Metal contamination and bioremediation of agricultural soils for food safety and sustainability

Deyi Hou[1], *David O'Connor*[1], *Avanthi D. Igalavithana*[2], *Daniel S. Alessi*[3], *Jie Luo*[4], *Daniel C. W. Tsang*[5], *Donald L. Sparks*[6], *Yusuke Yamauchi*[7,8], *Jörg Rinklebe*[9] *and Yong Sik Ok*[2]✉

Abstract | Agricultural soil is a non-renewable natural resource that requires careful stewardship in order to achieve the United Nations' Sustainable Development Goals. However, industrial and agricultural activity is often detrimental to soil health and can distribute heavy metal(loid)s into the soil environment, with harmful effects on human and ecosystem health. In this Review, we examine processes that can lead to the contamination of agricultural land with heavy metal(loid)s, which range from mine tailings runoff entering local irrigation channels to the atmospheric deposition of incinerator and coal-fired power-plant emissions. We discuss the relationship between heavy metal(loid) biogeochemical transformations in the soil and their bioavailability. We then review two biological solutions for remediation of contaminated agricultural land, plant-based remediation and microbial bioremediation, which offer cost-effective and sustainable alternatives to traditional physical or chemical remediation technologies. Finally, we discuss how integrating these innovative technologies with profitable and sustainable land use could lead to green and sustainable remediation strategies, and conclude by identifying research challenges and future directions for the biological remediation of agricultural soils.

Bioavailability
The proportion of a soil constituent that interacts with living organisms.

Soil is a non-renewable resource, generated at a rate of a few centimetres per thousand years[1]. It plays a critical role in supporting ecosystems and human society through providing a habitat for the majority of Earth's species and serving as a medium for crop production[2,3]. However, anthropogenic activities are causing widespread soil contamination and degradation[4,5]. A 2018 study predicted the presence of 2.8 million sites in the EU potentially contaminated with soil pollution[6], and, in China, 19% of agricultural soils contain harmful pollutants at levels exceeding environmental quality standards[7].

The United Nations has set 17 Sustainable Development Goals to be reached by 2030, and eight of these goals rely on a healthy soil environment (FIG. 1). Soil and permafrost together hold the largest terrestrial pool of carbon, storing an estimated 4.1 trillion tonnes — nearly five times the estimated mass of atmospheric carbon[8]. Contaminated soil might not be able to fulfil its role in the carbon cycle, thus, aggravating climate change. The degradation of agricultural soil and the resultant loss of crop yield are particularly alarming, as they put the most vulnerable people on the planet at greater risk of poverty and malnutrition[9]. Moreover, soil pollutants can cause seriously impaired neurological development and life-threatening cancers[10,11] after entering the human body through ingestion of contaminated crops, inhalation of contaminated soil dust or inadvertent ingestion of contaminated soil.

Heavy metals and metalloids, henceforth, referred to as heavy metal(loid)s, are important agricultural soil pollutants due to their toxicity, ubiquity, non-biodegradability and bioavailability for crop uptake, and are a major threat to global food safety and food security[12]. Global agricultural production must double by 2050 to meet the projected demand of a growing population with improved living standards[13]; however, the industrialization of developing countries is causing widespread heavy metal(loid) pollution of agricultural land. The most commonly encountered heavy metal(loid)s in soil include cadmium, arsenic, copper, mercury, lead and chromium; a 2014 national soil survey in China showed that these respectively accounted for 43%, 17%, 13%, 10%, 9% and 7% of all soil quality exceedances[14]. Cadmium is the most widespread and bioavailable heavy metal(loid) in rice paddy soils, leading to concerns that the rice produced is cadmium contaminated[15]. Millions of hectares of agricultural land

✉e-mail: yongsikok@korea.ac.kr

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Key points

- Agricultural soil is a non-renewable natural resource that requires careful stewardship in order to achieve the United Nations' Sustainable Development Goals.
- Global agricultural soil pollution by heavy metal(loid)s represents one of the biggest challenges to sustainable development, particularly in developing countries.
- Bioremediation, including phytoremediation and microbially mediated bioremediation, is a promising nature-based solution for treating heavy metal(loid) contamination.
- It is imperative that the international community realizes the seriousness of the heavy metal(loid)s contamination in soils, takes actions to prevent further pollution and instigates the remediation of contaminated sites with environmentally friendly techniques.
- Policymakers should foster a bioremediation-enabling environment through policy instruments and increased field-based research funding.

are now being taken out of production due to cadmium pollution[15,16], and, despite the increasing stringency of environmental protection regulations, cadmium pollution continues to accumulate[16].

