2.0 pg/g lipid in liver) compared to smolts from other hatcheries. The fact that chinook smolts at Robertson creek exhibited the highest contaminant concentrations (even though fry concentrations were lowest) is likely due to exposure of higher concentrations present in the EWOS aquafeeds.

In some cases, contaminant concentrations in liver tissue were significantly different ($p<0.05$) than concentrations observed in carcass samples (Table 3). For example, ∑PCBs in smolt livers at Robertson Creek (756±47 ng/g lipid) were 3 times higher than ∑PCBs observed in carcass samples of those smolts (303±161 ng/g lipid).

After the loss of the egg sac hatchery fry smolts are fed a series of starter feeds generally from starter # 0 to starter # 3. This feeding regime is designed to enhance nutritional value in series by increasing dietary lipid as the fish mature prior to release. Consequently, wet weight contaminant concentrations in the feed generally increase over the course of the rearing process (Fig. 5). For example, at the Chilliwack hatchery, lipid content of the feed increased from 16% (Starter # 0) to 21% (Starter # 3), resulting in ∑TEQ increases in those feeds from 0.84 to 1.14 pg/g wet wt. (Fig. 5A). Interestingly, at one hatchery (Big Qualicum) contaminant concentrations in the final feed (starter # 3) was substantially lower than the prior three feeds (starter #s 0–2), even though lipid contents increased. This is due to a change in aquafeed brand for the starter # 3 feed (i.e., EWOS for starter #s 0–2 and Moore–Clarke for starter # 3). ∑TEQs in the Moore–Clarke feed (0.57 pg/g) were approximately 2 times lower than the previous EWOS starter # 2 feed (1.24 pg/g) used at the Big Qualicum hatchery, (Fig. 5D). Although dietary exposure to smolts increased over this period, TEQs in the smolts were relatively low. For example, chinook smolts at Chilliwack exhibited ∑TEQs of 0.14±0.1 pg/g wet wt., which is approximately 8 times lower than the final consumed feed (1.14 pg/g wet wt.).

Calculated biomagnification factors (BMFs) for selected PCB and PCDD/F congeners varied between 0.2 and 3 in these smolts. No distinct relationship between BMFs and chemical K_{OW} (i.e., hydrophobicity) was apparent from the data (Fig. 6).

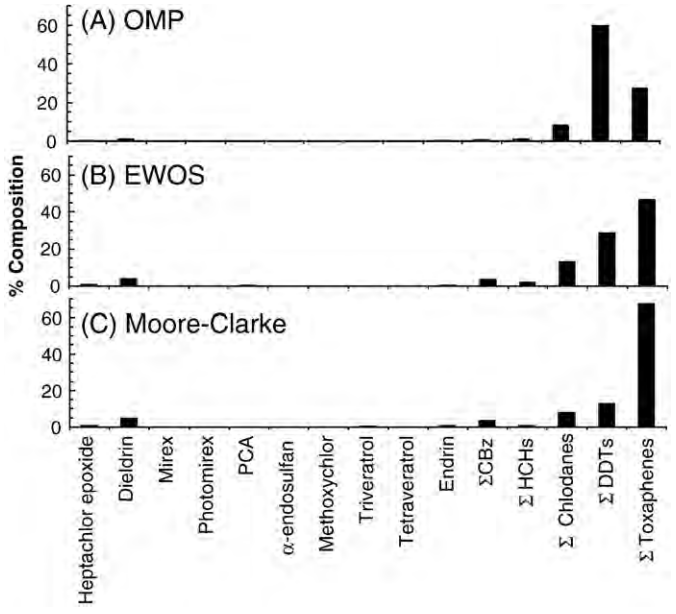

Fig. 3. Percent composition (% of ΣPesticides) for various pesticides in commercial aquafeeds used at BC salmonid hatcheries, including (A) OMP, (B) EWOS and (C) Moore–Clarke feeds.

3.4. Biochemical responses

To assess the potential biological effects in hatchery salmon caused by exposure to all aryl hydrocarbon (Ah) receptor inducers (i.e. PCBs, Dioxins, PAHs, etc.), hepatic EROD activity and CYP1A protein content were measured in samples of chinook smolt livers from Robertson Creek

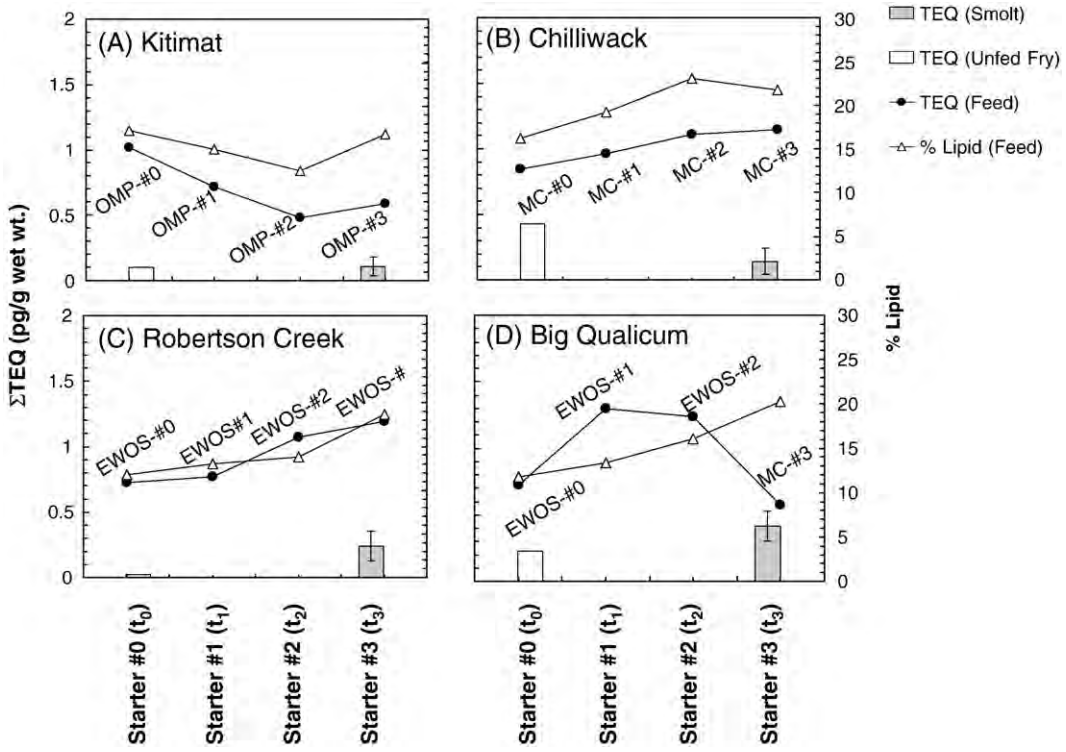

Fig. 5. Plot showing changes in lipid content (% lipid) and ΣTEQs (pg/g wet weight) in various starter feeds provided during the rearing process as well as ΣTEQ concentration (pg/g wet weight) in unfed fry carcass (t_0) and post-feed salmon smolt carcass (t_3) at (A) Kitimat, (B) Chilliwack, (C) Robertson Creek and (D) Big Qualicum hatcheries.

hatchery. Although these smolts exhibited the highest TEQs of all the smolts in the present study, EROD activity (3.88±2.46 pmol//mg/min) and CYP1A levels (4.2±1.6 ROD/mg) were relatively low.

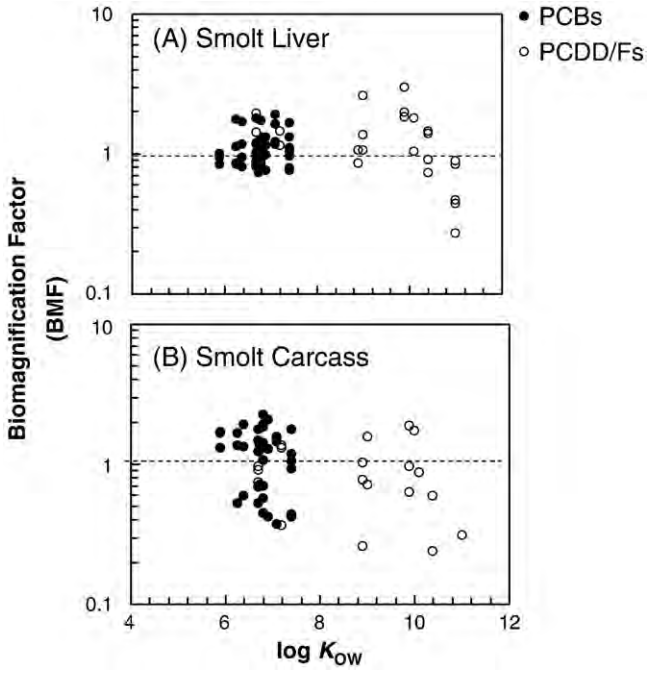

Fig. 6. Biomagnification factors (BMFs) of selected PCBs and PCDD/Fs observed in (A) liver and (B) carcass of Pacific salmon smolts from BC salmonid hatcheries versus the congener's octanol–water partition coefficient (log K_{OW}). Plotted BMF data represent ratios of lipid corrected concentrations of 14 selected PCB congeners and 10 PCDD/F congeners in hatchery smolts and their last consumed feed (i.e., starter # 3).

4. Discussion

Measured POP concentrations in aquafeeds from BC hatcheries (this study) are generally comparable to previously reported levels in commercial aquafeeds (Table 4). For example, ∑PCB levels in three different feed brands of aquafeeds from BC (Moore–Clarke, EWOS and Taplow) reported by Easton et al. (2002) were similar to concentrations observed in Moore–Clarke, OMP and EWOS feeds in the present study (Table 2). ∑TEQs we observed in Moore–Clarke (0.78±0.52 pg/g wet wt.), OMP (0.68±0.16 pg/g wet wt.) and EWOS (2.6±0.35 pg/g wet wt.) aquafeeds from BC hatcheries are similar to ∑TEQs (0.237±0.647 pg/g wet wt.) found in feeds from federal salmonid hatcheries in the United States (Maule et al., 2007). Conversely, concentrations of toxaphene in the three feed samples from the present study generally exceeded previously reported toxaphene levels. This may, in part, be due to the fact that our toxaphene analysis was full congener, whereas previously reported concentrations are based on 19 congeners. Relatively high toxaphene levels (375–590 ng/g lipid) have been reported in salmon feed from Scotland (Hites et al., 2004). Regardless, persistent organochlorine pesticides are present at appreciable levels (10 to 200 ng/g lipid) in commercial aquafeeds from British Columbia.

The data in Table 4 also show that POP concentrations can vary considerably between feeds of different brands and source locations. For example, Easton et al. (2002) found large differences in PCB conentrations between Taplow (70.2 ng/g lipid) and Moore–Clarke aquafeeds (322 ng/g lipid) from British Columbia. Hites et al. (2004) measured POPs in 13 fish feed samples obtained from Scotland, BC, Chile and Eastern Canada and found relatively higher levels in feeds from Scotland. Relatively high PCB concentrations in salmon feed from Scotland, reported by Jacobs et al. (2002), were as high as 3.8 μg/g lipid, an order of magnitude higher than all other reported PCB concentrations in aquafeeds.

The similar PCB and PCDD/F congener profiles observed between salmon smolts and aquafeed from BC hatcheries (Figs. 1 and 2), suggest contaminant burdens in these hatchery smolts are largely the

Table 4
Comparison of mean contaminant levels reported in commercial aquafeeds from North/South America and Europe

Country of origin	North/South America							Europe		
	Canada	Canada	Canada	USA	Canada	Canada	Chile	Norway	Scotland	Scotland
Feed brand	Moore–Clarke	EWOS	Taplow	Bio-Orgeon	Unknown brand	Unknown brand	Unknown brand	Unknown brand	Unknown brand	Unknown brand
PCBs (ng/g lipid)	322[a]	239[a]	70.2[a]	363[a]	15–220[d]	95[d]	30–170	–	382–3831[f]	160–300
Chlordanes (ng/g lipid)	24.6[b]	35.7[b]	15.0[b]	26.6[b]	–	–	–	22–46[e]	–	–
Toxaphene (ng/g lipid)	–	15.3[c]	183[c]	–	15–190[c]	–	3–220[c]	–	–	375–590[c]
Dieldrin (ng/g lipid)	33.6	5.36	3.27	21.1	3–30	–	3–20	–	–	20–40
Reference	Easton et al. (2002)	Easton et al. (2002)	Easton et al. (2002)	Easton et al. (2002)	Hites et al. (2004)[g]	Ikonomou et al. (2007)	Hites et al. (2004)[g]	Karl et al. (2002)	Jacobs et al. (2002)	Hites et al. (2004)[g]

[a] Sum of 112 PCB congeners.
[b] Sum of *cis*- and *trans*-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, heptachlor and heptachlor epoxide.
[c] 19 individual persistent chloroborane peaks.
[d] Full congener PCBs analysis.
[e] Sum of *cis*- and *trans*-chlordane, oxychlordane, and *trans*-nonachlor.
[f] Sum of 59 PCB congeners.
[g] Wet weight concentration data from bar graphs in Hites et al. (2004) were lipid corrected using mean lipid contents reported in Hamilton et al. (2005).

result of dietary exposure via consumed feeds. The overall PCB congener pattern found in feed and smolts was determined to be Aroclor in nature. Using a Direct Mixing Model (Sather et al., 2001) the source composition predicted for the feeds and smolts was 10% A1242: %35 A1254: 55% A1260), (Fig. S2, Supporting Information). These three Aroclors were sold and used extensively in industrial applications as dielectric fluids (e.g. capacitors, transformers), industrial fluids (e.g. hydraulic systems, gas turbines, vacuum pumps), fire retardants, heat transfer applications, and plasticizers (e.g. adhesives, textiles, surface coatings, sealants, printing, copy paper), (Hutzinger et al., 1979). Similarly, the PCDD/F profile in feed and smolts were comparable to the worldwide deposition of those compounds, with OCDD being the dominant congener found in the environment (Fig. S3, Supporting Information).

The role of lipids in the accumulation of hydrophobic organic contaminants such as PCBs, PCDD/Fs and DDTs in fish is well understood (Addison, 1982; Fisk et al., 2001; Kelly et al., 2007). Because hydrophobic chemicals are typically stored in fatty tissues, aquafeeds containing components (e.g. small fish, fish parts from processing plants and/or fish oil) with higher lipid contents are expected to exhibit the highest POP concentrations. The fact that the higher lipid content feed (Moore–Clarke) exhibited the lowest POP concentrations is likely due to contaminant level variation associated with geographic source locations and/or species composition differences between the feeds.

Previous studies have observed significant curvilinear relationships between BMFs and chemical K_{OW} (Gobas et al., 1993a; Fisk et al., 1998). In particular, less hydrophobic chemicals (log K_{OW}<5) tend to exhibit low BMFs due to efficient respiratory elimination of those relatively more water soluble compounds to water through gill ventilation, while high K_{OW} chemicals (i.e., log K_{OW}>7) tend to exhibit low BMFs due to reduced dietary absorption efficiency of those very hydrophobic compounds. Thus, chemical K_{OW} can be a useful predictor for determining contaminant concentrations in fish from known concentrations in consumed feed. However, no distinct relationship between BMFs and K_{OW} is apparent in these hatchery smolts (Fig. 6).

Observed BMFs of PCBs and PCDD/Fs in salmon smolts from the present study are relatively low compared to those BMFs reported in wild mature fish, which typically range between 5 and 10 (Gobas, 1993; Kelly et al., 2004). For example, the BMF of PCB-153 (the most recalcitrant congener (typically exhibiting the highest biomagnification potential) was often less than 1.0 in these hatchery smolts, indicating no biomagnification. The low degree of PCB and PCDD/F biomagnification in these smolts is likely the result of growth during this period, which can greatly diminish the biomagnification potential of organic chemicals (Gobas et al., 1999). The growth dilution effect is particularly important in hatchery fish consuming starter feeds, as those fish can grow several times in weight from ~1 g for fry to ~5 g for smolts over a short period of time. The high rate of tissue deposition during this growth period is likely more rapid than the rate chemical uptake, thereby counteracting the biomagnification effect.

In terms of toxicological significance, ∑TEQs in salmon smolts from all four hatcheries (0.21±0.19 pg/g wet wt.) were far less than the LD_{50} of 333 pg/g reported for rainbow trout egg mortality (Walker and Peterson, 1991). ∑TEQs in smolts were also generally below the 30% mortality threshold level of 0.3 pg/g previously reported in salmonid eggs (Giesy et al., 2002). Additionally, ∑PCB levels (6.9–45.9 ng/g wet wt.) were only a fraction of the relative effects threshold (RET) level of 2,400 ng/g wet wt., which is based on various end-points including thyroid activity, vitellogenin and decreased growth rates in salmonids from fry to adults (Meador et al., 2002).

Although Robertson Creek smolts exhibited the highest TEQs of all the smolts, EROD activity (3.88±2.46 pmol//mg/min) and CYP1A levels (4.2±1.6 ROD/mg) in those smolts was relatively low. Previous studies have reported relatively high hepatic CYP1A levels in hatchery-reared chinook (1+ years) of approximately 10 ROD/mg (Wilson et al., 2001). Also, Wilson et al. (2000) reported EROD the range found for several juvenile chinook (~18 months old) sampled from various rivers in BC was 20–45 ROD/mg. Furthermore, EROD and CYP1A levels are well below levels observed in laboratory exposure studies. For example, Servizi et al. (1993) reported EROD activity between 40 and 60 pmol/mg/min in chinook fingerlings following exposure to bleached kraft mill effluent, which is over 10 fold higher than EROD activity we observed in chinook smolts from BC hatcheries. Thus, the data indicate negligible effects associated with aryl hydrocarbon receptor (AhR) agonists in salmon smolts from these BC hatcheries.

5. Conclusions

Concentrations of several POPs, including PCBs, dioxins and furans, DDTs, toxaphenes, chlordanes, chlorobenzenes, hexachlorocyclohexanes, dieldrin, endrin and pentachloroanisole were measured (at ppt to ppb levels) in commercial aquafeeds used at four fish hatcheries in coastal British Columbia. Levels and patterns of POPs, in some cases, varied between different feed brands studied (i.e., Moore–Clarke, EWOS and OMP). Because young hatchery salmon smolts are sequentially introduced to increasingly lipid-rich diets during the rearing process (e.g., starting diets with ~10% lipid to final diets of

over 20% lipid), dietary exposure of hydrophobic organic contaminants like PCBs, PCDD/Fs and organochlorine pesticides generally increased during this period. However, tissue residue contaminant concentrations in smolts remained relatively low due to a low degree of chemical biomagnification (BMFs between 0.2 and 3), which is likely the result of growth dilution during this period. 2,3,7,8 TCCD toxic equivalent (ΣTEQ) concentrations in smolts were below levels associated with dioxin-like toxic effects. Biochemical response data (EROD activity and CYPIA levels) further indicated effects of aryl hydrocarbon receptor (AhR) agonists in these smolts were negligible. Aquafeeds investigated in the present study are similar in composition to those utilised in salmon farm operations, hence these data may be useful for fish farm operations using fish meal and fish oil based feeds. Our future work will focus on assessing the occurrence of various environmental contaminants of concern such as polybrominated diphenyl ethers (PBDEs), current-use pesticides (CUPs), pharmaceuticals and personal care products (PPCPs) and mercury in various commercial aquafeed formulations as well the toxicological implications associated with chemical bioaccumulation in Pacific salmon smolts from BC fish hatcheries.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.aquaculture.2008.08.035.

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[Article]

Contaminant Concentrations in Juvenile Fall Chinook Salmon from Columbia River Hatcheries

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Abstract.—Previous studies have reported elevated chemical contaminant concentrations in out-migrant juvenile salmon from the lower Columbia River. Hatchery rearing is a potential exposure pathway, as contaminants have been measured in hatchery fish and feed from other regions. In this study, we analyzed for polychlorinated biphenyls (PCBs), organochlorine pesticides including dichloro-diphenyl-trichloroethanes (DDTs), polycyclic aromatic hydrocarbons (PAHs), and polybrominated diphenyl ethers (PBDEs) in juvenile fall Chinook salmon *Oncorhynchus tshawytscha* and feed from eight hatcheries (Big Creek Hatchery, Oregon Department of Fish and Wildlife; Elochoman, Cowlitz, Washougal, Klickitat, and Priest Rapids hatcheries, Washington Department of Fish and Wildlife; Little White Salmon National Fish Hatchery; and Spring Creek National Fish Hatchery) that release fish into the lower Columbia River. In feed samples, the mean concentrations of summed PCBs, summed DDTs, and summed PAHs were 14, 27, and 370 ng/g wet weight, respectively. In Chinook salmon bodies, mean concentrations of summed PCBs, summed DDTs, and summed PAHs were 17, 9.0, and 30 ng/g wet weight, respectively; metabolites of PAHs were also detected in Chinook salmon bile. Other organochlorine pesticides were detected at low levels (<5 ng/g wet weight) in feed and Chinook salmon from all hatcheries. Concentrations of PBDEs in feed and fish from all hatcheries were low (<3 and <1 ng/g wet weight, respectively). Contaminant exposure levels in hatchery Chinook salmon were generally below those associated with adverse effects on salmon health and also lower than those in field-collected juvenile fall Chinook salmon of hatchery origin from the lower Columbia River, suggesting that the river is a more important source of contamination than are the hatcheries.

Recent studies show that threatened and endangered juvenile fall Chinook salmon *Oncorhynchus tshawytscha* (Myers et al. 1998) that rear and feed in the lower Columbia River are accumulating persistent organic pollutants (POPs) at concentrations that could potentially reduce their survival (Johnson et al. 2007a, 2007b; LCREP 2007). Thus, identifying contaminant sources and reducing exposure are priorities for the recovery of these stocks (LCREP 2007; USEPA 2009). The listed stocks include fish of hatchery origin, which cannot always be reliably distinguished from wild fish through genetic analyses or hatchery marking (Myers et al. 1998, 2006). This raises the possibility that contaminants absorbed during hatchery rearing may contribute to body burdens in juvenile fall Chinook salmon from the lower Columbia River. Concern about this issue has been heightened by reports of chemical contamination in farmed and hatchery salmon throughout Europe and North America (Easton et al. 2002; Parkins 2003; Hites et al. 2004a, 2004b), including salmon from some Pacific Northwest hatcheries (Johnson et al. 2007a). If such contaminants are present in hatchery-reared salmon released into the lower Columbia River, these fish could be contributing to the average contaminant body burdens for the juvenile salmon populations in the area, reducing the viability of hatchery stocks and acting as a source of contaminants for fish-eating predators.

In the present study, we measured concentrations of several classes of POPs in subyearling fall Chinook salmon from eight Columbia River hatcheries (Figure 1) before their release into the river in late May or early June. These hatcheries are thought to be major contributors to Chinook salmon populations in the lower Columbia River based on release data (CBR 2008) and genetic analyses of previously collected juvenile fall Chinook salmon collected from lower Columbia River sites (Johnson et al. 2007b). Altogether, the sampled hatcheries accounted for 74% of subyearling fall Chinook salmon released into the lower Columbia River in 2005, the year in which the present study was conducted (CBR 2008), and for about 79% of subyearling fall Chinook salmon released into the Columbia River in 2008 (CBR 2008).

The contaminants measured in Chinook salmon and hatchery feed included (1) polychlorinated biphenyls (PCBs), industrial chemicals that were banned in the USA in the 1970s but that are still common in urban

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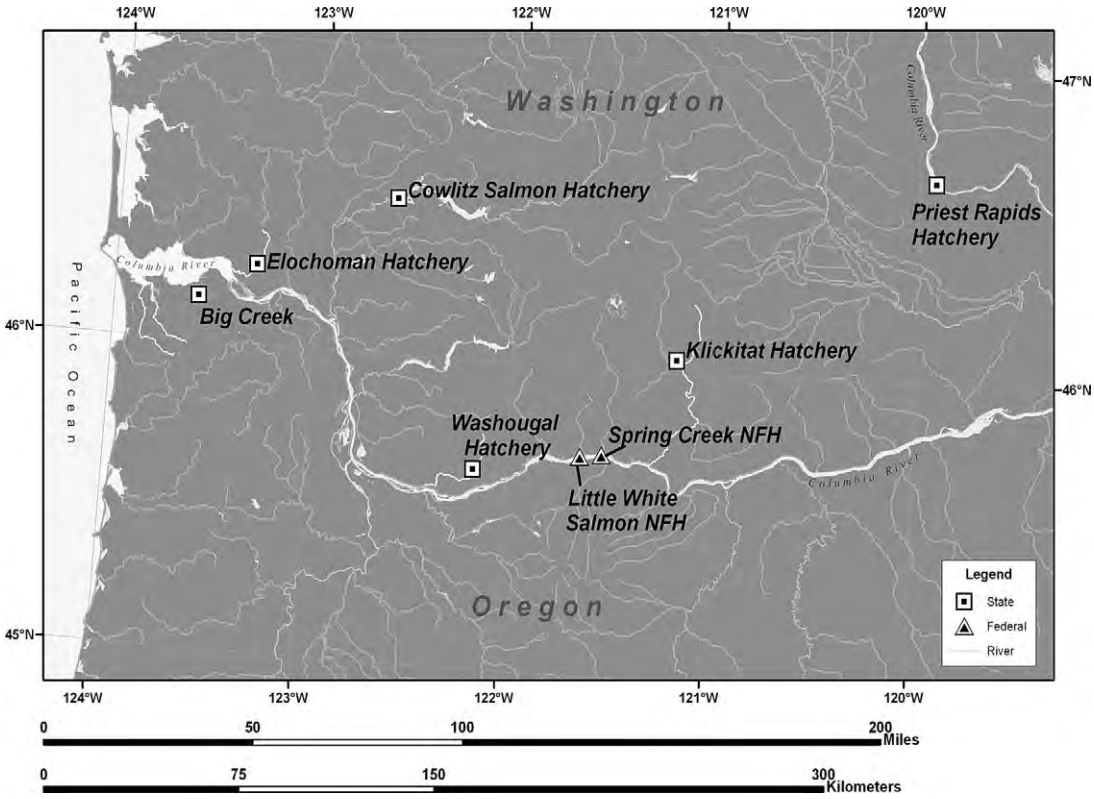

FIGURE 1.—Locations of Columbia River hatcheries in (Washington and Oregon) that were sampled as part of this study (NFH = National Fish Hatchery).

waterways (ATSDR 2000); (2) polybrominated diphenyl ethers (PBDEs), which are extensively used as flame retardants (ATSDR 2004); (3) dichloro-diphenyl-trichloroethanes (DDTs), insecticides that were banned in the United States in 1972 but that are still present in many agricultural areas (ATSDR 2002); and (4) polycyclic aromatic hydrocarbons (PAHs), which are derived from crude oils and various petroleum products (ATSDR 1995). These chemicals are associated with reproductive and developmental defects, immunosuppression, cancer, poor growth, and metabolic disorders in juvenile salmon and other fish (e.g., Incardona et al. 2005; Meador et al. 2006, 2008; Johnson et al. 2008). We also determined lipid content in Chinook salmon bodies because of its effect on contaminant uptake and toxicity (Elskus et al. 2005). The tissue concentration of a toxic lipophilic chemical is directly related to the amount of lipid in an organism, and when lipid content is high a higher proportion of the compound is associated with the lipid and is unavailable to cause toxicity (Lassiter and Hallam 1990; van Wezel et al. 1995).

Our objectives were to (1) determine whether POPs were present in Columbia River hatchery Chinook salmon and commercial feed at concentrations that could affect fish health or pose a threat to piscivorous wildlife and (2) evaluate the potential contribution of hatchery feed to contaminant body burdens of outmigrant fall Chinook salmon by comparing contaminant levels in hatchery fish with previously determined contaminant concentrations in juvenile fall Chinook salmon of hatchery origin from several sites the lower Columbia River (LCREP 2007).

Methods

Fish Collection

In May 2005, bodies of juvenile subyearling Chinook salmon were obtained from eight hatcheries along the Columbia River (Big Creek Hatchery, Elochoman Hatchery, Cowlitz Hatchery, Washougal Hatchery, Little White Salmon National Fish Hatchery [NFH], Spring Creek NFH, Klickitat Hatchery, and Priest Rapids Hatchery; Figure 1). Big Creek Hatchery is operated by the Oregon Department of Fish and Wildlife; Elochoman, Cowlitz, Washougal, Klickitat, and Priest Rapids hatcheries are operated by the

Washington Department of Fish and Wildlife; and Little White Salmon NFH and Spring Creek NFH are operated by the U.S. Fish and Wildlife Service. All collections were performed shortly before the release dates for juvenile subyearling Chinook salmon so that contaminant concentrations would be typical of those in juvenile fish when they entered the lower Columbia River.

In addition to fish, two feed samples of approximately 10 g each were obtained from hatchery personnel at the time the fish were sampled. The feed samples were taken from the type and lot of feed that the fish were currently consuming. The feeds came from various commercial suppliers based in the Pacific Northwest. The feed samples were placed in 118-mL (4-oz) glass jars rinsed with isopropyl alcohol and were stored in a cooler with dry ice for transport back to the Northwest Fisheries Science Center (NWFSC) laboratory in Seattle. At the laboratory, feed samples were stored at −20°C until chemical analyses were performed.

Approximately 30–40 individual fish at each hatchery were collected for necropsy. Fish were measured (to the nearest 1 mm), weighed (to the nearest 0.1 g), and then euthanized by a blow to the head. For each fish, bile was collected (when present) and the individual bile samples were composited into 4-mL glass vials containing glass 250-μL inserts. Bile samples from 10 to 20 individual fish per hatchery were pooled to obtain 3–5 μL of bile for measurement of PAH metabolites. The stomach and gastrointestinal tract were then removed, and stomach contents were extracted. These internal organs were then placed back into the visceral cavity of the carcass so that the tissue analyzed would consist of the whole body minus stomach contents. The carcasses containing the internal organs were individually wrapped in foil and labeled. Both body and bile samples were placed in a cooler with dry ice for transport back to the NWFSC laboratory in Seattle. At the laboratory, bile and body samples were stored at −80°C until chemical analyses were performed.

Sample Analyses

Lipid determination.—For lipid and chemical analyses, individual Chinook salmon bodies (carcass plus internal organs) from each hatchery were combined to produce composite samples consisting of 10 fish each. The amount of total, nonvolatile, extractable lipid (reported as percent lipid) in the body composites and feed samples was determined by gravimetric analysis as described in Sloan et al. (2004). Lipid classes were determined using thin-layer chromatography–flame ionization detection (TLC–FID) with Iatroscan analysis as described by Ylitalo et al. (2005). The TLC–FID analysis also provided an estimate of percent lipid content, which was compared with the values determined gravimetrically. Lipid measurements obtained by gravimetric analysis were used to normalize body contaminant concentrations for lipid content. With the exception of one feed sample from Big Creek Hatchery, lipids in all samples were measured by both the gravimetric and TLC–FID methods; in the feed sample from Big Creek Hatchery, lipid content was measured by TLC–FID only.

Chemical contaminants in feed and body samples.—Body composite and feed samples were analyzed by gas chromatography–mass spectrometry (GC–MS) for PCB congeners, DDTs, DDT isomers, and other organochlorine (OC) pesticides (hexachlorocyclohexanes [HCHs], hexachlorobenzene [HCB], chlordanes, aldrin, dieldrin, mirex, and endosulfans) as described by Sloan et al. (2005). The PBDEs were measured similarly and concurrently in the GC–MS analyses. A total of 47 individual PCB congeners were measured (International Union of Pure and Applied Chemistry [IUPAC] numbers 17, 18, 28, 31, 33, 44, 49, 52, 66, 70, 74, 82, 87, 95, 99, 101/90, 105, 110, 118, 128, 138/163/164, 149, 151, 153/132, 156, 158, 170/190, 171, 177, 180, 183, 187/159/182, 191, 194, 195, 199, 205, 206, 208, and 209). The HCHs measured included α-HCH, β-HCH, and γ-HCH (lindane). Dichloro-diphenyl-trichloroethanes measured included *p*,*p*′-DDT, *p*,*p*′-DDE, *p*,*p*′-DDD, *o*,*p*′-DDD, *o*,*p*′-DDE, and *o*,*p*′-DDT. Chlordanes and related compounds measured included heptachlor, heptachlor epoxide, γ-chlordane, α-chlordane, oxychlordane, *cis*-nonachlor, *trans*-nonachlor, and nonachlor III. A total of 10 individual PBDE congeners were measured (IUPAC numbers 28, 47, 49, 66, 85, 99, 100, 153, 154, and 183). In body samples, the limits of quantitation (LOQs) ranged from less than 0.059 ng/g wet weight to less than 0.34 ng/g wet weight for individual PCB congeners; from less than 0.23 ng/g wet weight to less than 0.35 ng/g wet weight for DDTs, aldrin, dieldrin, chlordanes, mirex, and HCHs; from less than 0.64 ng/g wet weight to less than 0.93 ng/g wet weight for endosulfan I; and from less than 0.28 ng/g wet weight to less than 0.34 ng/g wet weight for HCB.

