

# ORAU TEAM Dose Reconstruction Project for NIOSH

Oak Ridge Associated Universities I Dade Moeller & Associates I MJW Corporation

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## **PUBLICATION RECORD**

EFFECTIVE DATE	REVISION NUMBER	DESCRIPTION
10/15/2003	00	New document to establish TBD for occupational internal dose –
11/24/2004	01	Section 5. First approved issue. Initiated by Edward D. Scalsky.  Approved revision initiated to discuss historical limits and tolerance dose; discussion of separations plant (1944-46) and 231-Z (1945-46); clarified intakes in the 300 Area uranium fabrication and the laundry facilities; expanded information on <sup>241</sup> Am and <sup>241</sup> Am MDAs for 1946 and 1967-6/1969; added Section 5.2.4.1 on assignment of tritium doses; added Table 5.2.5-5 (MDAs for nonroutine uranium excreta analyses); revised Table 5.2.6-1 (routine fission product urinalysis detection levels); added discussion of <sup>214</sup> Bi and <sup>208</sup> Tl in whole-body counting (Section 5.3.1); expanded Section 5.7 on unmonitored workers to provide additional discussion, tables, and instructions to dose reconstructors for specific areas on the site; included listing of references; plus corrected miscellaneous typos, and table number changes. Incorporates formal internal and NIOSH review comments. Initiated by Edward D. Scalsky
06/22/2007	02	Approved revision initiated to revise or add information in Section 5. Section 5.1: updated tolerance air concentrations and assigned intakes for 1944-48; added summary of Ikenberry report on thyroid monitoring 1945-46; added fission product intakes for 1947. Section 5.2: added statement on types of urinalysis kits normally used. Section 5.2.1: added information about pure <sup>238</sup> Pu work in 325 Bldg.; added statement to use 5-yr-old Pu mixture for years prior to 1951. Section 5.2.2: added statement about <sup>241</sup> Am recovery operation in PFP in 1960-70s. Section 5.2.3: added stated about <sup>244</sup> Cm recovery in 325 Bldg. Section 5.2.5: added statement about work with 37% enriched U; changed period for 4 μg/L reporting level to 1974; increased Pu-239 contaminant level in RU to 0.8 nCi/gU; added information about U-233 production. Section 5.2.8: added information about promethium use. Section 5.2.10: added information about neptunium urinalyses. Section 5.2.11: added information about thorium urinalyses and <sup>35</sup> S urinalyses. Revised or added information to Section 5 tables. Introduction: added new boilerplate. all table numbers were reformatted. Section 5.1: removed reliance on air samples and referenced co-worker study (OTIB-0039), Section 5.2: added statement on types of urinalysis kits normally used. Sec. 5.2.1: added info. about pure Pu-238 work in 325 Bldg.; added statement to use fresh Pu for 1944-49 and 5-yr-old Pu mixture for 1950-54. Sec. 5.2.2: added statement about Am-241 recovery operation in PFP in 1960-70s. Sec. 5.2.3: added statement about Cm-244 recovery in 325 Bldg. Sec. 5.2.4: added information about tritium in the 400 Area drinking water. Sec. 5.2.5: Table 5-13: raised Pu-alpha contamination level to 0.8 nCi/gU; added statement about work with 37% enriched U; changed period for 4 μg/L reporting level to 1974; added information about U-233 production. Sec. 5.2.8: added some info. about promethium use. Sec. 5.2.9: removed discussion about background levels of Po-210. Sec. 5.2.10: added

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EFFECTIVE	REVISION	
DATE	NUMBER	DESCRIPTION
DATE	NUMBER	some info. about neptunium urinalyses. Sec. 5.2.11: added info. about thorium urinalyses and S-35 urinalyses. Sec. 5.3.1: revised Table 5.3.1-1 (5-22) to clarify MDAs, decision levels and reporting levels. Sec. 5.3.4: totally new section on thorium exposure and thorium monitoring. Sec. 5.4: replaced the old mixture rules with use of Table 5.7-2 (5-25); Sec. 5.6.2: (chronic intakes) deleted entire section. Sec. 5.7 (now 5.6): added information about tritium in the 400 area drinking water; added reference to OTIB-0039 and revised Table 5-25 accordingly. Moved old Appendix D into this document as Attachment A; took out all the tables on chronic intakes. Constitutes a total rewrite of the document. Incorporates NIOSH formal review comments. Many grammatical and format changes throughout. Table 5-12: removed the uranium specific activity data for DU and natural U; added statement to use IMBA defaults to footnote a. Section 5.3.4: added table showing buildings with thorium exposure; added text and table explaining adjustment factors for whole body measurements of Th-232 and associated intakes; added thoron doses for workers processing thorium in 3722 and 3732 bldgs. Section 5.6: clarified possible absorption types for Pu and U in the specific-facilities section; added thoron to Table 5-30; revised tritium intakes for unmonitored workers due a coworker analysis of tritium doses. New Tables 5-27 and 5-28 providing Th-232 intakes based on whole body counting at long times after end of chronic intakes. Section 5.3.4: revised thoron WLMs, removed reference to thoron dose with a reference to TIB-011, clarified use of OTIB-0039 for unmonitored thorium intakes. Section 5.6: removed references to 6-mrem tritium dose and stayed with intake values; clarified how the tritium intake values were determined for the 1955-60, 1970-71, and 1973-83 time periods; corrected a math error resulting in slightly increased intakes for 1984, 85, and 86. Includes Attributions and Annotations and additional references. Incorporates formal intern
		Initiated by Donald E. Bihl and Edward D. Scalsky.

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#### **ACRONYMS AND ABBREVIATIONS**

BPNNL Battelle - Pacific Northwest National Laboratory

CADW Chronic Annual Dose Workbook

CF commercial fuel

DAC derived air concentration
DIL derived investigation level
DOE U.S. Department of Energy
DOE-RL DOE Richland Operations Office

DU depleted uranium

EEOICPA Energy Employees Occupational Illness Compensation Program Act of 2000

ft feet

GeLi lithium-drifted germanium

GI gastrointestinal

GSD geometric standard deviation

HIE Hanford Internal Exposure (database)

HPGe high-purity germanium

ICRP International Commission on Radiological Protection

IMBA Integrated Modules for Bioassay Assessment

KPA kinetic phosphorescence analysis

LANL Los Alamos National Laboratory

LEPD Low Energy Photon Detector (also computer code to indicate use of the LEPD)

MDA minimum detectable activity or, for elemental uranium, minimum detectable amount

MDC minimum detectable concentration

min minute

MPBB maximum permissible body burden MPC maximum permissible concentration

NBS National Bureau of Standards

NCRP National Council on Radiation Protection and Measurements

NIOSH National Institute for Occupational Safety and Health

NU natural uranium

ORAU Oak Ridge Associated Universities

ORE Occupational Radiological Exposure (database)

PNL Pacific Northwest Laboratory

PNNL Pacific Northwest National Laboratory

POC probability of causation

PRTR Plutonium Recycle Test Reactor

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RDA Reliably Detectable Activity

Radiological Exposure (database) Richland Operations Office REX

RL

RU recycled uranium

SRDB Ref ID Site Research Database Reference Identification (number)

total propagated uncertainty thenoyl trifluoroacetone TPU TTA

U.S.C. **United States Code** 

UST **United States Testing Company** 

#### 5.1 INTRODUCTION

Technical basis documents and site profile documents are not official determinations made by the National Institute for Occupational Safety and Health (NIOSH) but are rather general working documents that provide historic background information and guidance to assist in the preparation of dose reconstructions at particular sites or categories of sites. They will be revised in the event additional relevant information is obtained about the affected site(s). These documents may be used to assist NIOSH staff in the completion of the individual work required for each dose reconstruction.

In this document the word "facility" is used as a general term for an area, building, or group of buildings that served a specific purpose at a site. It does not necessarily connote an "atomic weapons employer facility" or a "Department of Energy [DOE] facility" as defined in the Energy Employees Occupational Illness Compensation Program Act [EEOICPA; 42 U.S.C. § 7384I(5) and (12)]. EEOICPA defines a DOE facility as "any building, structure, or premise, including the grounds upon which such building, structure, or premise is located ... in which operations are, or have been, conducted by, or on behalf of, the Department of Energy (except for buildings, structures, premises, grounds, or operations ... pertaining to the Naval Nuclear Propulsion Program)" [42 U.S.C. § 7384I(12)]. Accordingly, except for the exclusion for the Naval Nuclear Propulsion Program noted above, any facility that performs or performed DOE operations of any nature whatsoever is a DOE facility encompassed by EEOICPA.

For employees of DOE or its contractors with cancer, the DOE facility definition only determines eligibility for a dose reconstruction, which is a prerequisite to a compensation decision (except for members of the Special Exposure Cohort). The compensation decision for cancer claimants is based on a section of the statute entitled "Exposure in the Performance of Duty." That provision [42 U.S.C. § 7384n(b)] says that an individual with cancer "shall be determined to have sustained that cancer in the performance of duty for purposes of the compensation program if, and only if, the cancer ... was at least as likely as not related to employment at the facility [where the employee worked], as determined in accordance with the POC [probability of causation¹] guidelines established under subsection (c) ... " [42 U.S.C. § 7384n(b)]. Neither the statute nor the probability of causation guidelines (nor the dose reconstruction regulation) define "performance of duty" for DOE employees with a covered cancer or restrict the "duty" to nuclear weapons work.

As noted above, the statute includes a definition of a DOE facility that excludes "buildings, structures, premises, grounds, or operations covered by Executive Order No. 12344, dated February 1, 1982 (42 U.S.C. 7158 note), pertaining to the Naval Nuclear Propulsion Program" [42 U.S.C. § 7384I(12)]. While this definition contains an exclusion with respect to the Naval Nuclear Propulsion Program, the section of EEOICPA that deals with the compensation decision for covered employees with cancer [i.e., 42 U.S.C. § 7384n(b), entitled "Exposure in the Performance of Duty"] does not contain such an exclusion. Therefore, the statute requires NIOSH to include all occupationally derived radiation exposures at covered facilities in its dose reconstructions for employees at DOE facilities, including radiation exposures related to the Naval Nuclear Propulsion Program. As a result, all internal and external dosimetry monitoring results are considered valid for use in dose reconstruction. No efforts are made to determine the eligibility of any fraction of total measured exposure for inclusion in dose reconstruction. NIOSH, however, does not consider the following exposures to be occupationally derived:

- Radiation from naturally occurring radon present in conventional structures
- Radiation from diagnostic X-rays received in the treatment of work-related injuries

<sup>1</sup> The U.S. Department of Labor is ultimately responsible under the EEOICPA for determining the POC.

#### 5.1.1 **Purpose**

This TBD documents the internal dosimetry program at the Hanford Site and provides the technical basis to be used to evaluate the internal occupational radiation dose for EEOICPA claims.

#### 5.1.2 Scope

The Hanford operations have played an important role in the development of the U.S. nuclear weapons program. This TBD is part of the overall Hanford Site Profile, which describes plant facilities and processes, historical information, and environmental data in relation to dose reconstruction for Hanford workers. It contains supporting documentation to assist in the reconstruction of occupational internal doses from these activities.

The methods and concepts of measuring occupational internal doses to workers have evolved since the beginning of operations at the Hanford Site. An objective of this document is to provide supporting technical data to evaluate internal occupational doses that can reasonably be associated with worker radiation exposures. In addition, this document presents the technical basis of methods used to prepare Hanford worker dose information for input to the NIOSH IREP (Interactive RadioEpidemiological Program) computer code.

When the first reactor became operational on the Hanford Site, there were no programs to monitor an employee for internal dose, with the exception of measuring particles in the air. The site was operating three reactors, a fuel manufacturing facility and four processing plants from 1943 to 1946 before a bioassay program was in place. The responsibility for personnel monitoring was with the Medical Department. The coworker analysis document, Internal Dosimetry Coworker Data for the Hanford Site, OTIB-0039 (ORAUT 2007), provides guidance on assigning intakes for the years prior to bioassay.

In the 1940s, the radiation protection community used the term "tolerance" to describe dose limits and other values such as air concentration values. This was before the National Council on Radiation Protection and Measurements (NCRP) introduced the terms Maximum Permissible Body Burden (MPBB) and Maximum Permissible Concentration (MPC). Air concentration tolerance levels were based on dose rates to significantly impacted organs. Examples at Hanford were 0.01 rep/day from alpha-emitters, 1 rep/day to the thyroid from beta/gamma emitters, or total accumulation in the body of a radionuclide with comparable effects to 0.1 µg of radium. The latter limit was used to establish the limit of 0.5 µg for total accumulation of plutonium in the body. The radiation protection program was directed toward preventing workers from acquiring a tolerance dose or being exposed to tolerance air concentrations. Workers were supposed to wear respiratory protection if an air concentration exceeded the tolerance level or if a tolerance level was anticipated to be exceeded for a given job. In some cases, a lower air concentration was established as the respirator-required air concentration. The tolerance air concentrations in 1945 were  $5 \times 10^{-10} \, \mu \text{g/cm}^3$  ( $4 \times 10^{-11} \, \mu \text{Ci/cm}^3$ ) for product (meaning plutonium),  $1 \times 10^{-8} \,\mu\text{Ci/cm}^3$  for fission/activation products, and  $1.5 \times 10^{-4} \,\mu\text{g/cm}^3$  (1.1 × 10 μCi/cm³) for uranium (Cantril 1945). By 1947, the respirator-required air concentrations were 2 ×  $10^{-11} \,\mu\text{g/cm}^3$  (2 ×  $10^{-12} \,\mu\text{Ci/cm}^3$ ) for plutonium, (1 ×  $10^{-9} \,\mu\text{Ci/cm}^3$ ) for fission products (assumed to mean particulate beta emitters), and  $5 \times 10^{-5} \, \mu \text{g/cm}^3$  (3.5 ×  $10^{-11} \, \mu \text{Ci/cm}^3$ ) for uranium (Parker 1947; Patterson 1949). Additional discussion on tolerance levels is provided in Attachment A.

According to the history compiled by R. H. Wilson (1987), one of the priority tasks for a special studies group formed in 1944 was to determine a way to measure plutonium in the body. Limits on the amount of plutonium in the body were set as early as 1944 and, after experimentation with various methods, routine urine sampling and analysis for plutonium were initiated in 1946. Urinalysis for

uranium seems to have also started in 1946 and was well established by 1948. Urinalysis for fission products started in this timeframe as well, although the Wilson document indicates that separation from <sup>40</sup>K was not always successful prior to 1949. Since then, monitoring for numerous radionuclides has occurred at Hanford because of the complex scope of work over the years, the many research projects, special "campaigns," etc. In addition, numerous techniques have been used because of improvements in techniques. The major sources of intakes have been plutonium, <sup>241</sup>Am either as an ingrown contaminant in the plutonium or as a separated waste product, uranium, fission products, activation products, and tritium. However, the records list a wide spectrum of radionuclides that were monitored and an even longer list of codes used to identify the radionuclides, groups of radionuclides, specific measurement techniques, or combinations of radionuclides and techniques. Many of the radionuclides apply to a small set of workers on a research project or to workers (for instance, radiation monitoring technicians) whose tasks "might" have exposed them to many different sources.

Table 5-1 provides a fairly exhaustive list of codes for analyses that dose reconstructors might encounter in the bioassay or internal dosimetry records for Hanford workers. Some of the codes were used for scheduling bioassay but not for reporting results of the bioassay. For instance, IPA is a code for performing plutonium and americium separation chemistry and alpha spectrometry on an excreta sample, but the results would normally be reported separately for <sup>238</sup>Pu, <sup>239</sup>Pu, and <sup>241</sup>Am. However, if the sample was not obtained or the results could not be reported due to analysis problems, the record will show just the IPA code with a reason for not obtaining a result [1]. Other codes refer to a type of *in vivo* count or a special type of sample analysis. For instance, LEPD is the code for performing an X-ray/gamma-ray analysis on an excreta sample using the low-energy photon detector (a thin-window germanium detector); however, if anything was detected, the actual radionuclide was reported [2]. The code GOK shows on *in vivo* count hardcopy records during the 1960s and 1970s. This refers to net counts per minute from an undetermined source in a low-energy region of the spectrum from Nal-based whole-body counters [3]

Other bioassay codes have been used to indicate the following:

- Sample type,
- In vivo count body location,
- Reason for the sample/count,
- Type of kit and some details about the sampling protocol,
- Laboratory used,
- Laboratory turnaround time versus analytical sensitivity,
- Units associated with the result, and
- Reason for not obtaining a valid excreta result or *in vivo* count.

In addition there are codes pertaining to the nature of the intake, including:

- Reason for an intake assignment,
- Source of intake (at Hanford or another site).
- Nature of intake, and
- Mode of intake.

Tables listing and explaining these codes are provided in Attachment A.

Table 5-1. Codes and radionuclides associated with bioassay at Hanford [4].

		les associated with bloassay at Haillold [4].
Code	Description	Comment
AAAA1	Americium	Probably Am-241
AAAA2	Americium	Probably Am-241
AAAA3	Americium	Probably Am-241
AAAA4	Americium	Probably Am-241
AAAA5	Americium	Probably Am-241
AAAA6	Americium	Probably Am-241
AAAA7	Americium	Probably Am-241
AC225	Actinium-225	
ACS	Actinium-227, thorium-227	Scheduling code
AC227	Actinium-227	
AC228	Actinium-228	
AG110	Silver-110	
AM241	Americium-241	
AM242	Americium-242	
AM243	Americium-243	
BA140	Barium-140	
BETA	Beta	
BI213	Bismuth-213	
BI214	Bismuth-214	
BK249	Berkelium-249	
BR 82	Bromine-82	
C 14	Carbon-14	
CE141	Cerium-141	
CE143	Cerium-143	
CE144	Cerium-144	
CF249	Californium-249	
CM242	Curium-242	
CM244	Curium-244	
CO 58	Cobalt-58	
CO 60	Cobalt-60	
CR 51	Chromium-51	
CS134	Cesium-134	
CS137	Cesium-137	
EU152	Europium-152	
EU154	Europium-154	
EU155	Europium-155	
EU156	Europium-156	
EV155	?	Probably a typographical error for Eu-155 that got left in the database
EV156	?	Probably a typographical error for Eu-156 that got left in the database
FE 59	Iron-59	1 result y a typograpmen error for 24 resound get for in the database
FP		
	Fission products	
GA	Gross alpha	
GB	Gross beta	
GELI	Gamma-GeLi detector	Excreta scheduling code for a gamma scan with a germanium detector
GOK		See text
GS	Gamma Nal detector	Excreta scheduling code for a gamma scan with a NaI detector
H 3	Tritium	g
I 125	Iodine-125	
I 129	lodine-129	
I 131	Iodine-131	
I 133	Iodine-133	
IAM	Isotopic americium	Excreta scheduling code for americium separation and alpha spectrometry
ICA	?	Probably scheduling code for americium and curium via alpha spectrometry
ICM	Cm isotopic	Excreta scheduling code for curium isotopes via alpha spectrometry
IEU	Eu isotopic	Excreta scheduling code for europium separation and isotopic analysis
IPA	Isotopic Pu and Am-241	Excreta scheduling code
IPIU	Isotopic Pu, isotopic U	Excreta scheduling code
IPS	Isotopic Pu and Sr	Excreta scheduling code
IPSA	Isotopic Pu, Sr tot & Am-241	Excreta scheduling code; Sr tot means radiostrontium by gross beta
IPSR	Seg Pu isotopic Sr-total	Excreta scheduling code for isotopes of Pu and radiostrontium
IPU	Isotopic plutonium	Excreta scheduling code
IPUB	Plutonium isotopic, Pu-241	Excreta scheduling code; Pu-241 separate anal. by beta counting
IPUBA		
	Pu isotopic, Pu-241, Am-241 Low-level isotopic Pu	Excreta scheduling code
IPUL	LI OW-IOVOLISOTODIC PIL	Pu-238 and Pu-239 using a 10,000-minute count

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Code	Description	Comment
IRA	Radium isotopic	Excreta scheduling code
IR192	Iridium-192	Excreta scheduling code
ISCP	Sequential Sr-90 Ce Pm	Excreta scheduling code
ISPEC	Gamma spectroscopy	Excreta scheduling code
ISR	Sr isotopic	Excreta scheduling code
ITH	Thorium isotopic	Excreta scheduling code
ITPAC	Seq isotopic Pu, Cm & Am-241	Excreta scheduling code
IU	U isotopic	Excreta scheduling code
IUPU	Isotopic plutonium/U-natural	Excreta scheduling code
K 40	Potassium	
LA140	Lanthanum-140	
LEPD	Low-energy photon detector	Excreta scheduling code for low-energy photon scan
MFP	Mixed fission products	
MN 54	Manganese-54	
MO 99	Molybdenum-99	
NA 22	Sodium-22	
NA 24	Sodium-24	
NAI	Gamma Nal detector	Excreta scheduling code
NB 95 NP237	Niobium-95	
NP237 NP239	Neptunium-237 Neptunium-239	
PB210	Lead-210	
PB210 PB212	Lead-210 Lead-212	
PM147	Promethium-147	
PO210	Polonium-210	
PR144	Praseodymium-144	
PU	Plutonium alpha	Total alpha from Pu isotopes after separation
PUMIX	Plutonium alpha	Total alpha from Pu isotopes and Am-241
PU238	Plutonium-238	
PU239	Plutonium-239	When pertaining to excreta samples, it's actually Pu-239+240
PU240	Plutonium-240	
PU241	Plutonium-241	
PU242	Plutonium-242	
QUS	U	Quick Uranium Soluble; excreta scheduling code for elemental U
QUS 1	U	Same as QUS
QUS 2	U	Same as QUS
RA224	Radium-224	
RA225	Radium-225	
RA226	Radium-226 Radium-228	
RA228 RH106	Radium-228 Rhodium-106	
RND	Radon daughters	
RU103	Ruthenium-103	
RU106	Ruthenium-106	
S 35	Sulfur-35	
SB124	Antimony-124	
SB125	Antimony-125	
SCP	Sequential Sr-total Ce Pm	Excreta scheduling code
SM153	Samarium-153	· ·
SR	Strontium	Total radiostrontium by beta counting
SR 89	Strontium-89	
SR 90	Strontium-90	When pertaining to excreta samples, Sr-90 by yttrium ingrowth
TAC	Total actinides	
TC 99	Technetium-99	
TH227	Thorium-227	
TH228	Thorium-228	
TH230	Thorium-230	
TH232	Thorium-232	
TH234 TL208	Thorium-234	
U	Thallium-208 Elemental uranium	
URAN	Elemental uranium	
U DEP	Depleted uranium	
U NAT	Natural uranium	
U 233	Uranium-233	See uranium discussion in text
	, <del></del>	2.

Code	Description	Comment
U 234	Uranium-234	Actually U-234 + 233, but usually U-234
U 235	Uranium-235	
U 236	Uranium-236	
U 238	Uranium-238	
UMIX	Uranium mix	Total uranium, used for intakes not bioassay
UMS	U-235 U-236 U-238 U-234	
US	U	
XX 0	Isotope will have no result	
ZN 65	Zinc-65	
ZR 95	Zirconium-95	

Attributions and annotations, indicated by bracketed callouts and used to identify the source, justification, or clarification of the associated information, are presented in Section 5.7.

# 5.2 IN VITRO MINIMUM DETECTABLE ACTIVITIES, ANALYTICAL METHODS, AND REPORTING PROTOCOLS

Most urinalysis records have, at some time, been entered into the electronic database(s). However, for some of the earliest urinalysis records, cases have been discovered where not all records were included in the electronic database. For any case where urinalysis might have been obtained prior to 1974, the hardcopy file for the case should be thoroughly reviewed for urinalysis results that might be missing in the electronic database. The Hanford Internal Exposure (HIE) database was implemented in 1974, followed by the Occupational Radiological Exposure (ORE) database in 1983, and the Radiological Exposure (REX) database in 1993. In principle, the REX database has all the information from the previous databases, but as stated above there could be isolated situations where some data was never entered in a database or did not get transferred from one database to another [5].

There is another anomaly found in the results from around 1946-1950. There is a urinalysis record with no result and no volume. This might indicate that the sample was not turned in or the analysis failed; however, experience has shown that this convention was also used to indicate a result that was a non-detection. In many cases the actual laboratory urinalysis results card is available in the worker's file and would show if the analysis was performed but the results were below detection or not [6].

Home sampling began very early in the program (1946) (Wilson 1987) and has continued throughout the history of Hanford. Home sampling was used to prevent contamination of samples in the workplace. The sampling protocol used most frequently (kit code 1) was a simulated 24-hr sample obtained by sampling from evening through morning on two consecutive nights (or equivalence for shift workers). Termination samples were often simulated 12-hr samples (kit code 2) [7]. The dose reconstructor needs to look at the kit code to determine if sample results should be treated as a day's excretion or adjusted to represent the daily excretion.

In vitro analyses were performed in house until the breakup of the main Hanford contractor (General Electric) in 1965. At that time, the U.S. Department of Energy (DOE) Richland Office established a contract for *in vitro* analyses with the United States Testing Company (UST), which built and operated a commercial low-level radiochemistry laboratory in north Richland until 1990. The responsibility for awarding and overseeing the contract was subsequently transferred to Battelle as operators of the Pacific Northwest Laboratory (PNL). Until 2005, with the exception of a period between 1990 and 1992, *in vitro* analyses were performed in the same facility. However, due to buyouts and mergers, the name of the laboratory changed in the following sequence: United States Testing, International

Technology Analytical Services, Quanterra Environmental Services, and Severn Trent Laboratories. The current *in vitro* laboratory is General Engineering Laboratory [8].

Battelle defaulted the contract with UST in June 1990, after which routine samples were collected and frozen (Lyon et al. 1991, 1992). Between September and November 1990, temporary contracts/ agreements were established and samples were analyzed at the following laboratories: Los Alamos National Laboratory (LANL; plutonium), TMA-Norcal (strontium), PNL-Analytical Chemistry Laboratory (325 Building; tritium), and Westinghouse Hanford Company (222-S Building; elemental uranium). In February 1991, International Technology Analytical Services began analyses for plutonium. americium, curium, and isotopic uranium. LANL was replaced by Oak Ridge National Laboratory and Reynolds Electrical and Engineering Company at the Nevada Test Site (plutonium) in April 1991. The contract with International Technology Analytical Services replaced the contract with UST, but the other laboratories continued to process samples until the backlog was worked off. Therefore, the work at the temporary laboratories was finishing during late 1991 through early 1992, with the last results received in March 1992. This interruption in the normal process of bioassay monitoring had two effects that will show in the bioassay records for this period: (1) changes in the minimum detectable activities for the various analyses reflecting capabilities at the interim laboratories; and (2) delays in collection, analysis, and follow-up to high routine results from the normal pattern (Lyon et al. 1991, 1992).

### 5.2.1 <u>Plutonium</u>

By far the most serious intakes at Hanford involved plutonium and <sup>241</sup>Am. Routine urinalyses for plutonium started in September 1946 (Wilson 1987). The first plutonium bioassay analysis consisted of lanthanum fluoride precipitation and thenoyl trifluoroacetone (TTA) extraction and gross alpha counting. Electrodeposition on a stainless steel disk combined with nuclear track emulsion (autoradiography) started in December 1952. Detection levels for these and subsequent procedures are listed in Table 5-2. The definition of "detection level" no doubt changed over the years, but the levels in Table 5-2 fit reasonably with the concept of limit of detection or Minimum Detectable Activity (MDA). For example, Wilson (1987) states,

From statistical evaluations of data collected in 1953, the true detection limit with nuclear-track film was determined. These evaluations showed 0.05 dpm was achievable within reasonable confidence levels. Occasionally recovery, counting, etc., allowed detection levels to be as low as 0.028 dpm and for a short period, a level of 0.027 dpm was reached and used as the detection level ... This practice [of recording lower detection levels] was discontinued and the more conservative 0.05 dpm was used routinely even though lower levels were possible part of the time.

Prior to October 1983, the recorded value was the total alpha activity from plutonium, so it would have included activity from <sup>238</sup>Pu, <sup>239</sup>Pu, and <sup>240</sup>Pu. The recorded results would not have accounted for any <sup>241</sup>Pu or <sup>241</sup>Am present in the urine [9].

The results might have been reported as Pu or  $^{239}$ Pu but, until October 1983, the result was really the total alpha activity from isotopes of plutonium. Results on plutonium urinalysis sheets were recorded in units of dpm/sample, but the same results were recorded in units of  $\mu$ Ci/sample in the electronic database. The units in the electronic database should have a unit code of 5, meaning  $\mu$ Ci/sample, but if the code is missing or unreadable, the units are still recognizable because the exponent is normally -7 or -8. A value of  $1.1 \times 10^{-8}$  was recorded for results for which plutonium was not detected (one-half of the nominal 0.05 dpm MDA). This method of recording was used through 1974. In 1975, the

Table 5-2. Routine plutonium urinalysis detection levels<sup>a</sup>.

•	MDA,	Decision level,	
Period	dpm/sample	dpm/sample <sup>'</sup>	Measured quantity
Prior to June 1949	0.96 <sup>b</sup>	0.66	Total Pu alpha
6/1949 to 11/1952	0.33		Total Pu alpha
12/1952 to 1/27/53	0.18		Total Pu alpha
1/28/53 to 3/26/53	0.15		Total Pu alpha
3/27/53 to 11/06/53	0.05		Total Pu alpha
11/07/53 to 12/04/53	0.07		Total Pu alpha
12/53 to 4/55	0.057		Total Pu alpha
5/55 to 8/55	0.027 <sup>c</sup>		Total Pu alpha
9/55 to 9/55	0.04 <sup>c</sup>		Total Pu alpha
10/55 to 9/30/83	0.05 <sup>d</sup>	0.025 <sup>d</sup>	Total Pu alpha
10/01/83 to 12/31/83	0.035	Not established, but	Each Pu-238, Pu-
		0.018 is reasonable	239
1/02/84 to 4/88	0.02		Total Pu alpha
5/88 to 5/90	0.02	0.01	Total Pu alpha
6/90 to 11/91	0.03	0.015	Total Pu alpha
11/91 to 4/2000	0.02	0.01	Total Pu alpha
5/2000 to 8/2001	0.02	$X_b + 2.05 \times TPU^e$	Total Pu alpha
9/2001 to present	0.02	2 x TPU <sup>e</sup>	Total Pu alpha

- a. From Wilson 1987 except as otherwise noted.
- b. Estimated from data in Table 1 in "Bioassay at Hanford" (Healy 1948).
- c. The values are probably closer to a decision level than an MDA. Recommend using 0.05 dpm as the MDA [11].
- d. During part of this period, results that were less than the detection limit were reported as 0.025. However, if net activity above background and above 0.025 was detected, the actual amount was recorded [12].
- e.  $X_b$  is mean of blanks and TPU is total propagated uncertainty. The  $X_b$  values used during that time were 0.0000 and 0.000487 dpm for Pu-238 and Pu-239 respectively. However, it is recommended that dose reconstructors use 2 x TPU as the actual decision level for both periods [13].

units were changed to dpm/sample (unit code 1) and 0.025 was recorded for results for which plutonium was not detected [10].

In October 1983 several changes were made. The lanthanum fluoride/TTA method was replaced by the use of anion exchange columns, alpha spectrometry analysis replaced autoradiography, and chemical yield was established for each sample separately by use of a <sup>242</sup>Pu tracer. The results of <sup>238</sup>Pu and <sup>239+240</sup>Pu have been reported separately since then. A 2,500-min count time has been used since 1984. A 10,000-min count time was introduced for special situations in 1996 but its use was rare [14].

Starting in the mid-1990s, the fecal procedure was enhanced to ensure improved oxidation of highly insoluble plutonium. Added steps included wet ashing with hydrogen peroxide and fusion with hydrogen fluoride. This procedure was tested with special high-fired plutonium oxide samples from the Idaho National Engineering and Environmental Laboratory and was found to work well [15].