The growing issue of soil pollution has caught the attention of national and international bodies, both governmental and non-governmental[17]. In 2013, the 68th session of the United Nations General Assembly declared December 5th to be 'World Soil Day' and 2015 as the 'International Year of Soils'. In 2017, the United Nations Environment Assembly (UNEA) adopted a resolution that requested a number of bodies to report on global soil pollution[18]. These bodies, including the World Health Organization (WHO) and the Food and Agriculture Organization (FAO), are required to assess the extent of the problem, monitor future trends and identify associated risks and impacts by 2021 (REFS[18,19]). Individual countries are also taking action; China, for example, revealed an ambitious action plan in 2016 to clean up approximately 700,000 hectares of seriously contaminated agricultural land by 2020 and make 95% of the nation's contaminated land safe for use by 2030 (REF.[15]).

In this Review, we discuss the latest findings on the sources of agricultural soil pollution and the natural and anthropogenic processes that influence the distribution of soil heavy metal(loid)s, ranging from local surface runoff to atmospheric transport and deposition. We illustrate how soil heavy metal(loid)s undergo biogeochemical transformation and bioaccumulation in the food chain. Lastly, we review the mechanisms and applicability of plant-based and microorganism-based remediation strategies (phytoremediation and microbial bioremediation) in treating contaminated agricultural soils, and conclude by identifying the challenges and outlook of implementing bioremediation strategies on a large scale.

Occurrence of soil heavy metal(loid)s

Heavy metal(loid)s have been extracted from minerals and used by humans for thousands of years[20] and, currently, are used in a wide variety of industrial, domestic and agricultural applications; chromium and cadmium are frequently used in metal plating, for example. Despite increasing awareness of the harm caused by heavy metal(loid)s in soils, their essential role in modern industry means that their production and use continue to increase. Over the past 50 years, global production of chromium and lead has increased by 514% to 37.5 Mt per year and by 232% to 11.3 Mt per year, respectively[21]. Heavy metal(loid)s are even required for renewable technologies in some cases[22]; cadmium and lead, for example, are used in lead–acid and nickel–cadmium battery cells[20,23,24], lead is used in perovskite solar cells and nickel is used in electric-car batteries[25,26]. Because of their intensive manufacturing, widespread usage and tendency to accumulate via adsorption, absorption and precipitation, heavy metal(loid)s have become the most widely distributed type of contaminants in agricultural soils[27]. Their occurrence in agricultural soils is associated with a wide variety of sources, which are discussed below.

Sources of soil pollution

Anthropogenic sources of heavy metal(loid)s pollution are associated with agriculture, industry and mining. These sources include surface runoff from mine tailings[28,29], soil treatment with impure mineral phosphate fertilizer[30,31] or sewage sludge[32] and irrigation of farmland with polluted water[33,34]. Heavy metal(loid)s present in dusts and aerosols released during mining and smelting activities[35], fossil-fuel burning[36], vehicle use[37], cement manufacture[38] and electronic-waste processing[39] can also enter the soil through atmospheric deposition (FIG. 2).

Different types of contamination are often associated with different sources. Elevated levels of lead, mercury, copper and zinc are often associated with anthropogenic sources[40], and lead is especially associated with transportation activity because of the historical usage of lead in gasoline and the abrasive wear of lead-containing vehicle components[41]. Land treatment with impure mineral fertilizers and manures is associated with cadmium[42,43], copper[42] and zinc[42] contamination in agricultural soil. Irrigation with contaminated wastewater also causes heavy metal(loid)s accumulation[44]. For instance, wastewater land irrigation in a region of Beijing, China approximately tripled soil chromium concentrations over the past 30 years, and increased lead and cadmium levels by factors of 18 and 84, respectively[45].