Summed (Σ) PCBs (ΣPCBs) were calculated by adding the concentrations of 17 commonly detected chlorobiphenyl congeners (IUPAC numbers 18, 28, 44, 52, 95, 101, 105, 118, 128, 138, 153, 170, 180, 187, 195, 206, and 209) and multiplying the result by two. This formula provides a good estimate of the total PCBs in a typical environmental sample of sediments or animals feeding on lower trophic levels (Lauenstein et al. 1993). The ΣDDTs were calculated by summing

the concentrations of *p*,*p*′-DDT, *p*,*p*′-DDE, *p*,*p*′-DDD, *o*,*p*′-DDD, *o*,*p*′-DDE, and *o*,*p*′-DDT. Summed chlordanes were determined by adding the concentrations of heptachlor, heptachlor epoxide, γ-chlordane, α-chlordane, oxychlordane, *cis*-nonachlor, *trans*-nonachlor, and nonachlor III. The ΣHCHs were calculated by adding the concentrations of α-HCH, β-HCH, and lindane (γ-HCH). The ΣPBDEs were calculated by adding the concentrations of the 10 PBDE congeners measured.

In addition to PBDEs and OC contaminants, feed and body samples were analyzed for low (2–3-ring) and high (4–6-ring) molecular weight PAHs using capillary column GC–MS (Sloan et al. 2005). Summed low-molecular-weight aromatic hydrocarbons (ΣLAHs) were determined by adding the concentrations of biphenyl, naphthalene, 1-methylnaphthalene, 2-methylnaphthalene, 2,6-dimethylnapthalene, acenaphthylene, acenaphthene, 2,3,5-trimethylnaphthalene, fluorene, dibenzothiophene, phenanthrene (PHN), 1-methylphenanthrene, and anthracene. Summed high-molecular-weight aromatic hydrocarbons (ΣHAHs) were calculated by adding the concentrations of fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[a]pyrene (BaP), benzo[e]pyrene, perylene, dibenz[a,h]anthracene, benzo[b]fluoranthene, benzo[k]fluoranthene, indenopyrene, and benzo[g,h,i]-perylene. The ΣPAHs were calculated by adding ΣHAHs and ΣLAHs. The LOQs for individual PAHs ranged from less than 0.13 ng/g wet weight to less than 0.47 ng/g wet weight in Chinook salmon body samples and from less than 0.087 to 0.37 ng/g wet weight in food samples.

To monitor the accuracy of the GC–MS method, a National Institute of Standards and Technology (NIST) Standard Reference Material (SRM) blue mussel *Mytilus edulis* homogenate (NIST SRM 1974b) and a fish tissue homogenate (NIST SRM 1947) were analyzed with each sample set and the results met laboratory criteria (Sloan et al. 2006). One (12.5%) out of eight feed samples was analyzed in duplicate to measure the precision of the method, and the laboratory quality assurance (QA) criteria were met for all analytes measured in the feed samples. Method blanks also met laboratory criteria. The QA procedures and criteria are described in detail by Sloan et al. (2006). The percent recoveries of the surrogate standards ranged from 75% to 106%.

To adjust for the influence of lipid on toxicity, we normalized body contaminant concentrations for lipid and relied primarily on lipid-normalized data to evaluate potential health effects of toxicants on juvenile salmon. Wet-weight data are also presented to facilitate comparison with other studies and to evaluate risks to predators that consume salmon with accumulated toxicants.

Polycyclic aromatic hydrocarbon metabolites in Chinook salmon bile.—Due to the relatively small volume of bile that can be collected from individual subyearling Chinook salmon, bile samples were composited from 30 individual fish per hatchery to provide an adequate sample volume (>25 μL) for high-performance liquid chromatography (HPLC)–fluorescence analysis. No PAH metabolite data were acquired for fish from Little White Salmon NFH because the volume of the bile composite was too small (<25 μL). Bile samples were analyzed for metabolites of PAHs using a HPLC–fluorescence detection method described by Krahn et al. (1984). Briefly, bile was injected directly onto a C-18 reverse-phase column (Phenomenex Synergi Hydro) and eluted with a linear gradient from 100% water (containing a trace amount of acetic acid) to 100% methanol at a flow of 1.0 mL/min. Chromatograms were recorded at the following wavelength pairs: (1) 260–380 nm, where several 3–4-ring compounds (e.g., PHN) fluoresce; and (2) 380–430 nm, where 4–5-ring compounds (e.g., BaP) fluoresce. Peaks eluting after 5 min were integrated, and the areas of these peaks were summed. The concentrations of fluorescent PAHs in the bile samples of juvenile fall Chinook salmon were determined using PHN and BaP as external standards and converting the fluorescence response of bile to PHN (ng PHN equivalents/g bile) and BaP (ng BaP equivalents/g bile) equivalents.

To ensure that the HPLC–fluorescence system was operating properly, a PHN–BaP calibration standard was analyzed at least five times, and a relative SD of less than 10% was obtained for each PAH. As part of our laboratory QA plan, two QA samples (a method blank and a fish bile control sample [bile of Atlantic salmon *Salmo salar* exposed to Monterey crude oil at 25 μg/mL for 48 h]) were analyzed with the fish bile samples (Sloan et al. 2006).

Biliary protein was measured according to the method described by Lowry et al. (1951). Biliary fluorescence values were normalized to protein content, which is an indication of feeding state and water content of the bile. Fish that have not eaten for several days exhibit higher biliary fluorescent aromatic compound values and higher protein content than fish that are feeding constantly and excreting bile more frequently (Collier and Varanasi 1991).

Fish condition factor.—To provide a measure of fish weight adjusted for size, Fulton's condition factor (Ricker 1975) was calculated for hatchery fish as $[(\text{gutted body weight, g})/(\text{fork length, mm})^3] \times 100$.

Estimation of hatchery contribution to in-river fall Chinook salmon body burdens.—To estimate the relative proportions of contaminants absorbed during hatchery rearing as compared with the proportions absorbed while fish were in the lower Columbia River, concentrations of PCBs, DDTs, and PBDEs in the hatchery fall Chinook salmon were compared with concentrations of these contaminants in composite body samples (carcasses and internal organs minus stomach contents) of juvenile fall Chinook salmon of hatchery origin collected in the lower Columbia River in 2005. This collection was part of a larger cooperative project conducted in collaboration with the U.S. Geological Survey, the lower Columbia River Estuary Partnership, and the Bonneville Power Administration (LCREP 2007). The subset of samples used for these calculations consisted only of marked (i.e., adipose-fin clipped) hatchery fall Chinook salmon from lower Columbia River stocks, so their contaminant body burdens are representative of fish that are released from lower Columbia River hatcheries and subsequently spend some time rearing and migrating in the river. However, the specific hatcheries from which the fish were released are unknown. In-river fish samples came from three sites: Warrendale, the Confluence, and Columbia City. The Warrendale site (45°36′45″N, 122°01′35″W) is located in a rural forested area at river kilometer (rkm) 227, near the town of Warrendale, Oregon, just downstream from Bonneville Dam. The Confluence site (45°38′27″N, 122°43′08″W) is located at rkm 163, just downstream from the major urban centers of Portland, Oregon, and Vancouver, Washington. The Columbia City site (46°09′96″N, 122°94′51″W) is located at rkm 134 in a rural area where forestry and agriculture are the primary land uses. However, this site is affected by municipal wastewater discharges and discharges from local industries. Additional information about the sites and the sampling and analysis procedures is given by the Lower Columbia River Estuary Partnership (LCREP 2007).

Estimated PCB, DDT, and PBDE contributions from the hatchery to fish body burdens for each site were calculated as described by Meador et al. (2002). For each contaminant, the first step in this analysis was to determine the total amount, in nanograms, for each source (i.e., the hatcheries and each field site). The data from all sampled hatcheries were averaged to calculate mean hatchery fish weight and contaminant concentrations. Totals for each source were calculated as follows:

$$\text{Total(ng)} = (\text{mean fish weight, g}) \times (\text{concentration, ng/g wet weight}).$$

The contributions from river exposure and hatchery exposure (in ng/g wet weight) to the contaminant concentrations in the samples from each source were then calculated:

$$\text{Contribution}_{\text{river}} = \frac{\text{total}_{\text{field site}} - \text{total}_{\text{hatchery}}}{\text{mean fish weight}_{\text{field site}}},$$

$$\text{Contribution}_{\text{hatchery}} = \frac{\text{total}_{\text{hatchery}}}{\text{mean fish weight}_{\text{field site}}}.$$

Finally, the percent hatchery contribution for each sample source was calculated as

$$\text{Hatchery contribution}\,(\%) = \frac{\text{contribution}_{\text{hatchery}}}{\text{concentration}_{\text{source}}} \times 100.$$

Statistical analyses.—Analysis of variance (ANOVA) and Tukey's multiple range tests were used to identify differences in length, weight, and condition factor among fish from different hatcheries and differences in contaminant concentrations among feed samples obtained from different suppliers (Zar 1984; Dowdy and Wearden 1991). Linear regression analysis (Zar 1984) was used to evaluate relationships between contaminant concentrations in hatchery feed, contaminant concentrations in Chinook salmon bodies, and PAH metabolites in bile. Regression analysis was also used to examine the correlation between lipid content values obtained through gravimetric analysis and those obtained through TLC–FID analysis. Before statistical analyses, data were normalized through log-transformation as necessary. In addition, data on contaminant concentrations in feed and fish bodies were used to calculate the percentage of dietary contaminant present in fish bodies.

Results

Fish Size and Condition

Mean length of juvenile fall Chinook salmon (Table 1) differed significantly among the hatcheries, with values ranging from 67 mm at Little White Salmon NFH to 86 mm at Spring Creek NFH (ANOVA: $P < 0.05$). Similarly, fish weight (Table 1) was lowest in Chinook salmon collected at Little White Salmon NFH (mean = 3.2 g), was highest at Spring Creek NFH (mean = 6.2 g), and differed significantly among all hatcheries. Condition factor (Table 1), on the other hand, was lowest in fish from Washougal Hatchery (mean = 0.91) and highest in fish from Big Creek and Little White Salmon hatcheries (mean = 1.04).

Lipid Content and Lipid Classes

Feed samples.—Lipid concentrations in hatchery feed samples ranged from 12% to 22% as determined

TABLE 1.—Mean (±SE) length, weight, and condition factor of juvenile fall Chinook salmon sampled from Columbia River hatcheries (Figure 1) in 2005. Values followed by letters not in common are significantly different (analysis of variance and Tukey's multiple range test: $P < 0.05$).

Hatchery	*n*	Length (mm)	Weight (g)	Condition factor
Big Creek	10	84 ± 0.7 zy	6.1 ± 0.2 z	1.04 ± 0.012 z
Elochoman	10	80 ± 0.7 xw	5.0 ± 01 y	0.99 ± 0.0012 yx
Cowlitz	10	78 ± 0.8 xw	5.0 ± 0.1 yx	0.99 ± 0.0014 zyx
Washougal	10	76 ± 0.7 w	4.1 ± 0.1 xw	0.91 ± 0.0014 v
Little White Salmon	10	67 ± 0.4 v	3.2 ± 0.06 v	1.04 ± 0.014 yx
Spring Creek	10	86 ± 1.1 z	6.2 ± 0.3 z	0.95 ± 0.014 xwv
Klickitat	10	71 ± 0.7 v	3.3 ± 0.1 wv	0.93 ± 0.0014 wv
Priest Rapids	10	81 ± 1.3 yx	5.3 ± 0.3 y	0.98 ± 0.0014 xw

gravimetrically and from 8.8% to 19% as determined by TLC–FID (Table 2). On average, values determined gravimetrically were about 29% higher than values determined with TLC–FID, but the values were significantly and positively correlated ($r^2 = 0.90$, $P = 0.001$, $n = 7$). Lipid content was highest in feed from Klickitat Hatchery and lowest in feed from Priest Rapids Hatchery. Triglycerides were the predominant class of lipids in most of the feed samples, accounting for 68–92% of total lipids in feed from all hatcheries except Cowlitz Hatchery. At Cowlitz Hatchery, triglycerides accounted for only 44% of total lipids, while phospholipids accounted for 53% of total lipids. In feeds from the other hatcheries, phospholipids accounted for 2.8–14% of total lipids. Cholesterol and free fatty acids were also present in feed samples in lower proportions, typically 1.4–3.5% of total lipids for cholesterol and 1.5–8.4% of total lipids for free fatty acids. The feed from the Little White Salmon NFH was unusual compared with feed from the other hatcheries, as it had a free fatty acid content of 23%.

Chinook salmon bodies.—Lipid concentrations in Chinook salmon body composites ranged from 2.6% to 6.2% as determined gravimetrically and from 2.5% to 4.8% as determined with TLC–FID (Table 2). On average, values determined gravimetrically were about 20% higher than those determined with TLC–FID, but the values were significantly and positively correlated ($r^2 = 0.81$, $P = 0.002$, $n = 8$). Free fatty acids and triglycerides were the predominant lipid classes present

TABLE 2.—Mean percent lipid content (determined gravimetrically and by thin-layer chromatography–flame ionization detection [TLC–FID] as described in Ylitalo et al. 2005) and lipid classes of juvenile fall Chinook salmon and feed sampled from Columbia River hatcheries (Figure 1) in 2005. Lipid determinations were made on composite samples of 10 fish/composite (NM = not measured).

Hatchery	Gravimetric lipid content (%)	TLC–FID lipid content (%)	Wax esters–sterol esters (%)	Triglycerides (%)	Free fatty acids (%)	Cholesterol (%)	Phospholipids and other polar lipids (%)
Chinook salmon bodies							
Big Creek	5.1	4.1	0.0	91.5	2.6	3.6	2.4
Elochoman	5.3	4.8	1.1	39.8	38.9	9.0	11.2
Cowlitz	4.7	3.6	1.1	32.6	51.3	7.9	7.2
Washougal	4.1	3.3	0.0	91.2	3.6	4.2	1.0
Little White Salmon	4.3	3.1	1.0	33.4	44.6	7.2	13.8
Spring Creek	2.6	2.5	0.87	35.9	48.1	8.2	6.9
Klickitat	6.2	4.7	0.90	35.6	27.0	9.7	26.8
Priest Rapids	5.1	4.7	0.91	31.5	51.8	7.3	8.5
All hatcheries (±SD)	4.7 ± 1.1 ($n = 8$)	3.9 ± 0.84 ($n = 8$)	0.74 ± 0.46 ($n = 8$)	49 ± 26 ($n = 8$)	33.5 ± 20.4 ($n = 8$)	7.1 ± 2.2 ($n = 8$)	9.7 ± 8.1 ($n = 8$)
Fish food							
Big Creek	NM	10	0	81	6.3	2.8	13.7
Elochoman	18	14	0.25	88	3.2	2.8	5.4
Cowlitz	20	17	0	44	1.5	1.4	52.7
Washougal	20	14	0	92	2.5	2.9	2.8
Little White Salmon	16	12	0	68	23	1.8	6.7
Spring Creek	18	15	0	76	7.4	2.2	14.0
Klickitat	22	19	0	88	2.5	1.7	7.8
Priest Rapids	12	8.8	0	79	8.4	3.5	9.4
All hatcheries (±SD)	18 ± 3.2 ($n = 7$)	14 ± 3.4 ($n = 8$)	0.031 ± 0.090 ($n = 8$)	77 ± 15 ($n = 8$)	6.9 ± 7.1 ($n = 8$)	2.4 ± 0.72 ($n = 8$)	14 ± 16 ($n = 8$)

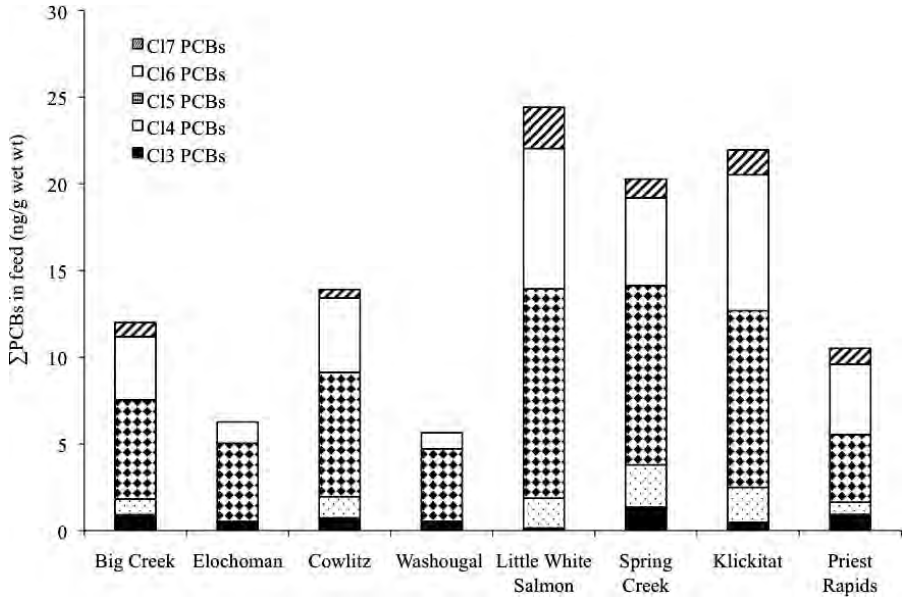

FIGURE 2.—Concentrations (ng/g wet weight) of summed (Σ) PCBs measured in fish food samples obtained from Columbia River hatcheries in 2005 (Cl3, Cl4, Cl5, Cl6, and Cl7 PCBs = PCB homologues with three, four, five, six, and seven chlorines, respectively).

in Chinook salmon bodies. Triglycerides accounted for 32–92% of total lipids in fish from the individual hatcheries, while free fatty acids accounted for 2.6–52% of total lipids. Cholesterol accounted for 3.6–9.7% of total lipids, while phospholipids accounted for 1.0–27% of total lipids. While statistical comparisons could not be made because only one composite sample was made per hatchery, the data on lipid classes given in Table 2 suggest that there might be differences among hatcheries in lipid profiles.

Contaminant Concentrations

Persistent organic pollutants in hatchery feed.—Measurable concentrations of DDTs, PCBs, and PAHs were found in feed from all eight hatcheries. Concentrations of ΣPCBs (Figure 2) ranged from 5.3 ng/g wet weight in feed from Washougal Hatchery to 25 ng/g wet weight in feed from Little White Salmon NFH. The predominant PCBs measured in feed were the pentachlorobiphenyls and hexachlorobiphenyls (e.g., PCBs 118, 138, and 153), with concentrations ranging from 0.23 to 3.3 ng/g wet weight (Figure 2). Of the PCB congeners, 13 congeners (PCBs 156, 158, 171, 177, 191, 194, 195, 199, 205, 206, 208, and 209) were less than the LOQs (<0.054–0.39 ng/g wet weight, depending on the congener and sample size) in feed samples from all hatcheries.

Concentrations of ΣDDTs in feed samples ranged from 9.8 ng/g wet weight in feed from Elochoman Hatchery to 39 ng/g wet weight in feed from Cowlitz Hatchery (Figure 3). In feed, *p*,*p*′-DDE and *p*,*p*′-DDD were the predominant DDTs, accounting for about 80% and 15% of ΣDDTs, respectively. In all feed samples, *p*,*p*′-DDT was also present, accounting for up to 5% of total DDTs. In addition to the *p*,*p*′-substituted isomers, low concentrations of *o*,*p*′-substituted DDTs were measured in most feed samples. In feed from all hatcheries, *o*,*p*′-DDD was measured; *o*,*p*′-DDE was measured in feed from Big Creek, Cowlitz, Klickitat, Little White Salmon, and Priest Rapids hatcheries, while *o*,*p*′-DDT was measured only in feed from Little White Salmon NFH and Klickitat Hatchery.

Concentrations of ΣPAHs ranged from 100 ng/g wet weight in feed from Washougal Hatchery to 610 ng/g wet weight in feed from Big Creek Hatchery (Figure 4). Of the HAHs measured in feed, fluoranthene, pyrene, and chrysene predominated, accounting for 60–80% of ΣHAHs in all samples (Figure 4A). Of the LAHs measured, dimethylnaphthalene, trimethylnaphthalene, and PHN predominated in feed, making up 60–80% of ΣLAHs. Overall, LAHs made up 90% or more of total PAHs in feed samples (Figure 4B).

Feed samples from the hatcheries contained low concentrations of several OC pesticides and PBDEs (Table 3). Chlordanes were measured in feed at all hatcheries except Priest Rapids Hatchery, with concentrations ranging from 0.69 to 4.6 ng/g wet weight. Concentrations of HCHs were less than LOQs in feed

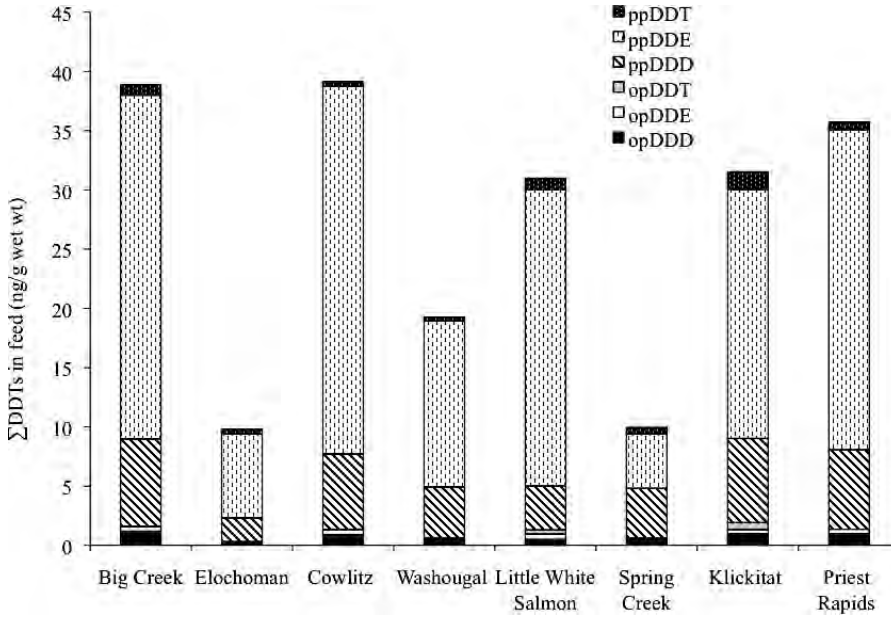

FIGURE 3.—Concentrations (ng/g wet weight) of DDT isomers and summed (Σ) DDTs measured in fish food samples obtained from Columbia River hatcheries in 2005.

from both Spring Creek NFH and Washougal Hatchery, whereas the levels ranged from 0.49 to 3.2 ng/g wet weight in feed from the other hatcheries. Dieldrin and HCB were measured in feed from some of the hatcheries, but the concentrations were less than 2 ng/g wet weight in all cases. Concentrations of mirex, aldrin, and endosulfan I were below LOQs at all hatcheries. In addition, concentrations of ΣPBDEs were low (<3 ng/g wet weight) in all feed samples. While statistical comparisons could not be made because only one feed sample was analyzed per hatchery, the POP concentrations in the samples we analyzed suggest there may be some differences in concentrations of DDTs, PAHs, and PCBs in feeds from the different hatcheries; concentrations of other contaminants were uniformly low.

Concentrations of contaminants were generally similar in feed samples obtained from the three different suppliers used by the hatcheries. Of the eight samples analyzed, four were from one supplier, three were from another supplier, and one was from a third supplier. Mean concentrations of ΣPCBs ranged from 23 to 31 ng/g wet weight, mean concentrations of ΣDDTs ranged from 7.8 to 22 ng/g wet weight, mean concentrations of ΣPBDEs ranged from 4.2 to 8.0 ng/g wet weight, and mean concentrations of ΣPAHs ranged from 150 to 480 ng/g wet weight. Concentrations were not significantly different among the suppliers for any of the contaminants measured (ANOVA: $0.10 < P < 0.80$).

Persistent organic pollutants in Chinook salmon bodies.—The primary contaminants found in Chinook salmon bodies were PCBs and DDTs. Concentrations (wet-weight basis) of ΣPCBs (Figure 5A) ranged from 7.3 ng/g wet weight in fish from Cowlitz Hatchery to 58 ng/g wet weight in fish from Priest Rapids Hatchery. On a lipid-weight basis (Figure 5B), body ΣPCB concentrations in Chinook salmon ranged from 170 ng/g lipid in fish from Washougal Hatchery to 1,200 ng/g lipid in fish from Priest Rapids Hatchery.

The bodies of hatchery Chinook salmon contained a wide range of PCB congeners (Figure 5), with pentachlorobiphenyls (Cl5 congeners) and hexachlorobiphenyls (Cl6) contributing greater than 65% to ΣPCBs. For example, PCBs 101, 110, 118, 138, and 153 were measured in all samples at concentrations ranging from 0.5 to 5.6 ng/g wet weight. Four congeners (PCBs 74, 128, 180, and 158) were found in fish from Priest Rapids only, while two congeners (PCBs 18 and 33) were found only in fish from Spring Creek NFH. The PCBs 156, 170, 171, 177, 183, 191, 194, 195, 199, 205, 206, 208, and 209 were below LOQs (generally <0.2–0.3 ng/g wet weight) in all samples analyzed in the present study.

Mean wet-weight concentrations of ΣDDTs (Figure 6A) ranged from 4.8 ng/g in bodies of Chinook salmon from Elochoman Hatchery to 15 ng/g in bodies of

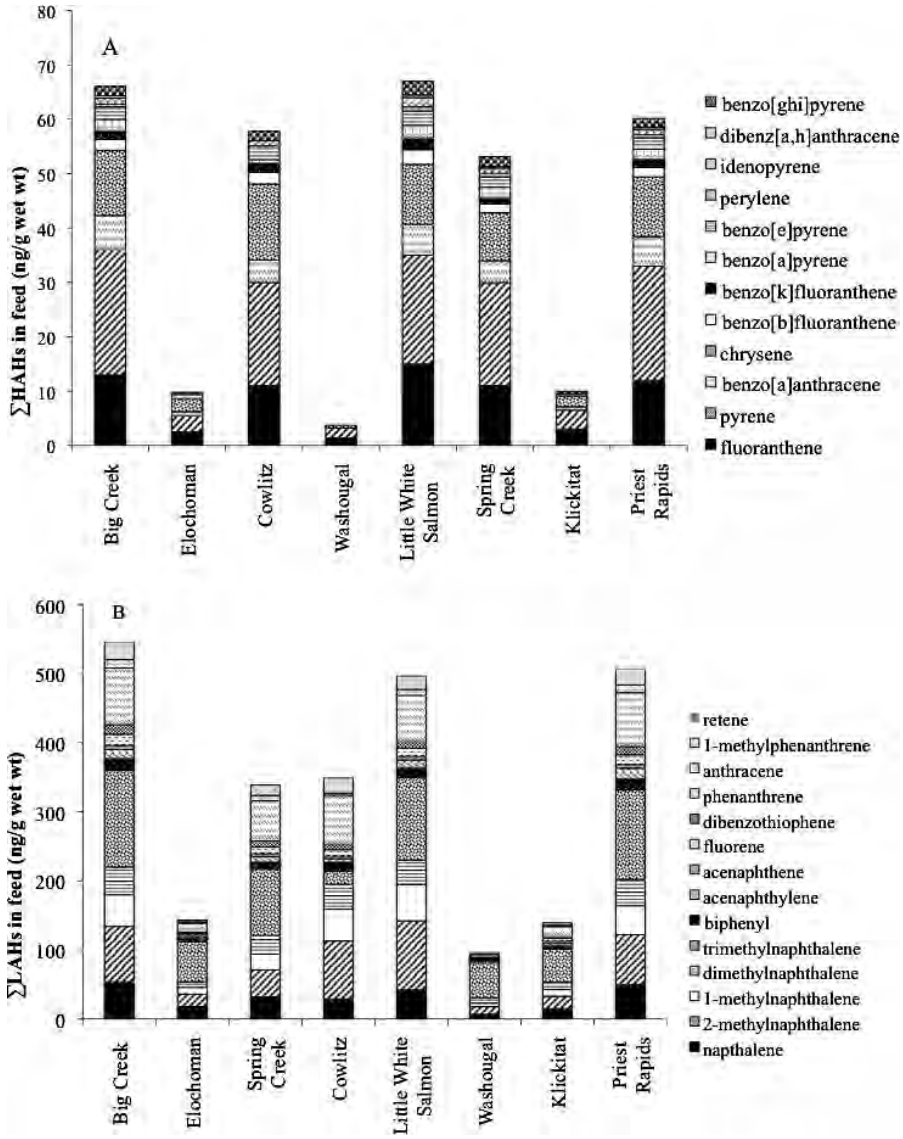

FIGURE 4.—Concentrations (ng/g wet weight) of **(A)** summed high-molecular-weight aromatic hydrocarbons (ΣHAHs) and **(B)** summed low-molecular-weight aromatic hydrocarbons (ΣLAHs) measured in fish food samples obtained from Columbia River hatcheries in 2005.

Chinook salmon from Priest Rapids Hatchery. When compared on a lipid-weight basis (Figure 6B), body ΣDDT concentrations ranged from 90 ng/g lipid in fish from Elochoman Hatchery to 380 ng/g lipid in fish from Big Creek Hatchery. Of the DDTs measured in Chinook salmon bodies, *p*,*p*′-DDE predominated, accounting for 75% or more of ΣDDTs measured. The second most prominent DDT was *p*,*p*′-DDD, which accounted for about 10–20% of DDTs measured. Concentrations of *o*,*p*′-DDD, *o*,*p*′-DDE, *o*,*p*′-DDT, and *p*,*p*′-DDT were generally low (<0.3 ng/g wet weight), accounting for 5% or less of the ΣDDTs in samples where they were measured.

In juvenile Chinook salmon bodies, levels of ΣPAHs ranged from 19 ng/g wet weight in fish from Washougal Hatchery to 42 ng/g wet weight in fish from Priest Rapids Hatchery (Figure 7). Concentrations of both LAHs and HAHs were measured, but HAH levels were below LOQs in most samples (Figure 7). Of the HAHs measured, fluoranthene was the most

common, with concentrations ranging from 0.35 ng/g in fish from Little White Salmon NFH to 0.58 ng/g in fish from Washougal Hatchery (Figure 7); other HAHs present included pyrene, benzo[e]pyrene, idenopyrene, and benzo[g,h,i]perylene. Concentrations of ΣLAHs in Chinook salmon bodies ranged from 16 ng/g wet weight in fish from Washougal Hatchery to 41 ng/g wet weight in fish from Priest Rapids Hatchery (Figure 7). Trimethylnaphthalene, dimethylnaphthalene, and naphthalene were the most common LAHs in bodies of Chinook salmon from most of the hatcheries.

In addition to DDTs, PCBs, and PAHs, OC pesticides and PBDEs were also measured at low concentrations in bodies of juvenile Chinook salmon from some of the hatcheries (Table 3). Chlordanes were measured in fish from all hatcheries except Cowlitz Hatchery; chlordane concentrations ranged from 0.38 to 3.0 ng/g wet weight, with highest levels found in fish from Big Creek Hatchery. Hexachlorobenzene was measured in fish from all hatcheries except Priest Rapids Hatchery and Spring Creek NFH, but concentrations were at or below 0.45 ng/g wet weight in all samples. Dieldrin, HCHs, and PBDEs were measured in fish from a few hatcheries, but concentrations were less than 1 ng/g wet weight in all cases. Mirex, aldrin, and endosulfan I were below LOQs in fish from all hatcheries.

Polycyclic aromatic hydrocarbon metabolites in Chinook salmon bile.—Levels of PAH metabolites in bile of hatchery fish ranged from 590 to 5,500 ng PHN equivalents/mg bile protein for LAH compounds fluorescing at PHN wavelengths and ranged from 24 to 210 BaP equivalents/mg bile protein for HAH compounds fluorescing at BaP wavelengths (Table 4). In fish from most hatcheries, levels of PHN metabolites were below 1,000 ng/mg bile protein and levels of BaP metabolites were below 100 ng/mg bile protein; only fish from Priest Rapids and Cowlitz hatcheries had higher levels.

Contaminant Concentrations in Feed versus Chinook Salmon Bodies and Bile

Although relationships between contaminant concentrations in feed and bodies tended to be positive, there were no statistically significant correlations between concentrations of ΣPCBs, ΣDDTs, or ΣHAHs in feed and concentrations of the compounds in Chinook salmon bodies (ΣPCBs: $r^2 = 0.064$, $P = 0.54$, $n = 8$; ΣDDTs: $r^2 = 0.02$, $P = 0.73$, $n = 8$; ΣHAHs: $r^2 = 0.05$, $P = 0.60$, $n = 8$). However, ΣLAHs in feed and fish bodies were significantly and positively correlated ($r^2 = 0.86$, $P = 0.0009$, $n = 8$). Neither mean fish length ($r^2 = 0.19$, $P = 0.28$, $n = 8$) nor mean body lipid content ($r^2 = 0.07$, $P = 0.50$, $n = 8$) were significantly correlated with body burdens of PCB, DDTs, or PAHs. Concentrations of LAHs in feed showed significant correlations with levels of PAH metabolites in bile fluorescing at PHN wavelengths (an estimator of exposure to LAHs); for LAHs in feed (ng/g wet weight) versus PHN metabolites (ng equivalents/g bile), the r^2 value was 0.91 ($P = 0.0008$, $n = 7$). However, concentrations of HAHs in feed showed no correlation ($r^2 = 0.25$, $P = 0.26$, $n = 7$) with levels of PAH metabolites in bile fluorescing at BaP wavelengths (an estimator of exposure to HAHs).