Fecal samples were usually not analyzed in total (were aliquoted after muffling, dry ashing, and wet ashing); hence, more than one analysis result for a given sample was possible and will often be found in the database [16].

The MDAs listed from 1983 to present are nominal MDAs based on contractual requirements. In general, the laboratory performed slightly better than the contractual MDA, but the true MDA varied

slightly over time and the contractual MDA was a reliable estimate [17]. Reporting of errors, which was the total propagated uncertainty including uncertainty associated with the determination of chemical yield, counting efficiency determination, and systematic errors, began in 1981 [18]. The implementation of a distinction between an MDA (type I and type II errors) and a decision level (type I error) occurred in April 1989 (Lyon et al. 1990). A fixed value of 0.01 dpm/sample was used initially for all results, being one half the nominal MDA. The decision level was allowed to become sample-specific based on the total propagated uncertainty in 2000, and an adjustment was made to the formula in 2001 (Lynch et al. 2000 and 2001).

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The MDAs listed in Table 5-2 apply to routine and priority processing of urine samples. Fecal sampling was used for special sampling after potential intakes, and other processing codes (emergency and expedite) have been available for special urine and fecal samples. The contractual MDAs for these samples are provided in Table 5-3.

Table 5-3. MDAs for nonroutine Pu excreta analyses [19].

	Fecal samples, MDA, dpm/sample			Urine samples, MDA, dpm/sample		
Period	<b>Emergency</b> <sup>a</sup>	Expedite	Priority	Emergency <sup>a</sup>	Expedite	
1/1965 to 10/1983 <sup>b</sup>	0.9-1.5	NA	0.1-0.15	0.5-0.7	NA	
10/1983 to 1/1985 <sup>c</sup>	9	NA	0.2	0.5	NA	
1/1985 to 6/1990	9	3	0.2	0.5	0.08	
6/1990 to 2/1991 <sup>d</sup>	20	4	NA	2	0.4	
2/1991 to present	9	3	0.2	0.5	0.08	

- a. At times the emergency category was called "rush" and the routine category was called "normal."
- b. MDAs varied according to sample size over the range shown; the lower value was generally applicable except for very large samples (Larson 1982). MDAs for this period apply to total Pu alpha.
- c. MDAs from this time forward apply to Pu-238 and Pu-239 separately.
- d. Emergency and expedited processing of urine and fecal samples was available through PNNL's Analytical Chemistry Laboratory. Priority fecal analyses were also available through the offsite laboratories but the MDA was not established, probably about 0.2-0.5 dpm/sample considering the state-of-the-art of those laboratories.

Fecal sampling was normally done in response to suspected intakes; however, routine fecal sampling was used for some high-risk plutonium workers, mostly operators at PUREX and the Plutonium Finishing Plant, from 1986 through June 1989. The special study showed that, when considered as a group, the mean fecal excretion was statistically significantly different from controls. Enhanced air sampling, initiated in response to the study, showed frequent-intermittent releases of plutonium in the workplaces, at levels below the detectability of normal air sampling. When modeled as chronic intake, the intakes and doses were low (less than 10 mrem committed effective dose equivalent), and were documented in the workers' records (Bihl, Buschbom, and Sula 1993; Lyon et al. 1988, 1989). When encountered in the workers' records, results for these fecal samples should be interpreted as associated with chronic intakes, not with an acute intake occurring many days prior to the sample dates.

With the exception of a few standards in radiochemistry laboratories and a <sup>238</sup>Pu purification experiment in the 325 Building C-cell around 1967 [20], plutonium at Hanford was comprised of a mix of radionuclides, namely <sup>238</sup>Pu, <sup>239</sup>Pu, <sup>240</sup>Pu, and <sup>241</sup>Pu. The activity of <sup>242</sup>Pu in plutonium mixtures at Hanford was too small to contribute significantly to dose. Hanford plutonium mixtures were categorized by their weight percent of <sup>240</sup>Pu. When the reactors were operated to produce plutonium for weapons, the target mixture was about 6% <sup>240</sup>Pu, a mixture referred to as weapons grade. N Reactor was also operated to produce electrical power for a local public power company. When the reactor was operated to produce power, the mixture in the fuel rods when removed from the reactor was nominally 12% <sup>240</sup>Pu, a mixture referred to as fuel grade. At any given time, individual fuel rods would have mixtures differing from these, as would individual batches of rods starting at the front end of the fuel rod dissolution and plutonium extraction processes. However, when refined and blended,

the target mixture was the weapons-grade mixture [21]. There is evidence at Los Alamos National Laboratory that plutonium produced at Hanford in the 1940s had less <sup>240</sup>Pu, perhaps closer to 3% (ORAUT 2004); however, documentation of that at Hanford has not been found. Tables 5-4 and 5-5 list the relative activities of plutonium isotopes and <sup>241</sup>Am, which grows in from <sup>241</sup>Pu, for 6% <sup>240</sup>Pu and 12% <sup>240</sup>Pu mixtures (from Carbaugh 2003a). In these tables, "aging" refers to the time since the <sup>241</sup>Am was separated from the plutonium then starts to build in again from decay of <sup>241</sup>Pu.

The values in these tables can help determine the total intake of plutonium and <sup>241</sup>Am if there are limited data on the composition of the source of the intake. For instance, only in rare large intakes

Table 5-4. Activity composition of Hanford reference weapons-grade plutonium mixture (6%).

Mixture designation:	Fresh	5-year	10-year	15-year	20-year	25-year	30-year
Years of aging <sup>a</sup> :	0	5	10	15	20	25	30
Specific activity in mixture (Ci/g)							
Pu-238	8.56E-03	8.23E-03	7.91E-03	7.60E-03	7.31E-03	7.03E-03	6.75E-03
Pu-239	5.77E-02						
Pu-240	1.36E-02						
Pu-241	8.24E-01	6.48E-01	5.09E-01	4.00E-01	3.15E-01	2.48E-01	1.95E-01
Pu-242	1.97E-06						
Am-241	0	5.83E-03	1.04E-02	1.39E-02	1.66E-02	1.87E-02	2.03E-02
Pu-239+240	7.13E-02	7.13E-02	7.13E-02	7.13E-02	7.12E-02	7.12E-02	7.12E-02
Pu-alpha	7.99E-02	7.95E-02	7.92E-02	7.89E-02	7.85E-02	7.83E-02	7.80E-02
Total alpha	7.99E-02	8.53E-02	8.96E-02	9.28E-02	9.52E-02	9.70E-02	9.83E-02
Activity ratios							
Pu-239+240:Am-241	NA	12.2	6.87	5.13	4.28	3.80	3.50
Pu-239+240:Pu-238	8.33	8.67	9.01	9.38	9.74	10.1	10.5
Pu-241:Pu-239+240	11.6	9.09	7.15	5.62	4.42	3.48	2.73
Pu alpha:Pu-239+240	1.12	1.12	1.11	1.11	1.10	1.10	1.10
Pu alpha: Pu-238	9.33	9.66	10.0	10.4	10.7	11.1	11.6
Pu alpha:Am-241	NA	13.6	7.62	5.68	4.73	4.19	3.84
Pu-241: Pu alpha	10.3	8.15	6.43	5.07	4.01	3.17	2.50

a. Time since separation of Am-241 from the plutonium mix.

Table 5-5. Activity composition of Hanford reference fuel-grade plutonium mixture (12%).

Mixture designation:	Fresh	5-year	10-year	15-year	20-year	25-year	30-year
Years of aging <sup>a</sup> :	0	5	10	15	20	25	30
Specific activity in mixture (Ci/g)							
Pu-238	1.71E-02	1.64E-02	1.58E-02	1.52E-02	1.46E-02	1.40E-02	1.35E-02
Pu-239	5.26E-02	5.26E-02	5.26E-02	5.26E-02	5.26E-02	5.26E-02	5.25E-02
Pu-240	2.72E-02	2.72E-02	2.72E-02	2.72E-02	2.72E-02	2.71E-02	2.71E-02
Pu-241	3.09E+00	2.43E+00	1.91E+00	1.50E+00	1.18E+00	9.29E-01	7.30E-01
Pu-242	3.93E-06						
Am-241	0	2.19E-02	3.89E-02	5.22E-02	6.24E-02	7.03E-02	7.63E-02
Pu-239+240	7.98E-02	7.98E-02	7.98E-02	7.97E-02	7.97E-02	7.97E-02	7.97E-02
Pu-alpha	9.69E-02	9.62E-02	9.56E-02	9.49E-02	9.43E-02	9.37E-02	9.32E-02
Total alpha	9.69E-02	1.18E-01	1.35E-01	1.47E-01	1.57E-01	1.64E-01	1.69E-01
Activity ratios							
Pu-239+240:Am-241	NA	3.64	2.05	1.53	1.28	1.13	1.04
Pu-239+240:Pu-238	4.67	4.86	5.05	5.24	5.46	5.69	5.90
Pu-241:Pu-239+240	38.7	30.5	24.0	18.8	14.8	11.7	9.16
Pu alpha:Pu-239+240	1.21	1.21	1.20	1.19	1.18	1.18	1.17
Pu alpha: Pu-238	5.67	5.87	6.05	6.24	6.46	6.69	6.90
Pu alpha:Am-241	NA	4.39	2.46	1.82	1.51	1.33	1.22
Pu-241: Pu alpha	31.9	25.3	20.0	15.8	12.5	9.91	7.83

a. Time since separation of the Am-241 from the plutonium mix.

was <sup>241</sup>Pu measured as part of the intake, so the activity of that isotope is almost never available. Americium-241 at the time of intake was also often not determined directly. Since 1983, <sup>238</sup>Pu and <sup>239+240</sup>Pu were measured separately, so the ratio of one to the other can be used to estimate the category of the plutonium mixture and, from the tables, to estimate the activities of <sup>241</sup>Pu and <sup>241</sup>Am. Prior to 1983, the measured quantity was total alpha from plutonium, which means the total of <sup>238</sup>Pu and <sup>239+240</sup>Pu. Therefore, unless <sup>241</sup>Am was measured or there is other information about the intake, there might be no way to tell from the bioassay how much <sup>241</sup>Pu and <sup>241</sup>Am were present at intake.

Most plutonium mixtures handled at Hanford were nominally weapons grade and, if the <sup>239+240</sup>Pu-to-<sup>238</sup>Pu ratio implies weapons grade (e.g., >8), dose reconstructors should use the ratios in Table 5-4. However, if no information is available about the nature of the plutonium mixture associated with an intake, an assumption of 10-year-old fuel-grade plutonium mixture would be favorable to claimants and reasonable, except it is recommended to assume fresh plutonium for the 1944 through 1949 and 5-year-old plutonium for 1950 through 1954. For intakes since about 1996, 20-year-old fuel-grade mixture could be assumed.

There was at least one project in the 1970s involving irradiated fuel rods from commercial power reactors (Nuclear Waste Vitrification Project). The work took place in the 324 and 325 Buildings and the material was stored in the 303C Building in the 300 Area [22]. Commercial fuel rods have a much higher degree of "burn-up," and those at Hanford were characterized by much more <sup>241</sup>Pu and nominally 26% <sup>240</sup>Pu. Table 5-6 provides the activity characteristics of the commercial fuel used in the Nuclear Waste Vitrification Project. In addition, the Plutonium Finishing Plant sometimes recycled plutonium from other DOE sites. This material would be rich in <sup>241</sup>Am. Plutonium from the West Valley commercial reprocessing site is also stored at Hanford. However, unless the records concerning the specific intakes being investigated have evidence of these unusual mixtures, the default mixtures mentioned above should be used.

Table 5-6. Activity composition of Hanford reference commercial power fuel-grade plutonium mixture (Carbaugh 2003a).

Mixture designation:	Fresh	5-year	10-year	15-year	20-year	25-year	30-year
Years of aging <sup>a</sup> :	0	5	10	15	20	25	30
Specific activity in mixture (Ci/g)							
Pu-238	1.71E-01	1.64E-01	1.58E-01	1.52E-01	1.46E-01	1.40E-01	1.35E-01
Pu-239	3.41E-02						
Pu-240	5.90E-02	5.89E-02	5.89E-02	5.89E-02	5.89E-02	5.88E-02	5.88E-02
Pu-241	1.34E+01	1.05E+01	8.28E+00	6.51E+00	5.12E+00	4.03E+00	3.17E+00
Pu-242	1.97E-04						
Am-241	0	9.49E-02	1.69E-01	2.26E-01	2.79E-01	3.04E-01	3.31E-01
Pu-239+240	9.31E-02	9.31E-02	9.30E-02	9.30E-02	9.29E-02	9.29E-02	9.29E-02
Pu-alpha	2.65E-01	2.58E-01	2.52E-01	2.45E-01	2.39E-01	2.34E-01	2.28E-01
Total alpha	2.65E-01	3.53E-01	4.20E-01	4.71E-01	5.10E-01	5.38E-01	5.59E-01
Activity ratios							
Pu-239+240:Am-241	NA	0.981	0.551	0.411	0.344	0.305	0.281
Pu-239+240:Pu-238	0.544	0.568	0.589	0.612	0.636	0.664	0.688
Pu-241:Pu-239+240	144	113	89.1	70.0	55.1	43.3	34.1
Pu alpha:Pu-239+240	2.85	2.77	2.71	2.63	2.57	2.52	2.45
Pu alpha: Pu-238	1.55	1.57	1.59	1.61	1.64	1.67	1.69
Pu alpha:Am-241	NA	2.72	1.49	1.08	0.857	0.770	0.689
Pu-241: Pu alpha	50.6	40.7	32.9	26.6	21.4	17.2	13.9

a. Time since separation of the Am-241 from the plutonium mix.

If some of the plutonium bioassay was obtained prior to October 1983 and some after, the two data sets are not compatible; the prior-to-October-1983 results are the sum of activities from <sup>238</sup>Pu, <sup>239</sup>Pu, and <sup>240</sup>Pu; the post-October-1983 results are separate for <sup>238</sup>Pu and <sup>239+240</sup>Pu. Use known ratios if

available or default activity ratios to determine the isotopic components of the total alpha results before combining the data sets for curve fitting.

Plutonium at Hanford could have existed as absorption type M or S. Dose reconstructors should model <sup>241</sup>Am that is a component of plutonium contamination in the lung the same as the plutonium matrix in which it has ingrown. In other words, the americium should be treated as the absorption type S if the plutonium is type S.

#### 5.2.2 Americium

Americium was usually a trace contaminant in plutonium mixtures as discussed in Section 5.2.1. However, americium was separated from plutonium at the reprocessing plants [e.g., T Plant and S Plant (REDOX) in the early years, PUREX from 1956] and at the Plutonium Reclamation Facility (a wing in the Plutonium Finishing Plant). There was also an americium recovery operation in the Plutonium Finishing Plant from the late 1960s until 1976. Therefore, waste tanks, transfer lines, and parts of the Plutonium Finishing Plant had <sup>241</sup>Am that was chemically separate from plutonium. This americium should be treated as americium (as opposed to trace americium atoms bound in a plutonium matrix) [23]. The International Commission on Radiological Protection (ICRP)recommended absorption type for americium is M (ICRP 1994).

It has not been discovered yet when americium analyses first started. There is no mention of americium excreta analysis in Healy (1948); no mention in a 1954 memo, "Bioassay Annual Report," that lists numbers of urinalyses for plutonium, fission products, and uranium; and no mention in a compilation of bioassay procedures, titled "Bioassay Procedures and Analysis (Old Bioassay Bible)" (no author or editor), dated April 10, 1961.

The first americium records in the database show 41 urinalyses for <sup>241</sup>Am in 1964 collected from 19 workers. No americium urinalyses were recorded in 1965 or 1966. The samples were collected over a short time period. The first samples, 12 of them, were collected on August 25 or 26. The last sample was collected on October 29. The results range from  $1.3 \times 10^{-6}$  to  $6.4 \times 10^{-6}$  [24]. No units are given, although the magnitude of the results suggest the units are µCi/sample or µCi/L. It is not clear whether the units are per sample or per liter. Hanford used per liter from 1967 through July 1969, and because the volumes can vary from less than a liter to more than a liter, no particular choice is technically better than another; therefore, assuming the units are µCi/L is reasonable [25].

One set of four samples for one worker was collected as follow-up to a contamination event. Among the remaining workers, most were sampled on August 25 or 26, and then had at least one additional sample a month or two later. Therefore, it is likely that the August 25/26 samples were baselines used to establish the bioassay program at the start of the experiments that were being conducted to support a new <sup>241</sup>Am separation process at the Plutonium Finishing Plant [26].

No information about the radiochemical analysis method or detection level has been found. Assuming the August samples were baselines, a cumulative probability analysis was performed on the results that provided a median value of  $1.9 \times 10^{-6} \,\mu\text{Ci/L}$  and a 95th percentile of  $2.3 \times 10^{-6} \,\mu\text{Ci/L}$ . Based on this analysis, the MDA for  $^{241}$ Am analyses in 1964 was assumed to be  $4.6 \times 10^{-6} \,\mu\text{Ci/L}$  [27].

The records show no americium bioassay in 1965 or 1966; there are 168 results in 1967. The latter are a combination of urine and fecal results, and most of the results seem to be duplicated twice in the database. The duplication appears to be a mistake in the database that does not show up in hardcopy listings of these bioassay results. The results are all from special samples for six workers who were involved in the same potential intake accident in 222-S on May 2, 1967. There are internal

dosimetry evaluations in the workers' files documenting the accident and bioassay results. Many of the urine sample results were listed as  $<5.41 \times 10^{-7} \, \mu \text{Ci/L}$ , so it is assumed that this was the MDA for the analysis. This is the same number that was handwritten in the margin of an unpublished article, "Evaluation of Internal Depositions of Americium Using Bioassay Samples," by R. C. Henle, dated May 1968. The handwritten note says, "5.41 ×  $10^{-7} \, \mu \text{Ci/L}$  detection level."

A memorandum to file from John J. Jech (1969a), Senior Development Engineer in the Personnel Dosimetry Services, states that according to a telephone conversation with Matt M. Lardy at UST, the new detection limit for <sup>241</sup>Am is 2.0 dpm/sample as of July 10, 1969. Mr. Lardy described the procedure as dibutyl N,N-diethylcarbamylphosphonate (DDCP) extraction to a planchet and gross alpha counting (Bihl 2003). According to a letter from Matt Lardy to Harold Larson, manager of Personnel Dosimetry Services, a reduction in the detection limit was implemented in 1974 (Lardy 1974). The new limit for <sup>241</sup>Am in urine was given as 0.1 pCi/sample at the 90% confidence limit. This limit was still listed in a statement of work with UST in 1979 and again in 1982, although it was stated as 0.2 dpm/sample. In the laboratory statement of work for a new contract starting October 1983 (Battelle 1983), the detection level was listed as 0.04 dpm/sample. This improvement was achieved by use of an alpha/gamma coincidence counter.

Until October 1983, the gross alpha count could have included <sup>242</sup>Cm or <sup>244</sup>Cm if any were associated with the intake. Assuming the results are <sup>241</sup>Am is favorable to claimants. However, sometime between October 1983 and October 1985, both the chemistry procedure and the counting technique were changed. The chemistry method was similar to that described in the Health and Safety Laboratory 300 manual (HASL 1985 or similar year) and commonly referred to as the "RICH-RC-50-80" method (Bihl 2003). This method involved sequential precipitation with calcium oxalate and iron hydroxide, removal of plutonium using anion exchange, loading on another column with nitric acid and methanol, and elution of the americium with HCl and methanol. Electrodeposition and counting by alpha spectrometry were also implemented at this time (Bihl 2003). The MDA in the 1985 statement of work (Battelle 1985) was listed as 0.02 dpm/sample consistent with the change to alpha spectrometry, and it has stayed there to the present. At present, Eichrom transuranic column exchange is used for the separation of the americium for urine; however, the MDA is the same (Bihl 2003).

Table 5-7 summarizes what has been uncovered concerning <sup>241</sup>Am MDAs for routine urinalysis.

Table 5-7. Routine <sup>241</sup>Am urinalysis detection levels [28].

Period	MDA, dpm/sample	Decision level, dpm/sample
1964	4.6E-6 µCi/L	Anything detected
1967 to 6/1969	5.4E-7 µCi/L <sup>a</sup>	Anything detected
7/1969 to 2/1974	2.0	Anything detected
3/1974 to 10/1983	0.2	Anything detected
10/1983 to 9/1985	0.04	Anything detected
10/1985 to 05/1988	0.02	Anything detected
05/1988 to 06/1990	0.02	0.01
07/1990 to 10/1991	0.03	0.015
11/1991 to 4/2000	0.02	0.01
5/2000 to 8/2001	0.02	$X_{b} + 2.05 \times TPU^{b}$
9/2001 to present	0.02	2 x TPU

- a. Not known if MDA or decision level.
- b.  $X_b$  is the mean of the blanks and TPU is total propagated uncertainty. The  $X_b$  value used during that time was 0.00276 dpm for Am-241. However, it is recommended that dose reconstructors use 2 x TPU as the actual decision level for both periods.

The MDAs listed in Table 5-7 apply to routine and priority processing of urine samples. Fecal sampling was used for special sampling after potential intakes, and other processing codes (emergency and expedite) have been available for special urine and fecal samples. The contractual MDAs for these samples are provided in Table 5-8. These analyses could have been used because of suspected intakes of pure <sup>241</sup>Am (such as the explosion of an americium exchange column at the Plutonium Finishing Plant in 1976) or to determine the activity of <sup>241</sup>Am in a plutonium mixture. There is evidence of a few intakes of pure <sup>241</sup>Am prior to 1969, involving unusual circumstances such as using a supposedly sealed source that had ruptured. These intakes were analyzed by urinalysis, so obviously a procedure existed at that time, although not part of the contract with UST [29]. On rare occasions for a serious intake, samples were analyzed for <sup>241</sup>Am using a low-energy photon detector (LEPD) prior to any chemistry. This technique came into existence in 1986 or 1987. Its detection level was about 5 dpm/sample (Battelle Northwest 1987). In general, the LEPD result was used just as a rapid indicator, and a more accurate result was obtained by wet chemistry/alpha spectrometry

Table 5-8. MDAs for nonroutine <sup>241</sup>Am excreta analyses [31].

	Fecal sam	Fecal samples, MDA, dpm/sample			, dpm/sample
Period	Emergency <sup>a</sup>	Expedite	Priority	Emergency <sup>a</sup>	Expedite
1/1967 to 2/1974	(b)	NA	(b)	(b)	NA
2/1974 to 1981	(c)	NA	4	(c)	NA
1982 to 9/1983	3.6-12	NA	1.2-5.0	0.7-1.0	NA
	(3.6 most probable) <sup>d</sup>		(1.2 most probable) <sup>d</sup>	(0.7 most probable) <sup>d</sup>	
10/1983 to 9/1985	200	NA	0.16	1.0	NA
10/1985 to 6/1989	20	6	0.1	1	0.08
7/1989 to 10/1991 <sup>e</sup>	20	4	NA	2	0.4
11/1991 to present	20	6	0.1	1	0.08

- a. At times the emergency category was called "rush" and the routine category was called "normal."
- b. Probably available but MDAs not found.
- c. Emergency analyses were available on request, but the statement of work [based on 1978 statement of work (DOE-RL 1978)] did not specify the MDAs. It implied that an MDA about 10 times the routine (or priority for fecal) MDA was expected.
- d. Varied according to sample size over the range shown; the lower value was generally applicable except for very large samples (Larson 1982).
- e. Emergency and expedited processing of urine and fecal samples was available through the PNNL Analytical Chemistry Laboratory. Priority fecal analyses were also available through the offsite laboratories, but the MDA was not established, probably about 0.2-0.5 dpm/sample considering the state-of-the-art of those laboratories.

#### **5.2.3** Curium

days later [30].

The curium isotopes of concern were 242 and 244. Most sources of curium at Hanford were minor calibration sources or minor constituents in an actinide mixture; however, extraction of <sup>244</sup>Cm from high-level waste occurred at the 325 Building sometime in the 1970s. The curium and americium procedure was the same so the results would have been reported as curium only if so requested through the bioassay request system, until alpha spectrometry was initiated. After 1985, the chemistry is the same as americium, but <sup>241</sup>Am, <sup>242</sup>Cm, and <sup>244</sup>Cm were reported separately if requested (Battelle Northwest 1987). The MDAs were not always identical with <sup>241</sup>Am, however. Routine urinalysis MDAs for curium are provided in Table 5-9 and nonroutine excreta analyses are provided in Table 5-10.

#### 5.2.4 Tritium

The history of tritium urinalysis at Hanford is not well documented. Tritium urinalysis was not mentioned at all in Wilson (1987). The earliest report found to date on tritium urinalysis at Hanford dates to 1949 by Jack Healy, the leading internal dosimetrist at Hanford for many years (Healy 1949). That procedure was based on "production of acetylene from the active water, with subsequent measurement of the ionization caused by the tritium beta particle" (Healy 1949). No detection level

Table 5-9. Routine Cm urinalysis detection levels [32].

Period	MDA, dpm/sample	Decision level, dpm/sample		
7/1969 to 1981	Not spe	Not specifically mentioned		
1982 to 9/1983	Listed for em	ergency processing only		
10/1983 to 4/1988	0.02	Anything detected		
5/1988 to 6/1990	0.02	0.01		
6/1990 to 10/1991	0.03	0.015		
11/1991 to 4/2000	0.02	0.01		
5/2000 to 8/2001	0.02	$X_b + 2.05 \times TPU^a$		
9/2001 to present	0.02	2 x TPU		

a.  $X_b$  is the mean of the blanks and TPU is total propagated uncertainty. The  $X_b$  values used during that time were 0.00206 and 0.0000 dpm for Cm-242 and Cm-244 respectively. However, it is recommended that dose reconstructors use 2 × TPU as the actual decision level for both periods. [33]

Table 5-10. MDAs for nonroutine Cm excreta analyses [34].

	Fecal samples, MDA, dpm/sample			Urine samples, MDA, dpm/	/sample
Period	Emergency <sup>a</sup> Expedite Priority		Emergency <sup>a</sup>	Expedite	
Prior to 1982	(b)	NA	(b)	(b)	NA
1982 to 9/1983	10 <sup>c</sup>	NA	NA	0.5-1.0 (0.5 most probable) <sup>c,d</sup>	NA
10/1983 to 9/1985	240	NA	0.8	10	NA
10/1985 to 6/1989	240	70	0.8	1	1.2
7/1989 to 10/1991 <sup>e</sup>	NA	NA	NA	NA	NA
11/1991 to present	240	70	0.8	1	1.2

- a. At times the emergency category was called "rush" and the routine category was called "normal."
- b. Probably available but MDAs not found.
- c. Total alpha; would have included any americium present also.
- d. Varied according to sample size over the range shown; the lower value was generally applicable except for very large samples (Larson 1982).
- e. Emergency and expedited processing of urine and fecal samples was available through the PNNL Analytical Chemistry Laboratory. Priority fecal analyses were also available through offsite laboratories, but the MDA was not established; it was probably about 0.2-0.5 dpm/sample considering the state-of-the-art of those laboratories.
- f. At times the emergency category was called "rush" and the routine category was called "normal."
- g. Probably available but MDAs not found.
- h. Total alpha; would have included any americium present also.
- Varied according to sample size over the range shown; the lower value was generally applicable except for very large samples.
- j. Emergency and expedited processing of urine and fecal samples was available through the PNNL Analytical Chemistry Laboratory. Priority fecal analyses were also available through offsite laboratories, but the MDA was not established; it was probably about 0.2-0.5 dpm/sample considering the state-of-the-art of those laboratories.

was mentioned in that letter, but one was mentioned in an internal memo (Parker 1950) that referred to the acetylene method for urinalysis and provides a sensitivity of about 1.2 μCi/L in water. However, that method apparently did not work well because a 1951 letter stated that, "Your problem on the determination of tritium in the urine samples is one that we have been working on for the last two years, and have finally obtained what appears to be a decent method for routine use" (Healy 1951). The copy of the letter is of such poor quality that the method described is hard to follow, but it definitely was not liquid scintillation counting. A 1961 report entitled *The Estimation of Whole Body Dose from Tritium by Urine Analysis* indicated that liquid scintillation was used by that time, but again no detection level was given. Liquid scintillation counting was implemented for tritium bioassay at the Savannah River Site in 1958 and it is reasonable to expect that Hanford did so at about the same time. In the above-mentioned interview with Matt M. Lardy (Bihl 2003), Mr. Lardy stated that liquid scintillation counting of a 1-ml aliquot of raw urine has been used since UST was awarded the bioassay contract in 1965.

Tritium intakes were accounted for as part of external dose until about 1987, when they were entered in the dose database as an internal dose [35].

Tritium was referred to as P-10 in the 1950s (for example, Parker 1950). The main source of tritium in the 1950s was 108-B, also called the P-10 Plant, in which tritium was extracted from irradiated Li-Al targets intermittently between August 1949 and 1955. A 1967 report states, "Battelle-Northwest and its predecessor at Hanford, the General Electric Company, have been involved in activities with tritium since about 1950, initially as a manufactured product for weapons applications and later as a byproduct of heavy water reactor operations. Our most recent experience is from operation of the Plutonium Recycle Test Reactor (PRTR)" (McConnon 1967). There was also some work on a tritium target program in the 1990s in the 300 Area and tritium light sources in the 1980s (involving just a few people), and there has been low-level use of tritium as a tracer in various biology experiments. Tritium in the groundwater was a source of ingestion for 400 Area workers, principally the Fast Flux Test Facility, from about 1978 to 2003. See Section 5.6 for a more detailed discussion and guidelines for assigning intakes. Tritium exposure was assumed to be chronic during the exposure period, unless a very large acute intake was known to occur.

Very little data on MDAs has been discovered. McConnon (1964) states that a tritium bioassay result exceeding 5  $\mu$ Ci/L will be reported to the Radiation Monitoring Office the day after the samples are picked up, indicating a level of concern probably well above the MDA. One P-10 Personnel Sample Analysis card, with entries in 1952, shows several values below 5  $\mu$ Ci/L, with the smallest value being 2.5  $\mu$ Ci/L. None of the values are listed as less-than values. The 1965 statement of work with UST (AEC-RL 1964) shows an MDA of 1  $\mu$ Ci/L (which is consistent with the MDA at Savannah River Site throughout the 1950s). Table 5-11 lists the most accurate compilation of MDAs for routine tritium urinalysis to date. From 1978 to the present, the MDAs were obtained from statements of work with the bioassay laboratory.

Table 5-11. Routine tritium urinalysis detection levels [36].

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Period	MDA			
1949 through 1960 <sup>a</sup>	≈ 5 µCi/L			
1961 through 1981	1 μCi/L			
1982 through 10/1991	10 dpm/ml			
11/1991 to present	20 dpm/ml			

Dates and MDA are best guesses. The change in 1961 was based on earliest reference to liquid scintillation counting.

#### 5.2.5 Uranium

Uranium exposure at Hanford involved principally three physical forms: depleted (DU), natural (NU), and slightly enriched (also called recycled) uranium (RU). Uranium-233 was also isolated from irradiated thorium at PUREX from 1966 to approximately 1971. Small numbers of researchers may have experimented with more enriched uranium at different times (e.g. metallurgy on commercial-grade fuel), but such exposure would have been to small groups for limited periods. For instance, there was a radiation work permit issued for work with "bare, enriched – 37.5% – solid form uranium" for the 306 Building effective for November 9, 1970, through November 9, 1971 (Battelle Northwest 1970; Jech 1970a). Table 5-12 provides the default uranium mixtures that have been used at the Hanford Site since the 1980s (Carbaugh 2003a). However, different batches of uranium can have different isotopic mixtures; therefore, the default mixtures in the Integrated Modules for Bioassay Analysis (IMBA) codes and the Dose Reconstruction Project tools are acceptable. In general, personnel working in the production facilities (e.g., fuel fabrication, the reactors, fuel dissolution and plutonium processing, waste management) were exposed to natural uranium during operation of the early reactors (through 1951) and recycled uranium starting in February 1952 at the UO<sub>3</sub> Plant and July 1952 at the 300 Area Fabrication plants (DOE 2002). Recycled uranium had impurities build up

and track with the uranium over time. Recommended impurity levels based on tolerance specifications or higher than normal concentrations in some drums are provided in Table 5-13 (DOE 2000). Plutonium-239 can be assumed for the plutonium alpha impurity and <sup>232</sup>Th can be assumed for the thorium [37].