Atmospheric deposition plays a major role in heavy metal(loid)s accumulation in agricultural soil[42,46]. In Europe, it was found that atmospheric deposition contributes more lead to soils than fertilizer application[43].

Author addresses

[1]School of Environment, Tsinghua University, Beijing, China.
[2]Korea Biochar Research Center, APRU Sustainable Waste Management Program and Division of Environmental Science and Ecological Engineering, Korea University, Seoul, Republic of Korea.
[3]Department of Earth and Atmospheric Sciences, University of Alberta, Edmonton, Alberta, Canada.
[4]College of Resources and Environment, Yangtze University, Wuhan, China.
[5]Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Kowloon, Hong Kong, China.
[6]Department of Plant and Soil Sciences and Delaware Environmental Institute, University of Delaware, Newark, DE, USA.
[7]Australian Institute for Bioengineering and Nanotechnology (AIBN), The University of Queensland, Brisbane, Queensland, Australia.
[8]International Center for Materials Nanoarchitectonics, National Institute for Materials Science (NIMS), Tsukuba, Japan.
[9]School of Architecture and Civil Engineering, Institute of Foundation Engineering, University of Wuppertal, Wuppertal, Germany.

Soil pollution and loss:

SDG1: no poverty
- Reduce crop yield and income, which jeopardizes health and cognitive potential

SDG2: zero hunger
- Lower food production per unit land
- Limit the available land for growing crops

SDG3: good health and well-being
- Reduce children's IQ due to lead exposure
- Cause kidney dysfunction, bone disease and cancers due to cadmium exposure
- Cause neurological damage due to mercury exposure

SDG6: clean water and sanitation
- Pollute groundwater
- Enter drinking-water sources via surface runoff

SDG15: life on land
- Threaten the health of plants and animals
- Reduce soil biodiversity

SDG13: climate action
- Reduce soil carbon storage potential
- Result in deforestation to obtain new farmland

SDG12: responsible consumption and production
- Make sustainable production in agriculture difficult

SDG11: sustainable cities and communities
- Result in derelict land, environmental displacement and iniquitous exposure

Fig. 1 | **The impact of soil pollution on SDGs.** Soil pollution negatively impacts sustainability, specifically, hindering progress on a number of the Sustainable Development Goals (SDGs) set out by the United Nations.

In England and Wales, atmospheric deposition is the main contributor of heavy metal(loid)s in most agricultural areas, accounting annually for 85% of the mercury, 78% of the lead, 60% of the nickel, 56% of the arsenic and 53% of the cadmium deposited in agricultural soils[46].

Heavy metal(loid)s in agricultural soils can also derive from geogenic sources[42,47,48]. Most existing studies suggest that soil parent material contributes to heavy metal(loid)s concentrations in agricultural soils. Certain heavy metal(loid)s tend to coexist in natural minerals; cadmium, for example, is often associated with zinc, lead or copper in sulfide forms[23]. Geogenic heavy metal(loid)s levels also correlate with soil properties, including clay content, carbonates and soil organic carbon[49]. The coexistence of chromium, cobalt and manganese in soil is indicative of soil heavy metal(loid)s being of lithogenic origin[40]. Some regional studies show that chromium contamination originating from soil parent materials can be associated with nickel[50,51], cadmium[52] and arsenic[53].

Heavy metal(loid)s distribution

Both geogenic and anthropogenic contaminants can accumulate over large spatial areas. Soil pollution in large areas of south-western China is mostly attributable to geogenic sources, and contamination in large areas of China's eastern developed coastal zone is mostly attributable to anthropogenic activity, for instance. However, differentiating the two remains a significant challenge[54]. Spatial distribution can also occur at a smaller scale, even within the same field. The spatial distribution of heavy metal(loid)s is dependent not only upon their sources but also natural factors that generate heterogeneity in soil properties, such as wet–dry cycles and anthropogenic processes such as soil tilling[40].