Generally, contaminant concentrations on a lipid weight basis were similar between feed and Chinook salmon bodies or were lower in feed than in fish bodies (Table 5). Depending on the hatchery, DDT concentrations in juvenile Chinook salmon were 96–380% of DDT concentrations in feed. For the majority of hatcheries, PCB concentrations in bodies were in a similar range (110–480% of the PCB concentrations in feed). However, in fish from Elochoman, Washougal, and Priest Rapids hatcheries, PCB concentrations in fish were much higher relative to PCB concentrations in feed than was typical. In fish from Washougal and Elochoman hatcheries, body PCB concentrations were 780% of those in feed; in fish from Priest Rapids Hatchery, body PCB concentrations were 1,300% of those in feed. Similarly, for chlordanes, juvenile

TABLE 3.—Concentrations (ng/g wet weight) of organochlorine pesticides and polybrominated diphenyl ethers (PBDEs) determined by gas chromatography–mass spectrometry in feed and bodies of juvenile fall Chinook salmon sampled from Columbia River hatcheries in 2005 (<LOQ = less than lower limit of quantitation; ΣHCHs = summed hexachlorocyclohexanes; ΣCHLDs = summed chlordanes; HCB = hexachlorobenzene; ΣPBDEs = summed PBDEs). Body samples are composite samples of 10 fish/composite.

Hatchery	ΣHCHs[a]	Dieldrin	ΣCHLDs[b]	HCB	ΣPBDEs
Fish food (ng/g wet weight)					
Big Creek	3.2	0.20	3.3	0.49	0.95
Elochoman	0.49	0.52	0.82	<LOQ	<LOQ
Cowlitz	2.0	<LOQ	1.9	0.63	1.0
Washougal	<LOQ	<LOQ	0.69	0.26	<LOQ
Little White Salmon	1.6	<LOQ	4.6	1.1	2.9
Spring Creek	<LOQ	2.0	3.8	0.32	2.6
Klickitat	0.52	1.4	2.8	0.9	0.79
Priest Rapids	0.98	<LOQ	<LOQ	0.45	0.95
Chinook salmon bodies (ng/g wet weight)					
Big Creek	0.46	0.19	3.0	0.34	0.78
Elochoman	<LOQ	<LOQ	1.1	0.26	<LOQ
Cowlitz	<LOQ	<LOQ	<LOQ	0.34	<LOQ
Washougal	<LOQ	<LOQ	1.7	0.28	<LOQ
Little White Salmon	<LOQ	<LOQ	1.1	0.31	<LOQ
Spring Creek	<LOQ	0.37	0.38	<LOQ	0.71
Klickitat	<LOQ	0.39	1.1	0.45	<LOQ
Priest Rapids	<LOQ	<LOQ	0.46	<LOQ	<LOQ

[a] Includes α-HCH, β-HCH, and γ-HCH (lindane).

[b] Includes heptachlor, heptachlor epoxide, γ-chlordane, α-chlordane, oxychlordane, *cis*-nonachlor, *trans*-nonachlor, and nonachlor III.

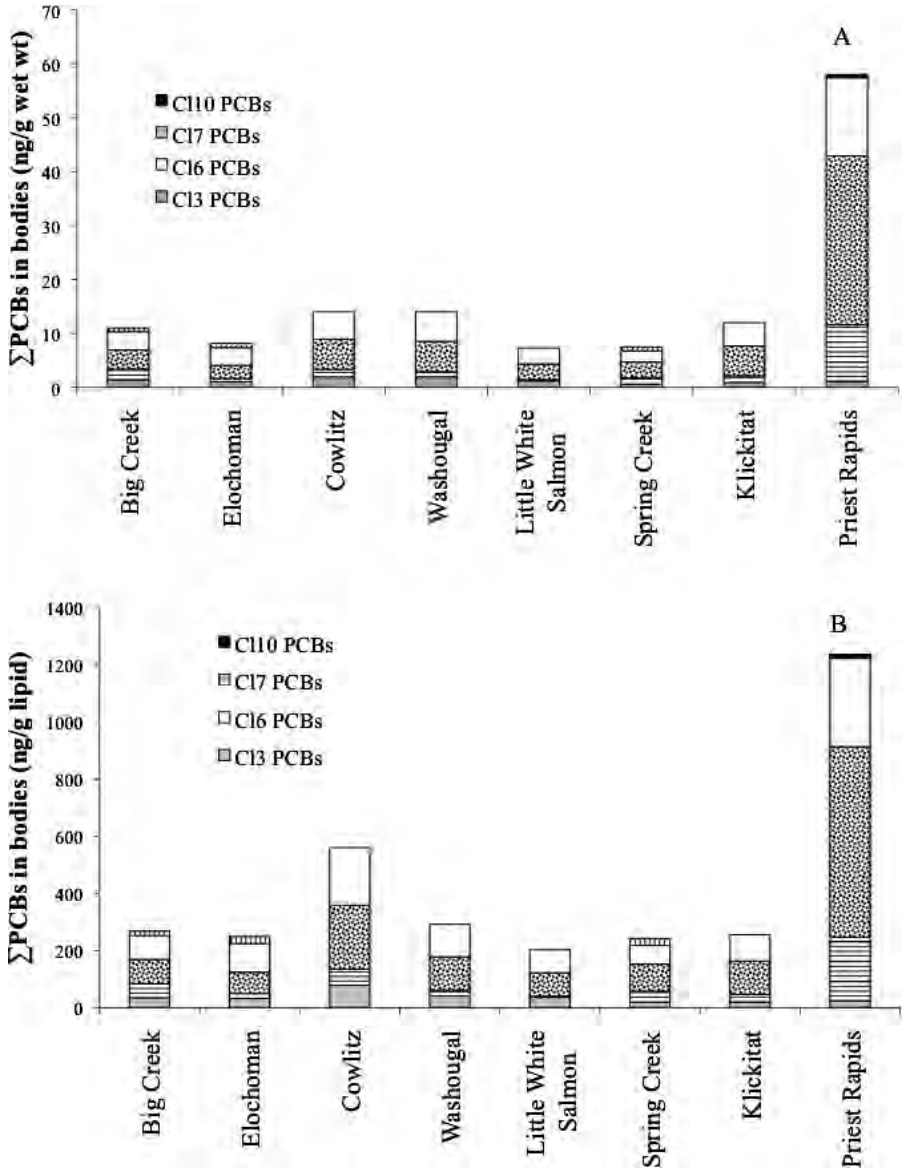

FIGURE 5.—Concentrations of summed (Σ) PCBs in **(A)** nanograms per gram of wet weight and **(B)** nanograms per gram of lipid, measured in body composites (10 fish/composite) of juvenile fall Chinook salmon collected from Columbia River hatcheries in 2005 (Cl3, Cl6, Cl7, and C110 PCBs = PCB homologues with 3, 6, 7, and 10 chlorines).

Chinook salmon typically contained from 44% to 140% of the levels in feed. At Big Creek, Elochoman, and Washougal hatcheries, however, chlordane concentrations in fish bodies were 230–1,200% of those in feed. Concentrations of HCHs, dieldrin–aldrin, HCB, and PBDEs were generally too low in both fish bodies and feed for comparisons to be calculated, but where they were present, concentrations in fish bodies ranged from 36% to 540% of the concentrations in feed, with values at Big Creek and Washougal hatcheries generally among the highest. Concentrations of LAHs and HAHs in bodies were considerably lower than those in feed. In Chinook salmon bodies, HAHs for the most part were barely detected, with maximum levels typically less than 7% of the concentrations found in feed. The exception was for fish from Washougal Hatchery, in which body HAH concentrations were 420% of the HAH concentration in feed. Concentrations of LAHs

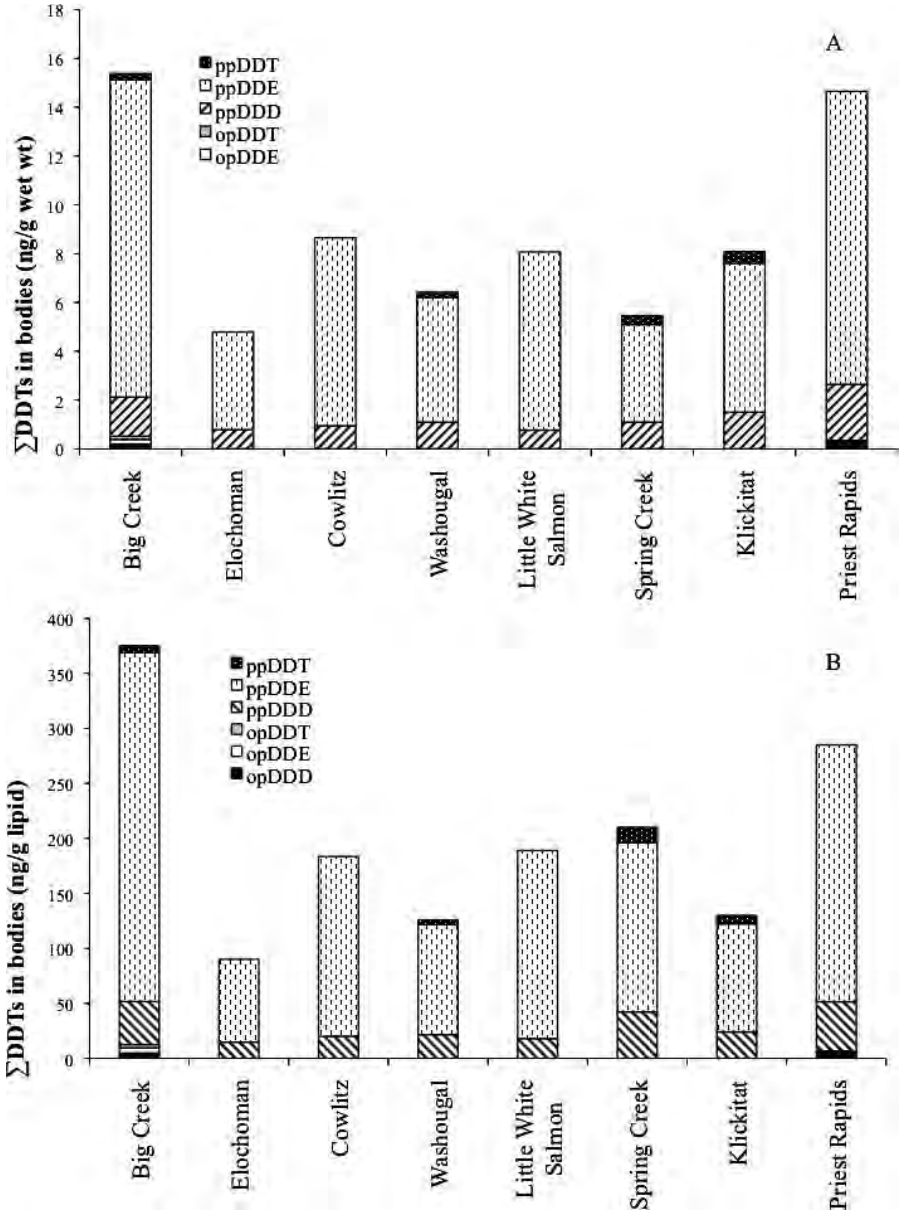

FIGURE 6.—Concentrations of DDT isomers and summed (Σ) DDTs in **(A)** nanograms per gram of wet weight and **(B)** nanograms per gram of lipid, measured in body composites (10 fish/composite) of juvenile fall Chinook salmon collected from Columbia River hatcheries in 2005.

were generally higher in bodies relative to feed than was the case for HAHs; body LAH concentrations ranged from 16% to 84% of concentrations in feed.

Hatcheries as a Source of Contamination in Lower Columbia River Fall Chinook Salmon

In juvenile fall Chinook salmon of hatchery origin from the lower Columbia River, the estimated proportions of PCBs, DDTs, and PBDEs absorbed during hatchery rearing were generally lower than the proportions absorbed in the river, although this varied with the contaminant and the site where the fish were collected (Tables 6–8). For PCBs, it was estimated that Chinook salmon from Warrendale would have accumulated only 4.6 ng PCBs/g of fish after leaving the hatchery, while those collected at the Confluence

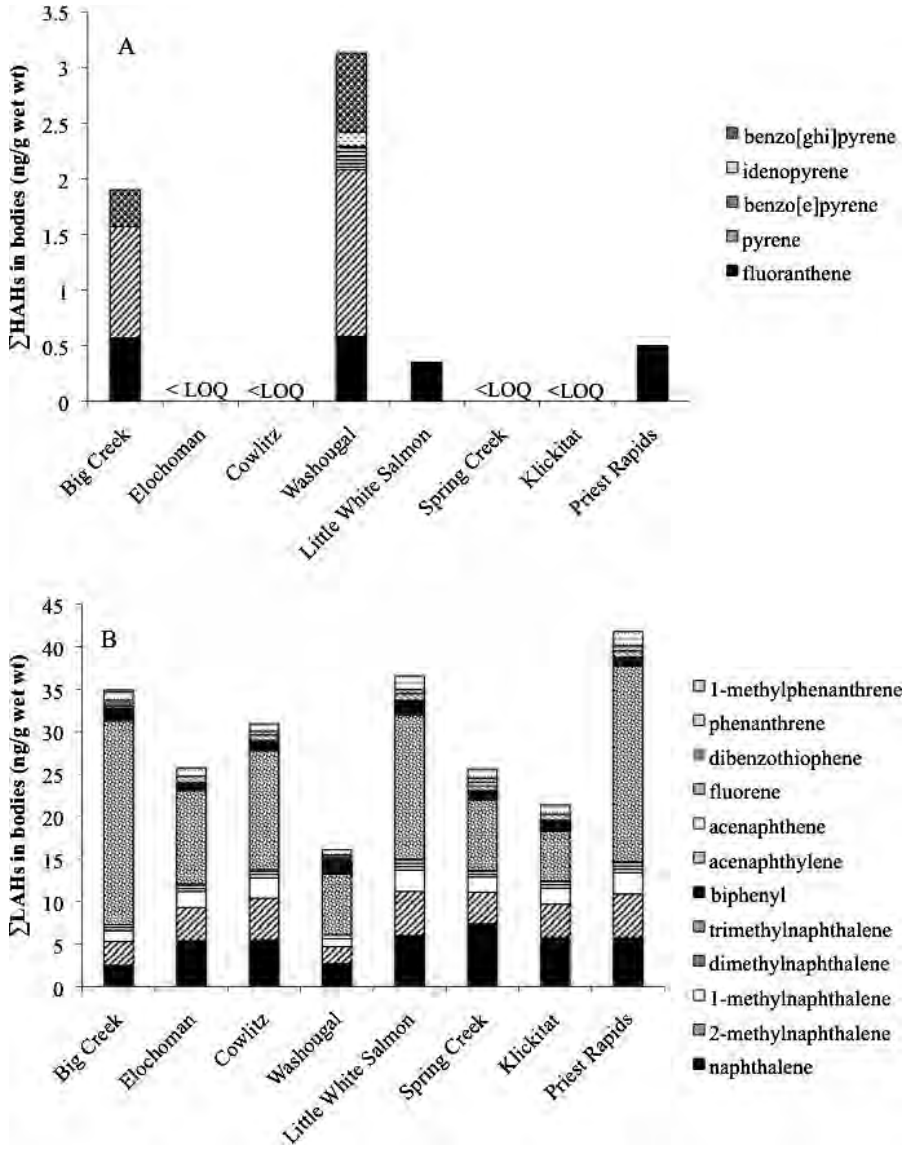

FIGURE 7.—Concentrations (ng/g wet weight) of **(A)** summed high-molecular-weight aromatic hydrocarbons (ΣHAHs) and **(B)** summed low-molecular-weight aromatic hydrocarbons (ΣLAHs) measured in body composites (10 fish/composite) of juvenile fall Chinook salmon collected from Columbia River hatcheries in 2005 (LOQ = limit of quantitation).

would have accumulated about 52 ng PCBs/g (Table 6). The estimated proportion of PCBs that could be attributed to hatchery exposure ranged from 76% in Chinook salmon from Warrendale to only 20% in fish from the Confluence. For DDTs (Table 7), it was estimated that fish from Warrendale and the Confluence would have accumulated 12–14 ng DDTs/g after leaving the hatchery, while fish collected at Columbia City would have accumulated about 30 ng DDTs/g (Table 7). The estimated proportion of DDTs that could be attributed to hatchery exposure ranged from 20% in Chinook salmon from Columbia City to 40% in fish from Warrendale. In the case of PBDEs (Table 8), the contribution of the hatchery to body burdens was minimal in fish from all sites, ranging from less than 1% to 4% total PBDEs.

Discussion

Our analyses indicate that chemical contaminants are present in the feed and bodies of juvenile fall Chinook

TABLE 4.—Concentrations (ng/mg of bile protein) of metabolites of high- and low-molecular-weight polycyclic aromatic hydrocarbons (PAHs) measured in bile of juvenile fall Chinook salmon sampled from Columbia River hatcheries (Figure 1) in 2005. Low-molecular-weight metabolites were measured at phenanthrene (PHN) wavelengths, while high-molecular-weight metabolites were measured at benzo[a]pyrene (BaP) wavelengths. Each bile sample was a composite of approximately 30 fish (NM = not measured).

Hatchery	PHN[a] (ng equivalents/mg bile protein)	BaP[b] (ng equivalents/mg bile protein)
Big Creek State	590	33
Elochoman	700	27
Cowlitz	2,000	71
Washougal	920	22
Little White Salmon	NM	NM
Spring Creek	930	43
Klickitat	600	24
Priest Rapids	5,500	210

[a] Concentrations in parts per billion (ng/g) based on total area compared with the fluorescence of the PHN standard at 260 and 380-nm wavelengths.
[b] Concentrations in parts per billion (ng/g) based on total area compared with the fluorescence of the BaP standard at 380 and 430 nm wavelengths.

salmon from a number of hatcheries in the Columbia River basin, although at relatively low concentrations. The most widespread contaminants were PCBs, DDTs, and LAHs, which were observed in fish and feed from all hatcheries. Some additional OC pesticides were also detected. The PBDEs, in contrast, were below LOQs in most samples.

Concentrations of POPs in feed samples were similar to or below concentrations reported in previous studies. In feed samples from U.S. Fish and Wildlife Service hatcheries, Maule et al. (2007) reported PCB concentrations in about the same range (<1–11 ng/g wet weight) as those we measured. Somewhat higher PCB concentrations were reported in commercial feed samples analyzed by Easton et al. (2002), Hites et al. (2004a), and Kelly et al. (2008), where a number of samples contained PCBs at concentrations in the range of 30–90 ng/g wet weight. Similarly, levels of PBDEs in the fish feeds we sampled were in the lower range of values reported by other investigators (<1–11 ng/g wet weight; Hites et al. 2004b; Montory and Barra 2006). Concentrations of DDTs in most of our samples were 30–40 ng/g wet weight, which is comparable with values in the range of 3–60 ng/g wet weight reported by Easton et al. (2002), Hites et al. (2004a), Maule et al. (2007), and Kelly et al. (2008). In our feed samples, *p*,*p*′-DDE and *p*,*p*′-DDD accounted for the majority of ΣDDTs measured, which is similar to results of previous studies (Easton et al. 2002; Hites et al. 2004a; Maule et al. 2007). However, low proportions of *o*,*p*′-DDT and *p*,*p*′-DDT, the parent forms of the compounds, were also present in most of our feed samples, suggesting that the feed contained products (e.g., fish oils, fish meals) from regions with relatively recent usage of DDTs (Jacobs et al. 2002).

Concentrations of PAHs in the hatchery feeds were higher than expected (up to 600 ng/g wet weight). More typical concentrations are below 200 ng/g wet weight (Easton et al. 2002; Johnson et al. 2007a), although concentrations over 1,000 ng/g wet weight have occasionally been reported (Easton et al. 2002). Commercial fish feeds, which consist primarily of fish meal and fish oil (Naylor et al. 2000), usually contain negligible levels of PAHs because these compounds are metabolized and eliminated by fish and do not accumulate in their tissues (Varanasi et al. 1989; Hom et al. 1996, 1999). Although the sources of the PAHs in the fish feeds we analyzed are not known, the composition of the PAHs measured in the feed (e.g.,

TABLE 5.—Contaminant concentrations (%) in fish bodies versus feed for juvenile fall Chinook salmon sampled from Columbia River hatcheries (Figure 1) in 2005. Values are body concentrations expressed as percentages of concentrations in feed. Calculations are based on lipid weight concentrations for both body and feed samples (Σ = summed concentrations; PCBs = polychlorinated biphenyls; HCHs = hexachlorocyclohexanes; CHLDs = chlordanes; HCB = hexachlorobenzene; PBDEs = polybrominated diphenyl ethers; LAHs = low-molecular-weight aromatic hydrocarbons; HAHs = high-molecular-weight aromatic hydrocarbons; <LOQ = less than lower limit of quantitation).

Hatchery	ΣPCBs	ΣDDTs	ΣHCHs[a]	Dieldrin	ΣCHLDs[b]	HCB	ΣPBDEs	ΣLAHs	ΣHAHs
Big Creek	230	96	36	<LOQ	230	170	200	16	7.2
Elochoman	780	160	<LOQ	<LOQ	450	<LOQ	<LOQ	59	<LOQ
Cowlitz	210	96	<LOQ	<LOQ	<LOQ	230	<LOQ	37	<LOQ
Washougal	780	170	<LOQ	<LOQ	1,200	540	<LOQ	84	420
Little White Salmon	110	96	<LOQ	<LOQ	88	100	<LOQ	28	1.9
Spring Creek	480	380	<LOQ	130	68	<LOQ	190	53	<LOQ
Klickitat	190	91	<LOQ	97	140	170	<LOQ	52	<LOQ
Priest Rapids	1,300	97	<LOQ	<LOQ	44	<LOQ	<LOQ	19	1.9

[a] Includes α-HCH, β-HCH, and γ-HCH (lindane).
[b] Includes heptachlor, heptachlor epoxide, g-chlordane, a-chlordane, oxychlordane, *cis*-nonachlor, *trans*-nonachlor, and nonachlor III.

TABLE 6.—Estimated accumulation of polychlorinated biphenyls (PCBs) in juvenile fall Chinook salmon sampled from the lower Columbia River and estuary. Fish from Warrendale, the Confluence, and Columbia City sites are marked hatchery released fish that were collected in the river in spring 2005. All fish belong to lower Columbia River stocks, but their hatcheries of origin are unknown.

Source	River kilometer	Mean weight of fish (g)	PCB concentration (ng/g)	Total PCBs (ng)	Concentration from river exposure (ng/g)[c]	Concentration from hatchery exposure (ng/g)	Percentage of PCBs from hatchery exposure
Warrendale[a]	227	5.2	19	99	4.6	14	76
Confluence[a]	163	5.6	65	364	52	13	21
Columbia City[a]	134	5.4	49	265	35	14	28
Columbia River hatcheries[b]		5	15	75	0	15	100

[a] Data on body PCB concentrations from these sites are from the Lower Columbia River Estuary Partnership (LCREP 2007).
[b] Mean of data from Elochoman, Spring Creek, Cowlitz, Little White Salmon, Klickitat, and Priest Rapids hatcheries (Figure 1).
[c] Concentrations of PCBs in fish from the estuary were determined by subtracting total nanograms for hatchery fish from the total nanograms in estuary fish and dividing by the weight of estuary fish (see Meador et al. 2002). All concentrations are reported on a wet-weight basis.

high proportions of naphthalene and alkylated naphthalenes) indicate that the feed or feed components were exposed to a petroleum product that contains these compounds, possibly at the hatcheries themselves.

The hatcheries sampled as part of this study obtained their feeds from several commercial feed suppliers based in the Pacific Northwest, so these feeds would probably be representative of those used by other trout and salmon hatcheries in the region. In fact, their contaminant concentrations were similar to those reported for feeds used by salmon hatcheries in British Columbia (Kelly et al. 2008). Our study was not designed to evaluate contaminant concentrations in feeds supplied by different companies, and the samples we collected would not be sufficient to characterize the various types of feed produced by these suppliers. However, we did find that the average concentrations of DDTs, PCBs, and PAHs were not significantly different in feeds supplied by different manufacturers.

For the most part, average body burdens of bioaccumulative POPs were similar in fish from hatcheries throughout the Columbia River area and somewhat lower than those previously reported for farmed salmon or juvenile fall Chinook salmon from other Oregon and Washington hatcheries (i.e., about 50 ng/g wet weight for PCBs and DDTs and up to 5 ng/g wet weight for PBDEs; Easton et al. 2002; Jacobs et al. 2002; Hites et al. 2004a, 2004b; Johnson et al. 2007a). In part, this is because the fish from the studies by Easton et al. (2002), Hites et al. (2004a, 2004b), and Johnson et al. (2007a) were generally older and larger than those examined in the present study and thus would have had a longer period to accumulate contaminants. Also, contaminant concentrations in feed were higher in some of the earlier studies. This appears to be a general trend; since the 1970s, levels of contaminants in fish feeds have tended to decline (Maule et al. 2007). In our study, Chinook salmon body contaminant concentrations comparable with those in fish from the studies cited above were found only in fish from Priest Rapids Hatchery, with body PCB levels that were almost 60 ng/g wet weight.

Concentrations of PCBs, DDTs, chlordanes, and other POPs measured on a lipid weight basis were generally up to 300% of concentrations in feed. However, contaminant concentrations in Chinook salmon bodies were not strongly correlated with concentrations in feed samples from the hatcheries where the fish were reared. This is perhaps not surprising, as a variety of factors that we could not

TABLE 7.—Estimated accumulation of DDTs in juvenile fall Chinook salmon from the lower Columbia River and estuary. Fish from Warrendale, the Confluence, and Columbia City sites are marked hatchery-released fish that were collected in the river in spring 2005. All fish belong to lower Columbia River stocks, but their hatcheries of origin are unknown.

Source	River kilometer	Mean weight of fish (g)	DDT concentration (ng/g)	Total DDTs (ng)	Concentration from river exposure (ng/g)[c]	Concentration from hatchery exposure (ng/g)	Percentage of DDTs from hatchery exposure
Warrendale[a]	227	5.2	20	104	12	8.1	40
Confluence[a]	163	5.6	22	123	15	7.5	34
Columbia City[a]	134	5.4	38	205	30	7.8	20
Columbia River hatcheries[b]		5	8.3	42	0	8.3	100

[a] Data on body DDT concentrations from these sites are from the Lower Columbia River Estuary Partnership (LCREP 2007).
[b] Mean of data from Elochoman, Spring Creek, Cowlitz, Little White Salmon, Klickitat, and Priest Rapids hatcheries (Figure 1).
[c] Concentrations of DDTs in fish from the estuary were determined by subtracting total nanograms for hatchery fish from the total nanograms in estuary fish and dividing by the weight of estuary fish (see Meador et al. 2002). All concentrations are reported on a wet-weight basis.

TABLE 8.—Estimated accumulation of polybrominated diphenyl ethers (PBDEs) in juvenile fall Chinook salmon from the lower Columbia River and estuary. Fish from Warrendale, the Confluence, and Columbia City sites are marked hatchery-released fish that were collected in the river in spring 2005. All fish belong to lower Columbia River stocks, but their hatcheries of origin are unknown.

Source	River kilometer	Mean weight of fish (g)	PBDE concentrations (ng/g)	Total PBDEs (ng)	Concentration from river exposure (ng/g)[c]	Concentration from hatchery exposure (ng/g)	Percentage of PBDEs from hatchery exposure
Warrendale[a]	227	5.2	2.9	15	2.8	0.12	4.0
Confluence[a]	163	5.6	8.6	48	8.5	0.11	1.2
Columbia City[a]	134	5.4	37	200	37	0.11	0.3
Columbia River hatcheries[b]		5.0	0.12	0.6	0.0	0.12	100

[a] Data on body PBDE concentrations from these sites are from the Lower Columbia River Estuary Partnership (LCREP 2007).
[b] Mean of data from Elochoman, Spring Creek, Cowlitz, Little White Salmon, Klickitat, and Priest Rapids hatcheries (Figure 1).
[c] Concentrations of PBDEs in fish from the estuary were determined by subtracting total nanograms for hatchery fish from the total nanograms in estuary fish and dividing by the weight of estuary fish (see Meador et al. 2002). All concentrations are reported on a wet-weight basis.

fully assess, including fish age, maternal transfer of contaminants, body lipid content, dietary moisture and lipid contents, contaminants in previous feeds, and feed ration, could affect absorption and body concentrations of contaminants. However, there were a few cases (e.g., ΣPCB concentrations in fish from Washougal, Elochoman, and Priest Rapids hatcheries) in which contaminant concentrations in fish bodies appeared unusually high for the dietary concentrations. These levels of contaminant uptake could not be explained by fish size or lipid content, raising the possibility that fish might be exposed to contaminants from sources other than the diet. Recently, there have been reports of fish in Montana and Washington state hatcheries being exposed to PCBs in paint (MFWP 2004; Cornwall 2005), but whether this is a possible source of exposure for the fish at the hatcheries we sampled is unknown.

In comparison with the OCs, accumulation of PAHs from feed into Chinook salmon tissues was low, with typical body concentrations of ΣLAHs and ΣHAHs below 84% and below 7.2%, respectively, of concentrations measured in feed. This is consistent with the extensive metabolism of PAHs in fish (Varanasi et al. 1989). Fish from Washougal Hatchery were an exception, as the ΣHAH concentration in tissues of these fish was 420% of that in feed. However, the reason for the relatively high uptake of HAHs in these fish is unknown. Concentrations of ΣLAHs in feed were highly correlated with levels of LAH metabolites in fish bile, indicating the importance of the diet as a source for these compounds. The relationship was not as strong for ΣHAHs, which were present at only low concentrations in feed samples.

In general, the concentrations of PCBs, DDTs, and PAHs measured in hatchery fish and feed in the present study were below levels associated with adverse biological effects in salmon. For PCBs, Meador et al. (2002) estimated a critical body residue of 2,400 ng/g lipid weight for protection against 95% of effects ranging from enzyme induction to mortality. Even in Chinook salmon from Priest Rapids Hatchery, PCB body burdens were well below this level. Similarly, DDT concentrations in hatchery Chinook salmon were well below the DDT effect threshold of 600 ng/g wet weight estimated by Beckvar et al. (2005), or approximately 6,000 ng/g lipid weight if we assume that most of the data on which the threshold was based were derived from laboratory-reared salmonids, which typically have a lipid content near 10% (Meador et al. 2002). Dietary levels of PAHs were also well below concentrations that have been associated with effects on growth, metabolism, or immune function in field and laboratory studies (Casillas et al. 1998; Palm et al. 2003; Meador et al. 2006), which are generally in the range of 5,000 ng/g wet weight and above.

Levels of PAH metabolites in bile of hatchery Chinook salmon were also generally below concentrations thought to be associated with toxic effects in juvenile salmon, estimated by Meador et al. (2008) as PHN metabolite levels in excess of 2 μg/mg bile protein. Levels of PHN metabolites in bile in this range were found only in fish from Cowlitz and Priest Rapids hatcheries. According to Meador et al. (2008), dietary PAH levels typically associated with bile metabolite levels like those in Cowlitz and Priest Rapids fish are 11–22 μg/g wet weight, much higher than levels in feed from these two hatcheries. This suggests that Chinook salmon from these two hatcheries may have been exposed to PAHs from other sources, possibly through the water column.

Lipid content in Chinook salmon and feed samples was measured by both gravimetric and TLC–FID quantitation methods. Previous studies have shown that TLC–FID lipid values are generally lower than those determined gravimetrically, probably because the gravimetric method measures lipids as well as other co-extracted biogenic materials that are not included in percent lipid values determined by TLC–FID (Delbeke

et al. 1995). In a recent study of juvenile Chinook salmon, TLC–FID lipid content values were about 15% lower than those obtained through gravimetric analysis (Johnson et al. 2007b). Results were similar for the samples we analyzed in the present study; on average, values determined by TLC–FID were 22% lower for feed and 28% lower for fish bodies than values determined gravimetrically. However, the percent lipids determined by both methods were highly correlated, as was also demonstrated in previous studies (Delbeke et al. 1995; Ylitalo et al. 2005).

Compared with that in field-collected juvenile Chinook salmon from the lower Columbia River and other Pacific Northwest estuaries (Johnson et al. 2007a, 2007b; LCREP 2007), the lipid content of the hatchery Chinook salmon we sampled was somewhat higher (3–6% versus 1–3%). This is typical of comparisons between wild and hatchery fish (Ackman and Takeuchi 1986). The hatchery Chinook salmon also had higher proportions of free fatty acids (up to 50% of total lipids) compared with those in field-collected juvenile fall Chinook salmon (<15% of total lipids; Johnson et al. 2007a, 2007b; LCREP 2007). This is unusual because most studies, including those with both wild and hatchery-reared salmonids, have found small amounts of free fatty acids in animal tissues (Sheridan 1989; Næsje et al. 2006; Pratoomyot et al. 2008). Diet can affect lipid profiles, including free fatty acid levels, in wild and hatchery-reared fish (Bergström 1989; dos Santos et al. 1993; Jobling and Bendiksen 2003). However, the lipid content and profile of the feeds we analyzed had no special characteristics and were similar to those reported for other feeds (Easton et al. 2002; Johnson and Barnett 2003; Hamilton et al. 2005). The implications of relatively high free fatty acid levels in hatchery fish are unclear, but high free fatty acid levels have been associated with diabetes, heart disease, and related health problems in humans and laboratory animals (Boden 2002).