Table 5-12. Radiological characteristics of Hanford uranium mixtures.

U	Uranium mixture <sup>a,b</sup>						
Weight percentage	Recycled (RU)	Commercial fuel (CF)					
U-234	0.0082	0.0300					
U-235	0.9700	2.9600					
U-236	0.0680	Negligible					
U-238	98.9500	97.0100					
Specific constituent ac pCi/µg) <sup>c</sup>	tivity in mixture (µ	ıCi/g, nCi/mg, or					
U-234	0.5125	1.8750					
U-235	0.0210	0.0639					
U-236	0.0440	Negligible					
U-238	0.3325	0.3260					
Total	0.9099	2.2649					
Specific constituent ac	tivity in mixture (d	dpm/μg) <sup>c</sup>					
U-234	1.1378	4.1625					
U-235	0.0465	0.1419					
U-236	0.0977	Negligible					
U-238	0.7381	0.7236					
Total	2.0200	5.0281					
Constituent fraction of	total uranium acti	ivity in mixture					
U-234	0.5632	0.8279					
U-235	0.0230	0.0282					
U-236	0.0484	Negligible					
U-238	0.3654	0.1439					
Total	1.0000	1.0000					

- a. CF data from Rich et al. (1988).
- RU data based on average of data presented by Sula, Carbaugh, and Bihl (1991).
- c. Can be used to represent specific alpha activity in the mixture as well.

Table 5-13. Impurities in recycled uranium at Hanford.

Constituent	Maximum allowed <sup>a</sup>	Observed range <sup>b</sup>	Recommended level <sup>c</sup>
Plutonium	10 ppb U	<1–2 ppb U	0.8 nCi Pu-alpha/g U
Neptunium	Not established	0.04-0.16 ppm U	0.4 nCi Np-237/g U
Thorium	750 ppm U	8–10 ppm U	5 pCi Th-232/g U
Tc-99	Not established	3–4 ppm U	0.2 uCiTc-99/g U
Ru-103, -106	<20 μCi/lb U	<6 μCi/lb U	40 nCi Ru-106/g U
ZrNb-95	<10 µCi/lb U	<4 μCi/lb U	20 nCi ZrNb-95/g U <sup>d</sup>
Other gamma emitters	<2 μCi/lb U	0.09–0.75 μCi/lb U	Negligible

- a. From UO<sub>3</sub> Plant operating specifications (Sula, Bihl, Carbaugh 1989).
- b. From analysis of uranium lots 88-1, 88-2, and 88-3 that were processed in 1988, and lots 93-01, 93-02, 93-03, 93-04, and 93-05, processed in 1993.
- c. The recommended levels are expected to result in a slight overestimate of dose compared to levels actually observed in 1988 and take into account other information presented in the DOE investigation of recycled uranium at Hanford (DOE 2000). The plutonium reference level was based on the 10 ppb specification, which was reached or exceeded in a few drums throughout Hanford history (DOE 2000).
- d. Interpret as 10 nCi each of the two radionuclides. [38]

Uranium compounds at Hanford ranged from very soluble uranyl nitrate and soluble  $UO_3$  to relatively insoluble  $UO_2$  and  $U_3O_8$ . Dissolution tests in simulated lung fluid were conducted on samples from the major uranium handling facilities. Results are shown in Table 5-14. Because the relationship between the old lung fluid studies and the ICRP Publication 66 absorption types (ICRP 1995) is not established, Table 5-14 also shows recommended absorption types for intakes from the listed facilities, which dose reconstructors should use unless person-specific data are available. These absorption type assumptions should be applied to the impurities as well.

Table 5-14. Inhalation class for Hanford uranium compounds.

ICRP 30 inhalation class from lung fluid studies	Compound and location	Recommended ICRP 66 lung absorption type
80% D	Hanford UO <sub>3</sub> Plant smear sample dissolution study in 1984 <sup>a</sup>	(b)
20% W	(UO <sub>3</sub> powder)	
10% D	Hanford 303-M Building air sample dissolution study <sup>c</sup> (300	(b)
90% Y	Area Uranium Fuel Production Facilities)	
29% D	Hanford 333 Building air sample dissolution study <sup>c</sup> (300 Area	(b)
71% Y	Uranium Fuel Production Facilities)	
20% D	Hanford 306-W Building Machine Shop air sample dissolution	(b)
80% Y	study <sup>c</sup>	
	Uranyl nitrate at PUREX or UO₃ Plant	F
	UCI <sub>4</sub>	M <sup>d</sup>
	U carbonate (assumed form after discharge to the soil)	F <sup>d,e</sup>

- a. Sula, Bihl, and Carbaugh (1989).
- b. Because the conversions from the solubility studies to the ICRP (1995) absorption types are not exact, the dose reconstructor can use the same percentages for D to F, W to M, etc., or can just use the predominant form to maximize dose to the organ of concern; for instance, the 303-M Building uranium might be considered 10% F, 90% S, or all type S.
- c. Fisher (1986).
- d. Long (1993)
- e. Cooke and Holt (1974). [39]

A note about sampling of  $UO_3$  Plant workers: Because chemical toxicity was the principal concern for uranium exposures at the  $UO_3$  Plant, one sampling scheme used was to obtain both a Friday evening sample and Monday morning sample. The period of this sampling scheme was not established, other than in the 1970s and maybe earlier. This scheme was changed to Monday-morning-only sampling about the early 1980s. Changeover should be clear in the records.

The Friday/Monday sampling scheme was also used in 1962 and 1963 for 313 and 314 Building workers.

Wilson (1987) states that the uranium urinalysis program prior to 1948 was not reliable. The fluorometric method, which fused uranium from raw urine with sodium fluoride and measured the fluorescence when the compound was exposed to ultraviolet light, was implemented sometime during the first half of 1948 (Healy 1948; Wilson 1987). This method was used for elemental uranium analyses, with refinements over the years including some upfront chemistry on the raw urine, until about 1991, when it was replaced by kinetic phosphorescence analysis (KPA; Bihl 2003). (Note: Mr. Lardy said "about 1990" but other evidence indicates late 1991.) Lardy (1970) describes two procedures: one with wet ashing with nitric acid and hydrogen peroxide, then acidification and counting of a 100 µL aliquot with a detection level of 0.5 µg/L; the other with extraction (after wet ashing) with methyl isobutyl ketone and ammonium hydroxide. The detection limit for the latter was listed as 0.05 µg/L but the recoveries were about the same for both methods, so the latter must have used a 10 times larger aliquot. Based on requirements in later statements of work, it is assumed that the first method was used for routine analyses. Lardy (1970) also describes a third method – a radiometric procedure using the same separation chemistry as the second procedure, but the sample "is measured by a gas flow proportional counter or a ZnS (Ag) scintillation counter." The detection limit was given as 0.5 dpm/sample. A 1989 description of the chemistry was wet ashing with HCl and

extraction with hexone (Carbaugh 1989). A 100-ml aliquot was used, but the results were reported as per total sample. The chemistry for the KPA involves a 50-ml aliquot that is wet-ashed with acid, passed through an ion exchange column, then eluted with weak acid. Results are reported as per total sample.

When alpha spectrometry was introduced in 1983, two uranium urinalyses procedures were offered: the elemental procedure discussed above and the alpha spectrometric procedure to provide isotopic results. In general, the elemental procedure was used for workers exposed to natural or slightly enriched forms of uranium, and the isotopic procedure was used for depleted or more than slightly enriched forms of uranium. In general, personnel working in the production facilities were monitored by the elemental analysis, whereas PNL workers were monitored by the isotopic analysis because of the wide scope of research projects that occurred over the years [40].

Alpha spectrometry cannot differentiate between <sup>233</sup>U and <sup>234</sup>U. Prior to 1994, the results for this region of the alpha spectrum were reported as <sup>233</sup>U; they have been reported as <sup>234</sup>U from 1994 to the present unless it was specifically determined that the worker was exposed to <sup>233</sup>U. Work with <sup>233</sup>U was rare after the early 1970s, long before alpha spectrometry came into use for bioassay. Therefore, unless specifically mentioned in an intake investigation report, assume <sup>233</sup>U results since 1983 are actually <sup>234</sup>U [41].

Table 5-15 summarizes the routine urinalysis detection levels and Table 5-16 summarizes nonroutine detection levels.

Table 5-15 Routine uranium urinalysis detection levels

	Eleme	ntal	Iso	topic
Period	MDA, μg/L	Decision level, µg/L	MDA, dpm/sample	Decision level, dpm/sample
Prior to 1948	Not specifically	/ mentioned	NA	NA
1948 through 1949	10	Anything detected	NA	NA
1950 through 1974 <sup>a</sup>	4 (reporting level)	(b)	NA	NA
1974 through 1981	0.4	Same as MDA	NA	NA
1982 through 9/1983	0.05 – 0.25 (0.1 most probable) <sup>c</sup>	Same as MDA	NA	NA
10/1983 through 12/1983	0.03 μg/sample	0.5 μg/sample <sup>d</sup>	0.035	Same as MDA
1/1984 through 8/1985	0.03 µg/sample	Not known	0.02	Same as MDA
9/1985 through 6/1990	0.03/0.5 μg/sam <sup>e</sup>	0.2 µg/sample <sup>t</sup>	0.02	Same as MDA
6/1990 through 10/1991	0.2/0.5 μg/sam <sup>e</sup>	0.2	0.03	0.15/0.015 <sup>g</sup>
11/1991 through present	0.06/0.5 μg/sam <sup>e</sup>	0.2 <sup>†</sup>	0.02	0.15/0.010 <sup>9</sup>

- Wilson 1958. Also Lardy (1970) indicates that the MDA from at least 1970 on was 0.5 µg/L but the database shows that the use of 4 µg/L continued as a reporting level.
- b. In 1958-1966, values were reported well below the 4 μg/L value but it is not known why (i.e., was the MDA actually much lower or was the laboratory just reporting all values as measured without truncation).
- MDAs were based on sample size, but 0.1 μg/L applied to most sample sizes (Larson 1982).
- d. Values below this were recorded but not followed up as occupational intakes.
- e. The larger value is the MDA for a special (rapid) analysis for UO<sub>3</sub> Plant workers based on potential chemical toxicity. The need for this special analysis ceased in 1994 after the last processing in the UO<sub>3</sub> Plant [42].
- f. Based on upper level for natural background excretion. See text for discussion.
  g. First value applied to <sup>234</sup>U and <sup>238</sup>U; second value applied to <sup>235</sup>U based on natural background in urine. In 2002, the <sup>235</sup>U decision level was lowered to 0.007 dpm (Carbaugh 2003b).

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Table 5-16. MDAs for nonroutine uranium excreta analyses [43].

	Analysis	Fecal samples, MDA, dpm/sample			Urine samples, MDA, dpm/sampl	
Period	type	Emergency <sup>a</sup>	Expedite	Priority	<b>Emergency</b> <sup>a</sup>	Expedite
1980 <sup>b</sup>	Elemental	NA	NA	NA	10 μg/L	NA
10/1983 to 9/1985	Elemental	8 μg/sample	NA	0.3 µg/sam	7 μg/sam	NA
	Isotopic <sup>c</sup>	12 dpm/sam	NA	0.3 dpm/sam	1 dpm/sam	NA
10/1985 to present	Elemental	8 μg/sam	5 μg/sam	0.3 µg/sam	7 μg/sam	0.5 µg/sam
	Isotopic	12 dpm/sam	4 dpm/sam	0.3 dpm/sam	1 dpm/sam	0.12 dpm/sam
6/1990 to 10/1991 <sup>d</sup>	Elemental	20 μg/sam	4 μg/sam	NA	20 μg/sam	4 μg/sam
	Isotopic	20 dpm/sam	4 dpm/sam	NA	2 dpm/sam	0.4 dpm/sam
11/1991 to present	Elemental	8 μg/sam	5 μg/sam	0.3 µg/sam	7 μg/sam	0.5 µg/sam
	Isotopic	12 dpm/sam	4 dpm/sam	0.3 dpm/sam	1 dpm/sam	0.12 dpm/sam

- a. At times the emergency category was called "rush" and the routine category was called "normal."
- b. Earliest mention found in a contract with the laboratory. The ability to perform analyses on fecal samples was also mentioned but an MDA was not specified.
- c. U-234, U-235, U-238.
- d. Emergency and expedited processing of urine and fecal samples was available through PNNL's Analytical Chemistry Laboratory. Priority fecal analyses were also available through the offsite laboratories but the MDA was not established, probably about 0.2-0.5 dpm/sample considering the state-of-the-art of those laboratories.

Starting about 1995, mass spectrometry has been used as an investigational tool to discriminate between natural background uranium and recycled uranium through measurement of  $^{236}$ U. The presence of  $^{236}$ U confirms an occupational intake of recycled uranium; the detection limit for  $^{236}$ U is such that urinary excretion of uranium greater than 0.2  $\mu$ g/L (see discussion of natural background excretion below) from an intake of recycled uranium should have a detectable amount of  $^{236}$ U (Carbaugh 2003a, MacLellan 1995).

Natural uranium from nonoccupational intakes (primarily food and water) is excreted in urine at levels above the analytical MDAs for either the elemental uranium analysis or the alpha spectrometry analysis. The  $^{234}$ U-to- $^{238}$ U ratio can be used to distinguish depleted uranium from natural uranium but, considering uncertainties in analytical results, that ratio cannot be used to distinguish recycled uranium. Three studies were conducted, in 1985, 1990, and 1995, to establish the range of natural background excretion in unexposed persons living near the Hanford Site. The third study purposely looked for possible geographic and seasonal differences in the background. All studies found natural excretion to be lognormally distributed. Although the 50th percentiles and slopes of the excretion curves were different in the studies, each study found 0.2  $\mu$ g/d to be about 99 to 99.9 percentile. The 1995 study had one result that greatly exceeded the 0.2  $\mu$ g/d value (Carbaugh 2003a).

Therefore, 0.2  $\mu$ g/d was established in 1985 and continues to be used at present as the environmental decision level for exposures to natural or recycled uranium. Only urinary excretion values greater than 0.2  $\mu$ g/d, which converts to 0.15 dpm/d for <sup>234</sup>U and <sup>238</sup>U and 0.007 dpm/d for <sup>235</sup>U, are considered indicative of a potential occupational source. Nevertheless, the one result in the 1995 study and many worker-specific investigations of urinary results exceeding 0.2  $\mu$ g/d have shown that results well above the environmental screen level do occur from natural sources. Some of these were shown to be due to a specific home water well; others occurred from workers on city water from wells (but apparently not all wells).

It is reasonable to use the urinary excretion values of  $0.2 \,\mu\text{g/d}$  for elemental analyses,  $0.15 \,\text{dpm/d}$  for  $^{234}\text{U}$  and  $^{238}\text{U}$  and essentially anything detected for  $^{235}\text{U}$ , to distinguish between natural background and potential occupational exposure for natural and recycled uranium, unless the worker's file shows  $^{236}\text{U}$  results or other studies show the excretion was from natural sources. These environmental decision levels should apply to the entire history of Hanford. Prior to 1985, there will undoubtedly be excretion values exceeding the environmental screening levels that were nevertheless due to natural sources, but it is unlikely there will be data available to prove it.

Background excretion of uranium in feces probably varies over an even larger range than urinary excretion; however, a definitive study for the Hanford area has not been conducted. Fecal samples were rarely obtained for potential uranium intakes; when they were, the investigation report should discuss how the results were interpreted.

Separation of <sup>233</sup>U from thorium occurred in three distinct campaigns: a small process test in 1965 and production runs in 1966 and 1970. A total of 820 kg of <sup>233</sup>U were produced (Walser 1978, Isochem Inc. 1967). See Section 5.3.4 for a discussion of the program to create <sup>233</sup>U by irradiation of thorium.

The  $^{233}$ U was recovered at PUREX in the nitrate form so it would have been absorption type F. Conversion to  $U_3O_8$  at Z Plant was discussed in the planning stage but whether that was actually done has not been verified yet (Tomlinson 1964). The Hanford bioassay database shows that a few urinalyses were obtained for  $^{233}$ U, mostly in 1970. No information has been found about how the analyses were performed or the units of the recorded results. Copies of an earlier database show the units to be  $\mu$ g/L, consistent with the units for all uranium analyses at that time; however, the recorded values are much too small to realistically be in  $\mu$ g/L, with the exception of the single value in 1969. The recorded values appear to be similar to plutonium alpha measurements at that time; and it is probable but not verified yet that autoradiography was used for these bioassay because of the high specific activity of the  $^{233}$ U and the major contaminant  $^{232}$ U [44].

There were contaminants in the <sup>233</sup>U. Specifications for contaminants in the <sup>233</sup>U product were more lax than for RU. The <sup>232</sup>U contaminant specification was stated as 6 ppm in the Tomlinson (1964) planning document but was listed as 8 ppm in the specifications for the first campaign (Isochem, Inc. 1967). However, the <sup>232</sup>U component would have been accounted for in the bioassay if the bioassay was in terms of either micrograms or total alpha activity (less than 2% of the total uranium activity would have come from the <sup>232</sup>U). The <sup>228</sup>Th (half-life 1.9 yr) progeny of <sup>232</sup>U would have built up slowly in the final product, but this would have occurred after the product was packaged so it would not have been a significant source of intake during processing or packaging. Other contaminant specifications were also stated in the Tomlinson planning document and updated by the Isochem document. If an intake of <sup>233</sup>U is assigned for an Energy Employee, add intakes of 62 pCi <sup>239</sup>Pu, 2 pCi <sup>95</sup>ZrNb, 4 pCi <sup>106</sup>Ru, 1 pCi <sup>233</sup>Pa, and 0.21 pCi <sup>237</sup>Np per microgram of <sup>233</sup>U (6.4 × 10<sup>-3</sup> pCi <sup>239</sup>Pu, 2.1 × 10<sup>-4</sup> pCi <sup>95</sup>ZrNb, 4.1 x10<sup>-4</sup> pCi <sup>106</sup>Ru, 1.0 x10<sup>-4</sup> pCi <sup>233</sup>Pa, and 2.2 × 10<sup>-5</sup> pCi <sup>237</sup>Np per pCi <sup>233</sup>U). [Note: the <sup>106</sup>Ru actuals exceeded the specification in about 15% of the batches as reported in the Isochem document hence the highest actual was used.]

See Section 5.6 for directions on how to apply intakes of <sup>233</sup>U for unmonitored workers, bearing in mind that only a few workers were monitored specifically for <sup>233</sup>U and only in 1970. [Workers may have been monitored via normal uranium urinalyses, but that method would not have had sufficient sensitivity for intakes of pure <sup>233</sup>U.]

#### 5.2.6 <u>Fission Product Analysis</u>

Fission product urinalysis was the method used to monitor for intakes of fission products until the implementation of whole-body counting in 1960. Routine fission product urinalyses started in January 1947, but ferrous hydroxide precipitation was used on the supernatant from the plutonium lanthanum fluoride procedure, and the results were erratic with occasional breakthrough of <sup>40</sup>K. Therefore, data prior to 1948 should be considered unreliable and ignored. The procedure initiated in 1948 was to add strontium carrier to the aluminum oxide solution for the plutonium procedure, then precipitate lanthanum hydroxide. This procedure was shown to extract the rare earths and strontium with yields ranging from 90% for cerium to 23% for strontium. The dried planchet was counted for beta activity with an approximate detection level of 30 dpm (Healy 1948; Wilson 1987). The same procedure was

in use in 1954 with the addition of a cerium carrier. It was also listed in the compilation of procedures referred to as the "Old Bioassay Bible" in 1961, but that same compilation had a separate procedure for  $^{90}$ Sr in urine. A memo in the Old Bioassay Bible discusses the start of use of a gas-flow, beta proportional counter in November 1958, which resulted in increased counting efficiency. The new detection limit was stated as  $1.4 \times 10^{-5} \,\mu$ Ci/sample, based on the counting efficiency of  $^{90}$ Sr. "Gross fission products" are also mentioned by Lardy (1970) with a brief description that seems to imply the same procedure was still available, although probably not used much. The detection level was given as 5 dpm/sample based on the beta counting efficiency for  $^{90}$ Sr. Table 5-17 summarizes the most accurate detection levels for the fission product urinalysis found to date.

Table 5-17. Routine fission product urinalysis detection levels [45].

Period	MDA
1948 to 2/1956	60 dpm/sample <sup>a</sup>
3/1956 to 10/1964	70 dpm/sample <sup>b</sup>
1965 to approx. 1969	31 dpm/sample
1970 <sup>c</sup>	5 dpm/sample

- Reported as 30 dpm/sample but that value was more of a decision level than an MDA.
- b. Recorded as 3.1 or 3.17  $\times$  10<sup>-5</sup>  $\mu$ Ci/sample.
- c. Listed in the bioassay contract but probably not used; replaced by whole body counting and <sup>90</sup>Sr urinalyses.

It is a challenge to interpret the fission product urinalysis in a way that is meaningful as representative of all the possible fission products and activation products to which a worker could have been exposed. The procedure separated and counted radionuclides of alkaline earths and rare earths, such as strontium, yttrium, barium, lanthanum, cerium, europium, promethium, zirconium and niobium. It did not account for radionuclides of ruthenium, cesium, zinc, cobalt, or manganese. The radiochemical yield of the elements that were carried through to the final planchet varied, however, from about 23% for strontium to about 90% for cerium (Healy 1948). The abundances of all the fission products, relative to each other, varied (considerably) as a function of the time from when the reactor fuel was removed from the core and allowed to cool to when the contamination was inhaled or ingested. OTIB-0054, Fission and Activation Product Assignment for Internal Dose-Related Gross Beta and Gross Gamma Analyses, provides guidance on how to interpret fission product urinalysis results. See also Section 5.4 for a discussion about interpreting fission product mixtures.

After whole body counting came into routine use, regular use of the fission product urinalysis continued for many workers at facilities such as B Plant and Semi-Works, where intakes of pure <sup>90</sup>Sr were possible. Therefore, it was apparently being used as a <sup>90</sup>Sr bioassay. The records show the use of fission product analysis in this way until early 1964. The same workers show actual <sup>90</sup>Sr analysis results starting in 1965, probably starting with the new contract with UST. For 1960 through 1964, fission product urinalysis results can be interpreted as <sup>90</sup>Sr [46].

### 5.2.7 Strontium

Records of <sup>90</sup>Sr urinalyses, both routines and specials, begin to appear in the database in 1965. However, the compilation of procedures called the Old Bioassay Bible, 1961, had a procedure specific for strontium in urine and fecal salts that included counting total strontium and then allowing for <sup>90</sup>Y ingrowth, yttrium separation, and counting of <sup>90</sup>Y to account for <sup>90</sup>Sr separate from gross strontium beta, if desired. This procedure was also mentioned in a memorandum, dated July 1963, documenting discussions between the Analytical Laboratories and Internal Dosimetry clarifying logistics of handling these samples and reporting <sup>90</sup>Sr results. Handwritten notes on this memo

indicate that the detection level is about 20 dpm. Nevertheless, the database records show the use of fission product urinalyses into 1964 and <sup>90</sup>Sr urinalyses apparently starting in 1965. The database contains frequent entry values of  $1.67 \times 10^{-5} \,\mu\text{Ci/L}$  (37 dpm/L) during 1965 and 1966, which seems to be the reporting level. This is consistent with a draft of the first contract with UST (the official version has not been found), dated August 1964, that listed a detection limit for 90Sr as 25 pCi/1.5L, which converts to 56 dpm/1.5L or 37 dpm/L (AEC-RL 1964). Lardy (1970) states that the detection limit is 1 pCi/L (2.2 dpm/L) (at 90% confidence), and describes the procedure as precipitation as the oxalate, then nitrate, removal of yttrium and barium, then reprecipitation as the carbonate and gross beta counting on gas flow proportional counters. A 1974 letter discussing terms of the statement of work with UST shows an "analytical limit" (defined as ±25%) at 50 dpm/sample and a reporting level of 2 dpm/sample (ERDA 1975). These values appear again in the 1978 statement of work (DOE-RL 1978) except the analytical limit is defined as ±100%. A letter (Robinson 1979) to R. B. Swoboda (UST bioassay supervisor) requests changes for <sup>90</sup>Sr urinalyses to lower the analytical limit (±100%) from 50 to 5 dpm/sample, increase the reporting level from 2 to 5 dpm/sample, and add an emergency analysis capability with an analytical limit of 10 dpm/sample and reporting level of 5 dpm/sample. In 1982, the detection limit was listed as 2.5 dpm/sample for <sup>90</sup>Sr and 5 dpm/sample for <sup>89</sup>Sr. The contract starting in October 1983 (Battelle Northwest 1983) lists the detection limit as 2.0 dpm/sample, and it stayed at that value until 1992 when it was raised to 10 dpm/sample (Battelle Northwest 1992). However, the procedure stayed the same throughout this period and the true MDA probably held at about 2 dpm/sample.

Beginning in 1982, actual analytical results were recorded in the database; that is, results were not truncated at a reporting level.

The results of the <sup>90</sup>Sr procedure usually were reported as <sup>90</sup>Sr although sometimes a value for <sup>89</sup>Sr was also reported. Sometime in the 1980s a shortcut was added to the procedure that allowed skipping the <sup>90</sup>Y ingrowth portion of the procedure if the first beta count was less than 1 dpm [47]. When this happens, the result is reported as Sr total or SRTOT, but the result may be interpreted as <sup>90</sup>Sr. These results were below the required detection level anyway. Table 5-18 summarizes the routine urinalysis detection levels for <sup>90</sup>Sr procedure.

Table 5-18. Routine 90Sr urinalysis detection levels [48].

Period	MDA or MDC
Prior to 1965	Might have been available but MDA not known
1965 to 1969	1.67E-5 μCi/L (37 dpm/L)
1970 to 1974	1E-6 μCi/L (2.2 dpm/L)
1975 to 3/1979	50 dpm/sample <sup>a</sup>
4/1979 to 1981	5 dpm/sample
1982 to 9/1983	2.5 dpm/sample
10/1983 to 6/1990	2 dpm/sample
9/1990 to 11/1991	30 dpm/sample
11/1991 to present	10 dpm/sample <sup>b</sup>

- Based on an unusual definition of "analytical limit" and probably conservative on the high side. Results <2 dpm were reported as 2 dpm; results >2 dpm were reported as measured.
- b. Decision level was 5 dpm/sample. Prior to that time, the MDA was also used as the decision level.

Dose reconstructors should consider all strontium at Hanford to be absorption type F [49]. It is favorable to claimants to assume that <sup>90</sup>Sr and total radiostronium results are <sup>90</sup>Sr even though <sup>89</sup>Sr might be present.

#### 5.2.8 Promethium

Hanford was involved in the manufacture of heat sources using <sup>147</sup>Pm. The period seems to start in 1966 and continue into the early 1970s (Howell and King 1968). The high activity work (kilocuries) took place in the 325 Building, but some exposure apparently occurred as early as 1962 or 1963 in the 222-S Chemistry Laboratory [50] and the 325 Building and as late as 1971 in the 308 Fuels Laboratory (Howell, 1969; Glover 1964). The database shows only seven <sup>147</sup>Pm urinalyses from 1972 to 1975 but an upswing in the number of analyses from 1976 to 1979, so perhaps the project ended and restarted in 1976 for a few years. In addition, animal studies were conducted with <sup>147</sup>Pm as part of research to develop a human biokinetic model for the behavior of promethium in the body, so animal tenders might have been exposed to low levels of <sup>147</sup>Pm. A small human volunteers study (14 volunteers) using <sup>143</sup>Pm was conducted in 1967 or 1968 (Palmer, Crook, and Nelson 1969, 1970) (12 volunteers: injection of 0.1 μCi each; 2 volunteers: ingestion of 10 μCi each). Another study on the behavior of promethium used swine with <sup>148m</sup>Pm as the surrogate for <sup>147</sup>Pm (McConnon et al. undated).

The work on the heat sources involved converting promethium/cerium nitrates into Pm<sub>2</sub>O<sub>3</sub> by separation chemistry then calcining (Howell and King 1968). There was also one mention of coldpressed, sintered Pm<sub>2</sub>O<sub>3</sub> for heart implants. According to ICRP (1995), the nitrate form should be considered absorption type M and the oxide form absorption type S.

The units in the database are µCi/L for urine and µCi/kg for feces through 1974 and dpm/sample from 1975 to the present. Table 5-19 lists the <sup>147</sup>Pm minimum detection levels at various times.

Table 5-19. Routine <sup>147</sup>Pm urinalysis detection levels [51].

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Period	MDA or MDC
Prior to 1965	Might have been available but MDA not known
1965 to 3/25/1970	1.67E-5 μCi/L (37 dpm/L)
3/31/1970 to 1974	1.0E-5 μCi/L (22 dpm/L)
1975 to 1979	50 dpm/sample <sup>a</sup>
1980 to 1981	20 dpm/sample
1982 to 9/1983	5 dpm/sample
10/1983 to 6/1990	4.0 dpm/sample
11/1991 to present <sup>b</sup>	30 dpm/sample

- a. Based on an unusual definition of "analytical limit" and probably conservative on the high side. Results < 25 dpm were reported as 25 dpm; results > 25 dpm were reported as measured.
- b. No <sup>147</sup>Pm analyses were performed during 1990 and 1991.

Fecal samples were analyzed for <sup>147</sup>Pm for some of the potential intake events in the late 1960s. The MDA or at least the lowest reporting level appears to be 1.67 x 10<sup>-5</sup> uCi/kg [52]. An MDA for fecal samples does not appear in laboratory statements of work during the 1970s, but reappears in the 1980s: 28-110 dpm/sample in 1982 depending on sample size (roughly 400 dpm/kg); 220 dpm/sample from 1983 to the 1990s [53].

Only one description of the procedure was found, and that procedure appeared in documents dated 1970, 1974 and 1977 (Lardy 1970, UST 1974, 1977). Promethium and rare earths were precipitated as the fluoride. Interferences such as zirconium, scandium, and IV actinides were removed by extraction by TTA in xylene, first at pH less than 1, then at pH about 4. The final sample was counted by liquid scintillation. Remaining rare earths were distinguished from <sup>147</sup>Pm by proper setting of the counting window on the liquid scintillation spectrometer.

## 5.2.9 Polonium

Considerable activity toward initiating a bioassay procedure and establishing a biokinetic model for <sup>210</sup>Po was found in the files from about 1968 through the mid-1970s. There is an indication of work with pure <sup>210</sup>Po in the 308 Building in 1968 and again in 1975. Whether the work in the 308 Building was continuous through that period or just in those two years was not determined. Inference can be made that there was work somewhat prior to 1968 based on a handwritten note documenting a telephone conversation in November 1967 during which it was stated that the <sup>210</sup>Po starts in the process in the soluble form but is converted to the insoluble form (Henle undated). However, UST was asked to develop a bioassay procedure in March 1968 and did so shortly thereafter, so apparently concern for possible intakes became important in early 1968. In addition, work with <sup>210</sup>Po in the 325 Building started in June 1972 and was scheduled "to run for 3.5 years" (Jech 1972).

The procedure developed for <sup>210</sup>Po by UST in March 1968 was as follows: for urine, gold, mercury, platinum, and tellurium were removed by reduction in hydrazine in an HCl solution. Iron was removed by reduction with ascorbic acid. The polonium was then removed from solution by deposition on silver film by heating at 95°C for 2 hr. The silver film was counted by alpha proportional counting. Fecal samples were first wet-ashed in concentrated nitric acid and peroxide then treated the same as urine samples. Sometime between 1968 and 1974, copper foil replaced silver foil and alpha spectrometry counting replaced proportional counting (UST 1974). Detection limits for routine urinalysis are shown in Table 5-20 and for nonroutine excreta bioassay in Table 5-21.