Variability in heavy metal(loid)s concentrations between fields in the same region can be attributed to chemical transformation[55,56], transportation[57,58], dilution[59] and accumulation[60]. Such processes tend to operate on larger scales than single agricultural plots, reflecting the heterogeneous mineral composition among fields and random distribution of exogenous soil particles with elevated heavy metal(loid)s contents[15,61–63]. Inter-plot variation can also depend on plot distance from pollution sources[64,65], with heavy metal(loid)s concentrations being highest in plots closest to polluted sources of irrigation water, such as contaminated wastewater conveyance channels or polluted natural watercourses[45]. Vehicular pollutants also cause elevated heavy metal(loid)s concentrations in agricultural fields adjacent to major roads[66]. Fields used for growing different crops can differ in heavy metal(loid)s constituents, owing to variations in agricultural cropping and irrigation practices; for example, vineyards in the Piedmont region of Italy have elevated copper and zinc levels due to the application of copper-containing and zinc-containing foliage spray to combat fungal disease[50]. Similarly, sewage-sludge application to individual agricultural plots in England and Wales, despite being conducted under regulatory constraints, was found to supply high levels of heavy metal(loid)s, including zinc, copper, nickel, lead, cadmium, chromium, arsenic and mercury, to the soil[46].

The spatial distribution of heavy metal(loid)s at the regional scale is driven by factors that differ from those controlling variability within and between plots. Such factors include geogenic differences[40], regional atmospheric deposition[67], land-use distribution[68] and the presence of major anthropogenic-emission sources. Heavy metal(loid)s occur naturally in the Earth's crust, and tectonic and weathering processes that result in stratigraphic and sedimentological features largely explain regional variability in topsoil heavy metal(loid)s levels[69]. Atmospheric deposition of parent materials, for example, by dust storms, coupled with anthropogenic atmospheric emissions can result in unique spatial distribution features along wind channels; a study in a major metropolitan area of northern China suggested that the south-eastern winds during the summer season were a key source of heavy metals for the region[67]. Indeed, agricultural fields close to large metropolitan areas are

Geogenic
Resulting from natural geological processes.