Although chemical contaminants were present in Columbia River hatchery fish and feed, our analyses suggest that hatcheries are unlikely to be a major source of contaminants for most juvenile fall Chinook salmon in the lower Columbia River. In the case of PBDEs, concentrations were extremely low in both fish and feed from all of the hatcheries we sampled. Concentrations of PCBs, DDTs, and PAHs or their metabolites in hatchery fish and feed were comparable with concentrations in stomach contents, bodies, and bile of juvenile fall Chinook salmon from rural estuaries in the Pacific Northwest (Johnson et al. 2007a) but were significantly lower than concentrations measured in bodies and stomach contents of juvenile fall Chinook salmon from several sites along the lower Columbia River (LCREP 2007). These findings were reflected in our analysis of in-river and hatchery contributions to contaminant body burdens in Columbia River out-migrant juvenile Chinook salmon of hatchery origin. The river was identified as the primary source of PBDE exposure for all juvenile fall Chinook salmon. Similarly, in fish from all sites, the river was estimated to be the greatest source of DDTs, although hatchery rearing accounted for a significant percentage of DDT uptake (20–40%). It appeared that hatchery rearing could account for most of the PCB body burden of fish from rural sites (e.g., Warrendale) where PCB contamination is minimal, but for fish rearing at or downstream from major urban centers such as Portland (i.e., fish collected from the Confluence and Columbia City sites), the river was by far a more important source of PCBs. In summary, the results suggested that high contaminant body burdens in out-migrant hatchery fall Chinook salmon are probably due to exposure to contaminants in the Columbia River rather than to rearing practices at regional hatcheries.

However, even if contaminant levels in hatchery feeds or the hatchery environment are below concentrations likely to affect fish during hatchery rearing, their accumulation in prerelease hatchery fish may be a management concern. If contaminant body burdens are already above background levels when fish leave the hatchery, they have an increased risk of reaching concentrations that could reduce their likelihood of survival during estuarine residence. Moreover, the lipid content of hatchery fish when they are released is relatively high, and when these lipids are metabolized during downstream migration, contaminants stored in body fat may also be released and exert toxic effects (Elskus et al. 2005). Finally, salmon with elevated contaminant levels would contribute toxicants to the environment and to the food chain (Missildine et al. 2005; O'Toole et al. 2006). This represents a hazard for birds and other piscivorous wildlife, which are known to be at risk from exposure to bioaccumulative contaminants (e.g., PCBs and DDTs) in the lower Columbia River (Anthony et al. 1993; Thomas and Anthony 1999; Henny et al. 2003; USFWS 2004; Buck et al. 2005.

Although contaminant concentrations were relatively low in all of the feeds we analyzed, we recommend routinely testing feeds for contaminants because other studies have shown that the types and concentrations of chemicals in feeds can vary substantially from lot to lot (Ylitalo et al. 2001; Easton et al. 2002; Hites et al. 2004a; Maule et al. 2007). The U.S. Fish and Wildlife Service already does chemical testing on feeds used in national fish hatcheries (e.g., Maule et al. 2007), and

the procedure should be encouraged for hatcheries managed by other agencies. The testing programs that have been implemented to date appear to have led to improvements in feed quality (Maule et al. 2007), and continued efforts will help to minimize any risks to fish, wildlife, and human health.

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Bioaccumulation of polychlorinated biphenyls in juvenile chinook salmon (*Oncorhynchus tshawytscha*) outmigrating through a contaminated urban estuary: dynamics and application

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Abstract A field study was conducted to examine bioaccumulation of polychlorinated biphenyls (PCBs) for hatchery-raised and naturally reared (wild) ocean-type juvenile chinook salmon outmigrating through the Lower Duwamish Waterway (LDW), a contaminated urban estuary in Seattle, WA, USA. These results show differences in bioaccumulation of PCBs over time and space in this estuary, which may also occur for any contaminant that is distributed heterogeneously in this system. Highly mobile, outmigrating salmon accumulated ~3–5 times more PCBs on the east side of the LDW than fish on the west side, which is supported by an almost identical difference in mean sediment concentrations. The tPCB concentration data suggest that for most of the spring and early summer, juvenile chinook were likely segregated between the east and west side of the LDW, but may have crossed the channel later in the year as larger fish. Additionally, we used biota-sediment accumulation factors to assess the relative degree of bioaccumulation and explore these factors as potential metrics for predicting adverse sediment concentrations. These results highlight the importance of time and space in sampling design for a highly mobile species in a heterogeneous estuary.

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Keywords PCBs · Bioaccumulation · Salmon · Spatial segregation · Toxicity guideline value

Introduction

Even though polychlorinated biphenyls (PCBs) were banned in the United States in 1979, they persist at high concentrations in sediments and aquatic foodwebs. The influx of cleaner sediments over time was expected to accumulate and bury these contaminants below the biologically active zone; however, these compounds still occur at very high concentrations in surface sediment and are biologically available to biota.

The Green River flows northwest from the western flanks of the Cascade Mountains near Mt. Rainier and travels ~150 km to Elliott Bay near downtown Seattle, WA, USA. For the last 19 km the Green River is called the Duwamish River and for the final 9 km it is known as the Lower Duwamish Waterway (LDW; Fig. 1). At river kilometer (rkm) 0 the river splits into the East and West Waterways around Harbor Island for 2 km before entering Elliott Bay. The LDW is a marine-influenced urban estuary that has been the focus of intense studies due to its highly contaminated sediment and water. The average width of the LDW is ~130 m and the water depth ranges from 3 to 20 m; however, most of LDW is maintained at 10 m depth (mean lower low water) by dredging. Even though most of the natural habitat has been severely altered, off-channel areas (e.g., Slip 4 and Kellogg Island) and a narrow shallow-slope intertidal habitat can be found along the waterway where outmigrating salmon likely forage and can be collected.

Past work has documented that sediment and organisms in the LDW are contaminated with PCBs, PAHs,

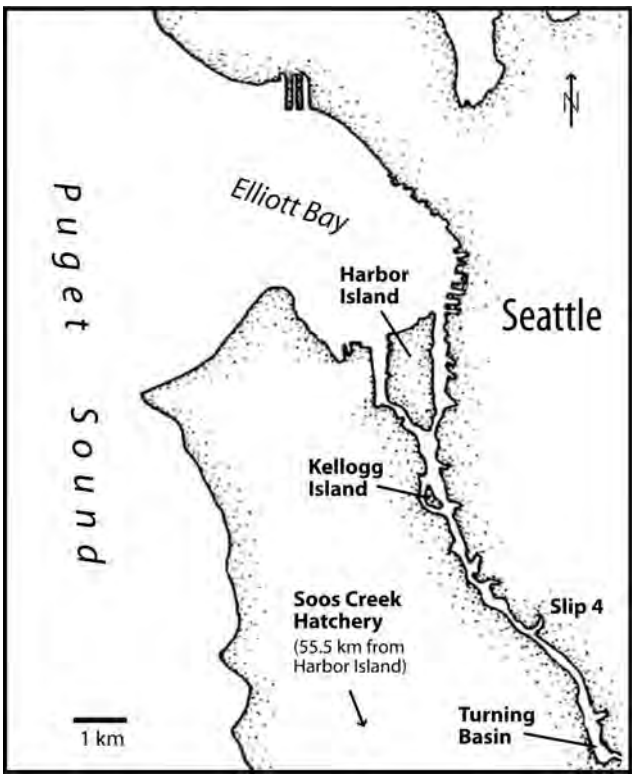

Fig. 1 Map of the Lower Duwamish Waterway

tributyltin, and other contaminants of concern (Varanasi et al. 1993; LDWG 2007). The entire LDW was listed as a Superfund site under the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) in 2001 and is currently progressing through the standard superfund remedial process. PCBs have been an important concern in the LDW for several years after they were discovered at high concentrations in sediment at several sites. We focused on PCBs because of elevated concentrations in the LDW, high potential for toxicity to juvenile salmon, low elimination rates in fish, and relative ease of assessing sediment and tissue concentrations.

Several salmonids including chinook (*O. tshawytscha*), coho (*O. kisutch*), chum (*O. keta*), and winter steelhead (*O. mykiss*), are raised in several hatcheries in this watershed and released every year. For most years, ~5–6 million fish have been released annually into the Green River and most of these (≈70%) are age 0+ (subyearling; age 0–1 year) ocean-type chinook (Sieler et al. 2002), which are protected in this watershed under the Endangered Species Act and were the target of our study. Juvenile chinook are released from three hatcheries on this system; however, 80% or more come from the Soos Creek hatchery. Additionally, ~1 million ocean-type chinook naturally rear (wild) in this system and also migrate through the LDW (Sieler et al. 2002). Since 2000, essentially all hatchery chinook released in this watershed have been marked by clipping their adipose fin. Because the error rate (bad clips) is generally low at ~4% (Ruggerone et al. 2006), this procedure has allowed us to distinguish hatchery from naturally reared fish with fairly high confidence. Juvenile salmonids migrate from relatively uncontaminated upstream waters into the Duwamish River and LDW during smoltification where they adjust to seawater, feed on relatively abundant invertebrates, and rear from a few days to several weeks before exiting to open water. The peak migration for age 0+ hatchery fish occurs from late May to mid June and wild fish are found in the Duwamish from mid January through late summer (Ruggerone et al. 2006).

The goal for this study was to examine PCB bioaccumulation in highly mobile, outmigrating juvenile salmon in this estuary, determine total amount accumulated, and examine the application of bioaccumulation factors to predict sediment concentrations that may result in adverse tissue concentrations. Our hypothesis was that juvenile chinook fish would migrate along the west or east bank of the river and reflect the contamination of each region. If fish freely crossed the waterway, the concentrations of PCBs and other contaminants in fish collected at Kellogg Island should be similar to the levels in fish collected at Slip 4. Small outmigrating salmonids tend to stay in shallow areas as they feed and migrate through an estuary (Healey 1991). On average, the west side of the LDW contains substantially lower concentrations of PCBs in sediment than those collected on the east side, which we hypothesized would be reflected in the amount bioaccumulated by the fish collected. Although not in our original design, we were also able to consider some temporal aspects of PCB bioaccumulation for juvenile salmonids because our sample dates spanned 11 weeks over late spring and mid summer.

Methods

The area of focus for this study is the lower Duwamish River occurring from the turning basin (rkm 7.6) to the confluence of the east and west waterways at the southern tip of Harbor Island (rkm 0; Fig. 1) and constitutes most of the marine influenced section of the Duwamish River. The surface area of intertidal and subtidal sediment in this section of river is ~142 ha (350 acres).

Fish sampling

Juvenile chinook were sampled from four locations in this river system. For the upstream sites in the Green River, fish were collected from the Soos Creek Hatchery on Big Soos Creek (a few km upstream of the confluence of the Green River and Big Soos Creek at rkm 54.4) for 3 years

(2000–2002) usually before they were released in late May, except in 2002 when fish were sampled from the hatchery on 8 August. Naturally reared fish (wild) were also collected one year (2000) from a screw trap at rkm 55.6, which is upstream from the Soos Creek hatchery and confluence of the Green River and Big Soos Creek. These fish were acquired live from personnel of the Washington Department of Fisheries and Wildlife (WDFW).

On the west side of the LDW, we collected fish at Kellogg Island, which is a semi-natural area off the main channel at rkm 1.3. On the east side we sampled fish at Slip 4 (rkm 4.3), which is a 1.5 ha (3.6 acre) blind inlet off the main channel. Historically, we have observed large numbers of migrating salmon and other fish species at these two locations. We sampled at both LDW sites over 4 years (2000–2002 and 2004). For the year 2000, we sampled fish in late May; ~5 days after the last group of hatchery fish had been released from the Soos Creek hatchery. For subsequent years, we collected fish at these sites from late June to early August. We also analyzed two composite samples of juvenile coho collected at Slip 4 in 2002 to determine if the values for whole body and stomach concentrations were similar to those found for chinook.

A 100-m beach seine was used in the LDW for sample collections and all fish were kept alive in coolers until processing at our laboratory. Samples were frozen at −80°C until analyzed. Stomach contents were removed from all fish; therefore the whole-body concentrations represent only the PCBs that were assimilated. Whole fish were analyzed as individuals or composite samples, each containing from 3 to 10 individuals. Samples for stomach contents were almost always composites of material from several individuals.

Analytical determinations for OCs and lipid in tissue

Whole-body fish and stomach content samples were analyzed for organochlorines (OCs), including dioxin-like PCBs, other selected PCB congeners, by a high-performance liquid chromatography/ultraviolet photodiode array (HPLC/PDA) method (Krahn et al. 1994). Sample extractions were split for PCB and lipid analyses. Prior to sample cleanup, a 1 ml portion of each whole-body extract was removed for percent lipid analyses by thin-layer chromatography/flame ionization detection (TLC/FID) (Ylitalo et al. 2005b). Lipid classes were measured by FID, but are not reported here. Percent lipid values were calculated by summing the concentrations of all lipid classes determined for each sample.

A separate study compared the tissue concentrations from sample splits for our HPLC/PDA method (NOAA lab) and those obtained with high resolution gas chromatography/mass spectrometry (GC/MS; Axys Analytical Services LTD, Sidney, British Columbia, Canada). The results for 30 samples (four species, whole body and muscle, range of 5–300 ng/g) indicated close agreement between methods, although 80% of the GC/MS values were higher than those for the HPLC/PDA method (Sandie O'Neill and James West, WDFW, personal communication). The overall mean (SD) percentage difference among all samples was 24 (0.22)%, which is very low. These results are supported by other studies that have shown close agreement for summed PCB concentrations obtained by the HPLC/PDA and GC/MS methods for a wide range of marine biota (Krahn et al. 1994; Ylitalo et al. 2005a).

Quality assurance for HPLC/PDA method

A method blank and a National Institute of Standards and Technology (NIST) blue mussel Standard Reference Material (SRM 1974a or 1974b) sample were analyzed with each sample set containing 8–12 field samples as part of a performance-based quality assurance program (Sloan et al. 2006). Results obtained for SRMs were in excellent agreement with the certified and reference values published for these materials by the National Institute of Standards and Technology. In addition, the other quality control samples met established laboratory criteria. Duplicate analyses were conducted for 10% of the tissue samples, with relative standard deviations ≤30% for more than 80% of analytes detected in the samples. Method blanks contained no more than four analytes that exceeded four times the limit of quantitation (LOQ), unless the analyte was not detected in the associated tissue samples in the set. The percent recovery of the surrogate standard ranged from 70 to 105%.

Sediment concentrations

A separate study of 326 sediment samples for PCBs in the Duwamish estuary (Industrial Economics 1998) was used to analyze bioaccumulation in fish (Table 1). This study conducted a comprehensive analysis of PCBs in sediment over the entire Lower Duwamish Waterway (142 ha sampled) from the turning basin to rkm 0 that included our fish collection sites. Total organic carbon and PCBs were determined for each sample, which allowed determination of the organic-carbon normalized sediment concentrations (sed_{oc}). The same method (HPLC/PDA) for PCB analysis described above for tissue was also used to quantify PCBs for these sediment samples. Of the sediment sites that were examined in detail, tPCBs from the LDW were mostly consistent with the Aroclor 1254 pattern or a mix of Aroclors 1254 and 1260 (>90% of samples).

The waterway was divided into five cross-river sections (intertidal and subtidal for the east and west sides and the navigational channel). The demarcation between the

Table 1 Concentrations of total polychlorinated biphenyls (tPCBs) in sediment

Regions and locations	Mean sediment (ng/g sed)	Mean sed_{oc} (μg/g OC)		Hectares	Sediment (ng/g sed)				
		Mean (SD)	Median		10th	25th	50th	90th	95th
West side	150 (20) 113	10.6 (1.5)	5.1	54.2	7	28	63	337	545
East side (to Slip 4)	500 (150) 95	33.5 (9.8)	7.8	22.9	6	11	107	1,038	1,987
Kellogg Island	190 (60) 35	8.9 (1.8)	5.3	28.9	11	28	69	444	756
Slip 4	1,200 (320) 42	88.8 (24.5)	35.1	16.5	74	190	450	2,700	4,511
East side—Slip 4 to opposite Kellogg Island	180 (40) 59	10.7 (1.6)	6.5	18.6	18	41	87	428	672

Values are mean and standard deviation (SD) for total PCBs in sediment and sed_{oc} (organic carbon (OC) normalized values; μg total PCBs/g OC in sediment). Several percentile values are also shown for each region and location. All values determined with minimum unbiased estimator for a lognormal distribution. Following SD denotes the number of samples per mean value. Data from Industrial Economics (1998)

subtidal areas and the channel was determined from navigation charts (Industrial Economics 1997). Within these major sections, numerous substrata were defined. A total of 90 substrata (nonoverlapping polygons of the sediment surface) were determined for the LDW. Some of the substrata represent discrete areas (e.g., slips, backwaters, noncontinuous intertidal areas, outfalls, and seeps). The overall intent for this sampling scheme was the primary efficiency criterion of stratification designs that concentrations within strata are more homogeneous than concentrations over the entire study area (Industrial Economics 1997).

Sediment sample sites within substrata were determined randomly and spaced less than 100 meters apart. Of the 54 substrata selected for our analysis, the mean (SD) size was 1.42 (1.45) ha. The mean (SD) number of samples for all substrata from that study was 2.2 (1.7) per hectare and no one area was overly represented. Substrata in the navigation channel were not included because we assumed that juvenile chinook would not occur in that area of the LDW or interact with this benthic environment that is frequently disturbed by river flow, tidal flux, and vessels.

To determine the mean sed_{oc} for the west side, all intertidal and subtidal samples from just north of the Turning Basin (rkm 7.6) to the southern tip of Harbor Island (rkm 0) were included. This value was used for the BSAF calculation for salmonids collected at Kellogg Island. Similarly, we choose all intertidal and subtidal sediment samples from just north of the Turning Basin to ~1,000 m north of Slip 4 on the east side for the BSAF equation for chinook collected at Slip 4. One sediment sample in Slip 4 was excluded because it was considered an outlier (Grubbs test, $P < 0.0001$). The tPCBs for this one sample was 25 μg/g, which was 50 times the mean value for all east side samples ($n = 96$) and was therefore not representative of values from this region. This hot spot represented a very small area and its inclusion would likely have skewed the BSAF values and conclusions. We also determined the sediment concentrations at the collection sites. For Kellogg Island, we included all inter- and subtidal sediment data from sampling sites around Kellogg Island and all sites ~1,000 m north and south of the island to calculate the mean sed_{oc}. The sediment concentrations for Slip 4 were determined in a similar fashion including all sites in Slip 4 and those inter- and subtidal sites 1,000 m to the north and south of this area.

Most of the PCB sediment contamination occurs on the east side of the LDW in inter- and subtidal areas from the Turning Basin to Slip 4 and is substantially more contaminated than the west side (Industrial Economics 1998). We determined that 56% of the sample sites on the east side contained PCB sediment concentrations >100 ng/g dry wt, which was higher than that for the west side (25%). Because we did not sample fish downstream of Slip 4 on the east side of the river those sediment concentrations were not included. The mean concentration for all sub- and intertidal sediment samples between Slip 4 and Harbor Island (rkm 0) on the east side was determined to be much lower than the upriver portion of the east side and very similar to the mean determined for the entire west side of the LDW (Table 1). This area contained one sample that was 23 times higher than the mean value and 10 times higher than any other concentration. It was determined to be an outlier based on Grubbs test ($P < 0.0001$) and was excluded for the same reasons stated above for the one Slip 4 value. If included, the mean tPCB sediment concentration would be 220 ng/g dry wt. a 25% increase, which was considered an undue influence for one of 60 samples.

Determination of PCB accumulation in the lower Duwamish

We used a mass balance approach to determine the total ng of PCBs accumulated per fish (body burden, bb) collected in the lower Duwamish.

$$PCB_{bb} = tPCB_{ld} \times WT_{ld} - tPCB_{u} \times WT_{u} \quad (1)$$

where PCB_{bb} represents the total ng of PCBs accumulated, $tPCB_x$ denotes the concentration of total PCBs (wet

weight), and WT_x is the wet weight for each fish or composite mean sampled. Subscripts for x are as follows: ld denotes fish collected in the Lower Duwamish and u denotes upriver fish (hatchery or wild). For all hatchery fish collected in the LDW we used the hatchery-collected fish for the upriver concentration in Eq. 1 ($tPCB_u$) and for all wild fish collected in the LDW we used the mean concentration of tPCBs measured in wild fish collected from the screw trap in 2000 ($tPCB_u$).

Biota-sediment bioaccumulation factors (BSAFs) were calculated to highlight differences and similarities among species and sites. The following equation was used:

$$\mathrm{BSAF} = \frac{[\mathrm{tissue}]/f_{\mathrm{lip}}}{[\mathrm{sediment}]/f_{\mathrm{oc}}} \quad (2)$$

where f_{oc} is the fraction of organic carbon (g/g dry wt.) and f_{lip} is the fraction of lipid (g/g wet wt). For the collection year 2000, specific site and type (wild or hatchery) lipid concentrations were used. For all other years a mean lipid value of 1.0% was determined from all remaining data and used for the BSAF calculations for chinook.

We assumed that fish had an equal chance of visiting (temporally and spatially) each of the sediment sites that were used for these calculations. We also assumed that each tPCB sediment concentration was proportional to the tPCB concentration for water and prey in the immediate area around the sample and that accumulation was proportional to the OC normalized sediment concentration (sed_{oc}). We calculated BSAFs using mean tissue and sediment concentrations, which we believe provided a better estimate of bioaccumulation than median values.

These BSAF values were used to determine a sediment concentration that would be expected to protect outmigrating juvenile salmon from adverse biological effects. This sediment quality guideline was calculated with Eq. 1 by solving for sed_{oc}. For these calculations we used a mean whole-body lipid content of 1% wet weight (Table 2) and the 50th percentile for organic carbon (OC), which was 1.6% dry wt for each side of the waterway. We selected the PCB tissue toxicity guideline of 2.4 µg/g lipid for salmonids from Meador et al. (2002) for conversion to sediment values.

Toxicity equivalents

We calculated the sum of toxic equivalents (ΣTEQs) for dioxin-like (dl) PCBs for each sample. Each TEQ was determined by multiplying a dl PCB concentration with its toxicity equivalent factor (TEF) for fish, which was obtained from van den Berg et al. (1998). Our analytical method quantified the dl-PCB congeners 77, 105, 118, 126, 156, 157, 169, and 189. The other four dl congeners (81, 114, 123, and 167) were not quantified due to problems with coelution by interfering compounds. The TEQ levels calculated in the current study are conservative values because of the higher limits of detection of the HPLC/PDA system compared to the GC/MS method and they do not include the contributions from polychlorinated dibenzodioxins (PCDDs) or dibenzofurans (PCDFs). In addition, when the concentration of a dioxin-like PCB was below the LOQ, a value of zero for the specific congener was used in the calculation, which was more conservative than the commonly used value of one-half the LOQ. These below-detection values were not used because our LOQ was relatively high (0.03–0.4 ng/g wet weight for most samples), which was due to low sample weights (<4 g).

Statistical analysis

Most of the concentration data reported here were log-normally distributed, which is very common for such data (Gilbert 1987). Because lognormally distributed data are skewed, a minimum variance unbiased (MVU) estimator is more appropriate for computing statistics, such as the mean, variance, and quantiles. We used the MVU estimator algorithms in Gilbert (1987) for estimating the mean, variance, and quantiles (Eqs. 13.1, 13.2, and 13.24) for all log-normally distributed data (TEQs, BSAFs, and whole-body, stomach, and sediment concentrations). This MVU algorithm was not used when sample sizes were <3. We used SYSTAT 11 to construct cumulative distribution functions (CDFs), perform regression analysis, and to examine distributions. Statview 5.0 was used to perform Analysis of Variance (ANOVA) and post-hoc testing. After performing the ANOVA, a post-hoc examination of treatment means was conducted with Fisher's Protected Least Significant Difference (PLSD) test. Log values for concentrations were used for ANOVAs and regressions. We also used Grubbs Test to examine datasets for statistical outliers. Standard deviation is shown to provide a measure of the range in data and standard error of the mean (SEM) was used to indicate variation about the mean.

Results

PCBs in salmon

Juvenile chinook from upstream areas (hatchery and screw trap) contained very low levels of tPCBs, except for hatchery fish in 2001 (Table 3). Mean tPCBs concentrations in fish collected from Slip 4 were always higher than those collected at Kellogg Island. Although variability was observed among individuals, it was likely due to a range in time spent in the LDW (Fig. 2). The differences between wild and hatchery fish collected in the LDW were mixed.

Table 2 Data for salmon collected in the Duwamish River and upstream

Year		Type	Wt (g)	Len (mm)	Lipid (%)	BSAF	BSAF median	N {N tot}
Kellogg Island								
2000	May	Chinook W	4.4 (1.1)	76.5 (6.8)	1.6 (0.3) 4c	0.18 (0.01)	0.18	17 {31}
	May	Chinook H	4.8 (0.2)	79.7 (0.3)	1.8 (0.1) 3c	0.21 (0.02)	0.21	3 {30}
2001	June	Chinook W	5.4 (3.0)	84.5 (18.9)	–	0.82 (0.53)	0.47	4 {4}
2001	August	Chinook W	12.1 (4.3)	106 (8.9)	–	0.35 (0.07)	0.20	35 {39}
2001	June	Chinook H	6.1	85	–	0.21	–	1 {1}
2001	August	Chinook H	12.3 (2.1)	111 (4.2)	–	0.89 (0.44)	0.48	6 {6}
2002	August	Chinook W	10.7 (5.2)	100 (12.3)	1.1 (0.3) 7i	2.9 (1.3)	1.4	7 {7}
	August	Chinook H	19.7	124	1.2 1i	3.9	–	1 {1}
2004	July	Chinook H	9.8 (1.0)	102 (2)	0.9 (0.7) 3c	1.2 (0)	1.2	3 {9}
	July	Chinook W	11.3	107	1.9	0.8	–	1 {3}
Slip 4								
2000	May	Chinook H	4.6 (1.0)	80.1 (5.6)	2.0 (0.1) 2c	0.30 (0.12)	0.20	7 {15}
	May	Chinook W	3.4 (0.1)	69.5 (0.7)	–	0.25 (0.3)	–	2 {2}
2001	June	Chinook W	3.5 (0.9)	72.3 (5.6)	–	1.1 (0.18)	1.0	12 {12}
2001	August	Chinook W	12.7 (4.3)	107 (11.0)	–	0.90 (0.6)	0.36	5 {5}
2001	June	Chinook H	5.0 (0.08)	82.7 (1.5)	–	0.55 (0.16)	0.50	3 {3}
2001	August	Chinook H	12.7 (3.3)	109 (7.3)	–	0.53 (0.1)	0.46	4 {4}
2002	August	Chinook W	7.3	8.8	0.9 (0.3) 2i	1.2	–	1
	August	Chinook H	20.5	120	1.1 1i	3.8	–	1
	August	Coho W	5.4 (0.7)	78.8 (4.5)	1.8 (0.1) 2c	0.8 (0.1)	–	2 {7}
Soos Creek								
2000	–	Wild	3.9 (0.8)	73.3 (5.5)	1.9 (0.4) 2c	–	–	14 {26}
	–	Hatchery	6.0	–	2.2 (0.6) 3i	–	–	–
2001	–	Hatchery	2.5 (0.07)	–	–	–	–	7 {7}
2002	–	Hatchery	9.4 (0)	–	1.6 (1.2) 2i	–	–	2 {2}

Values shown as mean and standard deviation and determined with algorithms for lognormal distributions (Gilbert 1987) for all $n \geq 3$. Type (*W* wild; *H* hatchery; *M* mix of both types). *N* is the number of samples for each mean and *n* total is the total number of fish measured for length, weight, PCBs and BSAFs. Sample sizes for lipids shown next to value. "i" indicates individuals and "c" indicates composite values (ci indicates a combination of composite and individual values). Composite samples contained 3–10 individuals

There were no significant differences between hatchery and wild fish collected at Slip 4 for all years combined. Concentrations of tPCBs in the hatchery origin fish collected from Kellogg Island were significantly higher than wild fish ($P = 0.04$) when all years were considered, which was mostly due to a pulse of upriver wild fish with low tPCBs in August 2001.

The tPCB values for the composite samples containing coho salmon were not different than those containing chinook from Slip 4 in 2002. The coho whole-body concentrations were 550 and 440 ng/g, which were lower than the mean value for the two individual chinook (725 ng/g). The stomach contents concentrations for the coho and chinook composite samples (one each) for 2002 from Slip 4 were essentially identical (750 and 770 ng/g), which is reflected in the mean value and low SD.

The temporal aspect of PCB bioaccumulation is also noteworthy. The fish collected in 2000 were sampled in late May, which was $\sim$5 days after the last release of fish from the Soos Creek hatchery. Total PCB concentrations in both wild and hatchery fish for the year 2000 were relatively low compared to the other sampling periods, which occurred later in the summer (Fig. 2). The Kellogg Island fish contained substantially lower concentrations of tPCB than Slip 4 fish for the years 2000 ($P < 0.005$) and 2001 ($P < 0.0001$; Table 3; Fig. 2). For 2002, the differences were far less substantial ($P = 0.12$), which may have been due to larger fish that were able to cross the waterway. The highest tPCB concentrations for Kellogg Island fish occurred in the largest fish collected, which may be the result of an increased ability to cross the waterway from the east side. Excluding all fish with tPCB concentrations <15 ng/g (these were considered background levels), the correlation between fish weight and tPCBs for Kellogg Island fish (all years) was highly significant ($P < 0.001$) with an $r^2 = 0.50$ ($n = 59$). There was no such correlation

Table 3 Total PCB concentrations in juvenile salmon collected in the Duwamish River and upstream

	Soos Creek hatchery	Soos Creek wild	Kellogg Island hatch	Kellogg Island wild	Slip 4 hatch	Slip 4 wild
Whole body						
24–31 May 2000	15 (1.1) 5i	7.8 (0.8) 14ci	40 (4) 3c	30 (1.3) 17 ci	203 (80) 7ci	131 (159) 2i
25 June 2001	50 (2.4) 7i	–	24 1i	94 (56) 4i	185 (59) 3i	376 (60) 12i
1 August 2001	–	–	94 (47) 6i	37 (7) 35 ci	177 (34) 4i	302 (195) 5i
7–8 August 2002	10 (0.1) 2i	–	445 1i	302 (151) 7i	725 (375) 2i M	495 (78) 2c ¥
29 July 2004	–	–	130 (0) 3c	180 1c	–	–

Stomach contents	Soos hatchery	Soos Creek wild	Kellogg Island mix	Slip 4 mix	Difference
2000	–	23 1c	57 (21) 3c	247 (30) 3c	4.3
2001	–	–	182 (138) 2c	445 (360) 2c	2.4
2002	12 Ø	–	260 (-) 1c	760 (14) 2c ¥	2.9

Values are mean and standard deviation (SD) ng/g. Following SD denotes *n* observations per mean value; "i" means individuals and "c" means composite values (ci indicates a combination of composite and individual values). Whole-body composite samples contained 3–10 individuals. M is mix for origin and mostly hatchery fish. Stomach contents were removed from these fish and used for separate analysis as composite samples containing 5–30 individuals. Date shows when in-river fish collected. Soos Creek fish (wild and hatchery) collected 18 May to 1 June, except for 2002 (8 August). Chinook in all samples except for ¥, which was two composite samples ($n = 3$ and 4 individuals) of juvenile coho and one comp for stomach contents (770 ng/g). Ø hatchery food. All values as wet weight, except fish food as dry wt (wet wt. equivalent for fish food $\approx$ 2.7 ng/g)

when all fish from Slip 4 were considered ($P = 0.42$, $r^2 = 0.02$, $n = 36$). Additionally, any whole-body tPCB value over 400 ng/g in fish from Kellogg Island was determined to be a statistical outlier ($P < 0.05$) in Grubbs test, which supports the contention that larger fish (>15 g) collected at Kellogg Island did not accumulate most of their PCBs from the west side of the LDW.

Concentrations of tPCBs in stomach contents of juvenile chinook collected at Kellogg Island and Slip 4 were substantially elevated compared to stomach contents in upriver wild fish and hatchery food (Table 3). These values also show site and year differences that are consistent with those for whole-body tPCBs. An analysis of the ratio for tPCBs in whole-body juvenile chinook and stomach contents (wet weights) for site/year combinations were relatively consistent with a mean (SD) of 0.77 (0.40) $n = 12$.

For the 2001 hatchery fish, we had sufficient data to estimate a likely growth rate. Five fish were sampled from the hatchery (mean (SD) 2.5 (0.1) g) on 7 June 2001 and compared to hatchery fish collected 54 days later at Kellogg Island and Slip 4 in the LDW. The mean weight (SD) for those fish was 13.7 (4.6) g $n = 10$. Based on a simple growth equation the mean growth rate was determined to be 3.2% bw/day (range = 2.6–4.4% bw/day). Fish were released from the hatchery between 18 May and 11 June 2001, therefore these values represent the maximum growth rate. If we assumed that all of the fish collected were from the earliest date (18 May) the mean growth rate would be 2.4%; however, these fish would have been smaller at the time of release.