Table 5-20. Routine <sup>210</sup>Po urinalysis detection levels [54].

Period	MDA or MDC
3/1968 to 1970	5.4E-7 μCi/L
1971 to 1974	5.0E-7 μCi/L
1975 to 1979	1 dpm/sample <sup>a</sup>
1980 to 9/1983	0.1 dpm/sample <sup>a</sup>
10/1983 to present	No longer listed in the contract except for expedited or
	emergency samples. Probably not used.

Based on an unusual definition of "analytical limit" and probably conservative on the high side. Reporting level listed as 0.5 dpm/sample.

Table 5-21. MDAs for nonroutine <sup>210</sup>Po excreta analyses [55].

	Fecal samples, MDA, dpm/sample			Urine samples,	MDA, dpm/sample
Period	<b>Emergency</b> <sup>a</sup>	Expedite	Priority	<b>Emergency</b> <sup>a</sup>	Expedite
3/1968 to 1973	NA	NA	5.4E-7 µCi/kg	NA	NA
1974 to 9/1983	NA	NA	(b)	NA	NA
10/1983 to 9/1985	340	NA	NA	0.8	NA
10/1985 to 6/1989	340	100	NA	0.8	0.1

a. At times the emergency category was called "rush" and the routine category was called "normal."

Because <sup>210</sup>Po is a natural radionuclide from the <sup>238</sup>U decay chain, <sup>210</sup>Po exists naturally in urine and feces. Nothing was found in the records indicating that a study on natural excretion levels for persons living around Hanford had been conducted.

Bismuth slugs were irradiated in Hanford reactors to produce <sup>210</sup>Po starting in 1945 and continuing into the early 1950s; however, it appears that the slugs were promptly shipped offsite (Tilley 1945, DOE 1997). No evidence has been found that extraction or handling of concentrated <sup>210</sup>Po was conducted at Hanford during the early years.

b. Probably available but not listed in the contract.

#### 5.2.10 Neptunium

At PUREX from 1958 through 1972, <sup>237</sup>Np was removed from the dissolved fuel, purified, and packaged for shipment offsite. It was downloaded from an ion exchange column and packaged in liquid form, but the chemical form has not been discovered yet. Although mostly <sup>237</sup>Np by mass, the small mass of <sup>238</sup>Pu in the product produced most of the radioactivity. Plutonium bioassay was considered sufficient to monitor for intakes [56].

The database contains only four bioassay samples for  $^{237}$ Np in the 1970s, all for PNNL workers; all four are baseline samples. No routine samples or ending work samples were collected. The value for all four samples was 1 × 10<sup>-7</sup> with no units shown. The practice at the time was to record the activity of excreta samples in units of  $\mu$ Ci, but it has not been determined to date if the units are  $\mu$ Ci/sample or  $\mu$ Ci/L. No information about the analysis method has been found [57].

A few sample results appear in the database in the 1980s, mostly for PNNL employees. The units are in dpm/sample [58]. The 1983 statement of work with UST (Battelle Northwest 1983) shows a nonroutine analysis for <sup>237</sup>Np with contractual detection levels of 0.02 dpm/sample for urine samples and 0.1 dpm/sample for fecal samples, which were consistent with alpha spectrometry for plutonium and americium at the time.

### 5.2.11 Other Limited-Exposure Radionuclides

Hanford has always been a center for research, first as part of Hanford Works, then (1965 to present) as part of Pacific Northwest National Laboratory. As such, small-scale (in terms of either the number of persons or activity of the source) use of various radionuclides not addressed above has occurred throughout the history of the Hanford Site. The following discussion, addressing <sup>14</sup>C, <sup>232</sup>Th, radon, <sup>90</sup>Y, <sup>227</sup>Th, <sup>227</sup>Ac, and <sup>32</sup>P, is probably not comprehensive.

**Carbon-14** exposure occurred at the 3731 Building in the mid-1950s when irradiated graphite samples were brought to the building from the operating reactors for destructive testing. No information has been uncovered about what bioassay, if any, was done. Carbon-14 was also used as a tracer in biological experiments. One documented study was conducted in the late 1990s in the Life Science Laboratory-II Building, involving a total of about 4 Ci of <sup>14</sup>C. Urinalyses were obtained on about 20 researchers. The MDA was 10 dpm/ml (Battelle Northwest 1987, Lyon et al 1991, Long 1997). Baseline samples were obtained from each worker because natural excretion levels had not been established. ICRP (1995, 1996) assigns <sup>14</sup>C in organic compounds to class SR2.

There are some **thorium** urinalysis samples beginning in 1979. These are identified as either TH or TH232. The TH urinalysis was a total thorium analysis by spectrophotometry, with a detection level of 1  $\mu$ g/L. The TH232 analysis was specific for <sup>232</sup>Th using alpha spectrometry. The detection level was listed as 0.5 dpm/sample in 1970 (Lardy) and 0.03 dpm/sample in 1983 (Battelle Northwest). Some work was also done with <sup>232</sup>Th slurries in the 3720 Building in the mid-1990s. The plan was to collect baseline urine samples on the few workers involved, then collect special bioassay samples if air samples exceeded a cumulative exposure of 40 Derived Air Concentration (DAC)-hr. The urinalysis MDA was stated to be 0.1 dpm/sample (Bihl 1994, Lyon et al. 1998).

There are some urinalysis results for  $^{35}$ S in 1967. According to Lardy (1970), analysis was by ashing then liquid scintillation counting with a detection limit of 10 dpm/sample; however, as with  $^{14}$ C, the results were reported as  $\mu$ Ci/L and the reporting level was apparently 1  $\mu$ Ci/L.

A **radon** generator was used for animal studies in the 108F Building; it was later moved to LSLII. Monitoring was probably just by air sampling; but no information has been discovered yet. There should have been only a few researchers potentially exposed [59].

Some unusual radionuclides were isolated in the 325 Building for nuclear medicine studies in the mid to late 1990s. One of these studies isolated <sup>90</sup>Y from <sup>90</sup>Sr, and packaged and shipped the <sup>90</sup>Y to users around the world. Only a few workers were involved. The work was monitored by air samplers and no loss of control of the material occurred, so no bioassay was obtained. The material was in an insoluble form so chest counting would have been the only possible bioassay because of the 64-hr physical half-life; however, the need to perform chest counting never arose (Carbaugh 1990, Bauman 1996).

Another project involved "milking" <sup>227</sup>**Th** from <sup>227</sup>**Ac** on an ion exchange column. A bioassay procedure was developed specifically for this project under the assumption that the project was going to continue for several years; however, the project ended after only a few milkings. Only a couple of researchers were involved (Bihl 1995). The bioassay procedure had a stated MDA of 0.1 dpm/sample for <sup>227</sup>Th.

**Phosphorous-32** was used for biological tracer studies and, according to one retired researcher, "pipetting was done by mouth in the old days" [60]. Such exposure would be limited to a few researchers and would have to be established through the claimant interview or by some indication of <sup>32</sup>P bioassay samples in the worker's record. More information might be found if such a case is encountered.

# 5.3 IN VIVO MINIMUM DETECTABLE ACTIVITIES, ANALYTICAL METHODS, AND REPORTING PROTOCOLS

*In vivo* counting equipment and techniques were developed in the late 1950s and have been in routine use for measuring X-ray and gamma-ray-emitting radionuclides since 1960. (Unless otherwise noted, the *in vivo* information below came from Wilson 1987 and Lynch 2001). The unit for all *in vivo* measurements is nCi.

### 5.3.1 Whole-Body Counters

The first whole-body counter started counting workers in mid-1959 and became a routine method in 1960. It consisted of a single Nal crystal (9.375-in. diameter and 4-in. thick) housed in a counting room with 10-in thick, pre-World War II steel plates on all six sides, and graded shielding on the inner surfaces (lead, cadmium, copper) (Wilson 1987; Roesch, McCall, and Palmer 1960). This was called the Iron Room. The counting geometry was a chair configured to simulate a 1-m arc. The original count time was 20 min, which was reduced to 10 min in October 1962 (Swanberg 1962). A second, same-sized Nal detector was added in 1963 (Brady 1964). According to personal recollection of H. E. Palmer, the two-detector system improved the detection capabilities somewhat. However, the MDAs quoted in a report in the fall of 1964 were the same MDAs listed in Mr. Palmer's Laboratory Record Notebook in 1960 [61], so apparently the difference between the systems was not great enough to warrant republishing the MDAs. Therefore, the MDAs shown in Table 5-22 are the only MDAs found for the 1960s and 1970s, and apparently were meant to apply generally to the various whole-body counters in operation during this period. Although they were referred to as detection limits, the calculations were more consistent with what is presently referred to as decision levels.

Shortly after the chair counter in the Iron Room became operational, an entirely new design called the shadow shield counter was developed. The shadow shield consisted of a bed shielded on the bottom

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Table 5-22. Routine whole-body counting detection levels.<sup>a</sup>

Na-22	(nCi)					
1960-1976    Na-24	·					
1960-1976    Cr-51						
Fe-59						
1980-1976   Co-60						
1-131						
Cs-137						
Na-22						
Na-22						
Na-24						
1977-1984   Cr-51						
Mn-54						
Fe-59						
1977-1984   Co-60						
1977-1984   Zn-65						
19/7-1984						
Ag-110m						
Ru-106						
Sb-125   3.0   10     I-131						
I-131						
Cs-137						
Ce-144						
Na-22						
Na-24						
Mn-54						
Fe-59						
1985-86   Co-60						
The content of the						
Tr-95   3   1   3     Ru-106   12   4   12     Eu-154   4.5   5   4.5     Cs-137   3   1   0.66     Na-22   1.5   Anything detected to the control of the						
Ru-106						
Eu-154						
Cs-137   3   1   0.66     Na-22   1.5   Anything detected is reported.    1987						
1987 <sup>f</sup> Na-22  1.5  Mn-54  Co-60  3  Fe-59  6  Cs-137  No changes for other radionuclides. Anything detected is reported.  New formalism for decision level calculation; "limit" in electronic database changed fro MDA to decision level.  Actual values, regardless of amount, reported for Co-60 and Cs-137, including negative.						
1987 <sup>f</sup> Mn-54  Co-60  Solution  Fe-59  Cs-137  No changes for other radionuclides. Anything detected is reported.  New formalism for decision level calculation; "limit" in electronic database changed fro MDA to decision level.  Actual values, regardless of amount, reported for Co-60 and Cs-137, including negative.	tod					
1987 <sup>f</sup> Co-60  Fe-59  6  Anything detected Cs-137  No changes for other radionuclides. Anything detected is reported.  New formalism for decision level calculation; "limit" in electronic database changed fro MDA to decision level.  Actual values, regardless of amount, reported for Co-60 and Cs-137, including negative.	leu					
Fe-59 6 Anything detector Cs-137 3 6  No changes for other radionuclides. Anything detected is reported.  New formalism for decision level calculation; "limit" in electronic database changed fro MDA to decision level.  Actual values, regardless of amount, reported for Co-60 and Cs-137, including negative.						
Cs-137 3 6  No changes for other radionuclides. Anything detected is reported.  New formalism for decision level calculation; "limit" in electronic database changed fro MDA to decision level.  Actual values, regardless of amount, reported for Co-60 and Cs-137, including negative.						
No changes for other radionuclides. Anything detected is reported.  New formalism for decision level calculation; "limit" in electronic database changed fro MDA to decision level.  Actual values, regardless of amount, reported for Co-60 and Cs-137, including negative.	tea					
New formalism for decision level calculation; "limit" in electronic database changed fro MDA to decision level.  Actual values, regardless of amount, reported for Co-60 and Cs-137, including negative						
MDA to decision level.  Actual values, regardless of amount, reported for Co-60 and Cs-137, including negative						
numbers.	Actual values, regardless of amount, reported for Co-60 and Cs-137, including negative numbers.					
Co-60 4 Every result						
Cs-137 4 Every result						
I-131 5 Every result						
. Mp-54 3 Every result						
1995–10/1999 Na-22 2 Every result						
Na-24 1 Every result						
Pr-144 (Ce-144) 230 Every result						
Other radionuclides Anything detection						

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Period	Nuclide	MDA (nCi)	Decision level (nCi)	Reporting level (nCi)
	Co-60	1.25	g	Every result
10/1999 to	Cs-137	1.3	g	Every result
present <sup>i,j</sup>	Eu-154	3.75	g	Every result
	Other radionuclides			Anything detected

- a. Nominal MDAs based on the phantom available at the time, the routine count time, and the least sensitive of various whole-body counters in operation at the time. Listing of an MDA for a given radionuclide does not necessarily mean that that radionuclide was frequently encountered. If smaller MDAs are listed in the database for a given count, use them.
- b. Based on 95% confidence of detection [62]. There were some special counts for thorium; see Section 5.3.4.
- See discussion on thyroid detectors.
- d. Based on 99% confidence of detection (Wilson 1987).
- e. The reporting level cutoff of 0.66 nCi for Cs-137 appears to start in 1975 [63].
- f. From Lyon et al. 1988.
- g. From Lyon et al. 1993.
- h. [64]
- i. Least sensitive of many options during the period. Much better sensitivities were available using the HPGe system (Lynch 1995 Tables 7.2 and 7.3).
- j. Physical configurations stayed essentially the same but ABACOS software introduced changes to the methodology for determining MDAs and decision levels. Decision levels were determined by software on a count-by-count basis (Lynch et al. 2000).

and sides by lead. The bed moved under a large Nal crystal (11.5-in. diameter by 4-in. thick) that was also shielded by lead with the exception of the downward-looking face that looked directly onto the body as it passed under the crystal. The shadow shield detector was mounted in a mobile trailer and moved to areas nearer the Hanford worksites. The trailer also had a thyroid detector and a wound counter. The shadow shield detector became operational in 1963 (Brady 1964). The mobile counter was described as having comparable sensitivity to the "larger, conventional whole body counters installed in massive iron rooms. There is, however, some decreased sensitivity in the lower energy region below about 300 keV, due to increased contribution to the background from scattered radiation" (Swanberg 1963).

A report on the radionuclides detected in workers at the whole-body counter facility in 1961 listed <sup>24</sup>Na, <sup>60</sup>Co, <sup>65</sup>Zn, <sup>95</sup>Zr, <sup>95</sup>Nb, <sup>99</sup>Mo, <sup>99</sup>Tc [presumably <sup>99</sup>Mo], <sup>103</sup>Ru, <sup>106</sup>Ru, <sup>131</sup>I, <sup>137</sup>Cs, and <sup>144</sup>Ce (Henle 1962). A similar report summarizing 1961–1963 results added <sup>46</sup>Sc, <sup>51</sup>Cr, and <sup>59</sup>Fe to the list (Brady 1964).

A shadow shield whole-body detector was added at the Whole Body Counting Facility in 1977. This assembly had two 35% GeLi detectors and a 4-in. by 4-in. by 16-in Nal detector. It ceased operation in 1987 when the two new counting rooms were added. Wilson (1987) provided a listing of MDAs that applied to 1980. These are used to represent this shadow shield detector.

By 1978, there were four shadow shield whole-body counters available for use: one at the Whole Body Counting Facility, two in mobile trailers, and one at the Emergency Decontamination Facility, the latter designated for use for large acute intakes with potentially high levels of external contamination.

A "standup" counter, put in operation in 1983 is still in operation today. It consists of five vertically stacked NaI crystals in a small lead-shielded area. The worker stands in front of the detectors with the detectors to his/her back; the detector array is raised or lowered to best fit the height of the worker. There are four 9.375-in.-diameter-by-4-in.-thick detectors and one 11-in.-diameter-by-4-in.-thick detector, the latter located behind the thoracic region. Count time is 200 sec. (Palmer et al. circa 1984).

A coaxial HPGe scanning array developed in July 1989 is still in operation today (Lyon et al 1990). For this system the person lies on a bed in a shielded room and the detector array moves under the

bed. The configuration of this system, in terms of number and size of the detectors, has changed many times. It started as four 68% HPGe detectors; one of the detectors was replaced with a 120% detector in late 1995; in May 1997 the system was upgraded to include seven detectors including three 120% detectors. As a four-detector array, the system was used only when a count on the "standup" counter had detectable activity of an occupationally related radionuclide. However, it was considered the count of record. In 1997, because of its greater resolution and lower decision levels, it started being used for routine counts for workers exposed to mixtures of <sup>137</sup>Cs and plutonium. The count time was usually 10 min; however, 20-min count times are used as confirmation of an initial count with detectable activity. As a consequence, the database will usually show a 10-min count and a 20-min count on the same day or a few days later if the first count had detectable activity (excluding <sup>40</sup>K or medical radionuclides) (Lyon et al 1998).

The radon progeny <sup>214</sup>Bi and the thoron progeny <sup>208</sup>TI were and are present in the background spectra from the shielded rooms used for *in vivo* measurements of workers at Hanford. To account for this contribution when calculating worker results, the background levels of these nuclides were estimated from background measurements in the rooms with the detectors shielded with an appropriate phantom. The environmental background count rates in addition to the continuum background count rates were subtracted from the gross count rates in the appropriate region of interest of the worker spectra to obtain an activity for these nuclides. This approach was implemented in May 1992 (Lynch 1992).

Even with the background contribution subtracted, activity levels of <sup>214</sup>Bi were measured periodically that were higher than the room background levels due to activity on the workers' clothes, skin, and hair. To compensate for this, "derived investigation levels" (DILs) were implemented in October 1995 for <sup>214</sup>Bi and <sup>208</sup>TI measurements using the coaxial germanium counting system. The DIL values were 6.47 nCi for <sup>214</sup>Bi and 0.75 nCi for <sup>208</sup>TI and represented the upper 99% confidence level of an unexposed population. Results were considered to be above detection and were reported to the REX database only if the value exceeded the DIL value. The use of the DIL values continued until November 1999 when a new software system was implemented. Since 1999, <sup>214</sup>Bi and <sup>208</sup>TI results have not been reported unless specifically requested.

Use of the first mobile counter at onsite locations stopped in the early 1980s. A new trailer was obtained in 1989 and reconfigured with a new standup counter consisting of five 4-in. by 16-in. by 4-in. thick NaI detectors plus one 4-in. by 8-in. by 4-in. thick detector. The trailer was parked in the 200 East Area and operated remotely starting in 1991 (Lyon et al. 1992). The sensitivity of the detector was comparable to that of the standup counter at the Whole Body Counter Facility. The use of this facility was infrequent, and it was discontinued in August 1995 (Lyon et al. 1996).

From 1960 to 1983, four radionuclides were reported routinely: <sup>24</sup>Na, <sup>40</sup>K, <sup>65</sup>Zn, <sup>137</sup>Cs [65]. Potassium-40 is strictly a natural source and can be ignored. Sodium-24 and <sup>65</sup>Zn were detectable in many whole-body counts in the 1960s. Most of these intakes came from sanitary water drawn from the Columbia River. The onsite reactors and many of the cities surrounding the Site used the Columbia River for sanitary water, although the <sup>24</sup>Na intakes came from the drinking water at the reactors and 200 Area (which received water from the 100 Area) because its short physical half-life greatly reduced the activity in city water supplies (Brady 1964). Therefore, intakes of <sup>24</sup>Na and <sup>65</sup>Zn can be considered chronic ingestions, with the exception of the highest body burdens that were statistically different from the general background in Hanford workers and had to come from inhalations. Guidance to distinguish acute inhalations from chronic ingestions based strictly on the measured activity level has not been developed; therefore, barring evidence in the employee's file tying the intake to an acute event, the dose reconstructor can assume either chronic ingestion or inhalation according to which is more favorable to the claimant for the organ of concern. Chronic

ingestion intakes would have been occurring prior to implementation of whole-body counting, probably from shortly after the startup of the first once-through-cooling reactor (August 1944), and would have ceased shortly after the shutdown of the 100 KE reactor (January 1971) [66].

Net counts in a fifth region of the spectrum were commonly calculated but not usually associated with a radionuclide. This was the low-energy portion of the spectrum noted as the GOK region. The technique was to calculate the activity of the higher energy radionuclides <sup>24</sup>Na, <sup>40</sup>K, etc., then subtract the Compton scatter contribution from those radionuclides and see if any counts were left over in the low-energy region. If sufficient counts were left over, they would have been investigated further to see if an occupational radionuclide was the source, recognizing that the low-energy region was also subject to increased electronic noise and general background scatter in the crystal [67].

If the hardcopy form (In-Vivo Counter Results) shows the "traces of xxx invalidate routine calculation" statement, some radionuclide other than the standard four was detected; this was often <sup>60</sup>Co. The activity of that radionuclide might be written on the form. Activities that exceeded 10 nCi or 1% of the MPBB were calculated and reported on a Whole Body Counter Evaluation form (Glenn 1968). See Section 5.3.6 for instructions.

Most workers in the early days of whole-body counting had detectable activities of <sup>137</sup>Cs. Most of this was attributed to fallout. Some workers had even higher levels of <sup>137</sup>Cs from consumption of wild game. A decision level used to establish the difference between occupational and nonoccupational sources of <sup>137</sup>Cs intake has not been found in the records, and might not have been developed as long as the <sup>137</sup>Cs measurement did not exceed 1% of an MPBB. Dose reconstructors can use the following guidance: [68]

- Consider the <sup>137</sup>Cs intake occupational if the same whole-body count detected other fission or activation products (excluding <sup>65</sup>Zn or <sup>24</sup>Na). Also consider it occupational if a fission product or radiostrontium urinalysis showed detectable activity and the sample was obtained within the period between the previous and next whole-body count.
- If an investigation record clearly shows that the intake was due to a nonoccupational source, disregard the <sup>137</sup>Cs.
- NCRP Report 94 provides mean body burdens of <sup>137</sup>Cs for the United States for the years most likely to produce interference with occupational whole-body count results (NCRP 1987). Table 5-23 lists those values. If no other fission or activation products are linked to the intake (excluding <sup>65</sup>Zn or <sup>24</sup>Na) and the <sup>137</sup>Cs result is less than the values in Table 5-23, the dose reconstructor may assume the <sup>137</sup>Cs result is due to fallout.

### 5.3.2 Chest Counters

In 1967, PNNL started using the original large Nal detector in the Iron Room for chest counting, with emphasis on uranium workers. The detector was placed directly over and nearly in contact with the chest region with the worker in the supine position. Count time was 30 min. MDAs were determined to be 6.7 nCi for "U natural," presumably based on <sup>234</sup>Th; 0.15 nCi for <sup>235</sup>U; and 0.33 nCi for <sup>241</sup>Am. However, in the next year a new counting room, called the Lead Room, was built specifically for chest counting. It had four 5-in.-diameter by 0.375-in-thick Nal detectors, two in front and two in back of the subject. Count time was 30 min. (Wilson 1987). A lung phantom with variable chest wall thickness was

Table 5-23. Mean body burdens of <sup>137</sup>Cs from fallout in the United States.<sup>a</sup>

Year	Body burden (nCi)
1953	0.27
1954	1.1
1955	2.2
1956	4.3
1957	5.1
1958	6.5
1959	8.1
1960	6.8
1961	4.6
1962	6.0
1963	11
1964	19
1965	16

Year	Body burden (nCi)
1966	9.7
1967	5.6
1968	3.5
1969	2.7
1970	2.7
1971	2.7
1972	2.7
1973	2.7
1974	1.6
1975	1.1
1976	1.6
1977	1.1

a. From NCRP (1987).

developed for calibration of the new system. MDAs were listed as 0.15 to 0.6 nCi for  $^{241}$ Am, 2.0 to 3.7 nCi for  $^{234}$ Th (assumed to be in equilibrium with  $^{238}$ U), and 0.17 to 0.37 nCi for  $^{235}$ U, depending on a subject's weight-to-height ratio (Andersen 1971, Wilson 1987). (Chest count MDAs are summarized in Table 5-24.) MDAs for direct measurement of  $^{238}$ Pu and  $^{239}$ Pu using the 17-keV X-rays were calculated at times, but the values were extremely large in relation to the Maximum Permissible Lung Burden, so primary reliance was placed on measuring  $^{241}$ Am and applying a plutonium-to-americium ratio. The chest counter was also calibrated to measure *bremsstrahlung* radiation from  $^{90}$ Sr or  $^{147}$ Pm, although these were probably not routine counts. MDAs for those counts were listed as 25 to 40 nCi and 0.5 to 1.5  $\mu$ Ci for  $^{90}$ Sr and  $^{147}$ Pm, respectively (Andersen 1971). A second chest counting system became operational in 1978 (Wilson 1987). A phoswich detector became available and was used occasionally for special chest counts but was never implemented on a routine basis [69].

A solid state germanium counting system using three planar HPGe detectors (shortly upgraded to six detectors) replaced the Nal detector in the Iron Room chair counter in 1983 (Palmer et al. 1984). The HPGe detectors provide better spectral resolution than the Nal detector, thus lower backgrounds in the region of interest and better discrimination against radon decay products and better detection of low-energy photon emitters in the presence of large activities of high-energy photon emitters (e.g., <sup>137</sup>Cs or <sup>60</sup>Co). A thin window on the end of the detector faces the chest for better transmission of low-energy photons. The detectors were positioned over the front of the chest (two over the right lung) with the subject in the supine position. Counting time was 2,000 sec. MDAs were quoted for "an average size person" as 0.1 nCi for <sup>241</sup>Am, 0.5 nCi for <sup>144</sup>Ce, 0.7 nCi for <sup>234</sup>Th (<sup>238</sup>U), and 0.05 nCi for <sup>235</sup>U (Palmer and Rieksts 1984). These values were quoted as being the RDA or Reliably Detectable Activity, which was defined as 3 standard deviations of the background continuum, and discernable by naked-eye inspection of the spectrum (Carbaugh et al. 1988). Special chest counts, as follow-up to high routine chest counts or on special request, were twice the normal counting time so the MDAs were somewhat lower.

Within less than a year, the three-detector system was upgraded to a six-detector array, which enabled reduction of routine counting times to 1,000 sec with nearly the same RDAs (Palmer et al. circa 1984, Carbaugh et al. 1988). A second HPGe detector array became operational in July 1989 in a new shielded cell called the Stainless Steel Room because the inner (i.e., visible) lining of the graded shield was stainless steel. Although intended to be a six-detector array, this counter had only four detectors at first because of operational problems with the detectors. Counting times were

Table 5-24. Routine chest counting detection levels. (Andersen 1971, Lyon et al. 1988, Lyon et al. 1989, Lyon et al. 1990, Lyon et al. 1993, Lyon et al. 1994, MacLellan et al. 1999, Lynch et al. 2000)

Period	Radionuclide	MDA (nCi)
1967	Am-241	0.33
	U-238 (Th-234)	6.7
	U-235	0.15
1968–1983	Am-241	0.15-0.6 <sup>a</sup>
	U-238	2.0-3.7 <sup>a</sup>
	U-235	0.17-0.37 <sup>a</sup>
	Sr-90	25-40 <sup>a</sup>
	Pm-147	0.5-0.15 <sup>a</sup>
1983–1986	Am-241	0.24 <sup>b</sup>
	U-238	1.1 <sup>b</sup>
	U-235	0.08 <sup>b</sup>
	Ce-144	0.78 <sup>b</sup>
	Eu-154	0.07
1987	Am-241	0.28 <sup>c</sup>
	U-238	1.8
	U-235	0.12
	Ce-144	0.6
	Eu-154	0.07
1988–6/1989	Am-241	0.28 <sup>c</sup>
	U-238	1.8
	U-235	0.12
	Ce-144	0.6
	Eu-154	0.07
7/1989–1991 <sup>d</sup>	Am-241	0.18 <sup>c</sup>
	U-238	1.2
	U-235	0.08
1992-5/1996 <sup>e</sup>	Am-241	0.18 <sup>c</sup>
	U-238	3
	U-235	0.2
6/1996–10/1999	Am-241	0.28 <sup>c</sup>
	U-238	1.6
	U-235	0.095
11/1999-present	Am-241	0.25 <sup>c</sup>
	U-238	1.5
	U-235	0.090

- a. Range for different weight-to-height ratios, a chest wall thickness adjustment for both front and back chest walls Use highest value for default to cover large persons.
- Assumed MDA = (RDA)(4.65/3). Am-241 adjusted for 95th percentile male chest wall (.2/.13) (Lynch, 2003)
- c. Adjusted for 95th percentile male chest wall.
   d. Cerium-144 and <sup>154</sup>Eu no longer automatically reported for chest counts because they now can be quantified in the Ge whole-body counter.
- Applies to the six-detector array. Better sensitivity was obtained by the four-large-area-detector array in the Stainless Steel Room.

increased to 2,000 sec for the six-detector array and 3,000 sec for the four-detector array (Lyon et al 1990).

In September 1994, the chest counter in the Stainless Steel Room was converted to a four-detector array using larger area detectors (Lyon et al. 1995). The same change was implemented in the Iron Room in June 1996 (Lyon et al. 1997). This configuration continues to the present. The routine counting time was increased to 3,000 sec for the larger area arrays in November 1995; special counts and recounts were 3,600 sec.

Ultrasonic measurements of chest wall thickness for workers who had activity in the lung began in about 1978 and continue today (Palmer and Rieksts 1979). Therefore, decision levels for nondetected activities use a weight-to-height ratio to estimate chest wall thickness, whereas detected activity is corrected for chest wall thickness using ultrasound [70].

Individual-specific decision levels were reported to the database for each count and each radionuclide starting in 1992 (Lyon et al. 1993).

For *in vivo* counting, the assumption was made that  $^{234}$ Th was in equilibrium with  $^{238}$ U [71]. This was a reasonable assumption at Hanford. Uranium recently separated from dissolved fuel was certainly not in equilibrium, and uranium being treated at the UO<sub>3</sub> Plant might have been in equilibrium depending on how long it had taken the material to go through the separation process and be transported to the UO<sub>3</sub> Plant. However, uranium in this part of the fuel cycle was very soluble and not important in relation to chest counting. Chest counts were used to monitor for intakes of insoluble forms of uranium, which were very old forms in terms of time since purification from decay progeny (e.g., machining on metal, uranium metallurgy studies) [72].

## 5.3.3 Thyroid Counters

Note: During some years, <sup>131</sup>I measured in workers who had radioiodine medical studies or treatment was recorded in the database; the workers' files should have a hard copy *in vivo* count form noting that the <sup>131</sup>I was due to a medical procedure. Dose reconstructors must look for such a note in the records of anyone who has a recorded <sup>131</sup>I measurement if the <sup>131</sup>I result has any impact on the probability of causation.

Thyroid counting appears to have started on a limited basis for high-risk workers at least as early as 1956. (See Section 5.6 for a discussion on thyroid counting in 1945 and 1946.) Wilson (1960) states, "At the present time routine thyroid monitoring is conducted on a limited basis in the REDOX and PUREX facilities. Generally the pattern for coverage in the PUREX facility includes about four to five employees weekly picked from the sampling crews, crane operators, and a Radiation Monitor assigned to the stack area. At the REDOX facility routine monitoring is accomplished on a weekly basis for the shift crane operators." (Wilson, 1960) The letter continues to discuss counts and other data obtained in 1959; however, there is no indication if those results were placed in workers' files. Radiation Monitoring data sheets from 1956 show that results below 10 nCi for <sup>131</sup>I were recorded as "less than."

The first mobile whole-body counter had a thyroid counter consisting of a 3-in. by 3-in. Nal detector (assumed to mean 3-in. diameter by 3-in. thick) that was positioned next to the neck. The MDA was 0.020 nCi for <sup>131</sup>I for a 30-min count (Wilson 1987). The same detector and MDA were included in a description of *in vivo* counting capabilities at the Whole Body Counting Facility (Andersen 1971) and again in Palmer (1985). In 1986, the first listing of MDAs in the database associated with each count occurred; because the MDA is different for each thyroid count, person-specific calculations were used.

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There is a cluster of thyroid counts with MDAs ranging from 0.020 to 0.050 nCi and another cluster ranging from 0.22 to 0.298 nCi. The second cluster occurred only in October 1986 and would seem to imply use of a different detector for some reason. The count-specific MDAs after October 1986 vary from 0.07 to 0.006 nCi [73].

The presence of <sup>131</sup>I would have been detected in a whole-body count but the recorded quantity would have been based on a whole-body calibration, not a thyroid calibration [74].

For counting <sup>125</sup>I in the thyroid, a thin, 2-in. diameter NaI crystal with a beryllium window was used starting at least as far back as 1967. The thickness of the crystal has not been determined yet. The MDA was listed as 0.11 nCi for a 1-min count or 0.07 nCi for a 10-min count, but there was no mention of which count time was regularly used. There probably were not many workers exposed to <sup>125</sup>I on a regular basis; however, there are indications of a contamination spread in 1978 involving several workers (Palmer 1979). The same counter is described for thyroid counting in 1982, except the "reporting level" is given as 0.020 nCi; it is not known if this better sensitivity came from a longer count time, better positioning, or an improved crystal.

By 1985, thyroid counting for <sup>125</sup>I was performed using two intrinsic germanium detectors, with an MDA of 0.005 nCi for a 2,000-sec count.

Thyroid counting for either of the iodine isotopes has been rare since 1987.

## 5.3.4 Thorium Exposure and Monitoring

(Note: Information on use of thorium at Hanford came from Gerber 1992 unless otherwise indicated.)

Thorium contamination was introduced into the 313 Building and surrounding fuel warehouses in the early 1950s, when a number of attempts were made to fabricate metallic thorium fuel targets to produce <sup>233</sup>U. Many problems connected with the rapid formation of a thick coat of oxide on the thorium metal targets led to experiments with a variety of bonding methods and coatings. After a technique was determined, powdered thorium oxide fuel targets (wafers) were fabricated in the 3732 Building from 1965 to 1967. Sintering of the wafers was part of the process in the 3732 Building. A new technique involving pelletized targets replaced the 3732 Building operations. The pelletized targets were fabricated in the 3722 Facility from 1968 through 1970. In addition, that building housed a furnace for the "recycling" (reduction) of depleted thorium oxide after it was processed in PUREX. Other buildings with thorium contamination included 3307 A and B (change houses), 306 (alloy and fabrication test and development activities), and 3706 (radiochemistry in support of fuel fabrication). It is reasonable to assume possible exposure to thorium in these facilities as shown in Table 5-25. It is reasonable to assume significant exposure, similar to exposure to uranium at Hanford, in the 3732 Building from 1965 through 1967 and the 3722 Building from 1968 through 1970. If building information is not available, but there is evidence that a person worked with uranium in the 300 Area, 1950-70, it is favorable to claimants to assume exposure to thorium.

Table 5-25. Buildings with thorium processing (Gerber 1992, Isochem 1967, Walser 1978).

Building, area	Date
313, 300	1950–1970
306, 300	1956-1970
3706, 300	1954–1963
3732, 300	1965–1970
3722, 300	1968–1970
202 A, PUREX, 200 East	1965–1970

In addition, the thorium was irradiated in a reactor and dissolved in PUREX, the residual thorium and freshly made <sup>233</sup>U were separated, and the thorium was recovered (Isochem 1967, Walser 1978). This work was performed in three distinct campaigns with extensive cleaning of the PUREX lines and tanks prior to introduction of the irradiated thorium wafers. The campaigns were a small test campaign conducted in 1965 and two production campaigns in 1966 and 1970. For the 1965 campaign the original dissolution was not conducted at PUREX but additional purification of the nitrate solution was (Isochem 1967, Walser 1978). Handling of the <sup>233</sup>U product and the recovered thorium was apparently done by the same crew, so thorium exposure should be assumed for PUREX workers if there is evidence of bioassay for <sup>233</sup>U (Jech 1970b). When not recycled to the 3732 Building, the thorium nitrate was stored in the WR Vault in the 200 West Area (Isochem 1967).

Thorium exposure would have been associated with uranium exposure. However, thorium was not associated with U Plant so uranium workers in that facility should not be assigned thorium intakes. (There were a few episodes of thorium handling at UO<sub>3</sub> plant in 1967, involving a few 100s of pounds, but this is considered to contribute insignificantly to dose considering that the UO<sub>3</sub> plant processed about 1,500 metric tons of uranium during 1967 [DOE 2000].)

Radiation protection practices were similar for thorium and uranium, so assume chronic intake. Because metal, oxide, nitrate, and perhaps other forms of thorium were being handled, either absorption type M or type S can be assumed (independently from uranium absorption types). Radium-228, the first progeny of <sup>232</sup>Th would have been removed, either during the processing of the ore at Fernald or during baking and sintering in the 300 Area, so the progeny would not have been in equilibrium (West 1965). Workers could have been exposed to thorium in a mixture of ages (i.e., time since purification), but most of the material would have been fairly young. It is favorable to claimants to assume younger material when whole body counting is the bioassay method (explained below); hence, the thorium was assumed to be 0.5 yr since purification. Therefore, for an intake of <sup>232</sup>Th by activity, assign <sup>228</sup>Ra at 0.058 times the <sup>232</sup>Th intake and assign <sup>228</sup>Th at 0.84 times the <sup>232</sup>Th intake [75]. The <sup>230</sup>Th specification for the thorium oxide was <1 ppm (Douglas United Nuclear 1965), which means that <sup>230</sup>Th contamination would have contributed <6% of the total alpha activity and can be ignored.

Routine monitoring for thorium intakes was by whole-body counting based on <sup>228</sup>Ac. **Note: use of** whole body counting for thorium monitoring is apparently different than at other AEC sites that used chest counting. Be sure to recognize that the following discussion and Tables 5-26-28 apply to whole body counts. Urinalyses were used for special investigations. Jech (1967) stated the detection limit for <sup>232</sup>Th was 1.2 nCi. In another letter, Jech (1969b), summarizing results of a worker's whole-body count, stated, " ... and no detectable thorium (<1 nCi)". The 1.2-nCi detection limit was probably a decision level, so the MDA would have been about 2.4 nCi [76]. Sometimes the whole-body count record shows <sup>232</sup>Th in the list of radionuclides if the worker was specially earmarked for possible exposure to <sup>232</sup>Th, but the dose reconstructor should not count on this for every exposed worker. If nothing was detected, no activity is shown. For a few of those workers no record of the whole-body count was found in the electronic database, but a paper record of the count was submitted as part of the DOE files. Although the in vivo records show the results as <sup>232</sup>Th, the measured activity was <sup>228</sup>Ac [77]. The two radionuclides would not have been in equilibrium; both disequilibrium at intake and separate biological processes on the <sup>232</sup>Th parent and the <sup>228</sup>Ra progeny mean that the <sup>232</sup>Th activity in the body would not have been the same as the measured <sup>228</sup>Ac. Assuming chronic intake and exposure to 0.5-yr aged mixture, the factors in Table 5-26 should be used to determine the intake of <sup>232</sup>Th per nCi of <sup>228</sup>Ac (reported as <sup>232</sup>Th) measured in a whole body count. Once the adjusted intake of <sup>232</sup>Th has been determined, the ratios given in the paragraph above must be used to determine the intakes of <sup>228</sup>Ra and <sup>228</sup>Th.

The dose reconstructor can use the whole-body count record plus urinalysis records, if there are any, to determine intakes of <sup>232</sup>Th. If a person worked in the buildings in Table 5-25 for the years listed, and was monitored for uranium but not thorium, it is reasonable to assume exposure to thorium and

Table 5-26. <sup>232</sup>Th intakes from whole body count measurements [78].

Period of chronic intake prior to whole	Ratio of Th-232 to Ac- 228 in whole body at time of measurement		e body at 228 (reported as Th-232) measur		
body count (days)	Type M	Type S	Type M	Type S	
182	23.17	11.33	1810	1310	
364	26.56	9.218	1150	648	
546	29.03	7.834	874	412	
728	30.40	6.898	704	297	
910	30.73	6.234	580	232	
1092	30.28	5.743	483	191	
1274	29.34	5.370	405	162	
1463	28.09	5.068	341	142	
1645	26.79	4.837	291	127	
1827	25.49	4.649	251	115	
2009	24.24	4.494	218	106	
2184	23.10	4.369	192	98.6	

use the coworker intakes provided in the most recent version of the coworker TIB (ORAUT 2007). For PUREX workers the preceding statement applies to workers with <sup>233</sup>U bioassay for 1965 through 1970.

Except for small scope, limited time projects that may have occurred occasionally, exposure to thorium at Hanford stopped in 1970. But later-day whole body counts might still be used as a check on the intakes estimated for exposure prior to 1970. Th-232 was not routinely reported in whole body counts, but a prominent <sup>228</sup>Ac peak would have been noticed and investigated. Tables 5-27 and 5-28 provide the ratios of <sup>232</sup>Th to <sup>228</sup>Ac in the whole body at times after cessation of chronic exposure. The MDA for <sup>228</sup>Ac or <sup>232</sup>Th was not listed in program documentation after the 1960s Jech letters (1967, 1969a) until 1979 when a few counts for <sup>232</sup>Th are listed with a detection limit of 0.5 nCi (based on a query of the REX database). This was probably a decision level. Twenty-one <sup>232</sup>Th counts are recorded in the 1980s with count-specific detection limits that vary from 0.19 to 0.93 nCi, with a median of 0.43 nCi (based on a query of the REX database). However, there was a difference between the detection level used for a radionuclide specifically targeted for identification by the software, and the trigger level used to flag a peak that was not specifically earmarked by the software. Stated another way, the criterion to identify and flag the <sup>228</sup>Ac peak would have been higher than just the decision level. With that in mind, it is reasonable to assume that from 1970 through July 1989, an <sup>228</sup>Ac body burden of 2 nCi would have been noticed and investigated but burdens less than that might have been overlooked [79].

Radon-220 (thoron) was driven off during the bake-out and sintering processes (West 1965, Bihl and Traub 2006), but would have grown back in quickly. Measurements of airborne thoron concentrations were not found. An upper bound estimation of thoron concentrations in the 3722 and 3732 Buildings was made based on the throughput of thorium during two campaigns (1964-1965 and 1969) and the following assumptions (Bihl and Traub 2006):

• 90% equilibrium between <sup>232</sup>Th and <sup>228</sup>Th in the material being processed in the 3722 and 3732 Buildings (upper bound calculation)

 100% release of thoron from the thorium metal or thoria during the processing (due in part to vaporization of <sup>224</sup>Ra)

Table 5-27. <sup>232</sup>Th to <sup>228</sup>Ac ratios in the whole body after end of intake period, type M [80]

Years after end of	Period of chronic intake, yr						
chronic intake	1	2	3	4	5	6	
0.5	34.1	34.0	31.0	27.5	24.4	21.8	
1	36.3	32.8	28.7	25.2	22.4	20.1	
1.5	33.9	29.5	25.7	22.7	20.3	18.4	
2	29.9	25.9	22.8	20.4	18.4	16.9	
2.5	26.1	22.8	20.3	18.4	16.8	15.5	
3	22.8	20.3		16.8	15.5	14.4	
4	18.3	16.6	15.4	14.3	13.4	12.6	
5	15.3	14.2	13.3	12.5	11.9	11.3	
6	13.2	14.5	11.8	11.3	10.8	10.4	
7	11.8	11.2	10.7	10.3	9.91	9.58	
8	10.7	10.2	9.87	9.53	9.22	8.96	
9	9.84	9.50	9.19	8.92	8.67	8.44	
10	9.17	8.89	8.64	8.41	8.20	8.01	
11	8.63	8.39	8.18	7.98	7.80	7.64	
12	8.17	7.97	7.79	7.62	7.46	7.31	
13	7.78	7.61	7.45	7.30	7.16	7.03	
14	7.44	7.29	7.15	7.02	6.89	6.77	
15	7.14	7.01	6.88	6.77	6.65	6.54	
16	6.88	6.76	6.64	6.54	6.43	6.33	
17	6.64	6.53	6.43	6.33	6.23	6.14	
18	6.42	6.32	6.23	6.14	6.05	5.97	
19	6.23	6.13	6.05	5.96	5.88	5.81	
20	6.04	5.96	5.88	5.80	5.72	5.65	

Table 5-28. <sup>232</sup>Th to <sup>228</sup>Ac ratios in the whole body after end of intake

period, type S [81].

Years after end of	Period of chronic intake, yr						
chronic intake	1	2	3	4	5	6	
0.5	6.34	5.23	4.60	4.19	3.94	3.76	
1	5.00	4.36	3.96	3.70	3.54	3.41	
1.5	4.23	3.82	3.55	3.38	3.30	3.18	
2	3.73	3.45	3.26	3.15	3.06	3.00	
2.5	3.39	3.19	3.07	2.98	2.92	2.88	
3	3.15	3.01	2.92	2.86	2.82	2.80	
4	2.86	2.79	2.75	2.72	2.71	2.70	
5	2.72	2.68	2.67	2.66	2.66	2.66	
6	2.65	2.64	2.64	2.64	2.64	2.65	
7	2.62	2.63	2.63	2.64	2.65	2.66	
8	2.63	2.64	2.65	2.66	2.67	2.68	
9	2.64	2.65	2.67	2.68	2.69	2.71	
10	2.66	2.68	2.69	2.71	2.72	2.73	
11	2.69	2.71	2.72	2.74	2.75	2.77	
12	2.72	2.74	2.75	2.77	2.79	2.80	
13	2.75	2.77	2.79	2.80	2.82	2.83	
14	2.79	2.80	2.82	2.83	2.85	2.87	
15	2.82	2.83	2.84	2.87	2.90	2.91	
16	2.86	2.87	2.89	2.91	2.93	2.94	
17	2.89	2.91	2.93	2.94	2.96	2.98	

Years after end of	Period of chronic intake, yr						
chronic intake	1	2	3	4	5	6	
18	2.93	2.95	2.96	2.98	3.00	3.01	
19	2.96	2.99	3.00	3.02	3.04	3.05	
20	3.00	3.02	3.04	3.06	3.07	3.09	

- 10% of the airborne thoron is not exhausted outside by the oven hoods and escapes into the workspaces
- 5 building air exchanges/hr
- An equilibrium factor of 0.04.

The estimated working level months/yr (WLM/yr) were 43 (Bihl and Traub 2007).

These WLM are constant upper bounds associated with processing hundreds of tons of thorium in the 3722 and 3732 Buildings, 1964 through 1970. All workers associated with these buildings (might be just mentioned as thorium processing) should be assigned these WLM. The WLM can be prorated for partial year exposure. Use the thoron TIB (NIOSH 2006) to convert the WLM to doses to the ET1. ET2, and lung.

Ventilation in the PUREX canyon would have removed thoron to the main stack; work with small quantities of thorium in hoods prior to 1964 would not have produced significant thoron intakes by workers [82].

Although there was residual thorium contamination in the plants after 1970, the exposure to the workers in relation to their uranium exposure would have been negligible and can be ignored.

#### 5.3.5 **Head Counters and Other Counts**

Miscellaneous counts have been performed over the years at Hanford, including wound counts, head counts, liver counts, lymph node counts, and various longitudinal scans with collimated detectors to pinpoint the location of external or internal contamination. Results of these in the database will almost always be listed as special counts associated with known intakes [83].

Since at least 1978, for intakes of plutonium or americium, head counts have been used to correct chest counts for activity in the bones of the chest region (Palmer 1979). Since the mid-1990s, liver counts have been added to the protocol for correcting chest counts to account for possible shine from the liver [84].

Routine head counting for <sup>90</sup>Sr or <sup>147</sup>Pm did occur for a while in the 1970s. These were not very sensitive and there is the question about what a head count means in relation to the activity in the total skeleton. The same worker will hopefully have <sup>90</sup>Sr urinalysis results. The latter should be given preference for confirming or quantifying an intake.

#### 5.3.6 **General Notes About Items in the Database**

All in vivo results appear to be given in nCi [85]. "Limits" were MDAs, which were treated the same as decision levels until 1992. The decision level is listed under "limits" starting in January 1992 [86].

Sometimes a radionuclide is listed without a value or limit. This probably means a "trace" was found. More information might be available on the In Vivo Counter Results Form if it was sent to the worker's personal radiation exposure history file. If not, assume the result of the count is 100 nCi (Glenn 1968).

Prior to the advent of GeLi detectors, when a significant peak in a whole-body count of a radionuclide not <sup>24</sup>Na, <sup>137</sup>Cs, <sup>40</sup>K, or <sup>65</sup>Zn occurred, the activity of the trace or "interfering" radionuclide might have been quantified. In addition, the activity of one or more of the regular four radionuclides might have been marked as invalid because of overlap with the interfering peak or because of the impact of the interfering peak on the spectrum stripping calculations [87]. For the small activities involved, there is no merit in trying to recalculate or estimate actual quantities. It is favorable to claimants to use the activities of <sup>137</sup>Cs as given; include the activity of the interfering radionuclide as given as well. Use 100 nCi for the interfering radionuclide if not given directly (Glenn 1968).

The radionuclides routinely reported to the database changed over the years. From the beginning until 1983, <sup>24</sup>Na, <sup>40</sup>K, <sup>137</sup>Cs, and <sup>65</sup>Zn were the only routinely reported radionuclides. In 1983, as part of the switch to the ORE database, only <sup>40</sup>K and <sup>137</sup>Cs results (or the MDAs) were routinely reported; in late 1987, 60 Co was added [88]. In 1995, with the implementation of a new spectrum analysis software program (NEXEC), the standup counter's energy spectrum was divided into 12 regions and a radionuclide was assigned to each region, including more naturally occurring radionuclides such as <sup>214</sup>Bi and <sup>208</sup>TI [89]. During this time, if a worker had a count using the coaxial HPGe whole-body counter, as many as 20 radionuclides might have been listed in the records. The listing of that many radionuclides was simply a bookkeeping approach, and had nothing to do with the sources of exposure [90]. Because of the shutdown of the last reactor in 1986, radionuclides such as <sup>59</sup>Fe. <sup>24</sup>Na. <sup>22</sup>Na, <sup>144</sup>Ce/Pr, and <sup>131</sup>I had decayed away to negligible levels at Hanford (unless a researcher was using a small source for studies). The lack of the need to report all these radionuclides routinely, unless a peak was actually present, was recognized, and when NEXEC was replaced by Abacos (October 1999), the routinely reported list was reduced to <sup>40</sup>K, <sup>60</sup>Co, <sup>137</sup>Cs, and <sup>154</sup>Eu (Bihl 1999) [91]. Reporting of radionuclides at levels below the MDA or decision level should not be interpreted as implying exposure to those radionuclides.

For chest counting, the database usually lists <sup>234</sup>Th as the potentially measured radionuclide as an indicator of <sup>238</sup>U. Until recently, routinely reported radionuclides for anyone receiving a chest count were <sup>241</sup>Am, <sup>234</sup>Th, and <sup>235</sup>U. This does not imply exposure to both plutonium/americium mixtures and uranium. Very recently, workers have been scheduled for types of chest counts based on their exposure in the workplace, so for plutonium workers, for instance, only <sup>241</sup>Am results are determined and reported.

#### 5.4 MIXTURES

Except in a few facilities in the weapons production cycle (such as B Plant/WESF after 1968, UO<sub>3</sub> Plant), bioassay methods did not measure all the radionuclides in the intake mixture. The emphasis was on measuring exposure to radionuclides with the greatest impact in relation to radiation protection standards (for instance, MPBB or committed effective dose equivalent), or radionuclides that were most common. Unmeasured radionuclides generally do not have a big impact on dose but might target different organs or might have a larger relative impact over times less than 50 years. Therefore, this section attempts to estimate possible mixtures of radionuclides that might have been part of an intake that was indicated by a measured radionuclide. In all cases, where actual bioassay data are available, dose reconstructors should use those data in preference to the following conservative mixtures.

Plutonium isotopic mixtures and uranium isotopic mixtures are discussed in Sections 5.2.1 and 5.2.5, respectively.

## 5.4.1 <u>Fission and Activation Products</u>

For fission and activation product mixtures up through 1987 (when N Reactor shut down) use the intake mixture ratios presented in *Fission and Activation Product Assignment for Internal Dose-Related Gross Beta and Gross Gamma Analyses*, OTIB-0054. So if an intake of <sup>90</sup>Sr, <sup>137</sup>Cs or <sup>106</sup>Ru was being assigned to a worker, the other fission and activation products should be assigned as discussed in that document. OTIB-0054 includes guidance on intake mixtures that applies to workers in different types of work or facilities.

The fission product urinalysis procedure measured beta activity from any radionuclides of strontium, yttrium, barium, lanthanum, cerium, europium, and promethium. It was calibrated for the <sup>90</sup>Sr/<sup>90</sup>Y betas so would have underestimated soft beta emitters, but chemical yield was several times higher for cerium and yttrium than for strontium so the recorded result overestimated cerium and yttrium activity (Healy 1948; Wilson 1987). Urine results designated as FISPR or FP in the workers' records indicate participation in the fission product bioassay program.

At Hanford some fission products had greater prominence than indicated in OTIB-0054. These were <sup>65</sup>Zn for essentially all workers because of its presence in the Richland drinking water through about 1972, <sup>24</sup>Na for workers at the once-through-cooled production reactors through about 1971 (see Section 5.3.1), <sup>154</sup>Eu for workers at N Reactor because of the samarium-ball emergency shutdown system, and <sup>24</sup>Na for 400 Area workers (FFTF) for 1980 through 1994 [92]. From 1960 to date these radionuclides would have been measured by whole body counts. If their activity is not included in the whole body count record, use the standard missed dose procedure for assigning these intakes to workers at the applicable facilities using MDAs or reporting levels in Table 5-22. The N Reactor and FFTF intakes should not be applied unless it is certain the worker had exposure at these facilities because, relative to the Hanford workforce, few workers were actually exposed to these radionuclides at those times. Zinc-65 and <sup>24</sup>Na were not measured as part of the fission product urinalysis, however, so during the period predating whole body counting, intakes of these radionuclides should be added using guidance in OTIB-0039 (startup to 1959). <sup>154</sup>Eu is not addressed in OTIB-0039 nor are MDAs available in Table 5-22 for all years; when not listed in Table 5-22, assume the MDA is 3 times the MDA for <sup>60</sup>Co.(1964 through 1984) [93].

OTIB-0054 provides guidance on fission and activation product ratios for waste management workers. However, characterization of the contents of the high-level waste tanks shows that plutonium and americium are also present in ratios relative to fission products such that they might increase the dose to certain organs. Americium is usually associated with <sup>90</sup>Sr in the supernatant and is generally more available as a contaminant. Plutonium is usually associated with <sup>137</sup>Cs in the sludge (Carbaugh 1995, Boothe 1992). For workers in waste management facilities or 200 Area workers in general if work location is not known, if an intake of fission products is determined and the worker does not have bioassay for plutonium or americium after the intake date, also assume intake ratios of 0.001 for both <sup>239</sup>Pu and <sup>241</sup>Am relative to the highest fission product intake (Carbaugh 1995, Boothe 1992).

Tritium intakes may have occurred by reactor workers and fuel dissolution plants workers. Dose from tritium was accounted for as part of external dose records for 1949 through 1987. See Section 5.6 for guidance on tritium intakes for other years or if there is no evidence of monitoring for tritium in the external dose record for reactor or fuel dissolution plant workers.

## 5.5 INTERFERENCES, UNCERTAINTIES

### 5.5.1 Contamination of Samples

Home collection of excreta samples started very early in the bioassay program (Wilson 1987); hence, contamination of excreta samples can be assumed to be negligible. Laboratory contamination and mix-up of samples in the laboratory are a possibility, although laboratory Quality Control procedures and performance of test samples were designed to minimize this source of contamination.

It is likely that a contaminated sample will show up as an obvious outlier in the dataset for a given worker.

For *in vivo* measurements, contamination can occur as external to the body or, in the case of chest counting, as external to the lung. If a follow-up *in vivo* count obtained the same day or within a few days shows a dramatic decrease in activity or no detectable activity, assume external contamination. Radon progeny and medical diagnostic or therapeutic procedures involving radionuclides can cause interferences to *in vivo* measurements, especially for NaI detectors. However, unless the count was invalidated or noted as being influenced by such interferences, use the results as recorded.

## 5.5.2 Uncertainties

Uncertainties for the bioassay measurements were included in the database starting in late 1981 for excreta measurements [94]. These are listed in the database under Error and represent total propagated uncertainty (one sigma) including counting uncertainty, yield uncertainty, and various other systematic uncertainties (UST 1987). Use these when available. For excreta, uncertainty can also exist in the sample date. For routine samples, assume an uncertainty of ±2 weeks. This is because one sample date is used for the month regardless of when the sample was actually obtained. For special samples an uncertainty of ±2 days is reasonable unless the sample is within 2–3 days of a known intake.

The period the sample represents is also a source of uncertainty. Most urine samples at Hanford were 24-hr simulated samples (kit code 1), meaning the sample was collected over two evening-through-morning periods. Medley, Kathren, and Miller (1994) indicated that this sampling method produced only about half of a true 24-hr sample based on volume for a group of nine workers over a 3-day period; however, Hanford collection protocol was based on percent of day, not volume, so the true bias (when samples were collected according to procedure) was about 75% of a true 24-hr sample [95]. If a worker has enough urine samples to establish the individual-specific excretion pattern, a sample can be normalized to the individual's expected 24-hr excretion. In general, the error associated with collection period results from under-collection of a 24-hr volume. It is favorable to claimants to normalize a volume that is less than reference man or reference woman; however, volumes larger than reference man or reference woman should be considered 24-hr samples without normalizing. If no volume information is available for a urine excretion result, a reasonable maximizing assumption would be to double the uncertainty of the result [96].

For *in vivo* results, uncertainties were not reported until 1986 for detected radionuclides and 1993 for the default set of radionuclides. These were 1-sigma counting errors until 1995 [97]. Total propagated error has been determined and submitted to the records since then. The propagated uncertainty includes counting uncertainty, calibration uncertainty, and a generic 5% positioning error (for both whole body and lung). The calibration uncertainty includes the uncertainty in source activity, counting error, decay correction, and interpolation using the calibration curve [98]. Uncertainty associated with reproducibly positioning a person to get the same result was studied at Hanford and

found to be about 5% [99]. All calibrations use phantoms, and there is considerable uncertainty associated with the representativeness of phantoms versus humans. A recent study for whole-body counting at Hanford used a 95th-percentile reference man phantom. There was a low bias of about 25% for the coaxial HPGe detector system for 662 and 1332 keV gamma rays (Lynch, 2007). A similar value of uncertainty (±25%) can reasonably be assumed for the other whole-body detectors (1-m arc, shadow shield, and standup counters) [100].

Uncertainties associated with chest counting are reduced by use of different calibrations for different chest wall thicknesses and use of ultrasound to measure chest wall thickness. One study showed a 1-sigma uncertainty of about 20% for americium and uranium values in chest counting, not including correction for interferences from bone and liver (Palmer 1990 Table 7-4). Uncertainties would be much higher for an individual with activity in the bone and/or liver. The uncertainty in lung activity estimates affected by contributions from activity in the liver and skeleton would probably range from 100% or more for levels near or below the MDA to 50% or more for activity above the MDA [101]. The uncertainty in the estimate of chest thickness using the height/weight correction was at least 50% for the front/back lung counter.

Because the uncertainty in the <sup>241</sup>Am-to-Pu-alpha ratio can vary nearly two orders of magnitude, plutonium intakes should not be determined solely by chest counting data if possible. If no information concerning the isotopic mixture of an intake is available and a default mixture is assumed, a geometric standard deviation of 5 uncertainty should be associated with an intake determined by chest counting [102].

Based on the above discussion, the assumption provided in the *Internal Dose Reconstruction Implementation Guide* (NIOSH 2002), namely the standard deviation is 0.3 times the MDA or reporting level, is adequate with the exception of chest counts, for which the dose reconstructor should use 0.5 times the MDA. If actual standard deviations or other indications of error are reported with a bioassay measurement result that exceeds the above guidance, use the reported value.

#### 5.6 UNMONITORED WORKERS

For unmonitored workers with external dosimetry, use the intakes provided from the coworker analyses in OTIB-0039 (ORAUT 2007a). Short-lived radionuclides, such as <sup>144</sup>Ce, <sup>106</sup>Ru, <sup>95</sup>Zr, <sup>95</sup>Nb, <sup>54</sup>Mn, <sup>65</sup>Zn, <sup>140</sup>Ba, <sup>140</sup>La, <sup>51</sup>Cr, <sup>59</sup>Fe, <sup>110m</sup>Ag, should not be assigned to workers after December 31, 1988 [103]. The exception is for <sup>24</sup>Na for workers at the Fast Flux Text Facility, which operated intermittently from 1980 to 1984. Special consideration is also provided for workers involved in decontamination and decommissioning. See facility-specific details at the end of this section. The coworker intakes do not address tritium or radioiodines. Guidance for assignment of intakes of tritium and radioiodines by unmonitored workers is provided in this section. There is also guidance for assignment of intakes of <sup>233</sup>U and contaminants for unmonitored workers on the <sup>233</sup>U project.

Under certain conditions, airborne effluents from one facility became air intakes for other facilities. In addition, workers were exposed to diluted effluents when walking between buildings or parking lots or while driving on the Site. Therefore, workers in buildings who did not enter contaminated or airborne areas and construction workers almost anywhere could have incurred environmental-level intakes.

Before 1994, workers with even a remote chance of exposure to workplace external radiation or workplace airborne contamination (as opposed to exposure from effluents) wore a dosimeter. Through 1971, workers obtained film dosimeters before proceeding past badge houses at the various

facilities. From 1972 to 1994, even workers with only a small potential for incurring external dose wore a minimal dosimeter, called the Hanford basic dosimeter (ORAUT 2006a).

The unmonitored internal dose for workers with no bioassay (or just baseline or termination bioassay) and no evidence of ever having worn a dosimeter should be based only on environmental intakes [104]. Termination whole-body counts were often provided to any worker regardless of exposure risk and should not be considered evidence of being on a bioassay program.

Considerable (hundreds per month) thyroid scans were being done for workers in the fuel dissolution (canyon) buildings during 1945 through at least 1946. A review of the thyroid monitoring program was conducted for the Hanford Environmental Dose Reconstruction Project (Ikenberry 1991). The tolerance level for  $^{131}$ I in air had been established in October 1945 as 1 × 10 $^{-7}$  µCi/cm $^3$  (Cantril 1945) based on a permissible equilibrium amount in the thyroid of 2 µCi. The routine scanning program began in late May or early June 1945. The Ikenberry document reviewed over 7,900 thyroid checks from October 1944 through August 1946. The decision level for the measurement was estimated to correspond to a thyroid burden of about 27 nCi, and 93% of the measurements were below this activity. The highest measurement considered to be reasonably due to a thyroid burden (as opposed to external contamination) was 131 nCi.

Since the beginning of routine whole body counting, detections of <sup>131</sup>I have been rare. For instance, from 1962 through 1969 (peak production years), only about 130 workers had detectable <sup>131</sup>I out of about 14,500 whole body and thyroid counts; results ranged from 0.001 to 560 nCi, the highest measurement being a same-day measurement after an accidental exposure in the T Plant canyon. Ninety percent of the detected results were less than 15 nCi [105]. From these two data sets, the assumption was made that was favorable to claimants that at-risk workers had thyroid burdens less than 30 nCi, which would result from a daily intake of 20,000 pCi [106]. This intake would apply to all at risk workers prior to 1960. From 1960 to 1987, it is reasonable to assume that an unmonitored worker, (i.e., did not have a whole body count) did not incur intakes greater than the thousands of workers given whole body counts who showed no detection. Using a nominal detection limit for whole body counts (as opposed to more sensitive thyroid counts) of 5 nCi, unmonitored workers associated with reactors or fuel dissolution plants should be assigned <sup>131</sup>I intakes of 3400 pCi/d [107]. No intakes of <sup>131</sup>I should be assigned after 1987 because there was no significant source term, unless, of course, the worker's personal information indicates a special exposure, such as for a research project [108].