For each individual fish and composite sample we determined the amount of tPCB that was accumulated in the LDW, which is presented as a percentage increase in total body burden (Fig. 3). This plot shows the general trend of higher bioaccumulation for Slip 4 fish and compared to Kellogg Island fish. All fish exhibited a positive increase in the total amount of PCBs and most increases were substantial. For example, the median increase in total ng of PCBs for all juvenile chinook collected in this study was 11-fold, which is equivalent to a 1,000% increase.

The ΣTEQ values (PCBs only) for all salmonid samples were low exhibiting a mean (SD) of 0.012 (0.024) ng/g lipid. The relationship between tPCBs and ΣTEQs in juvenile salmonids was very strong ($r^2 = 0.90$, $n = 110$) indicating that the concentration of tPCBs is a good predictor for the toxic potential from the dioxin-like congeners (Fig. 4).

Lipids

Percent lipid content for whole-body juvenile chinook based on wet weight was similar for the years 2001–2004 but higher for the year 2000 (Table 2), which is consistent with the usual pattern of smoltification whereby fish lose lipid content as they transition to seawater (Brett 1995). The mean and SEM was 1.0% (0.1) for 16 individual and composite chinook samples collected over 2001–2004.

BSAFs

The *P*-values ($n = 6$) for all possible pair combinations for the year 2000 BSAFs from the PLSD multiple comparison test were high ($P > 0.57$) indicating no difference between regions or fish origin for this year (Table 2). The majority

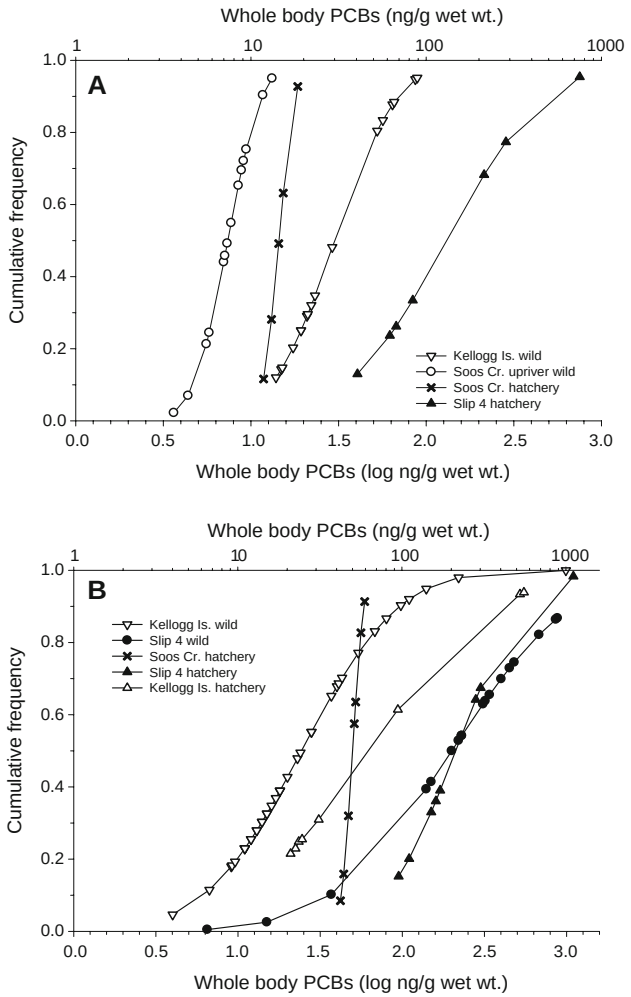

Fig. 2 Cumulative frequency distribution for total PCBs in juvenile chinook. Log_{10} concentrations are plotted. Upper *x*-axis show arithmetic equivalents. Location and origin (hatchery or wild) shown. **a** Data for the year 2000. **b** Data for the year 2001

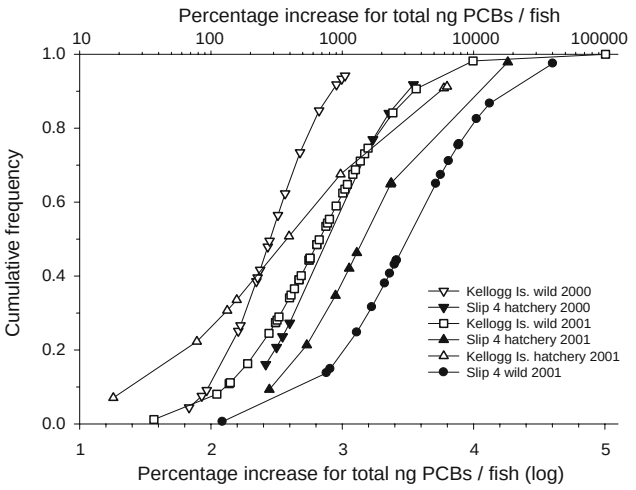

Fig. 3 Increase in total polychlorinated biphenyls (PCBs) in juvenile chinook. Cumulative frequency plot shows the percent increase in total nanograms of PCB per fish for the years 2000 and 2001. Data are based on individual fish or mean values for composite samples and plotted as log_{10} values. Arithmetic values shown on top *x*-axis. Location, fish origin (wild or hatchery), and year of collection indicated in legend

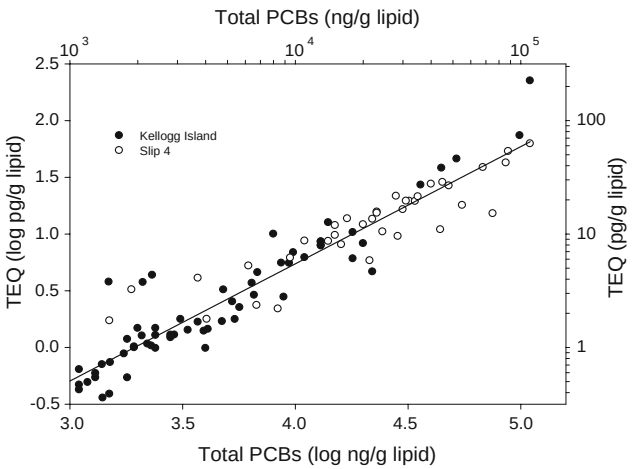

Fig. 4 Regression of total PCBs and PCB TEQs. Values are log_{10} total PCB concentrations in whole body juvenile chinook salmon and the sum of toxic equivalent quotients (TEQs) for the dioxin like PCBs. Arithmetic equivalents shown on the upper *x*-axis and right *y*-axis. The equation is $\Sigma TEQ = 3.39 + 1.03*tPCBs$, all concentrations as log_{10} ng/g or pg/g lipid

(74%) of all pairwise comparisons between year 2000 BSAFs and all other years were significantly different ($n = 26$). Fish collected for the years 2001–2004 were collected later in the summer, which provided potentially more time for bioaccumulation and higher BSAFs. Almost all comparisons among 2001–2004 BSAFs returned high *P*-values ($P > 0.1$), except for one low value for Kellogg Island wild fish for 2001.

Sediment guideline

We calculated the 50th, 90th, and 95th percentile sediment concentration associated with its respective BSAF for a given region for the years 2001–2004 (Table 4). These were calculated for all outmigrating juvenile salmon, except those from the year 2000 because of the short time spent in the lower Duwamish. If the year 2000 samples were included, the percentile values for the BSAFs would change slightly (e.g., 90th percentile, Kellogg Island = 1.4 and Slip 4 = 2.2) from the values presented in Table 4.

Discussion

PCBs in tissue

The variability in tPCB concentration in outmigrating juvenile chinook was high over time and space; however, a

Table 4 Proposed sediment values to protect against adverse effects in the Lower Duwamish Waterway

Qtile	Fish (tPCBs)		BSAF	Sediment guideline (tPCBs)	
	ng/g	µg/g lipid		µg/g OC	Sed ng/g
Kellogg Island/west side ($n = 58$)					
50th	39	3.9	0.36	7.1	106
90th	205	20.6	1.9	1.3	20
95th	331	33.1	3.0	0.83	13
Mean (sd)	88 (19)	–	0.81 (0.18)	–	–
Slip 4/east side ($n = 26$)					
50th	237	20.8	0.72	3.4	55
90th	789	90.7	2.4	1.0	16
95th	1111	138	3.4	0.70	12
Mean (sd)	360 (75)	–	1.1 (0.2)	–	–

Mean, SD, and various quantile values (Qtiles) determined with equation for lognormal distribution in Gilbert (1987). All fish for a given region over years (except 2000) were combined (years 2001–2004). Equation 2 used to determine sed_{oc} guideline values using BSAF and tissue guideline (2.4 µg/g lipid) for salmonids from Meador et al. (2002). Mean whole-body juvenile chinook lipid was 1% wet weight and 50th percentile for organic carbon (OC) for each side was 1.6% dry wt

few distinct patterns were detected. These data show that fish on the more contaminated east side of the LDW accumulated far higher amounts of tPCBs than those collected on the west side. Even though some benthic areas on the west side of the LDW contain high concentrations of tPCBs, it appears that the overall average concentration for the different sides is the more important metric for determining bioaccumulation in this mobile species. Based on these observations we conclude that the outmigrating fish probably follow the shallow areas of one side of the waterway or the other and are not likely to cross the channel until later in the summer when they achieve a larger size. One study (Ruggerone et al. 2006) sampled the mid channel area of the LDW from December through February 2005 with a purse seine and found no young-of-the-year chinook ($\sim$1.5 g individuals) in this habitat.

The concentrations of tPCBs in fish collected in the year 2000 were on average lower (two to tenfold) than for fish sampled in other years. This lower tPCB trend was not apparent for the year 2000 Slip 4 hatchery fish, which was due to due one individual fish out of 15 that comprise the mean. Without that one value, the mean drops 38% (from 203 to 125 ng/g). These lower values for the year 2000 fish may have been due to the relatively short time for exposure due to recent releases from the main hatchery, increased competition for prey items, or a change in the composition of their prey. The low tPCB concentrations in hatchery fish for the year 2000 may have been caused by the limited time these fish were in the LDW; however, this does not explain the lower values for wild fish, which may have been in the system longer. A plausible explanation for these differences is the expected high degree of competition for prey items among all fish during peak migration of the hatchery fish, which is supported by the lower concentrations for stomach contents for the year 2000 fish. The large release of hatchery fish and subsequent potential competitive interactions among these fish in the Duwamish for scarce resources has been proposed by Nelson et al. (2004) and Ruggerone and Jeanes (2004). This peak in abundance is relatively short-lived because most of the hatchery fish spend little time in this estuary (Nelson et al. 2004).

The low values for 2001 (August) Kellogg Island wild fish were considered atypical due to a number of large fish with near background concentrations. Based on this observation it appears that some juvenile salmon may reside upriver for extended periods before migrating into the contaminated lower estuary. This was observed by Nelson et al. (2004) for both wild and hatchery fish collected at rkm 21 in late June. Interestingly, the percentage of wild fish with low tPCBs (<25 ng/g) for both sampling dates (June and August) at Kellogg Island was far higher (58%) than what we observed at Slip 4 (13%), indicating that these newly arrived fish likely migrated down the west side of the waterway or spent very little time in the LDW before collection at Kellogg Island.

Wild fish are present in the Duwamish as early as January (Ruggerone et al. 2006; Nelson et al. 2004) and show two peaks in abundance, late February/early March for the fry migrants and late May for the fingerlings (Nelson et al. 2004). Based on these data, it is possible that wild chinook may spend several weeks in contaminated areas of the Duwamish accumulating PCBs. As discussed by Thorpe (1994), residence time in an estuary for juvenile chinook is variable and generally a function of season, fish size, and type of estuary; however, 30–90 days is not unusual.

All juvenile chinook increased their total PCB load as they outmigrated through the Lower Duwamish Waterway. As tPCB concentrations increased, fish also increased in

mass, which resulted in very high percentage increases in total PCB burden. Juvenile chinook in an estuary are capable of growing at rates of 3–5% body weight/day (Brett 1995; Healey 1991), which is consistent with our observed growth rates of ~3.2% bw/day for the 2001 fish and one study conducted in the LDW (Cordell et al. 2006). This very high rate of growth is due to a feeding rate of 12–20% body weight per day (Brett 1995), which is an important factor because these fish are likely accumulating contaminants at a high rate as a consequence of their high ingestion rate. The rate of prey consumption is an important kinetic parameter for any food web or bioaccumulation model.

One interesting observation is the percentage occurrence of wild versus hatchery fish in our collections. For the year 2000 the percentage of wild fish was 38%, which was most likely related to the recent releases of hatchery fish into the system. For the succeeding years, the percent occurrence of wild fish was far higher averaging 62%, including 1 year (2001) that averaged 83% wild fish. Studies have shown that hatchery reared fish will spend less time in the estuary than naturally reared fish (Levings et al. 1986), which is apparent from these data. This observation is important because we are more concerned with impacts to wild fish, including chinook salmon, under the Endangered Species Act than fish of hatchery origin. Due to the higher percentage of wild fish during the summer months and the higher levels of bioaccumulation observed for these fish compared to those earlier in the spring, the main focus should be on this group of fish that have spent several weeks in the estuary accumulating high levels of toxic compounds.

It is difficult to predict habitat usage by highly mobile, outmigrating juvenile chinook; however, we expected that a large percentage of fish would stay close to shore because of the generally higher abundance of prey and protection from predators. We believe that the higher tissue concentrations and relatively similar BSAFs for fish from the east versus west side of the waterway support this assumption of segregation within this system and indicate the need to consider appropriate geographic scales for bioaccumulation assessment for this (or any) fish species.

We found a very high correlation ($r^2 = 0.90$) between total PCBs and PCB TEQ values that could be used for predictions of toxicity. A few fish were elevated (PCB TEQ > 0.05 ng/g lipid); however, most were below the mean 95th percentile species protection benchmark for lethal effects (0.39 ng/g lipid) proposed by Steevens et al. (2005). When other dioxin-like compounds are considered, chinook at this life stage, and other species in the LDW, may exhibit TEQ values that are high enough to elicit toxic responses. It is known that dioxin-like compounds can impair the immune system, inhibit growth, cause thymic atrophy, and act as endocrine disruptors (Giesy and Kannan 1998), each an important function for estuarine fish.

BSAFs

As expected, the BSAFs for the year 2000 were generally lower because fish were collected in the spring, which is likely due to a short time period for accumulation, type of prey items available, or competition leading to reduced dietary uptake. For the other years, some of the juvenile chinook samples exhibited BSAF values that were surprisingly high. Based on their growth rate, juvenile chinook likely have a high rate of dietary accumulation and therefore would accumulate high tissue concentrations relatively rapidly. It is possible for these fish to exhibit high levels of accumulation and relatively high BSAFs after several days to a few weeks in the LDW. Additionally, salmonids have a high rate of ventilation, therefore uptake from the water column via the gills could be an important pathway for contaminant accumulation (Meador et al. 2008). The relative similarity for chinook BSAFs between the two regions for a given year (Tables 2, 4) and the high *P*-values between matched Kellogg Island and Slip 4 samples indicates that our selection of sediment concentrations for the BSAF calculations was appropriate for this species. This is also supported by the data in Table 4. If we had selected the sediment concentration at the collection sites, the tPCB tissue concentrations should have be tenfold higher in fish from the east side of the LDW compared to those from the west side. Additionally, using those Sed_{oc} values (8.9 and 88 μg/g OC) would have produced highly skewed BSAF values. Given the expected similar rates of ingestion and ventilation for these fish, plus a similar time frame for exposure, the BSAF values between the two sides of the LDW were expected to be similar.

Our intent was not to use BSAFs as an indicator of steady-state bioaccumulation or the theoretical bioaccumulation potential, but to allow for interconversion between tissue and sediment concentrations with the lowest achievable variance. The mean and various quantiles for the chinook BSAFs for both regions were relatively similar and varied by less than a factor of two, which was considerably less than the variability observed for whole-body tPCBs. We believe that many of these fish are far from steady state and that the rates of uptake (dietary and ventilatory) are the main factors controlling the levels of whole-body PCBs. For bioaccumulation, organismal lipid content is an important factor only for individuals at steady state and for chemicals that are not metabolized. While the numerator of the BSAF equation (lipid-normalized tissue concentrations) may not be an accurate indicator of bioaccumulation for fish in this study, we do consider the denominator (sed_{oc}) to be a reasonable indicator of the

bioavailable fraction from all sources available for uptake, which is primarily water and prey.

Determining a sediment guideline based on bioaccumulation

The determination of sediment concentrations that may result in adverse tissue concentrations can be accomplished with BSAF values (Meador 2006). For example, Meador et al. (2002) proposed that a tissue concentration of 2.4 μg tPCBs/g lipid was a protective tissue quality guideline (TQG) for salmonids. This TQG describes the 10th percentile of a variety of adverse biological responses for non-embryonic salmonids (fry to adult) that was compiled from several research studies. Using the BSAF (Eq. 2) and the TQG, we can solve for a sediment concentrations that should be protective against adverse effects. By examining the distribution of BSAF values observed in this study, we were able to determine sediment concentrations that could be used to protect a given percentage of the individuals. The values we provide in Table 4 would allow regulators to select appropriate percentile values that would be used to protect a given percentage of the population of outmigrant chinook salmon. For example, if the 90th percentile BSAF value was selected for chinook in the LDW, the sediment value to protect fish from bioaccumulating an adverse tissue concentration (≥2.4 μg/g lipid) would be 1.0 μg/g OC. The vast majority of juvenile chinook are from hatcheries and these fish move quickly through this estuary; however, it is the naturally reared juvenile chinook salmon that can spend considerable time in this system and likely accumulate high concentrations of PCBs and other contaminants that justifies this high percentage value.

The data we present here are just one example describing this application. Of course, several factors affect bioaccumulation and the BSAF, such as variable uptake and elimination rates, reduced bioavailability, reduced exposure, and insufficient time for sediment-water partitioning or tissue steady state. Because of these differences in bioaccumulation, a BSAF that is specific for a given estuary and species is recommended for a more accurate representation of bioaccumulation as a function of the above factors. Lipid content is also an important factor. Even though organismal lipid likely had little effect on the magnitude of bioaccumulation of PCBs for these fish (e.g., Stow et al. 1996), we believe that tissue lipids will be a factor in determining the toxic response. As proposed elsewhere (Lassiter and Hallam 1990), the lipid content of tissue controls the proportional availability of accumulated hydrophobic toxicants and therefore the magnitude of the toxic response, which is a factor we considered when developing the tPCB TQG for salmonids (Meador et al. 2002).

It is clear from these data that bioaccumulation of PCBs for a given area and time is highly variable. This is strong support for the importance of extensively sampling a given area at various locations and times to adequately characterize bioaccumulation, especially when considering population responses. These recommendations for other small estuaries include sampling in several locations, taking multiple samples over a species' potential residence time, and using a probabilistic approach for characterizing tissue concentrations that may lead to adverse effects. Obviously, a few composite samples from one or two randomly selected locations at one time period would severely underestimate the bioaccumulation potential for juvenile salmon as they rear in an estuary to accumulate mass and lipid stores before their first winter in open water. Additionally, these data indicate the importance of reducing sediment concentrations to effect reduced tissue concentrations to levels that are expected to be safe for fish and their prey. Assessing bioaccumulation in an iterative fashion after multiple rounds of sediment cleanup will provide needed information that remediation efforts are effective.

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ERRATUM

Erratum to: Bioaccumulation of polychlorinated biphenyls in juvenile chinook salmon (*Oncorhynchus tshawytscha*) outmigrating through a contaminated urban estuary: dynamics and application

James P. Meador · Gina M. Ylitalo · Frank C. Sommers · Daryle T. Boyd

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Erratum to: Ecotoxicology (2010) 19:141–152, DOI 10.1007/s10646-009-0399-x

In the original publication, the equation in the last sentence of Fig. 4 caption was published incorrectly. The correct version of the caption is given below.

Regression of total PCBs and PCB TEQs. Values are $\log_{10}$ total PCB concentrations in whole body juvenile chinook salmon and the sum of toxic equivalent quotients (TEQs) for the dioxin like PCBs. Arithmetic equivalents shown on the upper *x*-axis and right *y*-axis. The equation is $\Sigma TEQ = -3.39 + 1.03 * tPCBs$, all concentrations as $\log_{10}$ values (ng/g lipid or pg/g lipid).

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Draft Environmental Impact Statement on Two Joint State and Tribal Resource Management Plans for Puget Sound Salmon and Steelhead Hatchery Programs

July 2014

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Puget Sound Hatcheries Draft EIS

Appendix K

Chemicals Used in Hatchery Operations

Table of Contents

Hatchery operations routinely use a variety of chemicals to maintain a clean environment for the production of disease-free fish. These chemicals and safe handling requirements for the chemicals are described in this appendix. A brief description of commonly used chemicals in hatchery facilities and operations is provided below. In addition, a literature review is provided describing the potential for toxic contaminants in salmon and steelhead. This appendix provides information in support of Environmental Impact Statement (EIS) Subsection 3.7, Human Health, and EIS Subsection 4.7, Human Health.

1.0 Commonly Used Hatchery Chemicals

Common chemicals used in hatchery operations are disinfectants, therapeutics, anesthetics, pesticides and herbicides, and feed additives.

1.1 Disinfectants

Disinfectants are primarily used to clean equipment throughout hatchery facilities and may also be used to treat fish diseases. Hatchery facility workers are typically exposed to these chemicals through skin contact or inhalation during cleaning activities. However, Federal and state occupational safety and health programs (e.g., Occupational Safety and Health Act [OSHA], Washington State Industrial Safety and Health Act [WISHA]) ensure safe workplaces and require personal protective equipment and procedures (e.g., gloves, use of proper ventilation procedures, and/or respiratory protection in enclosed spaces). Following directions on product labels and using other hatchery-specific safety measures reduces chemical exposure to safe levels. Some common disinfectants used in hatchery operations are described below.

- **Chlorine (sodium hypochlorite).** Sodium hypochlorite is used for cleaning tanks and equipment and is the active component in chlorine. This compound may also be used to destroy fish fry that are infected with a disease.
- **Chloramine T.** Chloramine T is used for disinfecting tanks and equipment, and the treatment of bacterial gill diseases in salmon and steelhead. The active component is chlorine.
- **Formalin.** Formalin is a saturated aqueous solution of formaldehyde. It is used as a general disinfectant and is effective against fungal or parasitic infections.
- **Hydrogen peroxide.** Hydrogen peroxide is used as a general disinfectant and is effective against fish parasites (e.g., sea lice).
- **Iodophor.** Iodophor is a form of stabilized iodine employed as a general disinfectant. It is used to disinfect fish eggs and is effective against some bacteria and viruses.

- **Quaternary ammonium compounds (Hyamine).** Ammonium compounds or topical disinfectants are used to remove parasites from fish and have detergent and antibacterial properties.

1.2 Therapeutics

Therapeutics, which include antibiotics, are chemicals or veterinary medicines designed to be effective against parasitic, bacterial, or viral infections in fish. The most commonly used therapeutics in salmon and steelhead hatchery operations are:

- **Amoxicillin.** Amoxicillin is generally used as a veterinary antibiotic.
- **Erythromycin.** Erythromycin is generally used as a veterinary antibiotic.
- **Florfenicol.** Florfenicol is generally used as a veterinary antibiotic.
- **Oxytetracycline (terramycin).** Terramycin is widely used as an antibiotic. Oxytetracycline may be applied orally in fish feed or as a bath and is effective against a wide range of bacteria.
- **Potassium permanganate.** Potassium permanganate is primarily used as a bath treatment for fungal infections of finfish. It may also be used to alleviate acute oxygen shortage and to remove organic contaminants in fish ponds.
- **Penicillin.** Penicillin is generally used as a veterinary antibiotic.
- **ROMET®.** ROMET® is typically applied in fish feed and used to control a variety of bacterial infections.
- **Sulfamethazole trimethoprim.** Sulfamethazole trimethoprim is generally used as a veterinary antibiotic.
- **Vaccines.** Vaccines are generally used to treat viral diseases. There are a variety of vaccines available to treat animals in aquaculture. Salmon may be given vaccines to treat furunculosis, vibriosis, or yersiniosis. These vaccines are generally not considered a potential risk for human health since viral diseases of fish are typically not pathogenic to humans (World Health Organization [WHO] 1999), and the potential for exposure is minimal. The primary exposure pathway tends to be through accidental needle-stick injury (Douglas 1995; Leira and Baalsrud 1997).

Therapeutics typically are only applied when fish health specialists have determined that a disease is present in fish rearing in hatcheries. Human exposure to these chemicals typically would occur through skin contact by hatchery workers during application of the compound or through accidental needle pricks

during vaccinations. However, Federal and state occupational safety regulations (e.g., Occupational Safety and Health Act of 1970 [29 United States Code [USC] 651 et seq.]) are in place to prevent these types of accidents.

Outside of the use of therapeutic chemicals in the workplace, there are two primary environmental concerns with the use of therapeutics in hatchery facility operations:

1. Therapeutic substances are not 100 percent absorbed by the fish and may be excreted into the holding water (Texas Agricultural Extension Service 1994; Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection [GESAMP] 1997; Milewski 2001). Government agencies typically do not regulate disposal of chemicals in fish waste products; therefore, there is a potential for these chemicals to enter the environment surrounding the hatcheries (Texas Agricultural Extension Service 1994; GESAMP 1997; Milewski 2001). Federal Clean Water Act and state surface water regulations prevent the discharge of chemicals at concentrations that may pose a threat to human health. However, water quality regulations currently do not exist for all veterinary products, medicines, or their by-products when incompletely metabolized. The environmental persistence of therapeutic substances varies, and some may degrade in a few hours to a few months (GESAMP 1997). Antibiotics used at hatcheries have been detected in receiving waters downstream of aquaculture operations (Boxall et al. 2004; Pouliquen et al. 2009; Martinez-Bueno et al. 2009). Moreover, studies suggest these compounds may persist in sediments (Pouliquen et al. 2009; Martinez-Bueno et al. 2009).

   Therapeutics are typically applied infrequently and at low doses (GESAMP 1997). The use of therapeutics is governed by the Federal Drug Administration (FDA) through the Animal Medicinal Drug Use Clarification Act of 1994 (21 Code of Federal Regulations [CFR] 530), which does not permit extra-label use of drugs that are administered through feed (MacMillan et al. 2006). Currently, the volume of therapeutics released from hatcheries and the potential risks associated with these releases are unknown. Concentrations that have been reported in receiving waters near fish farms and hatcheries in other parts of the United States and in Europe are usually well below those toxic to fish and invertebrates (Boxall et al. 2004). It is expected that limited use of veterinary medicines following label instructions in U.S. fish hatcheries poses minimal risk to human health and the environment (GESAMP 1997; MacMillan et al. 2006), although locally high concentrations could occur depending on the nature of the receiving environment.

2. The use of antibiotics may increase the potential for the development of resistance in certain strains of bacteria (Burka et al. 1997; GESAMP 1997; WHO 1999). Therefore, overuse of antibiotics could render them ineffective for control of some bacteria. Resistant bacteria that infect fish have the potential to transfer resistant genetic material to bacteria that infect non-fish organisms (e.g., humans). Genetic bacterial resistance may occur by the movement of plasmids (i.e., genetic elements independent of the chromosome) between bacteria. This type of transfer has been demonstrated in a number of microorganisms (Burka et al. 1997; GESAMP 1997; WHO 1999; Cabello 2006). Therefore, the improper use of antibacterial antibiotics may cause resistance in bacterial pathogens that can infect humans (Burka et al. 1997; GESAMP 1997; WHO 1999; Cabello 2006). The use of therapeutics is governed by the FDA through the Animal Medicinal Drug Use Clarification Act of 1994 (21 CFR 530), which does not permit therapeutics for uses not specified in the drug's label (MacMillan et al. 2006). Adhering to this regulation and drug label recommendations minimizes the potential for the development of antibiotic resistance.

1.3 Anesthetics

Anesthetics are commonly used to immobilize fish during egg or milt collection, to calm fish during transportation, or during treatment with other therapeutics. They are typically applied or used at low concentrations and, thus, represent a low risk to human health (GESAMP 1997) when handled using general safety precautions (i.e., Federal OSHA or state WISHA regulations) and following label requirements. Some common anesthetics used in hatchery operations are:

- **Benzocaine.** Benzocaine is used during egg or milt stripping or during preparation for transport.
- **Tricaine methanesulfonate (MS-222).** MS-222 is used as a general sedative and applied as a bath in the holding tanks.

1.4 Pesticides and Herbicides

A wide variety of aquatic pesticides and herbicides is used in hatchery facility operations to protect fish from parasites and remove nuisance organisms, weeds, or algae. Due to their toxicity, a number of these chemicals are not approved for use in the United States. For hatcheries, pesticides and herbicides are typically highly toxic and are used in small concentrations to control algae growth or aquatic weed growth. Commonly used algaecides approved for use in the United States may contain various forms of copper. Some common aquatic herbicides include dichlobenil, diquat, endothall, fluridone, glyphosate, 2,4-dichlorophenoxyacetic acid, and 2-butoxyethyl ester. These products may be hazardous to human health if prolonged or accidental exposure (i.e., inhalation, ingestion, or dermal contact) occurs because

these compounds may be toxic at certain concentrations. Some of these products have bacteria as the active ingredient (e.g., Microbe Lift and Liquid Live Micro-organism) rather than a chemical ingredient to reduce the growth of pests. These products are typically less toxic to human health than synthetic chemicals. Safety measures on the product label and the material safety data sheet (MSDS) provide directions for proper use and applications. These safety measures, along with Federal OSHA and state WISHA regulations, serve to limit human exposure to potentially hazardous concentrations.

1.5 Feed Additives

While in hatchery facilities, hatchery-origin fish are fed with commercial diets containing fish oil and fish meal that can be from sources anywhere in the world. These feeds are known sources of toxic contaminants (Jacobs et al. 2002a; Carlson and Hites 2005; Maule et al. 2007; Johnson et al. 2010). The potential risk to human health from these contaminants is discussed further in Subsection 3.7.2, Toxic Contaminants in Hatchery-origin Fish.

Hatcheries may also use fish food that is supplemented with a variety of dietary additives. Fish raised and released from hatcheries are only fed (including dietary additives) while they are juveniles, which differs from fish raised in aquaculture farms that consume feeds and additives throughout their life. These additives may consist of artificial or natural pigments, fish oils, and/or vitamins. For example, astaxanthin and canthaxanthin are carotenoids commonly used in aquaculture to artificially color the flesh of salmon during the later stages of growth, since farm-raised fish tend to be less colorful than hatchery- or natural-origin fish. Vitamin C and Vitamin E are widely used to enhance the disease resistance of fish stocks. Exposure to feed additives from hatchery-origin fish is considered to be of low risk to human health because the concentrations used in hatcheries are typically below levels that would result in adverse health effects (GESAMP 1997).

1.6 Miscellaneous Chemicals

A variety of other chemicals are typically used at salmon and steelhead hatcheries. These chemicals are considered nonhazardous and, when used within the product label requirements and following OSHA regulations, are not expected to pose a risk to human health.

- **Anhydrous (3thyl) alcohol.** Ethyl alcohol is one of two chemicals used in a solution used to check the fertilization of eggs.
- **Lime (Type S).** Lime is widely used to neutralize acidity and increase total alkalinity of grow-out ponds.

- **Salt (sodium chloride).** Salt can be used to remove parasites or prevent stress during transport of fish.
- **Sodium thiosulfate.** Sodium thiosulfate is used to neutralize chlorine and iodophor prior to discharging wastewater.

2.0 Toxic Contaminants in Hatchery-origin Fish

Seafood consumption by humans is generally promoted due to the nutritional value of fish products. For example, fish contain elevated levels of omega-3 fatty acids, which are considered beneficial to the cardiovascular system (Mayo Clinic 2014). However, concerns have been raised that farm-raised and hatchery-origin fish may contain toxic contaminants (WHO 1999; Easton et al. 2002; Jacobs et al. 2002a; Jacobs et al. 2002b; Hites et al. 2004) that pose a health risk to consumers. Sources of contaminants in fish include chemicals or therapeutics, contamination of the nutritional supplements or feeds, and/or contamination of the environment where the fish are reared or released (Easton et al. 2002; Jacobs et al. 2002a; Jacobs et al. 2002b; Hites et al. 2004; Carlson and Hites 2005; Johnson et al. 2007; Maule et al. 2007; Kelly et al. 2008; Johnson et al. 2010). The contaminants of primary concern are those that are persistent in the environment and are known to accumulate in the tissues of fish (e.g., methylmercury, dioxins, dichlorodiphenyltrichloroethane [DDT] and its metabolites, or polychlorinated biphenyls [PCBs]) (Easton et al. 2002; Jacobs et al. 2002a; Jacobs et al. 2002b; Hites et al. 2004; Johnson et al. 2007; Maule et al. 2007; Kelly et al. 2008; Johnson et al. 2010).

Commercial diets fed to farm-raised and to hatchery-origin fish are known sources of toxic contaminants. Contaminant concentrations (e.g., pesticides, PCBs) measured in farm-raised fish are higher than in natural-origin fish (Hites et al. 2004; Hamilton et al. 2005), and the use of commercial feed in hatchery facilities may also contribute to higher concentrations of organic pollutants in hatchery-reared fish compared to their natural-origin counterparts (Johnson et al. 2007).