Tritium bioassay results are not included in the electronic database until 1984 [109]. Tritium doses were included in external dose reports from 1949 through 1987, so "monitored" workers would be determined by tritium dose listed in external dose records. A zero dose is indicative of a monitored worker, but there are only two of these in the database. No value listed at all under "tritium" might be indicative of either an unmonitored worker or a monitored worker with less-than-recordable dose. No tritium doses are listed on external dose reports for 1955 through 1960, 1970, 1971, and 1973 through 1984, so either there was no bioassay monitoring during these periods or the intakes were below some reporting criteria [110]. The 108B Tritium Extraction Facility shut down in 1955 so that probably explains the lack of tritium doses, and probably a lack of tritium monitoring, until the heavy-water-moderated Plutonium Recycle Test Reactor started up in 1960 (see Sections 2.1.6 and 2.2.8). If a worker's file does not show tritium dose in the external dose records (where a zero is considered a monitored dose), and if the worker might have had exposure to tritium as discussed below, then the unmonitored tritium intakes discussed below and provided in Table 5-29 should be applied.

Prior to 1949, it is assumed that tritium intakes were limited by the tolerance level. The earliest tritium tolerance limit found to date is an air concentration limit 0.01 µCi/L in 1947 (Parker 1947, Peterson 1949)(See Appendix 5A.2 for discussion on tolerance levels). Assuming chronic intake at this

concentration of 1.2 m³/hr for 2000 hr per year results in an intake rate of  $6.6 \times 10^7$  pCi/d. This produces annual organ doses of about 1,500 mrem [111], which is larger than all but a few recorded doses from 1949 through the 1960s. Hence, this intake rate is favorable to claimants for the highest risk workers, namely workers at the 108B Tritium Extraction Facility. Tritium would have been present at the fuel dissolution plants as well and to a lesser extent at the reactors, but certainly the tritium in the latter facilities would not have been as concentrated as at the 108B facility. It is assumed that tritium intakes at the reactors and fuel dissolution facilities would have been at most 10% of the intakes at 108B [112]. Therefore, daily intakes of  $7 \times 10^6$  pCi/d should be assigned to unmonitored workers at the reactors, fuel dissolution plants, or rovers.

Table 5-29. Statistical parameters of Hanford recorded tritium doses and associated intakes [113].

Year	Geometric mean (mrem)	GSD	Geometric mean doses normalized to present model (mrem)	Daily intakes resulting in the normalized doses ( × 10 <sup>6</sup> pCi/d) <sup>a</sup>
1949	29.9	2.9 (3.0) <sup>b</sup>	48.4	2.0
1950	43.6	2.8 (3.0)	70.6	2.9
1951	64.2	3.5	104	4.2
1952	67.1	4.0	109	4.4
1953	40.8	3.7	38.5	1.6
1954	104	3.0	98.2	4.0
1955–1960	Insufficient da	ta		
1961	62.4	1.9 (3.0)	58.9	2.4
1962	193	3.0	182	7.4
1963	185	3.8	175	7.1
1964	141	4.2	133	5.4
1965	134	3.8	127	5.1
1966	108	3.9	102	4.1
1967	185	4.7	175	7.1
1968	98.6	4.9	93.1	3.8
1969	26.5	2.3 (3.0)	25.0	1.0
1970–1971	Insufficient da	ta		
1972	58.9	2.3 (3.0)	55.6	2.3

a. Use as injection mode in IMBA or CADW.

To determine tritium intakes for 1949 through 1972, a coworker analysis was conducted on tritium dose data for that period. The statistical results are biased high because, with two exceptions, dose from tritium <10 mrem were not recorded; consequently, it is not known how many workers were monitored who had doses less than 10 mrem. The annual geometric means and geometric standard deviations (GSD) are shown in Table 5-29.

The methodology for calculating tritium doses from urine concentration has changed over time. The method introduced in ICRP Publications 2 and 10 (1959 and 1968) resulted in dose per unit activity in urine slightly higher than the method used presently. How the tritium doses were calculated prior to 1959 has not been found yet; however, Parker, head manager of radiation protection at Hanford in the 1940s and 1950s, referenced a method developed by Morgan in 1946 which shows that 40,000  $\mu$ Ci in the body produces the tolerance dose for the time (Morgan 1947). The tolerance dose in the 1940s was 100 mrep/d. Assuming the tritium is distributed in 42 L of body water, at equilibrium, 952  $\mu$ Ci/L [40,000  $\mu$ Ci/42L] produced 100 mrem/d; or, stated another way, a dose of 0.105 mrem/d would have been calculated from 1  $\mu$ Ci/L in urine. The 1953 body burden of tritium to produce a maximum

b. A default GSD of 3.0 should be substituted for a calculated GSD of less than 3.

permissible dose of 300 mrem/wk was 10,000  $\mu$ Ci (NBS 1953). Hence, 238  $\mu$ Ci/L [10,000  $\mu$ Ci/42L] would have produced 42.0 mrem/d; or 0.18 mrem/d would have been calculated from 1  $\mu$ Ci/L in urine. In the present model (ICRP 1997), 1.2 × 10<sup>6</sup> Bq/L results in 20 mSv/yr or 0.17 mrem/d dose is calculated from 1  $\mu$ Ci/L in urine. So the recorded doses for 1953 through 1972 are slightly higher per unit activity in urine than would be calculated using the present model. Assuming Hanford used the Morgan model through 1952, those recorded doses were increased by 0.17/0.105 to be consistent with the present model.

Because the doses shown in Table 5-29 were based on only the non-zero recorded doses, they were judged to be most applicable to unmonitored workers at the 108B facility through 1954 and the Plutonium Recycle Test Reactor, 1960 through 1969. It is assumed that tritium doses at the reactors and fuel dissolution facilities would have been at most 10% of the doses in Table 5-29 [114]. The GSDs in Table 5-29 should also be used with a minimum GSD of 3.

There was insufficient dose data in two periods, 1955 - 1960 and 1970 - 1971, for statistical analysis. Disregarding improvements in radiation protection regulations and practices, tritium exposure at the reactors and fuel dissolution plants could have been correlated with production. Using tons of uranium times fuel burn-up as a figure of merit for production (see Figure 1 in ORAUT 2007), average yearly production in 1955 through 1960 was 4 times greater than for 1949 through 1954. The average of the median daily intakes for unmonitored reactor and fuel dissolution plant workers during 1949 through 1954 was  $3.2 \times 10^5$  pCi/d (one tenth of the average of the intakes in the fifth column in Table 5-29). Four times this intake,  $1.3 \times 10^6$  pCi/d, should be used for unmonitored reactor and fuel dissolution plant workers during 1955 through 1960. The largest GSD of 4.0 can be applied to the 1955 through 1960 intakes. Production was essentially equal during 1970 and 1971 compared to 1949 through 1954 so the  $3.2 \times 10^5$  pCi/d intake should be applied to unmonitored reactor and fuel dissolution plant workers during 1970 and 1971 (GSD = 4).

No fuel dissolution was occurring during 1973 through 1983 and only one reactor was operating. The  $3.2 \times 10^5$  pCi/d intake can also be applied to unmonitored reactor workers during this period. A small set of urinalyses were collected from reactor workers in 1982-83. The set was too small for a complete statistical analysis, but the median result was about 4 pCi/ml. This results from daily intakes of about  $1.2 \times 10^4$  pCi/d [115] indicating that the  $3.2 \times 10^5$  pCi/d intake rate is favorable to claimants.

Tritium bioassay results are available for 1984 through 1986 for 100 and 200 Area workers. A coworker analysis was performed on these data [116]. The 50-percentile urinalysis results were 2.6, 0.37, and 0.21 pCi/ml for 1984, 1985, and 1986, with GSDs of 13, 3.3, and 6.2, respectively. The associated daily intakes were 7,600, 1080, and 610 pCi/d, respectively [117]. N Reactor shut down in 1987; PUREX ran intermittently in the late 1980s and was shut down in 1992 (See Section 2.2.3). The 1986 value can be applied to workers at those facilities through their respective shutdown dates [118].

By 1992 there was little exposure to tritium at Hanford except for special projects (e.g., tritium target program), low-level tracers, or in 400 Area drinking water discussed below.

Drinking water for workers in the Hanford 400 Area, of which the principal facility was the Fast Flux Test Facility (FFTF), was obtained from the unconfined aquifer which had low-level tritium contamination. The FFTF was started in 1980. The exact date well water was provided for drinking at the site has not been discovered, but it is reasonable to assume that it predated the start-up of FFTF by a year or so. An analysis of the tritium concentrations in the wells providing the drinking water and estimation of the tritium intakes from that source was performed (Bihl 2005). Some 400 Area workers were on routine tritium bioassay and the DRs should use those data when available; for other 400

Area workers, calendar day ingestion intakes of 48,000 pCi/d for 1978 through 1985 and 14,000 pCi/d for 1986 through present should be assigned (Bihl 2005). However, the annual doses from the post 1985 intakes are less than 1 mrem and can be ignored [119].

Operators, radiation monitors, and equipment maintenance workers at PUREX involved in the <sup>233</sup>U campaigns might have had intakes without monitoring specific for <sup>233</sup>U. The highest urinalysis result (4.39 pCi/d) from a batch of PUREX workers monitored in 1970 was used to estimate potential chronic intakes for the period 1965 through 1970 [120]. Because the material was absorption type F, urinary excretion equilibrates quickly so the same daily intake can be applied for 1 to 5 years chronic exposure. The daily intake producing 4.39 pCi/d uranium in urine was 16 pCi [121]. See the end of Section 5.2.5 for the contaminants that need to be added to this intake. These same workers might have been exposed to the thorium from which the <sup>233</sup>U was extracted and which was recovered and recycled. See Section 5.3.4 for a description of monitoring for thorium.

When information about what facility or type of facility an employee worked at is available, guidance on how to assign mixtures of the various radionuclides is given below. Note: ingestion intake of <sup>65</sup>Zn should be assigned to all workers per OTIB-0039.

#### 100 Area Reactors

- While operating, 1944–1987: Use fission/activation products from OTIB-0039 and <sup>131</sup>I and tritium intakes from Table 5-30.
- When not operating, 1988—present: Use fission/activation products from OTIB-0039 only; no <sup>131</sup>I or tritium; no short-lived fission/activation products after 1988 for best estimates.

**300 Area Test Reactors, 1944–1978:** Use fission/activation products from OTIB-0039 and tritium for PRTR from Table 5-30.

**400 Area, 1980–1994 (1978–1985 for tritium):** Use fission/activation products (reactors) from OTIB-0039 except use <sup>24</sup>Na (F) as the additional radionuclide for best estimates. Note that this reactor continued to be used intermittently until 1994. Use tritium ingestion from drinking water per Table 5-30.

#### 200 Area Fuel Separation Facilities

- T-Plant, 224-T, 1945–1956; B-Plant, 224-B, 1945–1952; REDOX, 224-S, 1952–1967; PUREX 1956–1992: Use fission/activation products, plutonium (M or S), and uranium (F) from OTIB-0039, except can ignore <sup>24</sup>Na from OTIB-0039 for best estimates. Use thorium intakes for PUREX 1965-70 from OTIB-0039 and <sup>233</sup>U intakes for the same workers from Table 5-30. Use tritium and <sup>131</sup>I intakes from Table 5-30. No short-lived fission/activation products after 1988 for best estimates.
- B-Plant, 225-B, 1953-present: Use fission/activation products from OTIB-0039, except can ignore <sup>24</sup>Na and the "additional radionuclide" for best estimates. Use <sup>65</sup>Zn ingestion-only intakes for 1953 1972. In other words exposure at these facilities was only to <sup>137</sup>Cs and <sup>90</sup>Sr, and <sup>65</sup>Zn via the ingestion pathway.

**Plutonium Finishing Plants (various Z buildings and 233-S), 1945–present:** Use plutonium intakes from OTIB-0039 (M or S).

**221-U, 224-U (UO**<sub>3</sub> **Plant), 1952–1993:** Use uranium type F intakes from OTIB-0039.

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# 108-B Tritium Extraction Facility, 1949–1956: Use tritium intakes from Table 5-30.

# **C-Plant (Hot Semiworks)**

• 1949–1956: Use fission/activation products (fuel dissolution) from OTIB-0039/OTIB-0054, except can ignore <sup>24</sup>Na for best estimates. Use plutonium (M or S) and uranium (F) from OTIB-0039.

Table 5-30. Intakes for workers with external monitoring but essentially no bioassay [122].

Period		Distribution	
		I .	
Through 1988		See OTIB-0039	
See OTIB-0039	See OTIB-0039	See OTIB-0039	
Through 1988	See OTIB-0039	See OTIB-0039	
All	See OTIB-0054	See OTIB-0054	
1944 - 1959	2 E4	Lognormal, GSD =3	
1960 - 1987	3.4 E3	Lognormal, GSD =3	
1988 – present	0		
1944 - 1948	6.6 E7	Lognormal, GSD = 3	108B
1944 - 1948	7 E6	Lognormal, GSD = 3	Reactors, fuel dissolution,
			rovers
1949 - 1954	From Table 5-29	Lognormal, from Table 5-29	108B
1949 - 1954	0.1 times Table 5-	Lognormal, from Table	Reactors, fuel dissolution,
	29	5-29	rovers
1955 - 1960	1.3 E6	Lognormal, GSD = 4	Reactors, fuel dissolution,
			rovers
1960 - 1969	From Table 5-29	From Table 5-29	PRTR
1961 - 1969	0.1 times Table 5- 29	From Table 5-29	Reactors, fuel dissolution
1970 - 1971	3.2 E5	Lognormal, GSD = 4	Reactors, fuel dissolution
1972	From Table 5-29	From Table 5-29	Reactors, PUREX plant
1973-83	3.2 E5	Lognormal, GSD =4	N Reactor
1984	7.6 E3	Lognormal, GSD = 13	N Reactor, PUREX
1985	1.1 E3	Lognormal, GSD = 3.3	N Reactor, PUREX
1986-87	6.1 E2	Lognormal, GSD = 6.2	N Reactor, PUREX
1988-92	6.1 E2	Lognormal, GSD = 6.2	PUREX only
1978 - 1985	4.8 E 4	Lognormal, GSD = 3	Ingestion of drinking water in the 400 Area
1965, 1966,	16	Constant	PUREX
1970			
	See discussion,		
•	last paragraph in		
	this section.		
	Period Through 1988 Through 1988 1964 through 1970 Through 1988 See OTIB-0039 See OTIB-0039 Through 1988 All  1944 - 1959 1960 - 1987 1984 - 1948 1944 - 1948 1944 - 1948 1949 - 1954 1955 - 1960 1960 - 1969 1961 - 1969 1970 - 1971 1972 1973-83 1984 1985 1986-87 1988-92 1978 - 1985	Period         Daily intake (pCi/d)           Through 1988         See OTIB-0039           1964 through 1970         See Section 5.3.4 for doses           Through 1988         See OTIB-0039           See OTIB-0039         See OTIB-0039           See OTIB-0039         See OTIB-0039           See OTIB-0039         See OTIB-0039           See OTIB-0039         See OTIB-0039           Through 1988         See OTIB-0039           Through 1988         See OTIB-0039           All         See OTIB-0039           Through 1988         See OTIB-0039           Through 1988         See OTIB-0039           Through 1988         See OTIB-0039           See OTIB-0039         See OTIB-0039           Through 1988         See OTIB-0039           See OTIB-0039         See OTIB-0039           Through 1988         See OTIB-0039           1944 - 1948         7.6           1944 - 1948	Period         (pCi/d)         Distribution           Through 1988         See OTIB-0039         See OTIB-0039           Through 1988         See OTIB-0039         See OTIB-0039           1964 through         See Section 5.3.4         Constant           1970         for doses         Constant           Through 1988         See OTIB-0039         See OTIB-0039           See OTIB-0039         See OTIB-0039         See OTIB-0039           See OTIB-0039         See OTIB-0039         See OTIB-0039           See OTIB-0039         See OTIB-0039         See OTIB-0039           All         Lognormal, GSD = 3           1944 - 1959         2 E4         Lognormal, GSD = 3           1944 - 1959         2 Fed         Lognormal, GSD = 3           1944 - 1948         7 E6 <t< td=""></t<>

 1962–1967: Use fission/activation products from OTIB-0039/OTIB-0054 (fuel dissolution), except can ignore <sup>24</sup>Na and the "additional radionuclide" for best estimates. Use <sup>65</sup>Zn ingestion intakes only.

**300 Area Fuel Fabrication Facilities (303, 306, 313, 314, 333), 1944–1988:** Use insoluble uranium intakes and thorium intakes from OTIB-0039.

**209-E and 120 Critical Mass Laboratories, 1950–1986:** Use plutonium intakes (M or S) from OTIB-0039.

**308 Plutonium Fuels Pilot Plant, 1960–1990:** Use fission/activation products (fuel dissolution), plutonium (M or S), and uranium (M or S), and <sup>147</sup>Pm intakes from OTIB-0039/OTIB-0054. Use <sup>131</sup>I intakes from Table 5-30. May ignore thorium and <sup>24</sup>Na intakes for best estimates.

**324 Building, 1966–2001:** Use fission/activation products (fuel dissolution) from OTIB-0039/OTIB-0054. The fuel was well-aged before arriving so may ignore <sup>24</sup>Na and other short-lived radionuclides in OTIB-0054 for best estimates, and may use <sup>65</sup>Zn ingestion intakes only.

**325 Building, 1953–present:** Use fission/activation products (fuel dissolution), plutonium (M or S), and <sup>147</sup>Pm intakes from OTIB-0039/OTIB-0054. May ignore <sup>24</sup>Na for best estimates; however, because of multiple uses of this facility, short-lived radionuclides may have been present past 1987.

**327 Building**, **1953–1987**: Use fission/activation products (fuel dissolution) and plutonium (M or S) intakes from OTIB-0039/OTIB-0054. May ignore <sup>24</sup>Na for best estimates.

**Tank Farms and Evaporators, 1944–present:** Use fission/activation products (waste management) and plutonium (M or S) and americium intakes at 0.001 times the fission product intake (to account for a small alpha component in the waste) from OTIB-0039. May ignore <sup>24</sup>Na for best estimates. May ignore <sup>24</sup>Na, <sup>65</sup>Zn, and the "additional radionuclide" intakes after 1988.

**D&D** and Remediation, 2002 – present: For employees of the site's remediation contractor whose job description indicates potential for exposure to airborne contamination, assign 40-DAC-hrs of unmonitored intakes per year, based on the investigation level of the lapel air sampling program. Various radionuclides are possible; however, choose the most favorable to claimants of the following (chose one): 260 pCi/d of either <sup>90</sup>Sr (type F) <sup>137</sup>Cs (type F), <sup>152</sup>Eu (type M), <sup>14</sup>C (particulate type M or S) or 0.26 pCi/d of <sup>239</sup>Pu (Type M or S) [123].

**Rovers:** Could have been exposed at many different facilities. Use the same intakes as for the fuel separations facilities, except do not include thorium or <sup>233</sup>U.

**Unknown facility:** Use judgment based on any available information and OTIB-014. If can associate the work with the 100 Area, assume exposure at a reactor; if can associate with the 200 Area, then may have to assume the worst case, which would be the fuel separations facilities; if can associate the work with the 300 Area, it would be helpful to determine if the work was associated with fuel fabrication or one of the research facilities. After 1964, fuel fabrication was done by the same contractor(s) as operated the 100 Area reactors (e.g., Douglas or United Nuclear Corporation). Assume 308 building if no other 300 Area information is available. If the work location is listed as the 600 Area, assign environmental intake only.

### 5.7 ATTRIBUTIONS AND ANNOTATIONS

Where appropriate in this document, bracketed callouts have been inserted to indicate information, conclusions, and recommendations provided to assist in the process of worker dose reconstruction. These callouts are listed here in the Attributions and Annotations section, with information to identify the source and justification for each associated item. Conventional References, which are provided in the next section of this document, link data, quotations, and other information to documents available for review on the Project's Site Research Database.

Donald Bihl served as the initial Subject Expert for this document. Mr. Bihl was previously employed at Hanford and his work involved management, direction or implementation of radiation protection and/or health physics program policies, procedures or practices related to atomic weapons activities at the site. This revision (and earlier revisions) have been overseen by a Document Owner who is fully responsible for the content, including all findings and conclusions. Mr. Bihl continues to serve as a Site Expert for this document because he possesses or is aware of information relevant for reconstructing radiation doses experienced by claimants who worked at the site. In all cases where such information or prior studies or writings are included or relied upon by Mr. Bihl, those materials are fully attributed to the source. Mr. Bihl's Disclosure Statement is available at www.oraucoc.org.

- [1] Bihl, Donald E. Battelle Pacific Northwest National Laboratory (BPNNL). Principal Health Physicist. October 2003.

  This is based on personal experience. These were the protocols for reporting results from the bioassay laboratory to the dosimetry database.
- [2] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.

  This is based on personal experience. These were the protocols for reporting results from the bioassay laboratory to the dosimetry database.
- [3] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.

  This is based on discussions with former *in vivo* counting program staff: E. Palmer and G. Rieksts. The GOK region also shows on whole-body count forms from the 1960s and 1970s.
- [4] Bihl, Donald E. BPNNL. Principal Health Physicist. From a query on the REX database by the REX custodian.
- [5] Bihl, Donald E. BPNNL. Principal Health Physicist.

  The dates for HIE and ORE are from Wilson (1978). The other remarks are from personal experience and discussions with various program managers of the Radiological Records Program.
- [6] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This is based on personal experience and review of the bioassay database.
- [7] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This is based on personal experience and review of the bioassay database.
- [8] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.

  Information in the previous paragraph came from personal experience, common knowledge passed down from previous managers of the Hanford Internal Dosimetry Program, and review of the various statements of work with the bioassay laboratory.

- [9] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This is due to the nature of the analysis, which involved separation of the plutonium then gross alpha analysis. Plutonium -241 is a beta emitter, and <sup>241</sup>Am would have been removed during the chemistry.
- [10] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This is based on personal experience. In addition, most of this information is readily noticeable based on queries of the database, which show the nature of how the data were recorded and when the changes in recording practices occurred. Detection levels are also discussed in Wilson (1987). The recording of 0.025 dpm/sample for nondetections during 1975 to 1982 and recording of actual results for measured results >0.025 dpm/sample was also verified when the former Personnel Dosimetry Program Manager, K. Heid, was asked that direct question shortly after his retirement in the late 1980s.
- [11] Bihl, Donald E. BPNNL. Principal Health Physicist. November 2004.

  This information is based on discussion in Wilson (1987) and general understanding of how the concepts of detection level and MDA have changed over the years.
- [12] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This is based on personal experience. In addition, most of this information is readily noticeable based on queries of the database, which show the nature of how the data were recorded and when the changes in recording practices occurred. Detection levels are also discussed in Wilson (1987). The recording of 0.025 dpm/sample for nondetections during 1975 to 1982 and recording of actual results for measured results >0.025 dpm/sample was also verified when the former Personnel Dosimetry Program Manager, K. Heid, was asked that direct question shortly after his retirement, in the late 1980s.
- [13] Bihl, Donald E. BPNNL. Principal Health Physicist. November 2006.
  This information was provided by J. A. MacLellan, the Bioassay Laboratory Contract Technical Administrator, and C. L. Antonio, Internal Dosimetrist in the Hanford Internal Dosimetry Program. It is also discussed in a letter to the Hanford Internal Dosimetry Program file (Antonio 2002).
- [14] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This is based on personal experience.
- [15] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.

  This is based on personal experience and discussions with J. A. MacLellan, the Bioassay Laboratory Contract Technical Administrator.
- [16] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.

  This is based on personal experience. In addition, a summary of bioassay procedures is contained in Carbaugh (1989). That two results were occasionally recorded for the same sample, because both splits were analyzed, is also evident from a query of the database.
- [17] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.

  The MDAs were extracted from various statements of work with the bioassay laboratory. The true MDAs for various times were determined as part of the laboratory's quality control program and from a double-blind oversight program operated by the Hanford Internal Dosimetry Program.

- [18] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.

  The requirement to report total propagated uncertainty shows in the 1983 statement of work (Battelle Northwest 1983). A query of the database shows that reporting of the uncertainty was sporadic in 1980 and became routine in 1981.
- [19] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This information was extracted from various statements of work with the bioassay laboratory, including AEC-RL (1964), ERDA (1975), DOE-RL (1979), and Battelle Northwest (1983, 1985, 1987, 1992).
- [20] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.

  There is some mention of the <sup>238</sup>Pu operations in Gerber (1992), although that reference does not give the date of 1967. The original reference (an incident write-up) using the year 1967 has not been relocated.
- [21] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This is based on discussions with PUREX engineers in the 1980s and from Sula et al. (1989).
- [22] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.

  The composition of the plutonium used in the Nuclear Waste Vitrification Project was added to the Internal Dosimetry Program technical basis document based on an internal memorandum from D. Bihl to Program staff, "Unusual Plutonium Mixture" (Bihl 1992), which in turn was based on a description of the mixture in a more formal document, only a portion of which was found in the files.
- [23] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. In mixtures that are mostly americium by mass or where the americium atoms have been separated from the plutonium atoms, the americium behaves as americium, not as plutonium.
- [24] Bihl, Donald E. BPNNL. Principal Health Physicist. November 2004. Information in the previous five sentences was based on a query of the REX database.
- [25] Bihl, Donald E. BPNNL. Principal Health Physicist. November 2004. No information was found to indicate whether the units were activity per sample or activity per liter during 1964. Guidance was required; therefore, the units used in 1967 to 1969 were assumed.
- [26] Bihl, Donald E. BPNNL. Principal Health Physicist. November 2004. This section is a continuation of the discussion about the 41 analyses introduced in the previous paragraph. This discussion is leading to an estimation of the MDA for the americium analysis in 1964.
- [27] Bihl, Donald E. BPNNL. Principal Health Physicist. November 2004.
  This is based on calculations by the author on the 1964 dataset in an attempt to determine the MDA of the procedure at that time.
- [28] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.

  Most of the information in this table is discussed in the text. The values during 1990 and 1991 resulted from the use of several DOE laboratories after the default of the contract with the commercial laboratory. The values from October 1991 are the same as for the plutonium

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- analysis. In addition, see Table 5-2, the discussion in the plutonium section, and Attribution 13.
- Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. [29] This is based on personal experience, results of a query of the REX database, and research on some of the cases in the files.
- [30] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This is based on personal experience.
- [31] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This information was compiled from the various statements of work with the bioassay laboratory, including AEC-RL (1964), ERDA (1975), DOE-RL (1979), and Battelle Northwest (1983, 1985, 1987, 1992).
- [32] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This information was compiled from the various statements of work with the bioassay laboratory, including AEC-RL (1964), ERDA (1975), DOE-RL (1979), Larson (1982), Battelle Northwest (1983, 1985, 1987, 1992), and Carbaugh (2003a, Appendix B).
- [33] Bihl, Donald E BPNNL. Principal Health Physicist. October 2003. This information was provided by J. A. MacLellan, the Bioassay Laboratory Contract Technical Administrator, and C. L. Antonio, Internal Dosimetrist in the Hanford Internal Dosimetry Program. It is also discussed in a letter to the Hanford Internal Dosimetry Program file (Antonio 2002).
- [34] Bihl. Donald E. BPNNL. Principal Health Physicist. October 2003. This information was compiled from the various statements of work with the bioassay laboratory, including AEC-RL (1964), Lardy (1970), ERDA (1975), DOE-RL (1979), Larson (1982), and Battelle Northwest (1983, 1985, 1987, 1992).
- [35] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This is based on personal experience. It is also evident from a query on the dosimetry database. It was standard practice to include the tritium doses as external dose until the mid-1980s.
- [36] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This information was compiled from the various statements of work with the bioassay laboratory, including AEC-RL (1964), Lardy (1970), ERDA (1975), DOE-RL (1979), Larson (1982), and Battelle Northwest (1983, 1985, 1987, 1992).
- Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. [37] Plutonium impurity would have resulted from incomplete separation of plutonium from the irradiated uranium, of which <sup>239</sup>Pu was the isotope with the highest activity. The thorium impurity was probably a mix of <sup>232</sup>Th, <sup>228</sup>Th, and <sup>234</sup>Th because all three isotopes were in PUREX; however, the assumption of 100% <sup>232</sup>Th is efficient and favorable to claimants.
- Bihl, Donald E. BPNNL. Principal Health Physicist. November 2006. [38] Because both the DOE Hanford RU document (DOE 2000) and the Hanford internal dosimetry manual (Carbaugh 2003b) express the entity as ZrNb, it is hard to know for sure if the activity is for each radionuclide or a total of both; however, DOE (2000) also refers to

- 106RuRh. In the latter case it is clear that the activity refers to the sum of the parent and progeny, so it was deemed reasonable to assume that the 20 nCi of ZrNb also refers to the sum of the parent and progeny. Because the radionuclides are usually found close to equilibrium, the recommendation was made to use 10 nCi <sup>95</sup>Zr and 10 nCi <sup>95</sup>Nb.
- [39] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. Hanford Internal Dosimetry Program staff member M. Long did some research on uranium discharged to the soil by asking various Hanford and PNNL soil chemists. Ms. Long concluded that uranium would be in a carbonate complex that most likely would be quite soluble (Long 1993).
- [40] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This is based on personal experience.
- [41] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. For some reason the reporting of activity in the <sup>233/234</sup>U energy region was set up as <sup>233</sup>U early on; the switch to reporting as <sup>234</sup>U was personally negotiated with the laboratory by Mr. Bihl.
- [42] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.

  The rapid method was coded QUS for "quick uranium soluble." Sula et al. (1989 Appendix C) refers to it as the "less sensitive method." The kit code for the QUS was 7.
- [43] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This information was compiled from the various statements of work with the bioassay laboratory, including AEC-RL (1964), Lardy (1970), ERDA (1975), DOE-RL (1979), Larson (1982), and Battelle Northwest (1983, 1985, 1987, 1992).
- Bihl, Donald E. BPNNL. Principal Health Physicist. October 2005. The  $^{233}\text{U}$  results were recorded on the same data sheets as for regular uranium analyses, and the units were printed on the form. However, the magnitude of the results argues against units of  $\mu\text{g/L}$ , and normal uranium analysis methods would not have produced results that small (ranging from 2 × 10<sup>-7</sup> to 4 × 10<sup>-6</sup>). A decision had to be made as to the units for the dose reconstructors to apply. It was reasoned that autoradiography was used to measure the tracks from the  $^{233}\text{U}$  alpha particles in the same manner as was being used for plutonium bioassay. As such the units would have been  $\mu\text{Ci/sample}$ . Someone probably forgot to mark the special units on the uranium bioassay form.
- [45] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.

  Data in the table was compiled from the references in the text and from a query of the bioassay database, which showed the distinct reporting levels used at different times.
- [46] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This is evident from a query on the bioassay data that shows use of the fission product urinalysis from 1960 to 1964. Microfiche records of several of the workers with fission product urinalysis were reviewed and showed evidence of working at B Plant and Semi-Works. The same workers showed <sup>90</sup>Sr urinalyses beginning in 1965. Therefore, it was concluded that the use of the fission product urinalysis after 1960 was for workers potentially exposed to radiostrontium.
- [47] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.
  This was explained to Mr. Bihl verbally during a regular contractual meeting between the

contract.

- [48] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.
  This information was compiled from the various statements of work with the bioassay laboratory, including AEC-RL (1964), Lardy (1970), ERDA (1975), DOE-RL (1979), Larson
- [49] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. No use of strontium titanate at Hanford has been discovered.

(1982), and Battelle Northwest (1983, 1985, 1987, 1992).