Hites et al. (2004) found that farm-raised salmon contained substantially more chemical pollutants than fish caught in the wild. This study suggested that these pollutants were originating from fish pellets that contain the dried and compressed body parts and toxicants from several whole fish, which they compared to a natural-origin salmon that eats a few bites of a single fish. In recent studies completed by Johnson et al. (2007), high concentrations of both PCBs and DDTs, comparable to those observed in farmed salmon, were found in juvenile hatchery-origin Chinook salmon. The authors attributed this effect in part to high body fat levels in hatchery-reared juveniles, which facilitates the uptake of lipid soluble contaminants, but concluded that there was too little information on contaminant concentrations in different lots of feed and in fish from different hatcheries, and concentrations were potentially too variable to determine how fish

feed affects contaminant levels in hatchery-origin fish. The authors stated that more comprehensive sampling of fish and feed from hatcheries would be needed to determine the extent of the problem in the Pacific Northwest (which includes the project area). In a more recent study (Johnson et al. 2010), subyearling Chinook salmon were sampled from eight hatcheries that release juvenile salmon into the Columbia River. Concentrations of PCBs and DDTs were found to be lower than those reported from previous studies (i.e., in Johnson et al. 2007), and were generally comparable to levels observed in juvenile salmon from minimally contaminated rural estuaries. Contaminant concentrations were higher in the earlier study, in part, because the fish sampled were older and larger than those sampled in the more recent study, but the differences could also be related to differences in contaminant concentrations in feed or in the hatchery environment.

Various investigations have examined the amount of organic contaminants in commercial fish feeds, and found elevated levels of PCBs, polycyclic aromatic hydrocarbons [PAHs], and pesticides (Easton et al. 2002; Jacobs et al. 2002a; Jacobs et al. 2002b; Hites et al. 2004; Neergaard 2004; Carlson and Hites 2005). In a study of contaminants in fish feeds used at National Fish Hatcheries, Maule et al. (2007) found contaminants present, although generally at lower concentrations than those reported by the investigators cited above. The U.S. Geological Survey (USGS) and U.S. Fish and Wildlife Service (USFWS) have continued studying contaminants in feeds and fish (USGS 2012) at several Federal hatcheries in the USFWS Pacific Region to 1) evaluate and compare overall contaminant levels, 2) identify temporal differences in contaminant levels found in various feed forms, 3) evaluate contaminant levels and bioaccumulation rates of different commercial diets in various life-stage history classes, 4) assess the re-distribution of contaminants during smoltification, and 5) simulate the release of fish from a hatchery by fasting fish and monitoring the mobilization and re-distribution of contaminants.

Another potential source of contaminants for hatchery-origin fish includes construction materials found within hatcheries. For example, PCBs identified in fish from the Leavenworth National Fish Hatchery in the Columbia River basin were found to be related to the paint lining fish tanks (Cornwall 2005). Some hatchery facilities in Puget Sound were constructed in the early to mid-1900s and may contain chemicals in historical building materials (e.g., paint) that are banned in current materials. Other sampling for toxic substances is ongoing at national fish hatcheries (Cornwall 2005), and the U.S. Environmental Protection Agency's (EPA's) National Pollutant Discharge Elimination System (NPDES) general permit for Federal and tribal facilities requires hatcheries to include information on painted and caulked surfaces that regularly contact process water when they apply for general permit coverage (EPA 2009). While the potential for exposure of hatchery-raised fish to contaminants in building materials exists, further incidents have not been reported.

While hatchery-origin fish may contain chemicals of concern, the risks to humans from consumption of contaminants in hatchery-origin fish remain uncertain. The potential for human exposure to contaminants in fish is directly tied to the frequency of consuming fish (EPA 1999). Thus, consumer groups that eat large amounts of fish may have a higher potential for exposure to contaminants. Current information on consumption patterns suggests that some groups of people may consume greater quantities of fish than the general population (often termed subsistence consumers) (EPA 1999; ODEQ 2008; Ecology 2013). However, information is not available to determine what proportion of the diet of subsistence consumers comes from hatchery-origin or farm-raised fish. In addition, not all the contaminants in hatchery-origin fish are derived from hatchery facilities and their operation.

Migrating and rearing salmon and steelhead encounter and accumulate additional contaminants in the rivers, estuaries, and oceans that they inhabit (Missildine et al. 2005; Johnson et al. 2007). It is unknown what proportion of contaminants present in hatchery-origin fish originates from hatcheries and what proportion originates after release. It is also unknown whether those contaminant levels pose a risk to human health. Johnson et al. (2010) suggested that the greatest accumulation of contaminants in the bodies of hatchery-origin juvenile salmon that feed and rear in urban areas occurs after the fish are released from hatcheries. In contrast, for juvenile hatchery-origin fish that are released into relatively uncontaminated rural areas, hatcheries can be a primary source of contaminants. Contaminants accumulated during hatchery rearing would probably contribute very little to concentrations of contaminants in returning adult salmon, since concentrations acquired only during the relatively short juvenile rearing period would be diluted as the fish grew larger to adulthood. Studies suggest that, for returning adult salmon, most of the contaminants present in their bodies are acquired during their time at sea (Kelly et al. 2007; Cullon et al. 2009; O'Neill and West 2009). An exception would be resident Chinook salmon that rear in Puget Sound (about 4 percent of Chinook salmon releases), and may carry a heavier load of contaminants than other salmon that spend more time at sea. Outside of resident Chinook salmon, there is no available information that demonstrates hatchery-origin fish have a greater proportion of contaminates than natural-origin fish, and thus, it is assumed that hatchery-origin salmon and steelhead do not present a greater threat of contamination than natural-origin salmon and steelhead. The Washington Department of Fish and Wildlife currently monitors toxic contaminants in fish and other organisms, as a member of the Puget Sound Ecosystem Monitoring Program.

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The Role of Hatcheries in North American Wild Salmon Production

Key Points

✔ A significant share of the salmon caught by North American commercial fishermen are released from hatcheries. In recent years, hatchery fish have accounted for about 38 percent of total Alaska "wild" salmon catches, including about 40 percent of Alaska pink salmon catches and 69 percent of Alaska chum salmon catches. Most Alaska hatchery production is concentrated in Southeast Alaska and Prince William Sound. The importance is highlighted by ADF&G: "The ocean ranching program provides hundreds of Alaskans with seasonal jobs. It is now considered the largest agricultural industry in Alaska" (Farrington 2004 p. 2).

✔ The Alaska hatchery program faces significant economic and political challenges, including:

- Lower economic net return due to lower prices
- Declining state financial support for hatcheries
- Declining direct benefits to fishermen from hatcheries as the share of catches needed to cover costs of hatchery operations increases
- Opposition from fishermen dependent on natural wild salmon catches who argue that large-scale hatchery catches has depressed ex-vessel prices they receive
- Lack of markets for "dark" hatchery fish (fish that have physiologically changed as they move back to fresh water) in some years, leading to discarding of fish carcasses after extraction of salmon roe
- Concerns about potential adverse effects of hatchery releases on Alaska natural wild salmon runs.

✔ There are also significant hatchery programs in British Columbia, the U.S. Pacific Northwest and California, which account for significant shares of the commercial and recreational fisheries.

✔ Hatcheries add another dimension of complexity and ambiguity to the discussion over environmental, economic and social issues related to wild and farmed salmon. Some of the environmental and economic issues associated with salmon farming are also associated with commercial hatchery production.

Introduction[1]

It is common to think of salmon as either "wild" or "farmed." However, not all "wild" salmon are equally wild. A large share of the salmon returning to North American streams, and a large share of the salmon caught by North American commercial fishermen, are released from hatcheries and are considered 'ranched' salmon. However, most discussion is framed in a 'wild' salmon context which includes both 'natural wild' and 'ranched.'

In some ways, hatchery salmon are more like farmed salmon than natural wild salmon.[2]

- Like farmed salmon, hatchery salmon spend the first part of their lives in hatchery incubation systems and/or rearing containers, eating similar kinds of feeds.
- Like those farmed salmon which escape into the natural environment, hatchery salmon may potentially affect the genetic diversity of natural wild salmon stocks. This is particularly a concern in Washington, Oregon and California.
- Like farmed salmon, hatchery salmon compete in world markets with natural wild salmon.
- Like farmed salmon, there are significant costs in producing hatchery salmon, and the extent to which hatcheries are economically viable depends upon market conditions.

[1] A good deal of the discussion of this chapter, in particular the portion on Alaska's hatchery programs, is drawn from Knapp (1999).

[2] See footnote 2 in Chapter II.

Unlike farmed salmon, hatchery fish compete with natural wild fish for food. For these reasons, hatcheries add another dimension of complexity and ambiguity to the discussion over environmental, economic and social issues related to wild and farmed salmon.

Once thought of as a way to restore and enhance natural wild salmon runs, hatchery salmon are now recognized as potentially harmful to natural wild salmon runs because of genetic interactions and competition for food and habitat in freshwater and marine environments. There is an active debate among scientists, commercial fishermen and the public as to the appropriate role and scale of salmon hatcheries. This is particularly true in the U.S. Pacific Northwest.

In this chapter we review the role of hatchery salmon in North American commercial wild salmon fisheries, and the economic issues associated with hatchery salmon.

Overview of North American Hatchery Programs

Salmon hatcheries have been established in North America for many purposes including:

- Introducing salmon fisheries where none previously existed.
- Replacing or enhancing natural salmon runs which were extinct or diminished.
- Increasing abundance of salmon for sports fisheries
- Increasing abundance of salmon for commercial fisheries.

Hatcheries were first established in North America in the second half of the nineteenth century, motivated by the recognition that natural stocks of salmonids were in decline and the desire to introduce salmon and trout outside their native ranges (Thorpe 1980). The first hatchery propagation of Pacific salmon (*Oncorhynchus* spp.) took place in Canada in 1857 (Bardach et al. 1972). Soon after, salmon hatchery techniques were adopted in the United States. The first U.S. hatchery was opened in 1864 in New York State to raise brook trout (Calabi 1990). However, hatchery-based enhancement programs were introduced at a significant scale only after the 1950s. Hatcheries were introduced to Japan in 1877.

More than two billion Pacific salmon were released in 2000 by North American salmon hatcheries (Table IV-1). Alaska accounted for 69 percent of total releases, while Canada and the U.S. Pacific Northwest each accounted for about 16 percent (Table IV-2).

Alaska releases were mostly pink and chum salmon, western Canadian releases (mostly British Columbia) were mostly sockeye, chum and chinook salmon and U.S. Pacific Northwest releases were mostly chinook and coho salmon. Alaska accounted for the largest share of pink and chum salmon releases; Canada accounted for the largest share of sockeye releases, and the U.S. Pacific Northwest accounted for the largest share of chinook and coho releases (Table IV-2).

Table IV-1 **Salmon Fry Releases by Species, Region, and Area, 2000 (millions of fish)**

		Chinook	Sockeye	Coho	Pink	Chum	Total
Alaska		9.2	59.8	19.3	879.7	507.7	1479.7
Canada	Yukon	0.2	0.0	0.0	0.0	0.0	0.2
	Queen Charlotte	0.2	0.0	1.3	0.0	22.2	23.7
	North Coast	4.3	90.5	1.6	0.2	12.7	109.3
	West Coast Vancouver Island	17.5	0.0	2.7	0.0	31.8	51.9
	South Coast	29.2	39.3	14.8	16.9	30.6	130.7
	Interior B.C.	2.2	19.2	0.7	0.0	0.0	22.1
	Canada Total	53.5	148.9	21.1	17.0	97.3	337.9
Pacific Northwest	Washington	117.4	16.9	43.9	1.6	38.8	229.5
	Oregon	32.3	0.0	8.7	0.0	0.0	46.8
	California	43.8	0.0	0.6	0.0	0.0	46.8
	Idaho	6.8	0.1	0.5	0.0	0.0	15.3
	Pacific Northwest Total	200.3	17.0	53.7	1.6	38.8	338.4
TOTAL		**263.0**	**225.7**	**94.2**	**898.4**	**643.8**	**2156.0**

Source: North Pacific Anadromous Fish Commission, NPAFC Hatchery Release Data.

Note: Includes all juvenile salmon releases.

Table IV-2	Share of Salmon Fry Releases, by Region and Species, 2000

	Chinook	Sockeye	Coho	Pink	Chum	Total
Alaska	3%	26%	21%	98%	79%	69%
Canada	20%	66%	22%	2%	15%	16%
Pacific Northwest	76%	8%	57%	0%	6%	16%
Total	**100%**	**100%**	**100%**	**100%**	**100%**	**100%**

Source: NPAFC Hatchery Release Data

Table IV-3	Number of Fry Released per Kilogram of Commercial Catches, 1997-2001

	Chinook	Sockeye	Coho	Pink	Chum	Total
Alaska	2.3	0.7	1.5	5.5	6.3	4.3
Canada	53.4	18.3	115.6	2.3	15.3	14.2
Pacific Northwest	52.1	15.8	42.1	1.8	16.2	34.9

Note: Calculated by dividing average fry releases for the period 1997-2001 (thousands of fish) by average commercial catches for the period 1997-2001 (thousands of kilograms). For the Pacific Northwest, average fry releases for the period 1997-2000 were used becauses 2001 data were not available.

Table IV-3 provides a general indicator of the relative scale of hatchery releases in comparison to commercial harvests. For chinook, sockeye and coho salmon, hatchery releases per kilogram of commercial catches were much higher in Canada and the U.S. Pacific Northwest than in Alaska—suggesting that commercial fisheries for these species are relatively more dependent on hatcheries in Canada and the U.S. Pacific Northwest than Alaska. For pink and chum salmon, hatchery releases per kilogram of commercial catches were much more comparable among the three regions.

The Hatchery Process

The production of salmon in hatcheries recreates the early portion of the life cycle of the species in a protected environment (Willoughby 1999). Salmon hatcheries consist of both a freshwater and a marine phase. The freshwater phase encompasses the spawning cycle, egg production, hatching and first-feeding stages. As the fry develop, they turn into fingerlings (or parr as the Europeans tend to call them), and finally grow to become smolts. At this point the fish have become physiologically adapted to seawater conditions.

- Broodstock management: Broodstock are the fish from which the eggs and milt (sperm) are taken. Selection of the broodstock from adults returning to the hatchery has changed significantly over time. Until recently, little concern was given to such things as managing to maintain the genetic integrity of a river's native salmon. In recent years, scientists have determined that these needs must be addressed and have prescribed methods to choose broodstock in a more careful manner (National Research Council 1996).
- The hatchery: The hatchery phase is probably the most technically demanding, requiring a high degree of organization and planning. The objective of this portion of the cycle is to fertilize and hatch the eggs then raise the fry until release to open water. After hatching, the young fish feed on the contents of their yolk sac for several weeks and are called yolk-sac fry or alevins. A short time after hatching the yolk sac has been almost totally consumed and the alevins are generally developed enough to start feeding. Starter diets formulated with feed ingredients, such as fishmeals and fish oils, give rapid growth.
- Fry and fingerling development: When the alevins begin to feed they are known as fry. During this phase, growth is rapid. As they develop, fry become more accustomed to solid feed and increase their activity. When the fry are sufficiently developed, they are transferred into larger tanks. Once the fry reach an average weight of about 5 g, they are known as *fingerlings*.
- Smolt production: Once the larger fingerlings are sufficiently developed, they will undergo major physical and physiological changes to become smolts. These changes mark the transformation from a freshwater fingerling to a seawater fish (Fitzgerald et al. 2002). The smoltification process involves changes in most organ systems, morphological (silvery color), physiological (ATPase activity) and behavioral (swimming with the current), which will allow the fish to survive, grow and develop normally in the marine environment.

Hatcheries managed for stock enhancement of the commercial and sport fisheries, generally release fish to

the open water at either the fry, fingerling or smolt stage depending on species and management objectives. Pink and chum salmon are generally released at the fry stage with a large number of fry released. In British Columbia, the U.S. Pacific Northwest, where the purpose of the hatcheries are generally to ensure the survival of the stock, species such as chinook, coho and sockeye are released as smolts to increase the probability of survival in the wild.

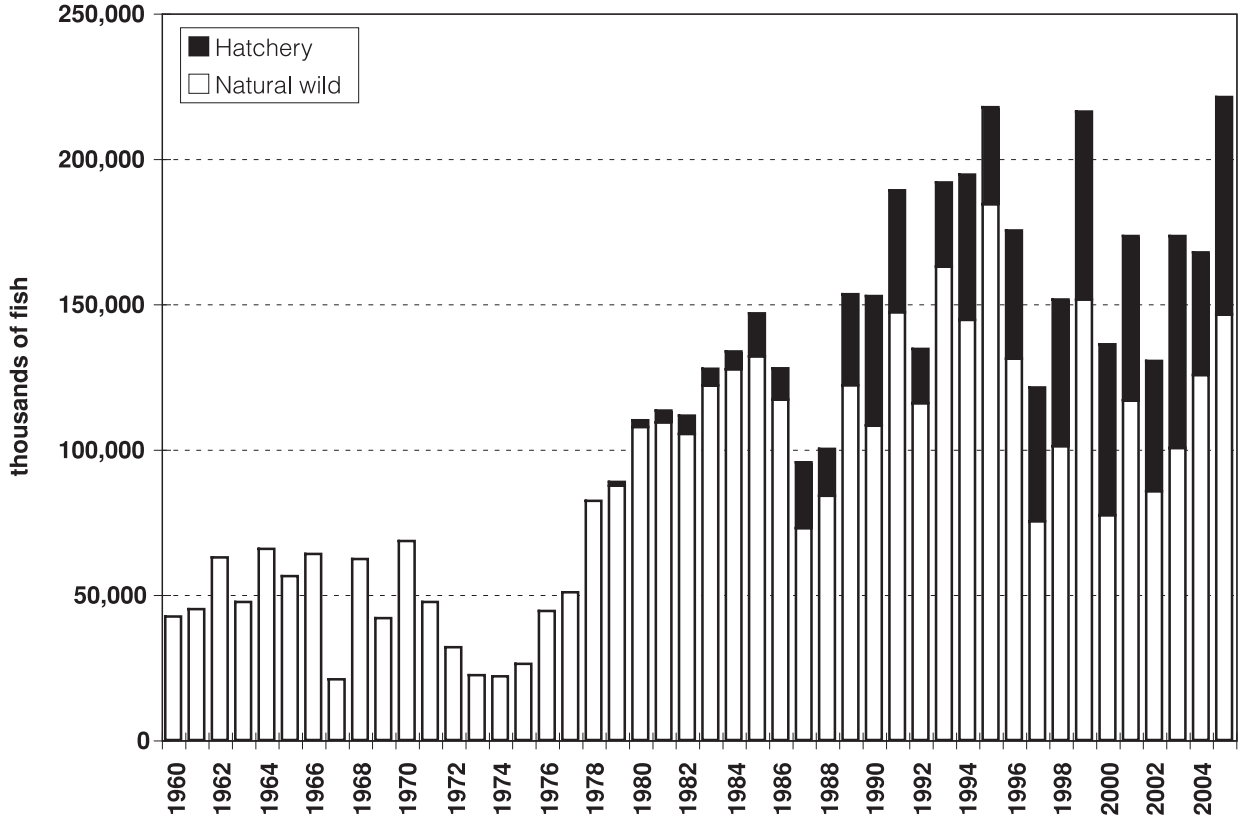
The Alaska Salmon Enhancement Program

Beginning in the 1970s, the State of Alaska supported the development of numerous salmon hatcheries, with the goal of increasing and stabilizing Alaska salmon returns.[3] State support of the Alaska salmon enhancement program was linked to the rapid rise in Alaska oil revenues following the discovery and development of oil on Alaska's North Slope. The State supported hatchery development by loaning money to private non-profit organizations for hatchery construction and operation, as well as by building and operating State-owned hatcheries which were later transferred to private non-profit regional aquaculture associations.

Beginning in the 1980s catches of both hatchery salmon and natural wild salmon increased rapidly. In 2002, the total catch of hatchery fish was 45 million salmon, about one-third of the total Alaska salmon catch (Figure IV-1).[4]

The relative importance of hatcheries varies between different Alaska salmon species. During the period 2000-2002, hatchery fish accounted for 69 percent of Alaska chum salmon catches, 40 percent of pink salmon catches and 12 percent of catches of other species (Table IV-2). Hatchery fish accounted for about

Figure IV-1 **Alaska Commercial Salmon Catches Since 1960: Natural Wild Salmon and Hatchery Salmon**



Source: Data for 1960-1978: ADFG Catch Data 1878-1981; Data for 1979-2005: ADFG Hatchery Data

[3] The Alaska Department of Fish and Game's annual reports on the Alaska Salmon Enhancement Program, available at www.cf.adfg.state.ak.us/geninfo/enhance/enhance.php, provide detailed information about the program.

[4] Hatchery fish are identified in several ways, including coded wire tags, fin clips and otolith marking (a process by which an identifiable microscopic colored ring sequence in fish ear bones is created by exposing fish to a series of planned temperature changes).

28 percent of the total ex-vessel value of Alaska catches. The importance is highlighted by ADF&G: "The ocean ranching program provides hundreds of Alaskans with seasonal jobs. It is now considered the largest agricultural industry in Alaska" (Farrington, C., ADF&G,. 2004 p. 2).

The relative importance of hatcheries also varies between different areas of Alaska. In 2002, Southeast Alaska and Prince William Sound accounted for about 80 percent of hatchery catches (Table IV-4).

Certain Alaska fisheries are overwhelmingly dependent on hatchery salmon, including the Southeast Alaska chum salmon fishery, the Prince William Sound chum salmon fishery and the Prince William Sound pink salmon fishery. In other major fisheries, such as western Alaska sockeye salmon fisheries and the southeast Alaska pink salmon fishery, hatchery fish account for only a small share of total catches. Note that the two highest value species, chinook and sockeye, are less dependent on hatcheries. Part of the explanation is the health of the natural sockeye stocks in Alaska, and the relatively high cost and time it takes to raise chinook smolts.

Although hatcheries have clearly increased Alaska salmon catches, they have not stabilized catches. Salmon catches by region and in the state as a whole still vary greatly from year to year, even with hatchery programs, because hatchery fish are subject to the same ocean conditions as wild salmon. This is illustrated in Figure IV-2. During the period 1990-2005, Alaska hatchery releases of pink salmon were relatively stable, ranging between 800 million and 1 billion fish. During the same period, returns of Alaska hatchery pink salmon ranged from 15 million to 69 million fish. The percentage of fish returning varied from 1.7 percent to 7.2 percent.

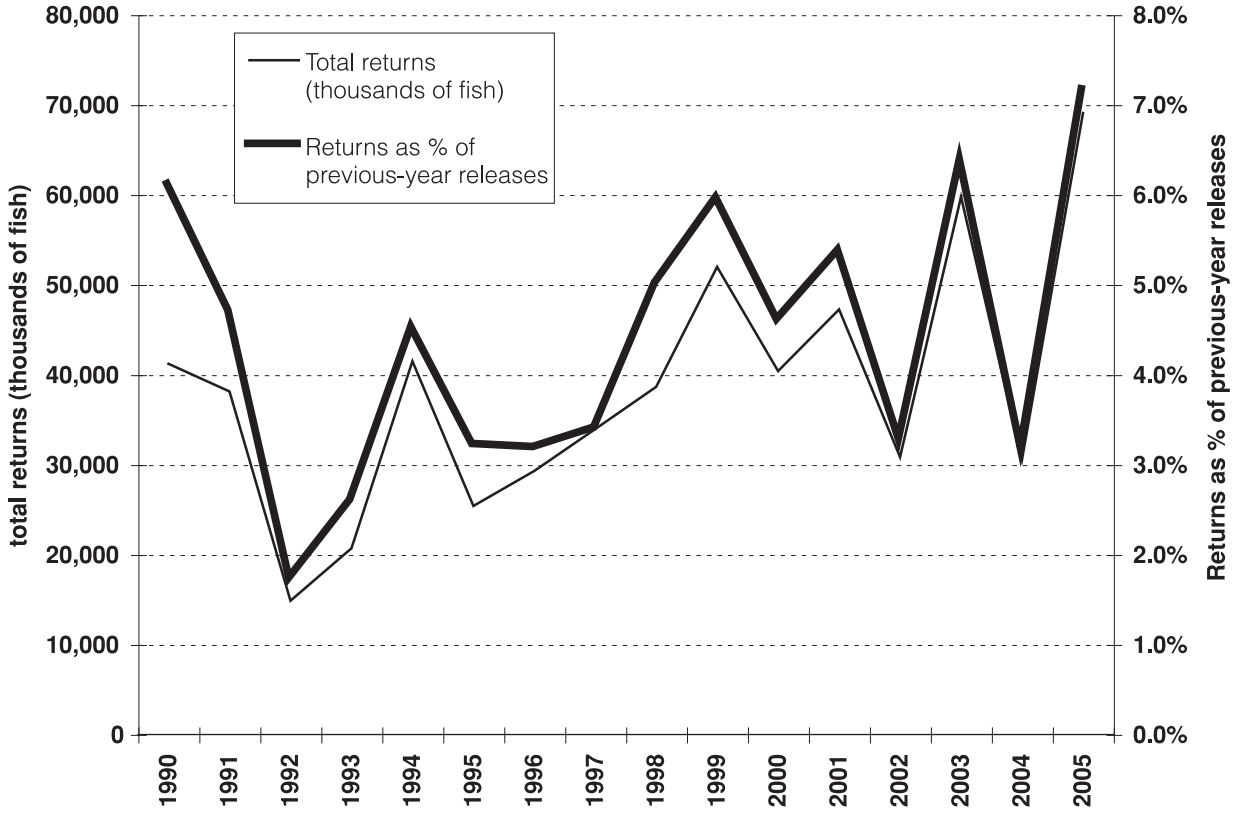
Large numbers of hatchery fish are caught by commercial fishermen prior to their return to the hatcheries. Near hatchery sites, boats hired by the hatchery catch additional large numbers of fish in the so-called 'cost recovery' fishery. All the proceeds from this fishery go to the hatchery. Any remaining hatchery fish are left to mill around the hatchery and die. They are not 'programmed' with a stream in mind to return to. Although some may stray may find a stream and spawn in it, this is neither intended nor desired.

Table IV-4 **Alaska Salmon Catches by Species and Region, Hatchery & Total, 2002**

	Area	Chinook	Sockeye	Coho	Pink	Chum	Total
Commercial catches of hatchery fish (000 fish)	Southeast	87	120	1,425	1,924	5,617	9,173
	Prince William Sound	0	1,164	36	18,772	6,112	26,084
	All other areas	1	1,466	217	7,747	88	9,519
	Alaska total	**88**	**2,750**	**1,678**	**28,443**	**11,817**	**44,776**
Share of total hatchery catches, by species	Southeast	1%	1%	16%	21%	61%	100%
	Prince William Sound	0%	4%	0%	72%	23%	100%
	All other areas	0%	15%	2%	81%	1%	100%
	Alaska total	**0%**	**6%**	**4%**	**64%**	**26%**	**100%**
Share of total hatchery catches, by area	Southeast	99%	4%	85%	7%	48%	20%
	Prince William Sound	0%	42%	2%	66%	52%	58%
	All other areas	1%	53%	13%	27%	1%	21%
	Alaska total	**100%**	**100%**	**100%**	**100%**	**100%**	**100%**
Total commercial catches (000 fish)	Southeast	372	787	2,986	45,612	6,294	56,051
	Prince William Sound	40	2,262	650	18,950	6,373	28,275
	All other areas	128	19,438	1,135	23,000	2,357	46,058
	Alaska total	**540**	**22,487**	**4,771**	**87,562**	**15,024**	**130,384**
Hatchery share of commercial catches	Southeast	23%	15%	48%	4%	89%	16%
	Prince William Sound	0%	51%	6%	99%	96%	92%
	All other areas	1%	8%	19%	34%	4%	21%
	Alaska total	**16%**	**12%**	**35%**	**32%**	**79%**	**34%**

Source: ADFG Hatchery Data.

Figure IV-2 **Estimated Returns of Alaska Hatchery Pink Salmon, 1990-2005**



Source: ADFG Hatchery Data.

Challenges for the Alaska Salmon Enhancement Program

The Alaska Salmon Enhancement Program consists of a variety of public and private sector salmon rehabilitation and enhancement projects. In 2002, these included 29 non-profit corporation hatcheries (by far the most significant component of the program), two state-operated hatcheries, two Federal or Bureau of Indian Affairs hatcheries and several streamside incubation and restoration projects (Farrington 2003).

The Alaska Salmon Enhancement Program has clearly succeeded in increasing total salmon catches, particularly in Southeast Alaska and Prince William Sound. However, the program faces a number of challenges which could affect the future scale of hatchery releases and thus total Alaska salmon catches, particularly of pink and chum salmon. Below, we briefly review these challenges.

Lower Prices

A fundamental problem for the Alaska Salmon Enhancement Program is that real (inflation-adjusted) prices have declined significantly since the start of the program, in particular for chum and pink salmon (Figure IV-3). As a result, investing in raising and releasing young salmon results in less of an increase in future catch value, for any given rate of ocean survival.

In theory, we might expect that as prices decline the net economic benefits of hatcheries would decline, and at some point total hatchery releases would begin to decline. However, this has not yet happened to any significant extent. Hatchery releases of pink and chum salmon stopped growing in the mid-1990s, but have not shown any significant decline (Figure IV-4).

In order to understand the relationship between salmon prices and hatchery releases, we must review the structure of hatchery operations and how they are financed. Most salmon hatcheries in Alaska are now operated by private non-profit (PNP) organizations, most of which received initial funding from state grants and capital and operating loans, to be repaid from hatchery revenues. There are two categories of PNP organizations: independent PNPs and regional aquaculture associations.

Hatcheries may earn revenues to cover operating expenses and repay state loans in two ways. First, hatcheries are authorized to catch a percentage of the adult salmon returning to terminal "special harvest

areas" for sale. These are referred to as "cost-recovery" catches." Typically cost-recovery fish are caught by just a few boats, catching very large volumes, working under contract to the hatcheries in the special harvest areas. All other returning hatchery salmon are caught in "common-property fisheries" by commercial, sport and subsistence fishermen.

Second, in management areas with regional aquaculture associations, fishermen may vote to assess an "enhancement tax" on the ex-vessel value of their salmon landings. These enhancement tax funds also support hatchery operations. Enhancement tax rates are presently 3 percent in southeast Alaska and 2 percent in Prince William Sound, Cook Inlet and Kodiak. No enhancement taxes are assessed in other areas.

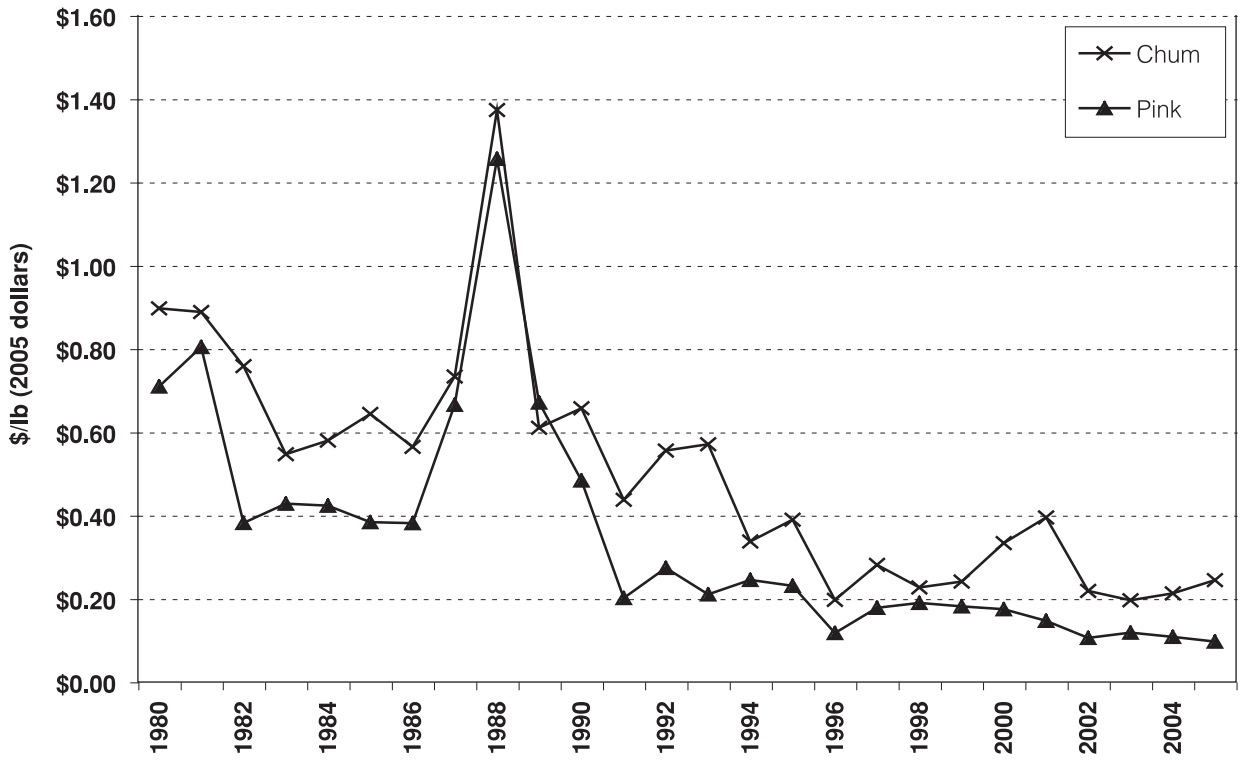
As ex-vessel prices have declined, enhancement tax collections have declined, so that the hatcheries have had to rely on cost-recovery catches for a greater share of their revenues. In addition, because prices are lower, hatcheries need to catch more fish in the cost-recovery fisheries to meet any given revenue target. As a result, as prices decline an increasing share of the hatchery returns have been caught in cost-recovery fisheries rather than by commercial fishermen in the common property fisheries. This trend is particularly evident for chum salmon, for which the cost-recovery share of catches increased from less than 30 percent in the early 1990s to more than 51 percent in 2003 (Figure IV-5).