- [50] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. Personal files show an incident involving potential intakes of <sup>147</sup>Pm in the 308 Building in 1971. Several workers were scheduled for routine <sup>147</sup>Pm bioassay due to work with the material in the 222-S Building in 1964; other notes hint at earlier work with the material, probably small-scale chemistry development of processes used later in the 325 Building.
- [51] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This information was compiled from the various statements of work with the bioassay laboratory, including AEC-RL (1964), Lardy (1970), ERDA (1975), DOE-RL (1979), Larson (1982), and Battelle Northwest (1983, 1985, 1987, 1992).
- [52] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This information is based on query of the database and review of the data.
- [53] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This information was compiled from the various statements of work with the bioassay laboratory, including AEC-RL (1964), Lardy (1970), ERDA (1975), DOE-RL (1979), Larson (1982), and Battelle Northwest (1983, 1985, 1987, 1992).
- [54] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This information was compiled from the various statements of work with the bioassay laboratory, including AEC-RL (1964), Lardy (1970), ERDA (1975), DOE-RL (1979), Larson (1982), and Battelle Northwest (1983, 1985, 1987, 1992).
- [55] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This information was compiled from the various statements of work with the bioassay laboratory, including AEC-RL (1964), Lardy (1970), ERDA (1975), DOE-RL (1979), Larson (1982), and Battelle Northwest (1983, 1985, 1987, 1992).
- [56] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. That <sup>238</sup>Pu represents the majority of alpha activity in the <sup>237</sup>Np source term has been tribal knowledge passed down through the Hanford Internal Dosimetry Program and is consistent with the assumption used at the Savannah River Site. However, proof in the form of analyses of the actual product has not been found.
- [57] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2005. This is based on queries of the REX database.

- [58] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2005. This is based on queries of the REX database.
- [59] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This is based on discussion with the project manager, Dr. D. Fisher, a PNNL employee.
- [60] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This is based on a casual discussion with Dr. M. Sikov, a retired PNNL radiobiologist, now deceased.
- [61] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.

  Mr. Bihl had access to a series of Laboratory Record Notebooks associated with the in vivo counting facility. Laboratory Record Notebooks are proprietary and permission was not sought to be able to copy them.
- [62] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. The earliest detection levels came from a slide in a training class on whole body counting at Hanford, circa 1964. The author was not stated, although it was most likely Earl Palmer, the In Vivo Monitoring Program Manager at the time (Author unknown 1964). Some of the detection levels were also shown, with calculations, in Earl Palmer's 1960 Laboratory Record Notebook. Based on the calculations, Mr. Bihl estimated that the recorded detection levels most closely approximated the modern-day concept of decision level. The 10-nCi recording level for some of the radionuclides came from "Non-Routine Whole Body Counter Calculations" (Glenn 1968).
- [63] Bihl, Donald E. BPNNL. Principal Health Physicist. July 2005. This is based on query of the REX database which shows a clear cutoff of reported results at 0.66 nCi for <sup>137</sup>Cs.
- [64] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.

  This is based on personal experience. It is also evident from query on the REX database that shows the start of reporting of negative numbers on March 29, 1993.
- [65] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. The reporting of these radionuclides is evident from a query on the database. It was also obvious from hard-copy printouts of *in vivo* data from the first computer databases used by the *in vivo* counting program. A yearly printout was made of all the counts for each calendar year and stored in the Radiological Records library. These printouts had hard-coded columns for <sup>137</sup>Cs, <sup>24</sup>Na, <sup>65</sup>Zn, and <sup>40</sup>K, whereas other radionuclides were noted in a comments field. For reasons not understood by Mr. Bihl, there were some counts for which one or more of the four regularly reported radionuclides were missing. So as a general rule they were nearly always reported, but clearly there are some exceptions in the database.
- [66] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. Although the persistence of burdens of <sup>24</sup>Na and <sup>65</sup>Zn were discovered when whole-body counting became routine and the sources were investigated over the next couple of years, it is favorable to claimants and reasonable to assume that the radionuclides were in the drinking water starting soon after the first once-through-cooling reactor started at Hanford. See also ORAUT (2007a).
- [67] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.

  This is based on personal discussions with former *in vivo* counting program staff, H. E. Palmer

- and G. A. Rieksts. Mr. Palmer was the *in vivo* counting program manager for many years, and Mr. Rieksts was a scientist in the program for many years. Both are retired. The GOK region is also shown on the early whole-body counting results forms.
- [68] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. Because nearly every U.S. resident had detectable burdens of <sup>137</sup>Cs in the 1960s and 1970s, the next three bullets were provided as guidance on when to assign occupational intakes of <sup>137</sup>Cs
- [69] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This is based on personal discussions with H. E. Palmer and G. A. Rieksts.
- Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. [70] This is based on personal experience and discussion with G. A. Rieksts. In addition, see for instance the 1992 "In Vivo Bioassay Statement of Work," page 3.6 (Lynch et al. 1992).
- [71] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This is based on discussion with program managers H. E. Palmer and T. P. Lynch. In addition, it is implied in most of the annual reports, such as Lyon et al. (1988) and others.
- [72] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This is based on personal experience and discussion with H. E. Palmer.
- Bihl, Donald E. BPNNL. Principal Health Physicist. October 2005. [73] This is based on queries on the REX database for <sup>131</sup>I results.
- Bihl, Donald E. BPNNL. Principal Health Physicist. October 2005. [74] This is based on discussion with T. P. Lynch and G. A. Rieksts.
- Bihl, Donald E. BPNNL. Principal Health Physicist. November 2006. [75] This is based on calculations by Mr. Traub; however, similar values can be extracted from the graph in West (1965).
- Bihl, Donald E. BPNNL. Principal Health Physicist. June 2005. [76] The terms "detection limit' or "less-than" during the 1960s appear to be most closely associated with the present concept of decision level. The MDA is approximately twice the decision level; therefore, the recommendation was made to assume that the MDA was twice the reported detection level.
- [77] Bihl, Donald E. BPNNL. Principal Health Physicist. November 2006. This is based on discussion with T. P. Lynch; however, this is an educated guess by Mr. Lynch based on present practices and his knowledge of the detection systems at the time. Documentation that the <sup>232</sup>Th body burdens in the 1960s and 1970s were based on <sup>228</sup>Ac has not been found. It is also not known if an adjustment for disequilibrium between <sup>228</sup>Ac and <sup>232</sup>Th was made before the results were reported. It was considered favorable to claimants to assume the disequilibrium adjustment was not made and to direct the dose reconstructors to revise the <sup>232</sup>Th results based on disequilibrium factors discussed in the text.
- Traub, Richard. BPNNL. Principal Health Physicist. November 2006. [78] This is based on calculations by Mr. Traub using MatlabTM. The calculations incorporated the latest ICRP biokinetic models with independent systemic biokinetics for the major progeny.

- [79] Bihl, Donald E. BPNNL. Principal Health Physicist. December 2006. Because <sup>228</sup>Ac is usually considered part of natural background, it is not routinely calculated or reported in a whole-body count spectrum. If it were known that the worker was exposed to thorium, then any detectable activity would have been calculated and reported, but otherwise it would take a more significant peak to prompt an investigation about possible thorium exposure. The exact level of activity that would have caused an investigation has not been found; the 2-nCi level was an assumption based on the fact that levels well below 1 nCi were being recorded in the early 1970s and detection levels generally improved over time. Therefore, 2 nCi is favorable to claimants.
- [80] Traub, Richard. BPNNL. Principal Health Physicist. December 2006.
  These were calculated by Mr. Traub using MatlabTM. The calculations incorporated the latest ICRP biokinetic models with independent systemic biokinetics for the major progeny.
- [81] Traub, Richard. BPNNL. Principal Health Physicist. December 2006.

  These were calculated by Mr. Traub using MatlabTM. The calculations incorporated the latest ICRP biokinetic models with independent systemic biokinetics for the major progeny.
- [82] Bihl, Donald E. BPNNL. Principal Health Physicist. December 2006. Because of the very high activities of fission products handled in the PUREX canyon, the ventilation system was built to ensure exhaust of contaminants through the canyon filtration and exhaust system. The air flow rate through the canyon exhaust stack was nominally 100,000 ft<sup>3</sup>/min. Fuel with high thorium and <sup>233</sup>U content was dissolved in the canyon same as normal fuel and thoron was exhausted out the canyon stack.
- [83] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.

  This is based on personal experience and also evident from a query of the REX database.
- [84] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.

  This is based on personal experience and discussion with T. Lynch, *in vivo* monitoring program manager. The correction for shine from the liver is also mentioned in Palmer (1990).
- [85] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.

  This is based on review of the REX database and a sampling of whole-body count results forms in the 1960s and 1970s.
- [86] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.

  This is based on discussion with T. Lynch, *in vivo* monitoring program manager. Document research has not found a reference that states this precisely, but Lynch (1992) shows the detailed work being done in relation to decision levels at that time.
- [87] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This is evident from a review of various whole-body count results forms.
- [88] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. These changes were evident from review of the REX database.
- [89] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. Implementation of NEXEC is discussed in Lyon et al. (1996). Lynch (1995, Table 4.1) shows the 12 regions for the stand-up counter, and Table 4.5 shows the 20 regions for the coaxial germanium counter.

- [90] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This is based on personal experience and discussion with T. Lynch.
- [91] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. The minutes of the November 1998 meeting of the Dosimetry Advisory Committee (Bihl 1998) show the proposal by T. Lynch to reduce the routinely reported radionuclides in whole-body counts to <sup>137</sup>C, <sup>60</sup>Co, <sup>154</sup>Eu, <sup>40</sup>K, and -<sup>24</sup>Na. By the time the ABACOS software was implemented in November 1999, <sup>24</sup>Na had been dropped because Hanford contractors believed there was no significant source of <sup>24</sup>Na left. Subsequent minutes of the Dosimetry Advisory Committee were scanned, but documentation of the decision to drop <sup>24</sup>Na. was not found.
- [92] Bihl, Donald E. BPNNL. Principal Health Physicist. May 2006. FFTF was sodium cooled so neutron activation of the sodium was a potential source of contamination. See Section 2.1.9.
- [93] Bihl, Donald E. BPNNL. Principal Health Physicist. March 2007. The principal <sup>154</sup>Eu photopeak is at 1.27 MeV with 35% abundance; the photopeak used to quantify <sup>60</sup>Co was at 1.33 MeV with 100% abundance. The detector efficiencies per emitted photon would have been nearly the same for the two photons; the background counts in the respective energy regions would also have been similar. Therefore, the MDAs would have differed by the ratio of the yields, or 1.00/0.35 ≈ 3.
- Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This is evident from a query on the database. Error limits are listed as "±10% at the 95% confidence level at 10 times the detection limits" in the earliest statement of work with the bioassay laboratory (AEC-RL 1964), and similar statements show up in subsequent statements of work, but uncertainties for each sample were not reported at that time. There was probably a change to the statement of work in 1981 that instigated or accompanied the change in that year to start reporting uncertainties for each sample but a copy of that statement of work has not been found. Propagation and reporting of uncertainty for each sample shows up in the 1983 bioassay laboratory statement of work (Battelle Northwest 1983) and in each statement of work thereafter.
- [95] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. The collection instructions are to collect starting with the last excretion before retiring for sleep, any other voidings during the night (or sleep period if on shift), and first excretion on rising in the morning for two consecutive nights. This covers about 18 hours or about 75% of a 24-hour period.
- [96] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. Routine urine samples with volume less than 500 mL were rejected (code IS for insufficient sample) and the worker was asked to resample. This was a quality control measure designed to prevent samples that were grossly less than a 24-hour sample. The volume available in the collection containers (which varied over the years from 1.5L to 3L) and the limited amount of time for collection provided an upper limit to the uncertainty associated with 24-hour collection. Therefore, the factor of 2 was considered reasonable for the range of the uncertainty. Dose reconstructors use an overall uncertainty for internal dose estimates of a GSD of 3, so the uncertainty associated with sample collection is considered part of the overall uncertainty.

- [97] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This is based on discussion with the *in vivo* monitoring program manager, T. Lynch.
- [98] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. This is based on discussion with the *in vivo* monitoring program manager, T. Lynch.
- [99] Bihl, Donald. BPNNL. Principal Health Physicist. October 2003.

  This is based on discussion with the *in vivo* monitoring program manager, T. Lynch.
- [100] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.

  The 95th-percentile phantom did not exist during the period that the 1-m arc or the shadow shield detectors were in use, and that phantom was not measured on the standup counter, so the impact of large workers on results from those detectors has not been quantified. Mr. Lynch believed the 25% bias was a reasonable figure to use for the latter detectors as well as the coaxial counter based on his general experience with the various detectors.
- [101] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003.

  This is based on discussion with the *in vivo* monitoring program manager, T. Lynch, and G. A. Rieksts.
- [102] Bihl, Donald E. BPNNL. Principal Health Physicist. November 2004. This is based on input by R. Falk during review of Rev. 01 of this document. A GSD of 5 covers a range of 2.3 orders of magnitude at the 95th-percentile confidence level.
- [103] Bihl, Donald E. BPNNL. Principal Health Physicist. December 2005. N Reactor, the last source of short-lived fission products, shut down in January 1987 after a brief operating period, and was also shut down for most of 1986. The December 31, 1988, date was arbitrarily chosen to allow time for the short half-life radionuclides to decay such that they were no longer significant contributors to contamination.
- [104] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. Workers without at least a basic dosimeter were not allowed into radiation areas or general areas of the site (200 Area, 300 Area, etc.) where there was some risk of exposure; some companies gave beginning work or termination bioassay to everyone regardless of their work location or tasks, and those bioassay are not indicative of exposure. Therefore, after consultation with other ORAUT experts (including J. Fix, E. Scalsky, and E. Brackett), the conclusion was reached that workers without at least a basic dosimeter were not at risk for occupational intakes, and the instruction was made to assign environmental intakes only.
- [105] Bihl, Donald E. BPNNL. Principal Health Physicist. December 2005.
  This is based on queries of the REX database and an incident record in the worker's file about the T Plant incident.
- [106] Bihl, Donald E. BPNNL. Principal Health Physicist. December 2005. Equilibrium thyroid burden from daily inhalation of 20,000 pCi of type F <sup>131</sup>I is 29.5 nCi after about 80 days. This calculation was performed using IMBA. This burden exceeds 93% of all the measurements made on at-risk workers in 1945 and 1946 and exceeds 90% of the measurements made in 1962 to 1969, so is was deemed to be favorable to claimants to apply the dose unmonitored workers in years before implementation of routine *in vivo* counts.

- [107] Bihl, Donald E. BPNNL. Principal Health Physicist. December 2005. The whole-body burden from daily inhalation of 3,400 pCi of type F <sup>131</sup>I is 5 nCi after only 10 days of intake and reaches an equilibrium burden of 7.2 nCi after about 80 days. This calculation was performed using IMBA.
- [108] Bihl, Donald E. BPNNL. Principal Health Physicist. December 2005.

  N Reactor shut down in May 1986 and restarted very briefly in January 1987. With an 8-day half-life, <sup>131</sup>I activities would have been negligible by the end of the 1987.
- [109] Bihl, Donald E. BPNNL. Principal Health Physicist. May 2006. This is based on query of the REX database.
- [110] Bihl, Donald E. BPNNL. Principal Health Physicist. May 2006. This is based on query of the REX database.
- [111] Bihl, Donald E. BPNNL. Principal Health Physicist. May 2006. This was calculated using IMBA.
- [112] Bihl, Donald E. BPNNL. Principal Health Physicist. May 2006.

  There were considerable differences in potential exposure to tritium between the 108B facility, which involved extracting and packaging curie quantities of tritium in a relatively small space, and exposures at large, heavily ventilated facilities with unconcentrated tritium such as the reactors and fuel separation facilities. Most of the tritium at the latter facilities was exhausted through the stacks or discharged to the ground in cribs. It is likely that the relative exposure to workers at the reactors or fuel separations plants was less than an order of magnitude relative to exposure at 108B. However, lacking air sample data, the order of magnitude ratio was considered reasonable and favorable to claimants.
- [113] Bihl, Donald E. BPNNL. Principal Health Physicist. Joseph Lochamy. Scientist. XCEL Engineering; and Richard Sparks. CDE Dosimetry Services. Principal Health Physicist. December 2006.

  The tritium dose data is from a query on REX by Mr. Bihl, a lognormal statistical analysis was performed by Mr. Lochamy in accordance with ORAUT-PROC-0095 (ORAUT 2006b), and a validation of statistical analysis was performed by Mr. Sparks.
- [114] Bihl, Donald E. BPNNL. Principal Health Physicist. May 2006.

  There were considerable differences in potential exposure to tritium between the 108B facility, which involved extracting and packaging curie quantities of tritium in a relatively small space, and exposures at large, heavily ventilated facilities with unconcentrated tritium such as the reactors and fuel separation facilities. Most of the tritium at the latter facilities was exhausted through the stacks or discharged to the ground in cribs. It is likely that the relative exposure to workers at the reactors or fuel separations plants was less than an order of magnitude relative to exposure at 108B. However, lacking air sample data, the order of magnitude ratio was considered reasonable and favorable to claimants.
- [115] Bihl, Donald E. BPNNL. Principal Health Physicist. November 2006. This was calculated using IMBA.
- [116] Bihl, Donald E. BPNNL. Principal Health Physicist; Joseph Lochamy. Scientist. XCEL Engineering, Inc.; and Richard Sparks. CDE Dosimetry Services, Inc. Principal Health Physicist. December 2006.

The tritium dose data is from a query on REX by Mr. Bihl, a lognormal statistical analysis was performed by Mr. Lochamy in accordance with ORAUT-PROC-0095 (ORAUT 2006b), and a validation of statistical analysis was performed by Mr. Sparks.

- [117] Bihl, Donald E. BPNNL. Principal Health Physicist. December 2006. These were calculated using IMBA.
- [118] Bihl, Donald E. BPNNL. Principal Health Physicist. December 2006.

  Although the tritium source term would have been smaller in the late 1980s and early 1990 because of reduced processing of fuel, it is favorable to claimants to use the values for 1984 to 1986 for the later years.
- [119] Bihl, Donald E. BPNNL. Principal Health Physicist. June 2005. These were calculated using IMBA. Ingestion of 40,000 pCi/d produces approximately 1 mrem/yr to all organs.
- [120] Bihl, Donald E. BPNNL. Principal Health Physicist. June 2006.

  There were not enough samples for a formal coworker analysis (n=11). The units were not known exactly; based on the magnitude of the results, microcuries per liter was assumed (see also Section 5.2.5).
- [121] Bihl, Donald E. BPNNL. Principal Health Physicist. June 2006. This was calculated using IMBA.
- [122] Bihl, Donald E. BPNNL. Principal Health Physicist. June 2006. This is a summary of information developed in the text.
- [123] Bihl, Donald E. BPNNL. Principal Health Physicist. June 2006.

  This is based on discussion with the D&D contractor's dosimetry expert, E. W. Carlson. It reflects radionuclides being encountered during D&D operations of old facilities, burial sites, liquid effluent discharge zones, etc. Because most of the work is outside, the contractor uses lapel air samplers to monitor for intakes. The 40-DAC-hr value comes from the requirement to initiate special bioassay if an individual's cumulative intake in a year as measured by the air samplers reaches or exceeds 40 DAC-hr (Carbaugh 2003b).
- [124] Bihl, Donald E. BPNNL. Principal Health Physicist. October 2003. The air concentration tolerance value as applied to fission products was deduced from the fact that other values were given for "product," <sup>131</sup>I, and uranium. In addition, the magnitude of the limit implies general fission products.

#### REFERENCES

- AEC- RL (U.S. Atomic Energy Commission Richland Operations Office), 1964, Draft statement of work with the commercial bioassay laboratory, August. [SRDB Ref ID 7788]
- AEC (U.S. Atomic Energy Commission), 1968, Manual Chapter 0524, "Standards for Radiation Protection," AEC 0524-01, November 8. [SRDB Ref ID 25866]
- Andersen, B. V. 1971, "Battelle Northwest In-Vivo Measurements," Pacific Northwest Laboratory, Richland, Washington, November 1. [SRDB Ref ID 17511]
- Antonio, C. L., 2002, Letter to Hanford Internal Dosimetry Program file, "Decision level calculation for excreta bioassay results using alpha spectrometry," Pacific Northwest National Laboratory, Richland, Washington, February 12. [SRDB Ref ID 609]
- Author unknown, 1964, pages from In-vivo Bioassay Methods and Sensitivities, Pacific Northwest Laboratory, Richland, Washington. [SRDB Ref ID 17510]
- Battelle Northwest. 1970, Radiation Work Procedure 306-4, Pacific Northwest Laboratory, Richland, Washington, November 18. [SRDB Ref ID 27673]
- Battelle Northwest, 1983, Statement of work with the commercial bioassay laboratory. Pacific National Laboratory, Richland, Washington, October. [SRDB Ref ID 7794]
- Battelle Northwest, 1985, Statement of work with the commercial bioassay laboratory. Pacific National Laboratory, Richland, Washington, October. [SRDB Ref ID 7796]
- Battelle Northwest, 1987, Statement of work with the commercial bioassay laboratory, third operational year, Appendix A . Pacific National Laboratory, Richland, Washington, September. [SRDB Ref ID 27676]
- Battelle Northwest, 1992, Statement of work with the commercial bioassay laboratory. Pacific National Laboratory, Richland, Washington, October. [SRDB Ref ID 7794]
- Baumann, B. L., 1996, Letter to S. B. Lamson and D. S. Mantooth, "Bioassay for 222-S Yttrium-90 Analysis," August 1.
- Bihl, D. E., 1992, Internal memo, "Unusual Plutonium Mixture", January 16. [SRDB Ref ID 30252]
- Bihl, D. E., R. L. Buschbom, and M. J. Sula, 1993, "Experience with a Routine Fecal Sampling Program for Plutonium Workers," *Health Physics*, 65(5). [SRDB Ref ID 13396]
- Bihl, D.E., 1994, letter to A.P. Mileham, "Is Bioassay Needed for Thorium Work in 3720 Building?" Battelle Pacific Northwest Laboratory, Richland, Washington, July 26.
- Bihl, D.E., 1995, letter to J. R. Hensley, "Intake monitoring for the Actinium-227 Project," August 7.
- Bihl, D. E., 1998, "Minutes of the Hanford Personnel Dosimetry Advisory Committee Held on November 18, 1998," Pacific Northwest National Laboratory, Richland, Washington.
- Bihl. D. E., 2003, Personal interview with M. M. Lardy. [SRDB Ref ID 606]

- Bihl, D.E., 2005. Letter to Edward D. Scalsky, "Intakes from Tritium Contaminated Drinking Water for the Hanford 400 Area Workers," January 11. [SRDB Ref ID 18103]
- Bihl, D.E. and R.J. Traub, 2007, Letter to file, "Revision to 'Basis for Thoron Concentration and Doses for Thoria Processing in 3722 and 3732 Buildings at Hanford' Based on Review by OCAS," January 3. [SRDB Ref ID 30120]
- Boothe, G. F., 1992, Letter to M. M. McCarthy, "Radiological Characterization of Waste," 24910-92-GFB-0002, Westinghouse Hanford Company, Richland, Washington, August 25.
- Brady, D. N. (editor), 1964, *Hanford Whole Body Counter Activities for 1961-1963*, HW-82409, Hanford Atomic Products Operation, Richland Washington. SRDB Ref ID 7771 (sub-doc, p. 5 of 75)
- Cantril, S. T., 1945, "Tolerance Limits," letter to file, October 18. [SRDB Ref ID 13428]
- Carbaugh, E. H., 1995, Letter to distribution dated June 1, 1995, "Bioassay for Spent Fuel and Waste Handling, Storage, and Analysis Facilities," Pacific Northwest National Laboratory, Richland, Washington. [SRDB Ref ID 7771 (sub-doc, p. 63 of 75)]
- Carbaugh, E. H., 1989, *Hanford Internal Dosimetry Project Manual*, PNL-MA-552, Rev. 0, Pacific Northwest Laboratory, Richland, Washington. [SRDB Ref ID 277]
- Carbaugh, E. H., 1990, Letter to D. P. Higby, "Bioassay for Yttrium-90," September 27.
- Carbaugh, E. H., 2003a, *Methods and Models of the Hanford Internal Dosimetry Program*, PNL-MA-860, Pacific Northwest National Laboratory, Richland, Washington. [SRDB Ref ID 11521]
- Carbaugh, E. H. 2003b, *Hanford Internal Dosimetry Project Manual*, PNL-MA-552, Pacific Northwest National Laboratory, Richland, Washington. [SRDB Ref ID 11521]
- Carbaugh, E. H., M. J. Sula, T. L. Aldridge, and H. B. Spitz, 1988, "The Impact of Upgraded In Vivo Lung Measurement Capability on an Internal Dosimetry Program," *Ann. Occup. Hyg.*, Vol 32, Supplement 1, pp. 885-892. [SRDB Ref ID 7774]
- Cooke, N., and F. B. Holt, 1974, "The Solubility of Some Uranium Compounds in Simulated Lung Fluid," *Health Physics*, Vol. 27 (July). [SRDB Ref ID 13357]
- DOE (U.S. Department of Energy), 1989, "Radiation Protection for Occupational Workers," Order 5480.11, Washington, D.C. [SRDB Ref ID 8073]
- DOE (U.S. Department of Energy), 1997, "Linking Legacies, Connecting the Cold War Nuclear Weapons Production Processes to Their Environmental Consequences," Washington, D.C. [SRDB Ref ID 11930]
- DOE (U.S. Department of Energy), 2000, Recycled Uranium, the Flow and Characteristics of Recycled Uranium Throughout the DOE Complex 1952-1999, Volume II: Part 1, Hanford Site, Site Assessment Team Report, DOE/RL-2000-43, Washington, D.C. [SRDB Ref ID 4971]
- DOE-RL (U.S. Department of Energy, Richland Operations Office), 1979, Statement of work with the commercial bioassay laboratory. Richland, Washington, February. [SRDB Ref ID 7791]

- Douglas United Nuclear, Inc., 1965, Engineering Specifications, Thorium Oxide Target Elements, RL-REA—2177, Specification number T-100, "Thorium Oxide," (effective date on the T-100 version obtained was April 22, 1968). [SRDB Ref ID 22756]
- ERDA- RL (Energy Research and Development Administration Richland Operations Office), 1975, Statement of work with the commercial bioassay laboratory. March. [SRDB Ref ID 7790]
- Fisher, D. R., 1986, "Particle Size Distribution and Solubility of Uranium Aerosols in 333 and 303 M Buildings at UNC and PNL's 306W Building," letter to Monte J. Sula. [SRDB Ref ID 15196]
- Gerber, M. S. 1992, Past Practices Technical Characterization Study 300 Area Hanford Site, WHC-MR-0388, Westinghouse Hanford Company, Richland, Washington. [SRDB Ref ID 13724]
- Glenn, D., 1968, "Non-Routine Whole Body Counter Calculations," unpublished set of instructions in the In Vivo Radioassay and Research Facility archives. [SRDB Ref ID 7771 (sub-doc. p. 30 of 75)]
- Glover, J. C., 1964, letter to H. V. Larson, "Bioassay Sampling and Analysis for Personnel Working with Pomethium-147, General Electric, Richland, Washington. [SRDB Ref ID 30238]
- Greninger, A.B., 1964, U-233 Production, RL-REA-354-DEL or HAN-90292, General Electric Hanford Atomic Products Operation, Richland, Washington. [SRDB Ref ID 20257]
- HASL (Health and Safety Laboratory), 1985 or similar year, HASL Procedures Manual, HASL-300. Energy Research and Development Administration, Health and Safety Laboratory, New York, New York.
- Healy, J. W., 1948, "Bioassay at Hanford," HW-10522, set of procedures and related discussion, Hanford Works, Richland, Washington. [SRDB Ref ID 4909]
- Healy, J. W., 1949, "Urine Analysis for Tritium Oxide," HW-13949, procedure, Hanford Works, Richland, Washington. [SRDB Ref ID 13422]
- Henle, R.C., undated note to Hanford Internal Dosimetry Program Po-210 file, author assumed based on comparison to other handwritten notes by Henle. [SRDB Ref ID 19314]
- Henle, R. C., 1962, "1961 Whole Body Counter Results," letter to F. Swanberg, Jr., January 9. [SRDB Ref ID 7771 (sub-doc p. 3 of 75)]
- Henle, R. C., 1967, handwritten note, "U.S. Testing <sup>210</sup>Po Bioassay Detection Capabilities, with followup notes on 2/14/1968 and 3/11/1968. Hanford Internal Dosimetry Program files, Battelle Pacific Northwest Laboratory, Richland, Washington. [SRDB Ref ID 30250]
- Henle, R. C., 1968, Draft presentation, "Evaluation of Internal Depositions of Americium Using Bioassay Samples," Hanford Internal Dosimetry Program files, Battelle Pacific Northwest Laboratory, Richland, Washington. [SRDB Ref ID 27677]
- Howell, W. P., and G. E. King, 1968, Radiation Protection Aspects of Work with Promethium-147, PNW-SA-1038, Pacific Northwest Laboratory, Richland, Washington. [SRDB Ref ID 13702]

- Howell, W. P., 1969, Letter to J. J. Jech, "Bioassay Sampling <sup>147</sup>Pm Workers," Battelle Pacific Northwest Laboratory, Richland, Washington, October 6. [SRDB Ref ID 30241]
- Ikenberry, T. A., 1991, Evaluation of Thyroid Radioactivity Measurement Data from Hanford Workers, 1944-1946, PNL-7254 HEDR, Pacific Northwest Laboratory, Richland, Washington. [SRDB Ref ID 14997]
- ICRP (International Commission on Radiological Protection), 1959, Report of Committee II on Permissible Dose from Internal Radiation, Publication 2, Pergamon Press, Oxford, England.
- ICRP (International Commission on Radiological Protection, 1975, *Reference Man: Anatomical, Physiological and Metabolic Characteristics*, Publication 23, Pergamon Press, Oxford, England.
- ICRP (International Commission on Radiological Protection), 1979, "Limits for Intakes of Radionuclides by Workers," *Annals of the ICRP (3/4)*, Publication 30, Pergamon Press, Oxford, England.
- ICRP (International Commission on Radiological Protection), 1994, "Human Respiratory Tract Model for Radiological Protection," *Annals of the ICRP 24 (1-3)*, Publication 66, Pergamon Press, Oxford, England.
- ICRP (International Commission on Radiological Protection), 1995, "Dose Coefficients for Intakes of Radionuclides by Workers," Replacement of ICRP Publication 61, *Annals of the ICRP 24 (4)*, Publication 68, Pergamon Press, Oxford, England.
- ICRP (International Commission on Radiological Protection, 1996, *Age-dependent Doses to Members of the Public from Intake of Radionuclides: Part 4 Inhalation Dose Coefficients*, Publication 71, Pergamon Press, Oxford, England.
- ICRP (International Commission on Radiological Protection), 1997, *Individual Monitoring for Internal Exposure of Workers*, Publication 78, Pergamon Press, Oxford, England.
- Isochem, Inc., 1967, *PUREX Plant Thorium Process Operation Report*, ISO-419, Isochem, Inc., Richland, WA. [SRDB Ref ID 31708]
- Jech, J. J., 1967, letter to C. D. Corbit, September 26. [SRDB Ref ID 14888]
- Jech. J. J., 1969a, letter to File, September 12, [SRDB Ref ID 17509]
- Jech, J. J., 1969b, letter to C.D. Corbit, December 17. [SRDB Ref ID 14888]
- Jech, J. J, 1970a, "Internal Deposition Surveillance Enriched Uranium Work," letter to J. H. Soehnlein, December 14. [SRDB Ref. ID 27675]
- Jech, J. J., 1970b, letter to G. E. Backman, November 10. [SRDB Ref ID 17504]
- Jech, J. J., 1972, "Internal Surveillance <sup>210</sup>Po," letter to K. R. Heid, March 27. [SRDB Ref ID 17503]
- Lardy, M. M., 1970, letter to J. P. Corley, April 23. [SRDB Ref ID 13399]
- Lardy, M. M., 1974, letter to H. Larson, March 27. [SRDB Ref ID 7787 (sub-doc, p. 189)]

- Larson, H. V., 1982, Hanford Dosimetry Evaluation Manual, Appendix L, "Collection and Radiochemical Analysis of Excreta and Other Samples," PNL-MA-575, Battelle Pacific Northwest Laboratories, Richland, WA. [SRDB Ref ID 30238]
- Long, M. P., 1993, internal memo to Don Bihl, "Uranium Compounds in Soil," October 19. [SRDB Ref ID 30251]
- Long, M. P., 1997, internal memo to E.H. Carbaugh, "Bioassay for LSL-II Animal Study," April 4.
- Lynch, T. P., 1992, "Program Change Record, Hanford Internal Dosimetry Program, WBC," Change Record Number 92003, May 21.
- Lynch, T. P., J. A. MacLellan, and D. E. Bihl, 1992, "In Vivo Bioassay Statement of Work," Pacific Northwest Laboratory, Richland, Washington.
- Lynch, T. P., 1995, *Whole Body Counting Manual*, PNL-MA-574 Rev. 2, Pacific Northwest Laboratories, Richland, Washington.
- Lynch, T. P., J. A. MacLellan, D. E. Bihl, R. K. Piper, M. L. Johnson, 2000, *Hanford Radiological Protection Support Services Annual Report for 1999*, PNNL-13238, Pacific Northwest National Laboratory, Richland, Washington.
- Lynch, T. P., J. A. MacLellan, D. E. Bihl, R. K. Piper, M. L. Johnson, 2001, *Hanford Radiological Protection Support Services Annual Report for 2000*, PNNL-13542, Pacific Northwest National Laboratory, Richland, Washington.
- Lynch, T. P., 2001, In Vivo Monitoring Project Manual, PNL-MA-574, Rev. 3. [SRDB Ref ID 13421]
- Lynch, T. P., 2003, electronic message to D. E. Bihl, "Hanford CWT Estimates," June 17.
- Lynch, T. P., 2007, "Effect of Phantom Size on Efficiency of Coaxial Germanium System," Hanford In Vivo Monitoring Program, Pacific Northwest National Laboratory, Richland, Washington, March 21.
- Lyon, M., J. J. Fix, J. L. Kenoyer, J. A. Leonowich, H. E. Palmer, and M. J. Sula, 1988, *Hanford Radiological Protection Support Services Annual Report for 1987*, PNL-6624, Pacific Northwest Laboratory, Richland, Washington. [SRDB Ref ID 15198]
- Lyon, M, J. J. Fix, J. L. Kenoyer, J. A. Leonowich, H. E. Palmer, and M. J. Sula, 1989, *Hanford Radiological Protection Support Services Annual Report for 1988*, PNL-6952, Pacific Northwest Laboratory, Richland, Washington. [SRDB Ref ID 15197]
- Lyon, M., D. E. Bihl, J. J. Fix, J. L. Kenoyer, J. A. Leonowich, and H. E. Palmer, 1990, *Hanford Radiological Protection Support Services Annual Report for 1989*, PNL-7417, Pacific Northwest Laboratory, Richland, Washington. [SRDB Ref ID 13423]
- Lyon, M., D. E. Bihl, J. J. Fix, R. K. Piper, T. J. Froelich, J. A. Leonowich, and T. P. Lynch, 1991, Hanford Radiological Protection Support Services Annual Report for 1990, PNL-7752, Pacific Northwest Laboratory, Richland, Washington. [SRDB Ref ID 13424]

- Lyon, M., D. E. Bihl, J. J. Fix, R. K. Piper, T. J. Froelich, J. A. Leonowich, and T. P. Lynch, 1992, Hanford Radiological Protection Support Services Annual Report for 1991, PNL-8236, Pacific Northwest Laboratory, Richland, Washington. [SRDB Ref ID 13425]
- Lyon, M., D. E. Bihl, J. J. Fix, R. K. Piper, T. J. Froelich, and T. P. Lynch, 1993, *Hanford Radiological Protection Support Services Annual Report for 1992*, PNL-8764, Pacific Northwest Laboratory, Richland, Washington.
- Lyon, M., D. E. Bihl, J. J. Fix, T. J. Froelich, R. K. Piper, and P. C. Olsen, 1994, *Hanford Radiological Protection Support Services Annual Report for 1993*, PNL-10047, Pacific Northwest Laboratory, Richland, Washington.
- Lyon, M., D. E. Bihl, J. J. Fix, R. K. Piper, T. J. Froelich, and P. C. Olsen, 1995, *Hanford Radiological Protection Support Services Annual Report for 1994*, PNL-10614, Pacific Northwest Laboratory, Richland, Washington.
- Lyon, M., D. E. Bihl, E. H. Carbaugh, J. J. Fix, R. K. Piper, and T. J. Froelich, 1996, *Hanford Radiological Protection Support Services Annual Report for 1995*, PNNL-11145, Pacific Northwest National Laboratory, Richland, Washington.
- Lyon, M., D. E. Bihl, J. J. Fix, M. L. Johnson, T. P. Lynch, and R. K. Piper, 1998, *Hanford Radiological Protection Support Services Annual Report for 1997*, PNNL-11881, Pacific Northwest National Laboratory, Richland, Washington. [SRDB Ref ID 30253]
- MacLellan, J. A., D. E. Bihl, M.L. Johnson, T. P. Lynch, and R. K. Piper, 1999, *Hanford Radiological Protection Support Services Annual Report for 1998*, PNNL-12191, Pacific Northwest National Laboratory, Richland, Washington.
- MacLellan, J. A., E J. Wyse, L.P. Scott, 1995, "Discrimination Between Occupational and Environmental Sources of Internal Uranium Exposure," poster presentation WAM B11, in Abstracts of Papers Presented at the Fortieth Annual Meeting of the Health Physics Society, Boston, Massachusetts, *Health Physics*, Supplement to Vol. 68, No. 6, June 1995.
- Morgan, K. Z., 1947, "Tolerance Concentrations of Radioactive Substances", Journal Phys. Colloid. Chemistry, Vol. 51, pp. 984 1003. [SRDB Ref ID 11537]
- McConnon, D.,1964, Untitled or numbered memo to PRTR Radiation Monitoring Personnel, Hanford Works, Richland, Washington, June 11. [SRDB Ref ID 13427]
- McConnon, D., 1967, "Tritium Experience at Pacific Northwest Laboratory," BNWL-SA-1026, presentation made at a symposium on instrumentation and health physics problems with tritium [title not given], Albuquerque, New Mexico, February 16-17. [SRDB Ref ID 13430]
- McConnon, D., C.R. Cole, V.H. Smith, undated, "The Comparative Metabolism of Intravenously Injected Promethium in Swine and Humans, BNWL-SA-3747, Pacific Northwest Laboratories, Richland, Washington. [SRDB Ref ID 17513]
- Medley, D. W., R. L. Kathren, and A. G. Miller, 1994, "Diurnal urinary volume and uranium output in uranium workers and unexposed controls," *Health Physics*. 67(2): 122-130. [SRDB Ref ID 13348]

- NBS (National Bureau of Standards), 1953, Maximum Permissible Amounts of Radioisotopes in the Human Body and Maximum Permissible Concentrations in Air and Water, Handbook 52, Superintendent of Documents, Washington, D.C.
- NBS (National Bureau of Standards), 1959, Maximum Permissible Body Burdens and Maximum Permissible Concentrations of Radionuclides in Air and Water for Occupational Exposure, Handbook 69, Superintendent of Documents, Washington, D.C.
- NCRP (National Council on Radiological Protection and Measurements), 1987, Exposure of the Population in the United States and Canada from Natural Background Radiation, Report 94, Bethesda, Maryland.
- NIOSH (National Institute for Occupational Safety and Health), 2002, Internal Dose Reconstruction Implementation Guideline, OCAS-IG-002, Office of Compensation Analysis and Support, Cincinnati, Ohio.
- NIOSH, (National Institute for Occupational Safety and Health) 2004, Estimation of Ingestion Intakes, OCAS-TIB-009, Rev. 0, Office of Compensation Analysis and Support, Cincinnati, Ohio.
- NIOSH (National Institute for Occupational Safety and Health), 2006, Lung Dose Conversion Factor for Thoron WLM, TIB-011, Office of Compensation Analysis and Support, Cincinnati, Ohio.
- ORAUT (Oak Ridge Associated Universities Team), 2004, Technical Basis Document for the Los Alamos National Laboratory – Occupational Internal Dose, ORAUT-TKBS-0010-5, Rev. 00, Oak Ridge, Tennessee, December 21.
- ORAUT (Oak Ridge Associated Universities Team), 2006a, Hanford Site Occupational External Dose, ORAUT-TKBS-0006-6, Rev. 02, Oak Ridge, Tennessee, November 21.
- ORAUT (Oak Ridge Associated Universities Team), 2006b, Generating Summary Statistics for Coworker Bioassay Data, ORAUT-PROC-0095, Rev. 00, Oak Ridge, Tennessee, June 5.
- ORAUT (Oak Ridge Associated Universities Team), 2007a, Internal Dosimetry Coworker Data for the Hanford Site, ORAUT-OTIB-0039, Rev. PC-2, Oak Ridge, Tennessee, January 31.
- ORAUT (Oak Ridge Associated Universities Team), 2007b, Fission and Activation Product Assignment for Internal Dose-Related Gross Beta and Gross Gamma Analyses, ORAUT-OTIB-0054, Rev. 00-C, Oak Ridge, Tennessee, March 9.
- Palmer, H. E., G. H. Crook, and T. C. Nelson, 1969, The Uptake, Distribution, and Excretion of Promethium in Humans and the Effect of DTPA on These Parameters, BNWL-SA-2432, Pacific Northwest Laboratory, Richland, Washington. [SRDB Ref ID 13431]
- Palmer, H.E. and G. A. Rieksts, 1979, "Hanford Whole Body Counter 1978 Activities," internal report, In Vivo Monitoring Program, Pacific Northwest Laboratory, Richland, Washington, March.
- Palmer, H. E., G. A. Rieksts, and H. B. Spitz, circa 1984, "Hanford Whole Body Counter 1983 Activities," internal report, In Vivo Monitoring Program, Pacific Northwest Laboratory, Richland, Washington. [SRDB Ref ID 17502]

- Palmer, H. E., and G. Rieksts, 1984, "The Use of Planar High-Purity Ge Detectors for In Vivo Measurement of Low-Energy Photon Emitters," *Health Physics*, Vol. 47, No.4, pp 569-578.
- Palmer, H. E., 1985, "Measurement Sensitivities for Radionuclides in the Body," memo to Monte Sula, April 1. [SRDB Ref ID 17512]
- Palmer, H. E., 1990, *Whole Body Counting Manual*, PNL-MA-574, Pacific Northwest Laboratories, Richland, Washington.
- Parker, H. M., 1945, "Proposed Revision of Tolerances for I<sup>131</sup>," letter to file, Hanford Works, Richland, Washington, December 17. [SRDB Ref ID 7766]
- Parker, H. M., 1947, "Introductory Lecture Series in HI [Health Instrumentation]," HW-7184 Pt 2, Lecture 16-21, Hanford Works, Richland, Washington. [SRDB Ref ID 24119]
- Parker, H. M., 1950, "P-10 Hazards," letter to A. B. Greninger, HW-15634, Hanford Works, Richland, Washington, January 10. [SRDB Ref ID 13420]
- Parker, H. M., 1960, "Radiation Protection Criteria and Standards: Their Use and Basis," Chapter 1 in Selected Material on Radiation Protection Criteria and Standards: Their Basis and Use," Joint Committee on Atomic Energy, Congress of the United States. Found in Herbert M. Parker, Publications and Other Contributions to Radiological and Health Physics, editors R. L. Kathren, R. W. Baalman, and W. J. Bair, Battelle Press, Columbus, Ohio. [SRDB Ref ID 27678]
- Patterson, C. M., 1949, "General Operating Tolerances," letter to Lauriston S. Taylor, National Bureau of Standards, HW-12710, Hanford Works, Richland, Washington, March 11. [SRDB Ref ID 7756]
- Rich, B. L., S. L. Hinnefeld, C. R. Lagerquist, W. G. Mansfield, L. H. Munson, and E. R. Wagner, 1988, *Health Physics Manual of Good Practices for Uranium Facilities,* EGG-2530, EG&G Idaho Inc., Idaho Falls, Idaho. [SRDB Ref ID 1781]
- Robinson, B., 1979, untitled letter to R. G. Swoboda discussing changes to <sup>90</sup>Sr analyses, Pacific Northwest Laboratory, Richland, Washington, May 10. [SRDB Ref ID 13395]
- Roesch, W. C., R. C. McCall, and H. E. Palmer, 1960, *Hanford Whole Body Counter 1959 Activities*, HW-67045, General Electric, Richland, Washington. [SRDB Ref ID 13426]
- Shapiro, J., 1990, Radiation Protection, third edition, Harvard University Press, Cambridge Massachusetts.
- Swanberg, F. Jr., 1962, memo to Internal Dosimetry Operations staff, "Whole Body Counter Examination Time, October 23. [SRDB Ref ID 7771, page 4.]
- Swanberg, F. Jr., 1963, *Hanford Mobile Whole Body Counter*, HW-80216, Hanford Atomic Products Operation, Richland, Washington. [SRDB Ref ID 7771 (sub-doc. p. 24 of 75)]
- Sula, M. J., E. H. Carbaugh, and D. E. Bihl, 1991, *Technical Basis for Internal Dosimetry at Hanford,* PNL-6866, Rev. 1, Pacific Northwest Laboratory, Richland, Washington. [SRDB Ref ID 270]

- Tilley, J.N., 1945, Letter to W.O Simon dated February 9, 1945, "Production of Polonium." [SRDB Ref. ID: 22757]
- Tomlinson, R.E., 1964, *Tentative U-233 and Thorium Nitrate Specifications*, RL-SEP-180, General Electric Hanford Atomic Products Operation, Richland, Washington. [SRDB Ref ID 20258]
- UST (United States Testing Company), 1974, "Promethium-147," internal procedure. [SRDB Ref ID 7787 (sub-doc, p. 187)]
- UST (United States Testing Company), 1987, "Plutonium Isotopic Determination All Matrices," internal procedure.
- Walser, R.S., 1978, "Purex Process Operation and Performance, 1970 Thoria Campaign," RHO-SA-37, presentation at the AIChE 84th National Meeting, Atlanta, Georgia, February 26 March 1, 1978. [SRDB Ref ID 27671]
- West, C.M., 1965, "Health Physics Considerations Associated with Thorium Processing," Y-KB-53, Y-12 Plant, Oak Ridge, Tennessee. [SRDB Ref ID 11596]
- Wilson, R. H., 1958, "The Hanford Uranium Bioassay Program," in *Symposium on Occupational Health Experience and Practices in the Uranium Industry*, held in New York City, October 15-17, 1958, HASL-58, Health and Safety Laboratory, New York, New York. [SRDB Ref ID 7886, p. 77]
- Wilson, R. H., 1960, "Review of the Thyroid Monitoring Program," letter to file, June 6. [SRDB Ref ID 7770]
- Wilson, R. H., 1987, *Historical Review of Personnel Dosimetry Development and its Use in Radiation Protection Programs at Hanford, 1944 to the 1980s, PNL-6125*, Pacific Northwest Laboratory, Richland, Washington. [SRDB Ref ID 262]

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### **GLOSSARY**

#### active

A term used in early writings at Hanford, circa 1940s and 1950s, to mean radioactive. Example, "production of acetylene from the active water, with subsequent measurement of the ionization caused by the tritium beta particle."

### aging

In the context of reactor fuel and mixtures of plutonium isotopes, the time since <sup>241</sup>Am was separated from the plutonium mixture.

## ashing

A chemistry term meaning driving off all moisture in a sample; ashing can be accomplished by heating in an oven or by dissolving the sample in a liquid (often nitric acid) then evaporating to dryness. The latter technique is called wet ashing.

### cooling

In the context of reactor fuel, the time since the fuel was removed from the reactor core.

## reliably detectable activity

Three standard deviations of the spectral continuum, plus has a peak discernable by the naked eye; used in *in vivo* counting circa 1980s.

### rep

Radiation Equivalent Physical. 83 ergs of energy from ionizing radiation per gram of tissue. This was redefined to be 93 ergs/g in the late 1940s or early 1950s. Later replaced by the rad.

#### simulated

In the context of urine sampling, collection of urine from about one-half hour before retiring to bed, through the sleep period, and for about one-half hour after rising for two consecutive nights to simulate a 24-hr sample or four consecutive nights to simulate a 48-hr sample.

## thoron

<sup>1220</sup>Rn.

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## A.1 CODES USED IN BIOASSAY AND INTERNAL DOSE RECORDS

(Adapted from Carbaugh 2003b)

These codes apply to information contained in the REX database and include present and historical uses. Different codes were implemented at different times according to the needs at the time; the early dates have few if any codes.

Table A-1. Sample type codes.

Code	Type of sample
В	Blood
F	Feces
S	Sputum
Т	Tissue
U	Urine

Table A-2. Bioassay reason codes.

Code	Name	Description
BL	Baseline	Measurement performed to establish reference level against which
		subsequent measurements will be compared. In general, could be for
		new employees, or for established employees, prior to commencing
		work with radioactive materials, beginning specific type of radiation
		zone work, or making offsite trip where potential intakes could occur.
PR	Periodic	Measurement performed at regularly scheduled interval.
EA	End of Assignment	Measurement performed following completion of specific work
		assignment, but not end of employment.
SP	Special	Measurement performed as part of specific investigation of potential
		internal dose. Could include response to off-normal work conditions or
		follow-up of abnormal periodic measurements.
CR	Contractor Request	Measurement requested by employer for reasons other than periodic,
		baseline, end of assignment, or special investigation.
RA	Reanalysis A	First reanalysis of sample by taking another aliquot and repeating same
		radiochemical or chemical analysis.
RB	Reanalysis B	Second reanalysis of sample by taking another aliquot and repeating
		same radiochemical or chemical analysis.
R1	Recount 1	First recount of original excreta sample or repeat in vivo examination.
R2	Recount 2	Second recount of original excreta sample or repeat in vivo
		examination.
QR	Quality and Research	Measurement performed as part of quality control, quality assurance, or
		research work.
TM or TS	Termination	Final bioassay at termination of employment.
12	Contract Work	In vivo measurement performed under contract to customers rather
		than for Hanford employees.
20	Source Count	In vivo source count made for system calibration or as function check,
		usually using known check source.
30	Background Count	In vivo system background measurement performed for system
		calibration or as functional check.

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Table A-3. Excreta sample kit codes<sup>a</sup>.

	it code <sup>b</sup>		
D/R	P/U	Media	Sample description
1	Р	Urine	Approximate 24-hr urine collection. Collected at home over 2-day period. Used for routine sampling and when larger volume sample is desired. Designated sample date is day after kit delivery to employee.
2	Q	Urine	Approximate 12-hr urine collection for termination sampling only.  Collected at home overnight. Designated sample date is day after date of kit delivery to employee.
3	R	Urine	Total 24-hr urine collection. Collected at home and at work (if necessary) to collect all urine voided during 24-hr period. Generally used for sampling immediately following occurrence or for work restriction sampling. Designated sample date is day after delivery or date on which sample collection began.
4	S	Urine	Single void (spot urine) collection. Collection in single bottle, used for initial indications of intake. Designated sample date is date of voiding.
5	T	Feces	Collection of single fecal voiding usually for investigation of potential intake. Sample date is day after kit delivery or date on which sample was actually voided.
6	U	Urine	Partial day or approximate 12-hr collection. Usually collected at home overnight. Used for collection following occurrence or when large-volume urine sample is necessary, such as for tritium or uranium determination. Designated sample date is date of delivery to employee.
7	V	Urine	Approximate 12-hr collection Sunday-Monday sample (Friday delivery only). Generally used for workers chronically exposed to soluble uranium. Designated sample date is Sunday in sampling period.
8	W	Urine prior to 1986. Feces starting in 1986.	Associated with urine sampling in 1950s through 1970s; used to mean undesignated or unknown. Starting in 1986: collection of single fecal voiding used for special program for plutonium oxide workers. Designated sample date for shift workers is Tuesday of long shift change, and for day workers is appropriate Sunday.
9	Х	Urine	Kit designed for collection of urine outside local service area.  Transportation handled by private carrier. Generally used for termination samples not collected locally.
A	Y	Urine	Simulated 48-hr urine collection. Collected at home over 4-day period. Used for IPUL sampling. Designated sample date is 2 days after kit delivery to employee.
В	Not applicable	Urine	12-hr urine collection for termination sampling only. Collected at home overnight. Kit delivered in normal manner, but brought to designated onsite location by worker for pickup by Contractor. Designated sample date is day after date of kit delivery to employee. Delivery Only, no home pickup required.

<sup>a. Prior to about 1983, kit codes were called collection codes.
b. D/R = Delivery and Retrieval; P/U = Pick-Up only (the latter series of codes were not used prior to about 1990, but</sup> should have no impact on dose reconstruction).

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Table A-4. In vivo body location codes.

Computer		
code	Body location	
ABD	Abdomen	
CHT	Chest result	
CH1	Chest result	
CH2	Chest result corrected by ultrasound measurement of chest wall thickness	
HED	Head	
HND	Hand	
KNE	Knee	
LG1	Lung result (chest result corrected for skeleton burden interference)	
LG2	Lung result (chest result corrected for skeleton and liver burden interference)	
LV1	Liver	
LV2	Liver result corrected for skeleton burden interference	
LV3	Liver result corrected for skeleton and lung burden interference	
LYM	Lymph nodes	
SKL	Skull (head) – old code no longer in use	
SK1	Total activity in skeleton based on head count	
SK2	Skeleton result based on something other than head count	
SPL	Special	
THX	Thorax	
THY	Thyroid	
TRT	Throat – old code no longer in use	
WBC	Whole body	
WND	Wound	

Table A-5. Excreta unit codes.

Computer code	Description of units
code	Description of units
1	dpm/sample
2	dpm/volume analyzed
3	μg/L until 07-01-82
	μg/sample after 07-01-82
4	μg/gram until 07-01-82
4	μg/sample after 07-01-82
5	μCi/sample
6	μCi/L
7	nCi
8	μCi

Table A-6. Excreta processing codes.<sup>a</sup>

Processing code	Description
R	Routine processing
Р	Priority processing
X	Expedite processing (added about 1985)
E	Emergency processing

Used in conjunction with contract with commercial laboratory starting in 1965; used to designate turnaround time and MDAs; that is, different processing codes had different MDAs.

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Table A-7. Excreta laboratory codes.

Code	Analytical laboratory
IT	IT Analytical Services, Richland
LA	Los Alamos National Laboratory
OR	Oak Ridge National Laboratory
PL	PNNL Analytical Chemistry Laboratory
QN	Quanterra
RE	REECo (Reynolds Electrical & Engineering Company,
	Nevada Test Site)
ST	Severn Trent Laboratories, Richland
TA	TMA/Norcal, Richmond, California
WH	Westinghouse Hanford Company, 222-S Laboratory

Table A-8. Excreta no-sample codes.

No-sample code	Description
CN	Kit not out. Sample kit not out at time of scheduled pickup.
CS	Cancelled sample/analysis.
CT	Sample lost due to bioassay analysis contract termination.
FA	Failed Analysis. Valid analytical result could not be obtained.
IS	Insufficient sample. Sample provided by worker but volume
	insufficient to meet contractual requirements.
LC	Lost container. Sample kit not retrieved.
ND	Not delivered. Sample scheduled but kit never delivered.
NS	No sample. Kit retrieved but no sample provided by worker.

Table A-9. In vivo invalid result codes.

	in vivo invalia result codes.
Code <sup>a</sup>	Reason for no results
С	External contamination other than radon detected on subject.
	Measurement invalid; no results obtained.
F	Failure of equipment or faulty setup of equipment. Measurement invalid;
	no results obtained.
I	Interference from localized activity in another part of subject's body.
	Measurement invalid; no results obtained.
L	Location of internal or external activity qualitatively determined by
	mapping, masking, or collimating. Could include one or more
	measurement counts. Measurements are qualitative for identifying
	location of activity and do not yield quantifiable estimates of activity.
M	Medically administered radioactivity interfered with measurement.
	Measurement invalid; no results obtained.
Р	Preliminary count, when followed by more quantitative record count.
	Used to indicate measurement taken, but not record count.
R	Radon interference from subject's clothing, hair, or skin. Measurement
	invalid; no results obtained.
S	Subject's actions interrupted completion of count. Measurement invalid;
	no results obtained.
Х	Measurement invalid; no results obtained. Other no-result codes do not
	apply. See comment field for brief description.

a. The comment field might contain a brief explanation in addition to the codes listed.

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Table A-10. INTERTRAC mode-of-intake codes.

Code	Mode of intake
ABS	Absorption
ING	Ingestion
INH	Inhalation
NON	None (no intake)
UNK	Unknown
WND	Wound

Table A-11. INTERTRAC evaluation reason codes.

Code	Reason for evaluation
Α	Annual chronic intake evaluation
С	Contractor-requested evaluation
Н	High routine bioassay evaluation
ı	Incident evaluation
N	New hire measurement or previous employment record
	indicated exposure prior to Hanford employment
R	Reevaluation

Table A-12. INTERTRAC source-of-intake codes.

Code	Source of intake		
DHE	Intake at DOE site while employed at Hanford		
HAN	Intake at Hanford		
NHE	Intake at non-DOE site while employed at Hanford		
NOC	Nonoccupational intake		
PTH	Intake occurred prior to Hanford employment		

Table A-13. INTERTRAC miscellaneous codes.

Code type	Code	Description
Intake confirmed	Y	Yes
	N	No
Nature of intake	Α	Acute
	С	Chronic
Recorded dose	Υ	Yes
	N	No
	0	Undetermined - old evaluation assessing body burden rather than dose, or evaluation in process
	Z	Recorded dose is 0 mrem
Source known	Y	Yes
	N	No
Type of evaluation	Р	Preliminary
	F	Final

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Table A-14. Special whole-body count resolution codes (RC) (used in 1983 only).

Code	Description
Α	Investigation in progress
В	Recent intake < or = to 1% Maximum Permissible Annual Dose
С	Previous deposition
D	Under investigation with additional examinations scheduled
E	Investigation completed, see radiation exposure records
F	Unresolved
G	Deposition from previous non-Hanford employment
Н	Exposure received offsite by Hanford employee
	Activity derived from medical diagnostic or therapeutic procedure

#### A.2 TOLERANCE DOSE AND TOLERANCE VALUES AT HANFORD

Various health physics textbooks and histories document the history of the tolerance dose. The description below was extracted from Jacob Shapiro's textbook (Shapiro 1990) and from *Radiation Protection Criteria and Standards: Their Basis and Use* (Parker 1960). Herbert M. Parker was the manager of the radiation protection activities at the start of Hanford and eventually became the manager of the Hanford Laboratories.

The tolerance dose was based on a rate of radiation exposure that produced no obvious harm. The basis for harm changed as understanding of health effects of radiation improved.

The first tolerance dose was established in 1934 by the International X-ray and Radium Protection Commission as 0.2 R/day. In 1936 the U.S. Advisory Committee on X-ray and Radium Protection reduced the tolerance dose to 0.1 R/day. This value was used at the Manhattan Project sites during World War II. For instance, Parker (1960) states that in 1945 the values accepted as a working basis for occupational exposure were:

- 100 mr/day for external X and gamma radiation
- 10–14 Ci/cc for radon in air of working rooms
- 0.1 µg of radium as the maximum allowable amount deposited in the body

In 1949, the NCRP recommended reduction of the permissible dose to 0.3 rem/week. In 1953, the NCRP developed maximum permissible amounts of certain radionuclides in the body and maximum permissible concentrations in air and water, published as National Bureau of Standards Handbook 52 (NBS 1953). At that point tolerance values were replaced with Maximum Permissible Body Burdens and maximum permissible concentrations.

Hanford's radiation protection standards were in accordance with these national and international standards. For instance, the following are examples of the "tolerance limits for prolonged exposure" at Hanford in 1945 (Cantril 1945):

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External gamma and X radiation	0.1 R/day
External beta radiation	0.1 rep <sup>a</sup> /day
Fast neutron radiation	0.02 rep/day
Slow neutron radiation	0.025 rep/day
Internal alpha radiation	0.01 rep/day
Radium deposition in body	0.1-µg total accumulation
Radio concentration in atmosphere <sup>b</sup>	1 × 10 <sup>-14</sup> Ci/cc
Radioactive I-131 in atmosphere	1.0 × 10 <sup>-13</sup> Ci/cc
Mixed fission products in drinking water	1.2 × 10 <sup>-9</sup> Ci/cc
Product <sup>c</sup> concentration in drinking water	10 <sup>-5</sup> μg/cc
Product concentration in atmosphere	5 × 10 <sup>-10</sup> μg/cc
Product deposition in body	0.5-µg total accumulation
Uranium dust in atmosphere	1.5 × 10 <sup>-4</sup> µg/cc

- a. rep (roentgen equivalent physical) was a unit of dose from particulate radiation invented by Herbert Parker. It was a precursor to the rad. It was defined as an absorbed dose of 83 erg per gram of tissue and was later changed to 93 erg/g.
- b. Assumed to refer to fission products [124].
- c. Product was a euphemism for plutonium.

By March 1949, the Hanford limits had changed to incorporate the 0.3 rem per week guidance from the NCRP. Examples are quoted below from Patterson (1949), but these same air concentrations limits were discussed as Hanford limits in a lecture series by Parker (1947). The Patterson letter refers to these values as permissible tolerances.

#### **External Radiation Limits**

Whole body – 0.3 rem per week or whatever the National Committee on Radiation Protection recommends

Hands only – 1.0 rem per week or whatever the National Committee on Radiation Protection recommends

#### **Internal Emitter Limits**

0.3 rem per week to the significant organ or whatever the National Committee on Radiation Protection recommends

## **Drinking Water**

Uranium – 100 μg/liter - believe this is high

Plutonium – 0.01 μg/liter – intend to revise to not more than 0.001 μg/liter

Mixed fission products  $-0.1 \, \mu c^*$ /liter - to be changed when new figure provided by K. Z. Morgen's subcommittee

#### **Air Contamination Limits**

Respiratory protection required Uranium - >0.05 μg/liter

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<sup>\*</sup> Assumed to mean microcuries.

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Plutonium - >2 x  $10^{-8}$  µg/liter to be revised perhaps to 2 x  $10^{-9}$  µg/liter Mixed fission products - > $10^{-6}$  µc/liter

Other air contamination limits

Tritium – gas 0.1  $\mu$ c/liter, vapor 0.01  $\mu$ c/liter Carbon – 0.02  $\mu$ c/liter Argon – 1.6  $\times$  10<sup>-3</sup>  $\mu$ c/liter lodine – 1.5  $\times$  10<sup>-6</sup>  $\mu$ c/liter Xenon – 0.01  $\mu$ c/liter

The tolerance values for air and drinking water were based on the dose to the significant organ and state-of-the-air understanding of the biokinetics of the element in the body. An assumption of continuous inhalation or drinking of water was usually made, although in the example below a tolerance value based on a single inhalation was also established. A sample calculation was found (letter to file dated December 17, 1945) that provides the calculation of the tolerance value for "potentially long-continued exposure" to  $^{131}I$  and a "one-shot tolerance value," the latter assuming a single 8-hr exposure (Parker 1945). For the chronic exposure, Parker assumed a thyroid radiation tolerance of 1 R/day, which resulted from an equilibrium thyroid burden of 1.95  $\mu$ Ci, which would result from a daily intake of 0.85  $\mu$ Ci. Parker assumed the volume of air breathed per 8-hr work day was 8 × 10 $^6$  cc. For the acute intake, Parker referenced a "Project Handbook" that condoned a dose of 100 R; however, he stated that this was too close to the dose given patients in the treatment of hyperthyroidism, so he arbitrarily reduced the dose by a factor of 10. He then calculated an air concentration for a single 8-hr exposure to be 1.2 × 10 $^{11}$  Ci/cc.