As the cost-recovery share of hatchery catches increases, the share of the benefits captured by commercial fishermen (other than those few who participate in the cost-recovery fishing) declines. Put differently, an increasing share of the fish goes to support the hatcheries, rather than the original concept of increasing the total volume of fish available to all fishermen.

Increasing the share of hatchery fish going to cost-recovery harvests has allowed the hatcheries to continue to operate despite lower salmon prices. However, over time, this may create a political problem for the hatcheries, which depend upon enhancement taxes paid by fishermen on all catches—not just catches of hatchery fish—and which also depend upon the political support of commercial fishermen to address other issues which they face (discussed below).

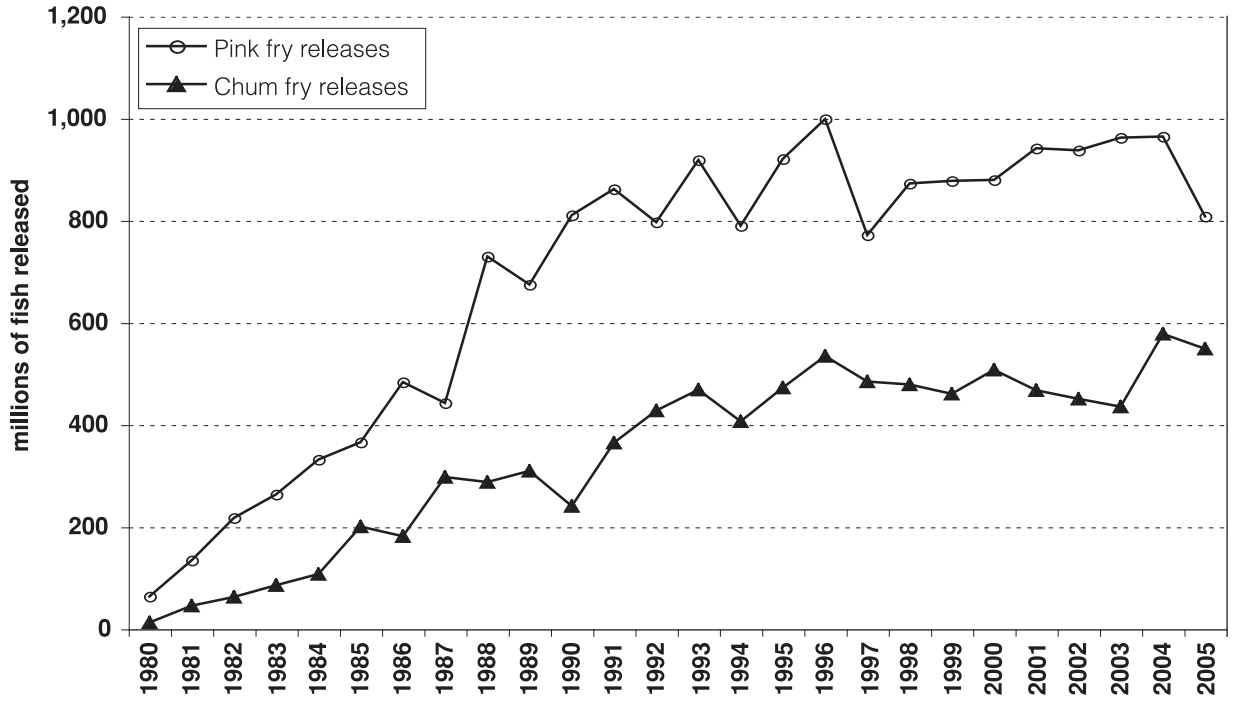
In addition to covering their operating costs, hatcheries also need to make payments on the loans they have received from the State of Alaska's Fisheries Enhancement Revolving Loan Fund. During the early 1990s, as ex-vessel prices declined, many hatcheries requested and received permission to reschedule loan repayments. As Alaska's oil revenues have declined, the State is less likely to extend this kind of assistance should hatcheries face financial difficulties in the future.

Figure IV-3 **Average Real Ex-Vessel Prices for Alaska Chum and Pink Salmon, 1980-2005**



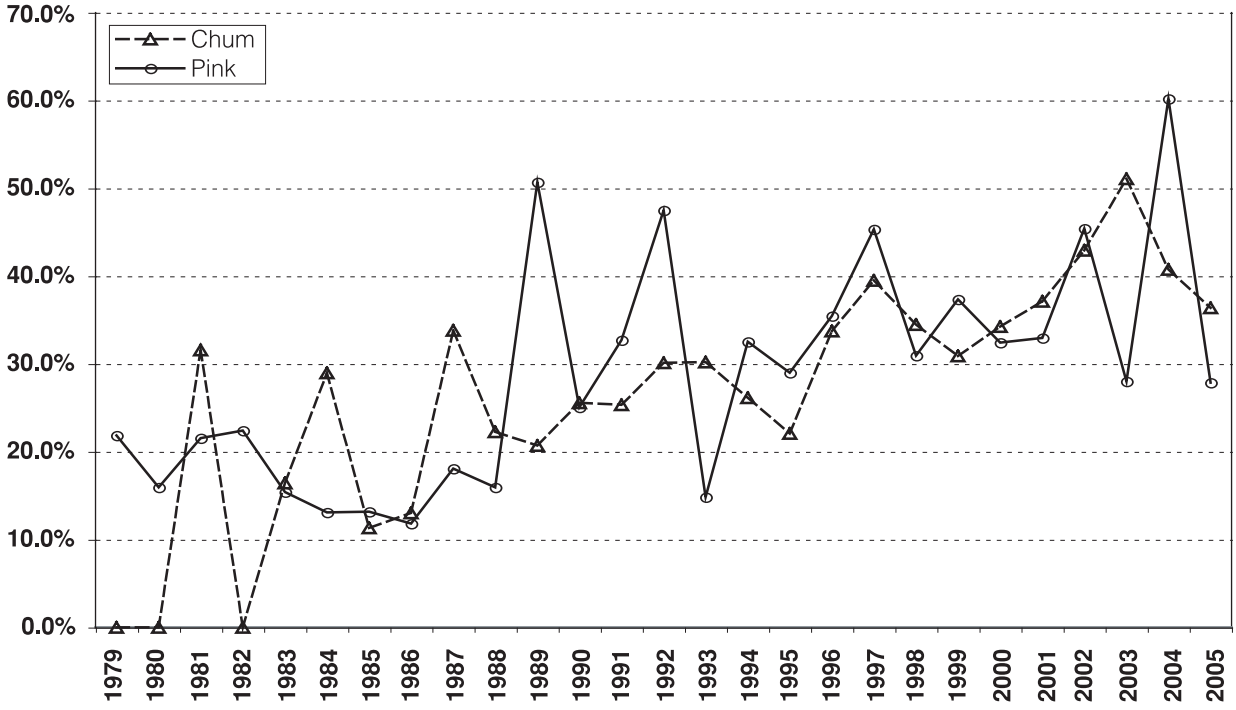
Source: ADFG Catch data. Adjusted for inflation based on Anchorage CPI.

Figure IV-4 **Alaska Hatchery Releases of Pink and Chum Salmon Fry, 1980-2005**



Source: ADFG Hatchery Data.

Figure IV-5 **Hatchery Cost-Recovery Share of Alaska Hatchery Salmon Catches**



Source: ADFG Hatchery Data.

Market Effects of Hatchery Production

As we discuss in future chapters, salmon prices are sensitive to total salmon supply.

During the 1990s, fishermen in regions of Alaska without hatchery production—in particular areas of interior and western Alaska dependent on chum salmon—argued that increased hatchery catches were responsible for the disastrous decline in prices which they had experienced. More generally, the question began to be raised whether Alaska salmon hatcheries were actually increasing the total value of Alaska salmon catches, or whether the value of the increased harvests was being offset by corresponding negative effects on prices.

How much Alaska hatchery catches may have depressed Alaska salmon prices, or whether or not hatcheries have actually increased the total ex-vessel value of Alaska salmon catches (not to mention net economic value after subtracting costs of hatchery operations) is not an easy question to answer. As we discuss in subsequent chapters, salmon markets are complex and are affected by many factors. In addition, they are subject to structural change, so that the effects of a given volume of hatchery catches on prices may have changed over time.[5]

In the short-term, higher catches in a given region in any given year tend to lower ex-vessel prices in that year. Over the longer term, prices are driven by world supply and demand rather than supply and demand from any particular region. If, as with hatchery production, other regions have the ability to respond to higher prices by increasing production, then higher or lower production by a particular region will not necessarily affect long-term world prices.

In general, it seems likely that Alaska hatchery production has had some negative effects on ex-vessel prices of chum and pink salmon, but that hatcheries are not the only factor contributing to lower prices. Clearly, hatcheries have benefited fishermen and processors in some areas (primarily Prince William Sound and Southeast Alaska) by greatly increasing catches. At the same time, hatcheries have not benefited, and may well have harmed, fishermen and processors in other areas without hatchery production. Thus, the Alaska salmon hatchery program has at times been an issue between different regions of Alaska.[6]

Roe "Stripping" or "Salvaging"

A particularly contentious issue associated with the Alaska salmon hatchery program has arisen as a result of declining prices for fresh, frozen and canned salmon while prices for salmon roe have remained strong. In some years the value of fresh, frozen and canned products have fallen below the costs of processing, particularly for lower-quality "dark" salmon caught in hatchery terminal areas after they have begun to undergo physiological changes associated with return to fresh water, and when unexpectedly large returns exceed local processing capacity. For these fish, the most economically profitable utilization is to extract the salmon roe but to dispose of the salmon carcass.

Normally, it is illegal to dispose of salmon harvested in Alaska without utilizing the fish, under a State law which bans the "waste" of commercially harvested fish. However, in some years hatcheries and processors have applied for exemptions from this law and have received permission to grind up and dispose of salmon carcasses at sea, after first removing valuable salmon roe. This practice is commonly referred to as "roe-stripping" or "roe-salvaging" depending on one's perspective on it.

This "dumping" of salmon has been strongly criticized by some segments of the Alaska salmon industry and the public who have argued that it is immoral to waste fish and that the "stripped" or "salvaged" roe competes unfairly with other roe production. Others have responded that utilizing the valuable salmon roe is better than the alternative of not harvesting the fish at all, in particular since returning hatchery fish provide no ecological benefit and large volumes of dead fish in hatchery terminal areas would pollute these areas.

One example of this issue occurred during the 2003 pink salmon season in Prince William Sound, when 49 million pink salmon were caught after a preseason harvest projection of 27 million fish. More than 4 million pink salmon (about 8 percent of the Prince William Sound pink salmon catch and about 3 percent of the total Alaska pink salmon catch) were ground and "recycled" after the eggs were removed (Tkacz 2003).

When low prices or lack of processing capacity lead to the disposal of hatchery fish after roe extraction, it usually contributes to adverse publicity for the salmon hatchery program and questioning whether the hatchery production is needed—adding to the other political issues faced by hatcheries.[7]

Effects of Hatcheries on Alaska Natural Wild Salmon

To minimize potential adverse effects of hatchery releases on natural wild runs, the State has established an extensive regional planning process for salmon enhancement and set strict conditions for egg collection, fish transport and release and management

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5 Market effects of the Alaska hatchery program were addressed by Boyce et al. (1993) and Herrmann (1993). These analyses were critiqued by Wilen (1993).

6 A different market-related issue is whether hatchery sales of cost-recovery catches may depress prices paid to local fishermen for both hatchery and natural wild fish caught in common-property fisheries.

7 In an *Anchorage Daily News* article, a Prince William Sound fisherman who is a former chairman of the Alaska Seafood Marketing Institute was quoted as commenting: "It's just disappointing. . . We've got the mother of all runs, and we can't sell all of the fish. I'm worried. I'm worried that some fishermen and legislators in other areas might think it's a mistake to be generating these pink salmon, but we're pretty grateful for them around here for all the opportunity they create. Nobody anticipated this kind of return. We should not do anything knee-jerk about occasionally having overproduction." (Loy 2003).

of enhanced stocks. Hatcheries may only use eggs collected originally from local wild salmon stocks.[8]

However, as in other areas, some critics still question whether the Alaska salmon hatchery program may adversely affect Alaska's natural wild salmon runs. One concern relates to the potential for competition for food between hatchery salmon and natural wild salmon, both for juvenile fish in near-shore waters as well as in the open ocean.

Another set of issues relate to the management of commercial fisheries in which fishermen are catching mixed stocks of hatchery and natural wild salmon. If large returns of hatchery fish are mixed with depleted runs of natural wild fish, there is the potential for over-harvests of natural wild fish runs.

Another concern relates to the "straying" of returning hatchery fish into streams with natural runs of wild salmon, with the potential for genetic change in the natural wild salmon populations. For all of these concerns, the scientific complexity of the issues, together with lack of data and research, makes it difficult to determine how serious the potential problems associated with the hatchery program may or may not be.[9]

"Wild" Image of Alaska Salmon

An issue which may grow in importance over time is the effect of Alaska's salmon hatchery program on the "wild" image of Alaska salmon fisheries. The salmon farming industry has been subject to growing criticism over alleged adverse environmental effects as well as market effects on wild salmon fisheries. As we discuss in later chapters, the argument has been made that because of these alleged adverse effects of farmed salmon, consumers should favor wild salmon over farmed salmon. Over time, some salmon farmers may respond to these criticisms by pointing out problems associated with wild salmon. One response is likely to be that not all Alaska salmon are fully "wild," and that there are environmental and market issues associated with hatchery salmon as well as farmed salmon.[10] If this caused Alaska's hatchery program to become a concern for some consumers in the future, it could possibly reduce political support within Alaska for the hatchery program.

It should be noted that Alaska chum salmon, which account for by far the largest share of United States consumption of fresh and frozen Alaska wild salmon, is also the species most dependent on the Alaska hatchery program.

The Future of the Alaska Salmon Enhancement Program

The issues discussed above are the subject of an intense and long-running political debate about the Alaska salmon hatchery program, between supporters of the program and those who argue for substantially scaling back hatchery releases. The debate is not widely understood outside of Alaska or the salmon industry.

A series of special studies and task forces and special studies have examined the issues related to hatcheries, and at various times proposals to limit hatchery production have been debated before the Board of Fisheries. In 1991, a committee of the Alaska Senate undertook a special review of fisheries enhancement in Alaska, in order to "assemble and analyze information about the program and the global context in which it operates," and to "serve as the first step in ensuring that current and future enhancement efforts will be economically and biologically sound, while fulfilling the goals for which the program was established" (Alaska State Senate 1992). In 1996, a "Hatchery Policy Group" was appointed to review and make recommendations on state-wide hatchery production policy and hatchery loan policy (Gardiner 1996). In 2002, the Alaska legislature established a Joint Legislative Salmon Industry Task Force to review issues facing the salmon industry and make recommendations to the legislature. The Task Force formed a number of subcommittees, including a 'Hatchery Subcommittee' which was charged with examining Alaska hatchery policy issues.[11]

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8 See McGee (1995) for a useful review of the planning process and state policies related to the hatchery program and protection of wild salmon.

9 These concerns were summarized in Environment and Natural Resources Institute (2001): "Alaska's ocean-ranching salmon hatcheries operate amidst considerable uncertainty. Perhaps the most striking feature uncovered by this review was the many gaps in the scientific data from which one could fairly draw conclusions of the effects hatcheries may or may not have on wild salmon. Alaska has been successful in augmenting salmon harvest with hatchery-produced fish, but whether or not salmon biodiversity has been adequately protected in the process is unanswered. . . . With respect to fish-culture practices, Alaska's hatcheries are among the best in North America. . . . Given the late date at which Alaska's ocean-ranching program was established, the state was able to benefit from mistakes made elsewhere. The program started on better footing by having genetic oversight of operations through fish transport permits, hatchery siting, egg takes, broodstock development, etc." Nevertheless, the report concluded that, as a result of mixed-stock management issues, competition for resources between hatchery and wild salmon stocks, and potential effects on genetic diversity of wild salmon populations, "industrial-scale hatchery salmon production . . . could be jeopardizing Alaska's wild salmon."

10 Dodd (2003) suggested that "the fish which the hatcheries produce for commercial fishermen undoubtedly eat sizeable quantities of prey species as they move up the feed chain towards harvest time, prey that would otherwise be available to truly 'wild' fish." Another example is provided by an article posted on the website of the Washington Fish Growers Association (www.wfga.net): "Salmon farming vs. salmon ranching is another interesting issue that likely doesn't make its way into the 'wild is good, farmed is bad' marketing campaign. In order to help maintain its commercial fishery, and enhance wild fish stocks, Alaska decided to forego the salmon farming route and do salmon ranching instead. Salmon ranching is a lot like salmon farming. Fish are raised in ocean-based pens, fed a steady diet of processed food (purchased in British Columbia, interestingly enough, and consumed at nearly six times the rate used in British Columbia fish-farm operations), fed some dyes important to their health and colour, also antibiotics. When they're big enough, they let them go. Alaska releases more than 1.5 billion "ranched" fish into the waters every year, and they happily swim away, competing for food with their natural-born cousins, and eventually get caught (along with the wild fish) in the commercial fishery. . ."

11 Information about the activities of the Task Force, including proposed legislation developed by the task force, was posted on the website of the United Fishermen of Alaska, at www.ufa-fish.org/taskforce/.

Earlier task forces and studies have not resulted in major changes to the Alaska's hatchery program policies or the scale of hatchery releases. However, the underlying political issues remain and the debate over the program continues, even expanding into new fora. With the Marine Stewardship Council's (MSC) certification of the Alaska salmon fishery as a sustainable fishery (see Chapter XVI for a more thorough discussion), one of the concerns brought up in the certification process in 2000-2001 was the hatchery program. In particular, the assessment team was concerned about the lack of research on the potential effects of salmon hatcheries on the wild stock gene pool and reproductive fitness (Scientific Certification Systems 2000). This concern remained in 2005 as the Alaska salmon fishery entered its new five-year assessment for re-certification under the MSC program.

It is possible that Alaska hatchery salmon releases and catches could decline significantly in the future due to lower economic return of hatcheries and/or changing political circumstances. It is difficult to predict whether such a decline will in fact occur or when it might occur. It could be that hatchery salmon—as opposed to natural wild salmon—would be most affected by changing economic circumstances in wild fisheries.

The British Columbia Salmonid Enhancement Program

In 1977, in response to declining British Columbia salmon runs, the Canadian federal Department of Fisheries and Oceans (DFO) launched a Salmonid Enhancement Program (SEP). The program included both the construction of hatchery facilities as well as a variety of other habitat enhancement projects such as spawning channels, incubation boxes and lake enrichment.

DFO estimates that about 10-20 percent of the British Columbia sport and commercial salmon catch originates from SEP projects, and about a dozen terminal fisheries are dependent on enhanced stocks (DFO 2000a). A terminal fishery is one that occurs at the place where the hatchery salmon were released into fresh water.

In a 2000 review of the Salmonid Enhancement Program, the Pacific Fisheries Resource Conservation Council (PFRCC 2000; DFO 2000b) concluded that:

> *In hindsight, it is difficult to say whether the Salmonid Enhancement Program and its predecessors, which have accounted for close to a half-billion dollars in public investments over the years, have produced any net return on investment, if measured by a net gain of salmon. There is evidence to suggest a net loss of wild salmon abundance, directly and indirectly because of enhancement initiatives. . .*
>
> *The Council's review of the Salmonid Enhancement Program leads inevitably to the conclusion that some facilities created by it have resulted in the displacement of wild salmon by hatchery-produced fish. This has occurred when hatchery salmon have attracted fishing effort that unavoidably produced unsustainably high rates of harvest on co-migrating wild salmon. It has also occurred because juvenile fish from wild populations have been subjected to competition from hatchery fish in rearing areas, and in the ocean phase of the salmon life cycle.*
>
> *Declines in numerous wild-salmon populations, concurrent with increases in production from a few large hatcheries, tend to create a situation in which salmon abundance is attributable to ever-fewer stocks. This places the salmon resource at an increasingly greater risk of random, catastrophic disruption.*

History of Salmon Hatcheries in the U.S. Pacific Northwest

Hatchery techniques for the artificial propagation of Pacific salmon were developed for the first time in Canada around 1857 and soon spread to the United States (Bardach et al. 1972).

The construction and operation of the first hatcheries for Pacific salmon in the United States began on the McCloud River in northern California in 1872 and in 1877 and 1878 on the Clackamas and Rogue Rivers in Oregon (Atkinson 1988). In 1883, the first Canadian hatchery for Pacific salmon was built at Bon Accord (near New Westminster, British Columbia) on the Fraser River (PCSF 2004). The first hatchery in Washington State was built on the Kalama River in 1895 (WDFW 2004). Four years later, the Washington Department of Fish and Wildlife began the construction of salmon hatcheries in the mid-Columbia River region, on the Wenatchee and Methow Rivers (Wahle and Pearson 1984).

Hatcheries were originally built to reverse the trend of declining populations of wild salmon and to compensate for land use decisions that permanently altered large areas of fish habitat (WDFG 2004). Emphasis was initially placed on chinook and coho salmon despite an incomplete understanding of the complex life history of these species. Hatcheries propagated and stocked salmon for many years without concrete evidence of the success and long-term implications of their efforts.

Large-scale construction of salmon hatcheries began in 1938, when Congress passed the Mitchell Act to provide federal money for construction of hatcheries as a way of replacing the thousands of acres of salmon spawning grounds that were blocked or flooded behind dams. Subsequently, more than 80 hatcheries were built in the Columbia River basin (Novak 1998).

Currently, the State of Washington has one of the largest artificial propagation systems in the world, with

a hatchery program that operates 24 complexes with 91 rearing facilities. Together they raise and release more than 201 million Pacific salmon, 8.5 million steelhead (salmon) trout and 22.6 million trout and warm-water fish (Maynard and Flagg 2001). Hatchery-bred fish help support the State's $850 million per year sportfishing industry (*The Wave News Network* 2004).

A group called the Hatchery Review Group unveiled a new blueprint for the State of Washington's hatchery programs on April 23, 2004 (*The Wave News Network* 2004). The blueprint cost $28 million to write and has more than 1,000 recommendations for improving the large salmon hatchery system. Examples include closing some hatcheries that are especially detrimental to wild stocks, and limiting the number of hatchery fish released so that they do not overrun wild stocks protected under the Endangered Species Act.

In addition, the state has 12 federal hatcheries and 35 tribal rearing facilities which produce another 50 million salmonids for release. In Oregon, the Department of Fish and Wildlife operates 34 hatcheries and 15 other rearing facilities, which release about 43 million Pacific salmon, 5.7 million steelhead (salmon) trout and 8.3 million trout. California has eight salmon and steelhead (salmon trout) hatcheries.

Depending on species and area, the salmon enhancement programs in the U.S. Pacific Northwest produce as much as 70 to 90% of salmon harvested in the commercial and recreational fisheries.

The potential for hatchery salmon to affect wild stocks went unrecognized for many years. Between the mid-1950s and early 1970s, scientists found increasing evidence that hatchery salmon was harming the remaining wild salmon runs. It seems clear now that hatcheries have had demographic, ecological and genetic impacts on wild salmon populations.

These effects include the reduction of genetic diversity within and between salmon populations, creation of mixed-population fisheries, altered behavior of fish, ecological imbalances due to the elimination of the nutritive contribution of carcasses of spawning salmon from streams, and the displacement of the remnants of wild runs (NRC 1996). As Hilborn (1992) notes:

> *Large-scale hatchery programs for salmonids in the Pacific Northwest have largely failed to provide the anticipated benefits; rather than benefiting the salmon populations, these programs may pose the greatest single threat to the long-term maintenance of salmonids… I argue that hatchery programs that attempt to add additional fish to existing healthy wild stocks are ill advised and highly dangerous.*

As a result, academic, environmental and salmon advocate groups have proposed a redesign of the traditional objectives of hatchery management, which needs to shift away from producing more fish for harvest towards providing a means for the recovery and conservation of wild salmon populations (LLTK 2004; NRC 1996).

It is worth noting that there have been a few attempts at private salmon ranching, such as Ore Aqua Foods, a subsidiary of Weyerhaeuser and Anadromous Inc., a subsidiary of British Petroleum, both operating in Oregon during the late 1970s and 1980s. Private salmon ranching is based on the premise that smolts released from the private hatchery will return and will be captured by the "owner" of the fish. These have been unsuccessful primarily because ocean mortality is high and uncertain, and property rights related to salmon released to the ocean are poorly defined. In addition to these problems, salmon enhancement (public or private) may undermine the management of wild stocks through direct and indirect competition.

The 2005 *Atlas of Pacific Salmon* summarized the breadth and complexity of the issues related to salmon hatcheries in the U.S. Pacific Northwest and elsewhere.

> *The benefits of hatcheries are compelling: they may offset losses in abundance in naturally spawning stocks and reduce harvest pressure on wild populations; they help stabilize commercial harvest; and they serve as laboratories for the study and preservation of biodiversity. Hatcheries also provide a solid economic base for salmon-dependent communities, including native peoples.*
>
> *Yet these benefits are counterbalanced with significant scientific uncertainty regarding freshwater and ocean carrying capacity, particularly within a trans-Pacific context . . . Interbreeding and brood stock transfer among rivers can challenge wild population viability and genetic integrity. Hatchery production can mask ecological problems at the heart of declines in wild populations. Artificial propagation can deprive rivers of marine-derived nutrients . . . essential to functioning freshwater ecosystems. Unfortunately, isolating impacts of hatchery fish on wild populations is extremely difficult, and so efforts to determine hatchery success or failure remain inconclusive.*
>
> *Two legislative debates—whether to count hatchery fish under endangered species legislation . . . and whether to allow surplus hatchery fish to spawn in the wild—have fulminated in recent years, underscoring the fact that hatchery management is among the most controversial issues in fisheries today.*

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Tribal Natural Resources Management

A report from the Treaty Indian Tribes in Western Washington
2015

06530

We, the Indians of the Pacific Northwest, recognize that our fisheries are a basic and important natural resource and of vital concern to the Indians of this state, and that the conservation of this natural resource is dependent upon effective and progressive management. We further believe that by unity of action, we can best accomplish these things, not only for the benefit of our own people, but for all of the people of the Pacific Northwest.

– PREAMBLE TO THE NWIFC CONSTITUTION

Member Tribes of the Northwest Indian Fisheries Commission

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Cover: Shawn Johnstone, left, and Skip Pickett, Quinault Indian Nation hatchery technicians, pull a coho from a net as part of the Quinault Indian Nation's hatchery operations on Lake Quinault. *Photo: Debbie Preston. Map: Ron McFarlane.*

US v. WASHINGTON 1974-2014 BOLDT 40

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Year in Review

Lorraine Loomis

As we celebrated the 40th anniversary of *U.S. v. Washington* (the Boldt decision) this year we also mourned the loss of Billy Frank Jr., our longtime chairman and good friend. Billy, 83, passed away on May 5, 2014.

From his first arrest at age 14, Billy spent his entire life fighting for the recognition of tribal rights reserved in treaties with the United States. The 1974 ruling by Judge George Boldt in *U.S. v. Washington* re-affirmed the tribal treaty right to harvest salmon and established the tribes as natural resources co-managers entitled to half of the harvestable salmon returning annually to western Washington waters.

Today tribes are leaders in the management of the region's salmon fisheries and other natural resources.

Billy Frank Jr.

I am honored and humbled to follow in Billy's footsteps as chair of the Northwest Indian Fisheries Commission. The tribes remain committed to Billy's legacy and direction to "stay the course" with salmon recovery. Our goal is to return all salmon populations to sustainable levels that can support harvest. We commit ourselves to this task with the recognition that we must act in the best interests of those who will follow us seven generations from now.

Treaty Rights at Risk

Sadly, ongoing loss and damage to salmon habitat has stalled salmon recovery and threatens tribal treaty rights. For those rights to have meaning, there must be salmon available for harvest. That is why we are continuing the Treaty Rights at Risk initiative begun in July 2011 by Billy and other tribal leaders.

Through this effort we are asking the federal government, our trustee, to align its agencies and programs and take charge of a more coordinated salmon recovery effort. We want the federal government to take charge of salmon recovery because it has the obligation and the authority to ensure both salmon recovery and protection of treaty rights.

We are disappointed with the federal government's slow response and lack of progress. There has been a lot of discussion, but little action by the federal government, in spite of its responsibility to protect tribes' treaty rights and recover salmon stocks listed as threatened under the federal Endangered Species Act (ESA).

We have requested that the Treaty Rights at Risk initiative be institutionalized in the U.S. government via President Obama's Council on Native American Affairs created several years ago. Addressing tribal natural resources concerns was supposed to be one of five main areas of work when the council was founded. Subgroups of the council already have been formed to focus on economic development, education, climate change and energy. We think a similar group should be formed to address tribal natural resources concerns, especially salmon recovery and treaty rights.

Importance of Hatcheries

Even as we struggle with the continual decline of salmon populations caused by lost and damaged habitat, hatcheries are under attack. Hatcheries were designed to make up for lost natural salmon production, and are essential to fulfilling tribal treaty rights, but federal funding has not kept pace with needed repairs and replacement of aging facilities. Tribes produce about 40 million salmon and steelhead annually.

The National Marine Fisheries Service has worsened the situation by delaying review and approval of permits required under the ESA for hatchery operations. The delays led to legal action that prevented the release last fall of nearly 1 million hatchery-raised steelhead in western Washington. Indian and non-Indian fishermen will feel the loss of those fish for years to come.

Hatcheries and the salmon they produce are absolutely necessary as long as lost and damaged habitat prevents salmon recovery. They deserve more support from all corners. Today, most of the chinook and coho harvested by Indian and non-Indian fishermen come from hatcheries.

Updated Water Quality Standards

Tribes continued their efforts to encourage the state of Washington to adopt a more realistic fish consumption rate as part of updating water quality standards. The higher the fish consumption rate, the cleaner the water must be.

For more than 20 years, the state has operated under water quality rules based on a fish consumption rate of 6.5 grams per day, or one 8-ounce serving a month. This was one of the lowest rates in the nation, even though Washington residents eat more fish and shellfish than people in other states, and most tribal members consume much more than that.

In July, Gov. Jay Inslee approved an increase to 175 grams per day, a compromise rate supported by the tribes, but still lower than the actual amount of fish and shellfish eaten by Indian people in western Washington. At the same time, Inslee increased the risk of getting cancer from water pollution from one in a million to one in 100,000. The tenfold increase in cancer risk effectively cancels out most of the benefits of the higher fish consumption rate.

As a result, the treaty tribes have approached their trustee, the U.S. Environ-

mental Protection Agency, to step in and enact new water quality rules for the state. The 1972 federal Clean Water Act requires states to implement standards that ensure waters are clean enough to support fish that are safe to eat.

Fossil Fuel Transportation

Proposals to build coal and oil export terminals in western Washington continued as major concerns in 2014. The planned increases in train and ship traffic threaten the health and safety of tribal members as well as treaty-protected rights and resources.

Coal export terminals proposed for Cherry Point near Bellingham and Longview on the Columbia River would be fed by hundreds of trains daily from coal fields in Montana and Wyoming. Coal dust from each train would be spread all along its route.

Also proposed is a plan to use mile-long crude-oil trains to feed massive new oil terminals in Grays Harbor. As with increased coal train traffic, tribes are deeply concerned about health, safety and environmental issues associated with the trains and ships transporting the oil.

Disastrous Fraser Sockeye Season

A high diversion rate of Fraser River sockeye through Johnstone Strait around the northern part of Vancouver Island led to poor catches for treaty tribal and non-tribal fishers in 2014. Nine treaty Indian tribes in western Washington harvest sockeye returning to British Columbia's Fraser River.

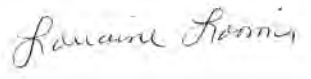
Typically, about half of the returning sockeye swim around Vancouver Island and through the Strait of Juan de Fuca where treaty tribal and non-Indian commercial fishermen can harvest them when they enter U.S. waters. By the end of August, Canadian fishermen had caught about five million fish; non-Indian commercial and treaty tribal fishermen harvested about 275,000.

Tribes will be requesting a declaration of natural disaster under the Stafford Disaster Relief and Emergency Assistance Act, and a fisheries economic disaster under the Magnuson-Stevens Act, which would provide services and financial assistance to fishermen.

Culvert Repairs Begin

As part of the favorable ruling for the tribes in the Culvert Case, talks began in 2014 to prioritize repair of culverts under state roads that are barriers to fish passage. The state was ordered by the federal court in 2013 to repair more than 600 state-owned culverts over the next 17 years. Fish-blocking culverts deny salmon access to hundreds of miles of good habitat in western Washington streams, affecting the fish in all stages of their life cycle. The treaty tribes and the U.S. filed the initial Culvert Case litigation in 2001 under *U.S. v. Washington*. The state has appealed the ruling.

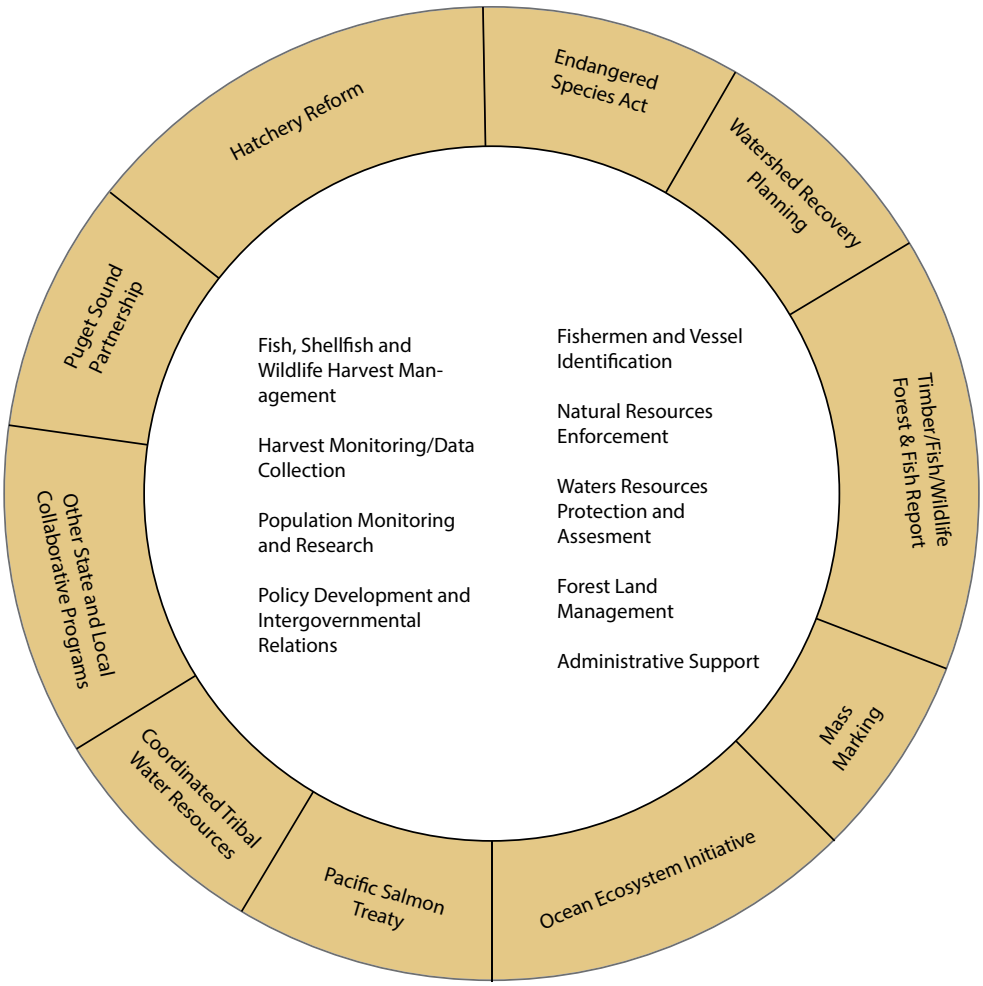
To find out more about these and other natural resources management issues important to the treaty tribes, visit the Northwest Indian Fisheries Commission website at *nwifc.org*.



Lorraine Loomis
NWIFC Chair

Tribal Natural Resources Management Core Program

Natural resources management functions and associated programs of the treaty tribes in western Washington:



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Habitat Management

Habitat protection and restoration are essential for recovery of wild salmon in western Washington. Tribes are taking action to recover salmon in each watershed.

- The tribes continue to support the Treaty Rights at Risk initiative, calling on the federal government to align its agencies and programs to better meet salmon recovery goals, particularly those for habitat protection and restoration. The initiative calls on the federal government to lead a more coordinated salmon recovery effort because it has both the obligation and authority to recover salmon and protect tribal treaty rights.

- The NWIFC Salmon and Steelhead Habitat Inventory and Assessment Program (SSHIAP) provides a "living database" of local and regional habitat conditions. SSHIAP has launched an interactive map to track repairs to state-owned culverts; a tool to map potential steelhead habitat; and a data exchange for research about the nearshore environment.

- Tribes continue to address the habitat concerns identified in the 2012 State of Our Watersheds report. The report, which documents ongoing loss and damage of salmon habitat, can be viewed at *nwifc.org/sow*. It will undergo a comprehensive update in 2015.

- Tribes conduct extensive monitoring of water quality for pollution, and ensure factors such as dissolved oxygen and temperature levels are adequate for salmon and other fish. To make limited federal funding work to its fullest, tribes partner with state agencies, industries and property owners through collaborative habitat protection, restoration and enhancement efforts.

- In western Washington, the National Oceanic and Atmospheric Administration's Pacific Coastal Salmon Recovery Fund has supported projects that have restored thousands of acres of forest, protected hundreds of acres of habitat and removed hundreds of fish passage barriers.

Lower Elwha Klallam Tribe Monitors Restored River

Tiffany Royal

With the Glines Canyon and Elwha dams completely removed, the Elwha River now flows freely.

While two massive fish-blocking dams on the Elwha River were being torn down between 2011 and 2014, the Lower Elwha Klallam Tribe was studying how the river, salmon and wildlife were responding to dam removal.

After the 108-foot-tall Elwha and 210-foot-tall Glines Canyon dams were built in the early 1900s, millions of cubic yards of sediment built up behind the structures, creating lakes Aldwell and Mills.

As the dams were deconstructed, sediment flowed downriver, changing the dynamics of the river and restoring the river mouth from cobblestone to sandy beach. Scientists have found forage fish and shellfish, such as Dungeness crab, using the new habitat.

In the estuaries, tribal staff are seining the ponds to examine fish populations and study the stomach contents of juvenile salmon.

After the Elwha Dam was completely removed by spring 2013, salmon were found spawning above the former dam site. The second dam, Glines Canyon, was completely removed by end of September 2014 and soon after, bull trout and chinook were detected beyond that dam site.

Biologists have been counting adult fish through scuba surveys and a sonar camera in the lower river. Since 2013, biologists have counted nearly 9,000 chinook and steelhead returning to the river.

The tribe's new hatchery was finished in 2010 and regularly spawns and rears coho and chum salmon. It also operates steelhead and pink broodstock programs.

Since lakes Aldwell and Mills were drained, crews from the tribe, Washington Conservation Corps and Olympic National Park have been eradicating invasive plants and replacing them with native trees, shrubs and grasses within the new open riverbeds. More than 50 engineered logjams have been installed to help slow the river's velocity and create pools and other salmon habitat.

The tribe's wildlife staff studied river otters and American dippers, looking at how the animals used the river for food and habitat and how those needs were impacted by dam removal. Post-dam removal, the wildlife staff is monitoring how elk, deer and small mammals are using the newly exposed lakebeds as habitat.

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Hatchery Management

Hatcheries must remain a central part of salmon management in western Washington as long as lost and degraded habitat prevents watersheds from naturally producing abundant, self-sustaining runs of sufficient size to meet tribal treaty fishing harvest rights.

- Treaty Indian tribes released more than 39 million salmon in 2013, including 10 million chinook, 16.5 million chum and 7.7 million coho.
- Most tribal hatcheries produce salmon for harvest by both Indian and non-Indian fishermen. Some serve as wild salmon nurseries that improve the survival of juvenile fish and increase returns of salmon that spawn naturally in our watersheds.
- Tribes conduct an extensive mass marking and coded-wire tag program. Young fish are marked by having their adipose fin clipped before release. Tiny coded-wire tags are inserted into the noses of young salmon. The tags from marked fish are recovered in fisheries, providing important information about marine survival, migration and hatchery effectiveness.

Tribal Programs Support Threatened Steelhead

Tribal hatcheries not only produce fish for harvest, but also provide a vital role in helping imperiled stocks. Some tribal facilities are the linchpin in restoring weak runs of steelhead. Puget Sound steelhead are listed as threatened under the federal Endangered Species Act.

Hatchery programs started by the Skokomish, Puyallup and Muckleshoot tribes in 2006 to rescue weak runs of steelhead have seen tremendous results.

The steelhead population in the Skokomish River has doubled since the Skokomish Tribe began its supplementation project as part of a 16-year-long project to boost the steelhead stocks in Hood Canal.

"The increase in the number of egg nests has given us an early indication that the project is working, but the long-term monitoring will be the true test of its success," said Matt Kowalski, the tribe's steelhead biologist.

The tribe spent the past eight years collecting 30,000 steelhead eggs annually from the Skokomish River. The eggs, collected between May and June, have been raised to smolts in a state hatchery. Most are released as juveniles, but 400 of the fish are transported to a federal hatchery where they are raised to 4-year-old adults before release to improve their chances of spawning in the river.

The Puyallup Tribe of Indians is continuing its successful steelhead broodstock program by releasing young steelhead from an acclimation pond in the upper White River.

"Acclimation ponds help ensure there are juvenile steelhead in the river each year to take advantage of the available habitat," said Blake Smith, the tribe's hatchery manager. The fish will be released at a pond on Huckleberry Creek, a tributary to the White River in the Puyallup watershed.

To help recover the declining run, the Muckleshoot and Puyallup tribes started the steelhead broodstock program eight years ago. Each year, the partners spawn up to 25 wild steelhead taken from an adult trap on the White River.

Up to 50,000 juvenile steelhead are produced annually at the Muckleshoot Tribe's White River hatchery. This year will mark the first release of hatchery steelhead from the acclimation ponds.

Tiffany Royal

Tribal and federal staff and volunteers collect steelhead eggs from the Skokomish River valley.

Harvest Management

Salmon

Treaty Indian tribes and the Washington Department of Fish and Wildlife co-manage salmon fisheries in Puget Sound, the Strait of Juan de Fuca and nearshore coastal waters.

- For decades, state and tribal salmon co-managers have reduced harvest in response to declining salmon runs. Tribes have cut harvest by 80-90 percent since 1985.
- Under *U.S. v. Washington* (the Boldt decision), harvest occurs only after sufficient fish are available to sustain the resource.
- The tribes monitor their harvest using the Treaty Indian Catch Monitoring Program to provide accurate, same-day catch statistics for treaty Indian fisheries. The program enables close monitoring of tribal harvest levels and allows in-season adjustments.
- Tribal and state managers work cooperatively through the Pacific Fishery Management Council and the North of Falcon process to develop fishing seasons. The co-managers also cooperate with Canadian and Alaskan fisheries managers through the U.S./Canada Pacific Salmon Treaty.

Fraser Sockeye Run Avoids U.S. Waters

Kari Neumeyer

Swinomish fisherman Landy James helps bring in Fraser sockeye during the tribes' ceremonial and subsistence fishery.

Tribal fishermen had high hopes for the 2014 Fraser River sockeye fishery, forecast to be more than 20 million fish.

The returning fish were the offspring of the record 2010 Fraser run of about 30 million fish.

Unfortunately, 96 percent of the sockeye were diverted into Canadian waters, out of reach for the nine tribes with treaty-reserved rights to harvest Fraser sockeye. The tribes are Jamestown S'Klallam, Lower Elwha Klallam, Lummi, Nooksack, Makah, Port Gamble S'Klallam, Suquamish, Swinomish and Tulalip.

This year's diversion rate was one of the highest on record. Usually, about half of the sockeye swim around Vancouver Island through the Strait of Juan de Fuca, where tribal fishermen can harvest them when they enter U.S. waters. But by the end of August, Canadian fishermen had caught about five million Fraser sockeye, while in the States, tribal and non-tribal fishermen had caught about 275,000 fish.

Tribes will be requesting a declaration of natural disaster under the Stafford Disaster Relief and Emergency Assistance Act, and a fisheries economic disaster under the Magnuson-Stevens Act, which would provide services and financial assistance to fishermen.

Before the commercial fishery opened in August, the Swinomish Tribe held a one-day ceremonial and subsistence fishery intended to harvest sockeye for both Swinomish and Tulalip tribal members to put away for the winter.

"The tribes take some of their quota to save for ceremonies, and to give to tribal members to cook or can," said Lorraine Loomis, fisheries manager of the Swinomish Tribe. "We don't have enough fish to get through the winter."

The tribes were targeting 35,000 sockeye to be caught for the nine sockeye tribes' ceremonial and subsistence use, but only 3,100 were caught.

Lummi Nation tribal members fished for Fraser sockeye with a traditional reef net in addition to their commercial purse seine and gillnet fleet.

"It's an imitation of the seafloor, like a reef," said Lummi fisherman Richard Solomon. "*Sxwole* is what our people called it."

The net is suspended from two canoes while tribal fishermen watch for salmon to swim into the simulated reef and then lift the net.

"We have to relive the path," said Lummi fisherman Troy Olsen. "Our journey back to the *sxwole*, our reef net, is in its infancy and we're just now starting."

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Shellfish

Treaty tribes harvest native littleneck, manila and geoduck clams, Pacific oysters, Dungeness crab, shrimp and other shellfish throughout the coast and Puget Sound.

- Shellfish from ceremonial and subsistence fisheries are for tribal use only, and are a necessary part of their culture and traditional diet.
- Tribal shellfish programs manage harvests with other tribes and the state through resource-sharing agreements. The tribes are exploring ways to improve management of other species, including sea cucumbers, Olympia oysters and sea urchins.
- Tribes continue to work with property owners to manage harvest on non-tribal tidelands.
- Tribal shellfish enhancement results in bigger and more consistent harvests that benefit both tribal and non-tribal diggers.
- Shellfish harvested in commercial fisheries are sold to licensed shellfish buyers. For the protection of public health, shellfish are harvested and processed according to strict state and national standards.
- In 2013, treaty tribes in western Washington commercially harvested nearly 900,000 pounds of manila and littleneck clams; more than 2.6 million pounds of geoduck clams; more than 4 million oysters; 8.5 million pounds of crab; nearly 271,000 pounds of sea cucumbers and more than 247,000 pounds of shrimp.

Jamestown S'Klallam, Partners Start Hatchery

Tiffany Royal

Hatchery technician Nicolas Rosales rinses oysters before placing them in a floating upwelling system.

The Jamestown S'Klallam Tribe is developing its own shellfish hatchery to benefit both tribal and non-tribal shellfish operations in Puget Sound.

The tribe plans to raise shellfish and grow seed to sell, said Kurt Grinnell, Jamestown S'Klallam Tribe vice-chair.

The tribe leased the former Washington Department of Fish and Wildlife shellfish hatchery in Quilcene in late 2013 and started rearing 800 Pacific oysters in March.

The tribe is working in partnership with Troutlodge, a private salmon and shellfish aquaculture company, and Jones Farm, a shellfish farm on Lopez Island. The tribe and its partners are working together because water chemistry has been an issue when sourcing seed from one location.

"We lower risk by partnering up with others," Grinnell said.

The primary focus will be growing manila clams, geoduck and oysters from seed to adult, plus the algae needed to feed everything, as well as selling seed to others.

"We want to create our own larvae and broodstock here and provide seed to others," Grinnell said. "We've had a need for something like this for a long time and to have it accessible to all the tribes. We're going to make this work, we just have a long ways to go. There is such a demand for seed and everything we grow will be sold."

Harvest Management (continued)

Marine Fish

Treaty tribes are co-managers of the marine fish resource. They work closely with the state of Washington, federal agencies and in international forums to develop and implement species conservation plans for all groundfish stocks in Puget Sound and along the Pacific coast.

- The Pacific Fishery Management Council, which includes the tribal and state co-managers, regulates the catch of black cod, rockfish and other marine fish. Halibut are managed through the International Pacific Halibut Commission, established by the United States and Canada governments. Tribes are active participants in season-setting processes and the technical groups that serve those bodies.
- The state of Washington, Hoh Indian Tribe, Makah Tribe, Quileute Tribe and the Quinault Indian Nation are working with the National Oceanic and Atmospheric Administration to develop research goals that look at changing ocean conditions and managing ocean resources.
- The tribes and state support ocean monitoring and research leading to ecosystem-based management of fishery resources. In 2013, the Quinault Indian Nation developed a nearshore ocean-monitoring system that uses sensors in crab pots to gather water quality information.

Halibut Hook Links Generations

Debbie Preston

Makah elder Jesse Ides examines a modern day halibut hook designed to test traditional fishing methods.

A fish hook has tied history, culture and the Makah community together in unexpected ways.

The *čibu·d* (pronounced "cha bood"), or halibut hook, became the subject of a student project during an internship with Makah Fisheries Management.

"I had a student, Larry Buzzell, come to me wanting to do a project that related to historical fishing methods," said Jonathan Scordino, marine mammal biologist for the Makah Tribe.

Historically the hooks were made of both wood and bone. As the tribe gained access to new materials, they also made hooks from metal.

"The goal of the project was to test if the *čibu·d* was more selective for catching halibut than contemporary circle hooks when fished on a longline," Scordino said.

Setting up the experiment was challenging because the study required 200 *čibu·d* to be made by hand. The Makah Cultural and Research Center opened its exhibit preparation space for several weeks to allow community members to come in and help make the hooks.

Through trial and error, a group of volunteers learned it was better to bend the metal hooks cold rather than heat the metal.

Elder Jesse Ides (*Hushta*) watched as young people learned to make the hook he used in his youth.

"It's terrific seeing them show the determination to make it and use it," Ides said.

He recalled his father hauling canoes out to the halibut grounds to fish.

"You'd catch just halibut with that gear, nothing else," he said.

"The *čibu·d* was known to not only fish selectively for halibut, but not catch too small or too big a halibut," Scordino said. "From a management perspective, that's exactly the size you want to catch so the older spawners remain and the young grow to be a harvestable size."

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Wildlife Management

The treaty Indian tribes are co-managers of wildlife resources in western Washington, which include species such as deer, elk, bear and mountain goats.

- Western Washington treaty tribal hunters account for a small portion of the total combined deer and elk harvest in the state. In the 2013-14 season, treaty tribal hunters harvested a reported 432 elk and 567 deer, while non-Indian hunters harvested a reported 7,246 elk and 27,448 deer.

- Tribal hunters do not hunt for sport, but for sustenance. Most do not hunt only for themselves. Tribal culture in western Washington is based on extended family relationships with hunters sharing game with several families. Some tribes have designated hunters who harvest wildlife for tribal elders and others unable to hunt for themselves, as well as for ceremonial purposes.

- All tribes prohibit hunting for commercial purposes.

- As a sovereign government, each treaty tribe develops its own hunting regulations and ordinances for tribal members. Tribal hunters are licensed by their tribes and must obtain tags for animals they wish to hunt.

- Many tribes conduct hunter education programs aimed at teaching tribal youth safe hunting practices and the cultural importance of wildlife to the tribe.

Tracking Deer and Elk Predators: Bobcats, Cougars

Olympic Peninsula tribes are tracking bobcats and cougars to find out whether they are the primary predators of deer and elk on the peninsula. Until now, there hasn't been much scientific evidence supporting or disproving that theory.

Several tribes are putting radio-signal transmitting collars on cougars to better understand their home ranges, diet and other behavior. The Makah Tribe is the only entity collecting similar data on bobcats.

"There really has been no research done on bobcats in Washington," said Rob McCoy, Makah wildlife division manager. The tribe has been conducting research on cougars since December 2010 and started radio-collaring bobcats in January 2012.

"We have really good data on cougars and male bobcats," McCoy said. "We're working to get more females into the study to better understand reproduction and size of litters and survival."

The tribe now has four male and four female bobcats with collars.

When a collared cat makes a kill, the radio signals show that it has stopped moving while it feeds. Biologists walk in and note the kill species.

"We're still gathering data, but right now, we just aren't seeing elk in the bobcat diet at all," McCoy said. "It's early in the study, but we aren't seeing a significant number of deer being killed by bobcats either. There is evidence they scavenge on deer opportunistically after a cougar kill or natural cause of death."

McCoy said that bobcats may actually survive on smaller prey such as mountain beavers, birds, rabbits, moles and mice.

Adult male bobcats have little overlap of home ranges as they are quite territorial.

"One of the things we want to know about female cats is whether their home ranges are larger or smaller and how territorial they are, comparatively," McCoy said.

Coupled with extensive research of elk and deer within their traditional hunting area, the tribe will use the research on cats to manage them in the future.

Debbie Preston

Rob McCoy, wildlife division manager for the Makah Tribe, applies eye-drops to a bobcat prior to fitting it with a radio collar.

Regional Collaborative Management

Puget Sound Partnership

The Puget Sound Partnership (PSP) was created in 2007 to recover Puget Sound's health by 2020. Tribes are actively involved in leadership and participation in a wide range of projects to improve the health of Puget Sound.

- U.S. Reps. Derek Kilmer and Denny Heck formed the Puget Sound Recovery Caucus in 2013. The congressional caucus coordinates action at the federal level and collaborates with stakeholders on efforts to improve the health of Puget Sound.
- The 2014-15 Action Agenda update focused on revisions to recovery activities that should begin or be completed within two years. New initiatives, priorities and strategies are not included in this update, but will be considered when substantial review and updating takes place in 2016.
- Tribal representatives are active in partnership efforts to protect salmon habitat. One approach seeks improved habitat protection through review and improvements to current regulatory processes.

Tribes Collaborate on Salish Sea Survival

Emmett O'Connell

Jed Moore, salmon biologist for the Nisqually Indian Tribe, takes zooplankton samples to better understand the food available to migrating juvenile salmon.

Fisheries managers studying poor ocean survival of salmon are concentrating their research on juvenile fish and their preferred prey. Several tribes collaborated on studies in 2014.

The Tulalip, Nisqually, Port Gamble S'Klallam, Lummi, Swinomish and Sauk-Suiattle tribes are among the collaborators sampling zooplankton throughout the region.

Zooplankton and ichthyoplankton are the preferred prey for juvenile salmon. Researchers want to find out whether prey availability has changed in the Salish Sea during the critical period of juvenile salmon development, leading to poor growth and survival.

"This effort will fill critical knowledge gaps in understanding the lower levels of the marine food web that affect juvenile salmon," said Paul McCollum, director of natural resources for the Port Gamble S'Klallam Tribe. "The data will contribute to the development of ecosystem indicators that have already been demonstrated to greatly improve adult salmon return forecasting."

In Hood Canal and Admiralty Inlet, the Port Gamble S'Klallam Tribe has been conducting nearshore research and monitoring of juvenile salmon and forage fish, using acoustics, trawl and beach seine methods, as well as zooplankton sampling.

"The increasing inability in recent years to accurately estimate annual salmon returns is impacting tribal treaty rights and implementation of the U.S./Canada Pacific Salmon Treaty," said Terry Williams, commissioner of fisheries and natural resources for the Tulalip Tribes. "It also impairs the critical decision-making necessary to achieve salmon recovery goals and sustainable fisheries."

The Tulalip and Nisqually tribes are partnering on a study of juvenile salmon in the Snohomish and Nisqually river watersheds and adjacent nearshore and offshore marine areas.

The study will examine the entire community structure of competitors and predators, including plankton and other fish species. Smolt traps operate continuously on both rivers from winter through summer to collect timing, size and abundance data for out-migrating salmon. Both tribes also sample juvenile fish use of nearshore marine areas and pocket estuaries using fyke nets and beach seines.

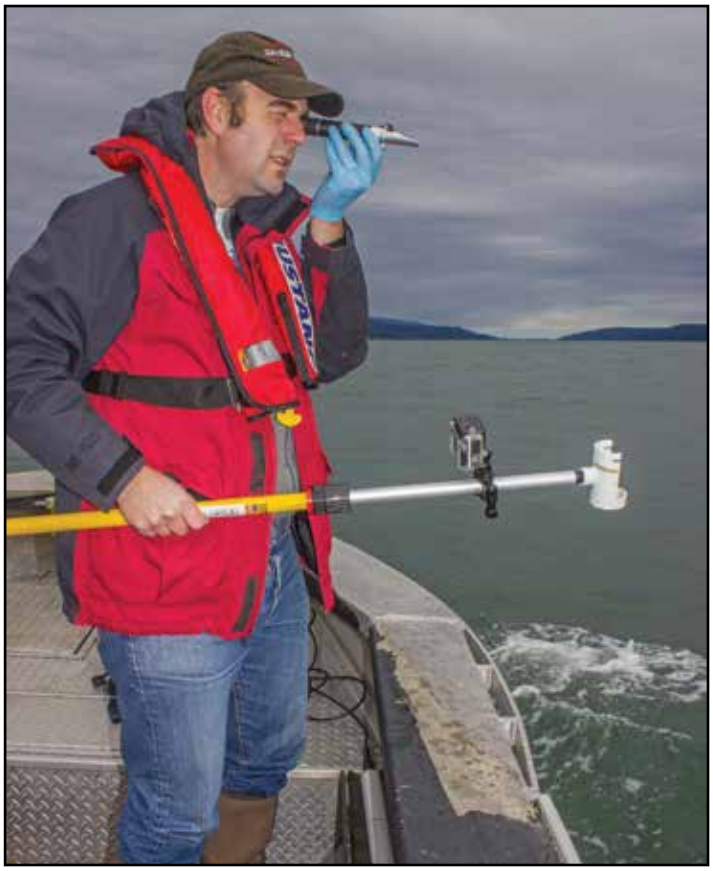
This sampling data should allow researchers to identify the life stage, timing and locations where growth of juvenile salmon is limited.

Ocean Ecosystem Management

The state of Washington, the Hoh, Makah and Quileute tribes and the Quinault Indian Nation work with the National Oceanic and Atmospheric Administration (NOAA) to integrate common research goals to understand changing ocean conditions and create the building blocks for managing these resources.

- In recognition of the challenges facing the Olympic Coast ecosystem, the tribes and state of Washington established the Intergovernmental Policy Council to guide management of Olympic Coast National Marine Sanctuary. Many of the research and planning goals established by tribes and the state mirror the recommendations of the U.S. Ocean Policy.
- Climate change and ocean acidification have been top priorities the past two years. Because of their unique vulnerability, coastal indigenous cultures are leaders in societal adaptation and mitigation in response to events driven by climate change.
- The tribes continue to work with the state of Washington and federal partners to respond to the findings of the state's blue ribbon panel on ocean acidification including prioritizing research to understand its effects on marine ecology and shared natural resources.

Tribal Environmental Protection and Water Resources Program



Tiffany Royal

Skokomish Tribe water quality biologist Seth Book measures the salinity of a water sample from Hood Canal near Hoodsport.

The Coordinated Tribal Water Quality Program was created by the Pacific Northwest tribes and the federal Environmental Protection Agency (EPA) to address water quality issues under the Clean Water Act.

- EPA's General Assistance Program (GAP) was established in 1992 to improve capacity for environmental protection programs for all tribes in the country. Many tribes are now participating in the pilot "Beyond GAP" project to build on the investments of the last 20 years by creating environmental implementation programs locally while supporting national environmental protection objectives.
- These programs are essential to combat the threats to tribal treaty resources such as declining water quality and quantity. In western Washington, climate change and urban development negatively affect water resources and aquatic ecosystems, and will get worse with a state population expected to rise by 1 million in the next 20 years.
- Tribal water quality resource program goals include establishing instream flows to sustain harvestable populations of salmon, identifying limiting factors for salmon recovery, protecting existing groundwater and surface water supplies, and participating in multi-agency planning processes for water quantity and quality management.

Regional Collaborative Management (continued)

Forest Management

Two processes, the Timber/Fish/Wildlife (TFW) Agreement and the Forests and Fish Report (FFR), provide the framework for an adaptive management process that brings together tribes, state and federal agencies, environmental groups and private forest landowners to protect salmon, wildlife and other species while providing for the economic health of the timber industry.

- Treaty tribes in western Washington manage their forestlands to benefit people, fish, wildlife and water.
- Reforestation for future needs is part of maintaining the healthy forests that are key to vibrant streams for salmon, and that enable wildlife to thrive.
- Forestlands are a source of treaty-protected foods, medicine and cultural items.
- A tribal representative serves on the state's Forest Practices Board, which sets standards for activities such as timber harvests, road construction and forest chemical applications. Tribes also are active participants in the FFR Cooperative Monitoring, Evaluation and Research Committee (CMER).

Upper Skagit Tribe Improves Elk Forage

Debbie Preston

Elk populations in the North Cascades have suffered as a result of degraded habitat. The Upper Skagit Tribe recently helped thin a forest and added mulch to the soil to improve forage quality.

The Upper Skagit Indian Tribe's natural resources department thinned and mulched forestland on Puget Sound Energy (PSE) property last fall to improve elk forage in the North Cascades mountains.

Degraded and disconnected habitat is one of the main causes of the decline in numbers of the Nooksack elk herd, which went from a population of more than 1,700 20 years ago to about 300 by 2003. Since then, tribal and state co-managers have improved elk habitat in the region. Annual population surveys indicate that the herd is showing signs of recovery.

"Elk need a corridor of habitat that is rich in forage to keep them from becoming nuisances in populated areas," said Scott Schuyler, natural resources director for the Upper Skagit Tribe.

PSE acquired the land from the Department of Natural Resources as part of the mitigation requirements of the 2008 relicensing agreement with the Federal Energy Regulatory Commission for the utility's Baker River Hydroelectric Project.

A crew used chainsaws to remove hundreds of trees on about 3 acres of land and 1,500 feet of road. The trees, mostly small Douglas fir, were then put through a wood chipper to mulch the dry, rocky soil.

"We needed to remove enough of the canopy to let light in so grasses can grow," said Upper Skagit timberland services manager Robert Schuyler. "The trees we left can be harvested later for a commercial crop."

The mulched ground was seeded with grasses, clover and small burnet.

"There's no forage out here, it's all knee-deep salal, Oregon grape and sword fern, which elk don't eat," said Tony Fuchs, PSE wildlife biologist. "Once we get grasses and clover established, elk will find a better place to forage."

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NWIFC Functions, Programs and Activities

Tiffany Royal

NWIFC fish pathologist Marcia House, left, and Lower Elwha Klallam Tribe hatchery manager Larry Ward discuss coho that returned to the hatchery in November 2014.

The Northwest Indian Fisheries Commission (NWIFC) was created in 1974 by the 20 treaty Indian tribes in western Washington that were parties to the *U.S. v. Washington* litigation that affirmed their treaty-reserved salmon harvest rights and established the tribes as natural resources co-managers with the state.

The NWIFC is an inter-tribal organization that assists member tribes with their natural resources co-management responsibilities. Member tribes select commissioners who develop policy and provide direction for the organization. The commission employs about 70 full-time employees and is headquartered in Olympia, Wash., with satellite offices in Forks, Kingston and Burlington.

The NWIFC provides broad policy coordination as well as high-quality technical and support services for its member tribes in their efforts to co-manage the natural resources of western Washington. The NWIFC serves as a clearinghouse for information on natural resources management issues important to member tribes. The commission also acts as a forum for tribes to address issues of shared concern, and enables the tribes to speak with a unified voice.

The NWIFC has coordinated the tribal Treaty Rights at Risk initiative that seeks to encourage the federal government to align its agencies and programs with salmon recovery goals and to lead a more coordinated salmon recovery effort. Tribes are calling on the federal government for assistance because it has both the obligation and authority to recover salmon and protect tribal treaty rights.

Habitat Services

- Coordinate policy and technical discussion between tribes and federal, state and local governments, and other interested parties.
- Coordinate, represent and monitor tribal interests in the Timber/Fish/Wildlife Forests and Fish Report process, Coordinated Tribal Water Resources and Ambient Monitoring programs. Analyze and distribute technical information on habitat-related forums, programs and processes.
- Implement the Salmon and Steelhead Habitat Inventory and Assessment Project.

U.S./Canada Pacific Salmon Treaty

- Facilitate inter-tribal and inter-agency meetings, develop issue papers and negotiation options.
- Inform tribes and policy representatives about issues affected by the treaty implementation process.
- Serve on the pink, chum, coho, chinook, Fraser sockeye and data-sharing technical committees, and other work groups and panels.
- Coordinate tribal research and data-gathering activities associated with implementation of the Pacific Salmon Committee.

Quantitative Services

- Administer and coordinate the Treaty Indian Catch Monitoring Program.
- Provide statistical consulting services.
- Conduct data analysis of fisheries studies and developing study designs.
- Update and evaluate fishery management statistical models and databases.

Fisheries Management

- Long-range planning, wild salmon recovery efforts and federal Endangered Species Act implementation.
- Annual fisheries planning: developing pre-season agreements; pre-season and in-season run size forecasts; monitoring; and post-season fishery analysis and reporting.
- Marine fish management planning.
- Shellfish management planning.

Enhancement Services

- Coordinate coded-wire tagging of more than 4 million fish at tribal hatcheries to provide information critical to fisheries management.
- Analyze coded-wire data.
- Provide genetic, ecological and statistical consulting for tribal hatchery programs.
- Provide fish health services to tribal hatcheries in the areas of juvenile fish health monitoring, disease diagnosis, adult health inspection and vaccine production.

Information and Education Services

- Provide internal and external communication services to member tribes and NWIFC.
- Develop and distribute communication products such as news releases, newsletters, videos, photos and web-based content.
- Respond to public requests for information about the tribes and their tribal natural resources management activities.
- Work with state agencies, environmental organizations and others in cooperative communication efforts.

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