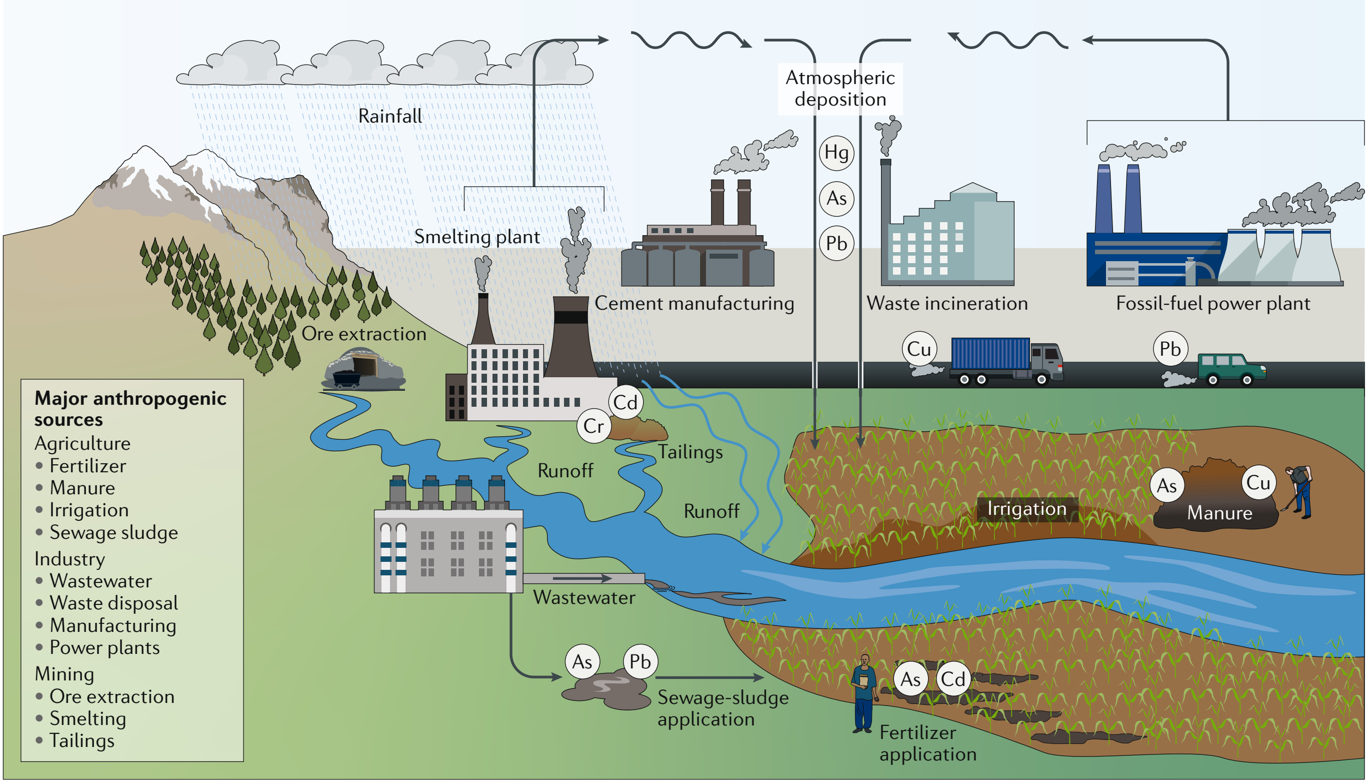

Fig. 2 | **Sources of heavy metal(loid)s pollution in agricultural soil.** Major anthropogenic sources can be classified into three categories: agricultural, industrial and mining. Heavy metal(loid)s can enter agricultural soil through atmospheric deposition, following release into the atmosphere from fossil-fuel burning, waste incineration or cement manufacture. Heavy-metal(loid)s-contaminated runoff from mining and industry can enter waterways and reach agricultural land. The use of manure or sewage contaminated with heavy metal(loid)s to fertilize crops can also contaminate agricultural land.

expected to have higher heavy metal(loid)s concentrations than those in remote areas, owing to more intensive anthropogenic emissions[42,66]. Moreover, major anthropogenic heavy-metal(loid)s-emission sources, such as mega-mining sites, large smelting facilities and large power plants and incinerators without adequate emission control, can result in elevated heavy metal(loid)s in agricultural soil on a regional scale[70,71].

Global mapping of heavy metal(loid)s distribution in agricultural soils is lacking, but airborne heavy metal(loid)s distributions provide an indication of their global distribution in soil. Studies conducted under the United Nations' Convention on Long-Range Transboundary Air Pollution show that atmospheric concentrations of lead, cadmium and mercury are the highest over China, followed by India, the Middle East and northern Africa (FIG. 3a–c). Densely populated areas of Europe and North and South America also have high atmospheric concentrations of these metal(loid)s[72].

Studies mapping soil heavy metal(loid)s concentrations have been conducted on both national and continental scales, though global mapping has not been undertaken. The first harmonized sampling programme of agricultural soils in the EU found that 137,000 km^2 of agricultural land requires further local assessment and remediation[27]. A meta-analysis of compiled regional data from south-western China indicates that high heavy metal(loid)s concentrations are present in agricultural fields in a region with high geogenic background concentrations and extensive mining activities[63,73]. In eastern China, industrial facilities that have operated for several decades have caused high heavy metal(loid)s concentrations in agricultural soils[74]. Models using stable mercury isotopic analyses and geospatial climate and vegetation data suggest that South America and East and Southeast Asia have relatively high mercury concentrations in soil[75] (FIG. 3d).

Heavy metal(loid)s bioavailability

Although spatial distributions of metal(loid)s provide useful data on their impact in agricultural soils, the toxicity of heavy metal(loid)s is contingent on their bioavailability, which is, itself, dependent on the oxidation state and specific chemical form of the metal(loid)s[76]. The bioavailability of a given metal(loid) can vary widely depending on the soil type. Only a small fraction of the heavy metal(loid)s in soils are freely available in soil pore water for plant uptake, and dissolved heavy metal(loid)s (usually present as free hydrated ions or complexed ligands) often reach a dynamic equilibrium with the bulk of heavy metal(loid)s existing in the solid phase of the soil[77]. The distribution equilibrium is affected by soil pH, moisture, organic-carbon content, redox conditions, carbonate content, sulfide content, clay minerals and metal-oxide content[23,78–80], factors that can be modified by anthropogenic pollution. For example, irrigation with wastewater can reduce soil pH and increase soil organic matter[45].

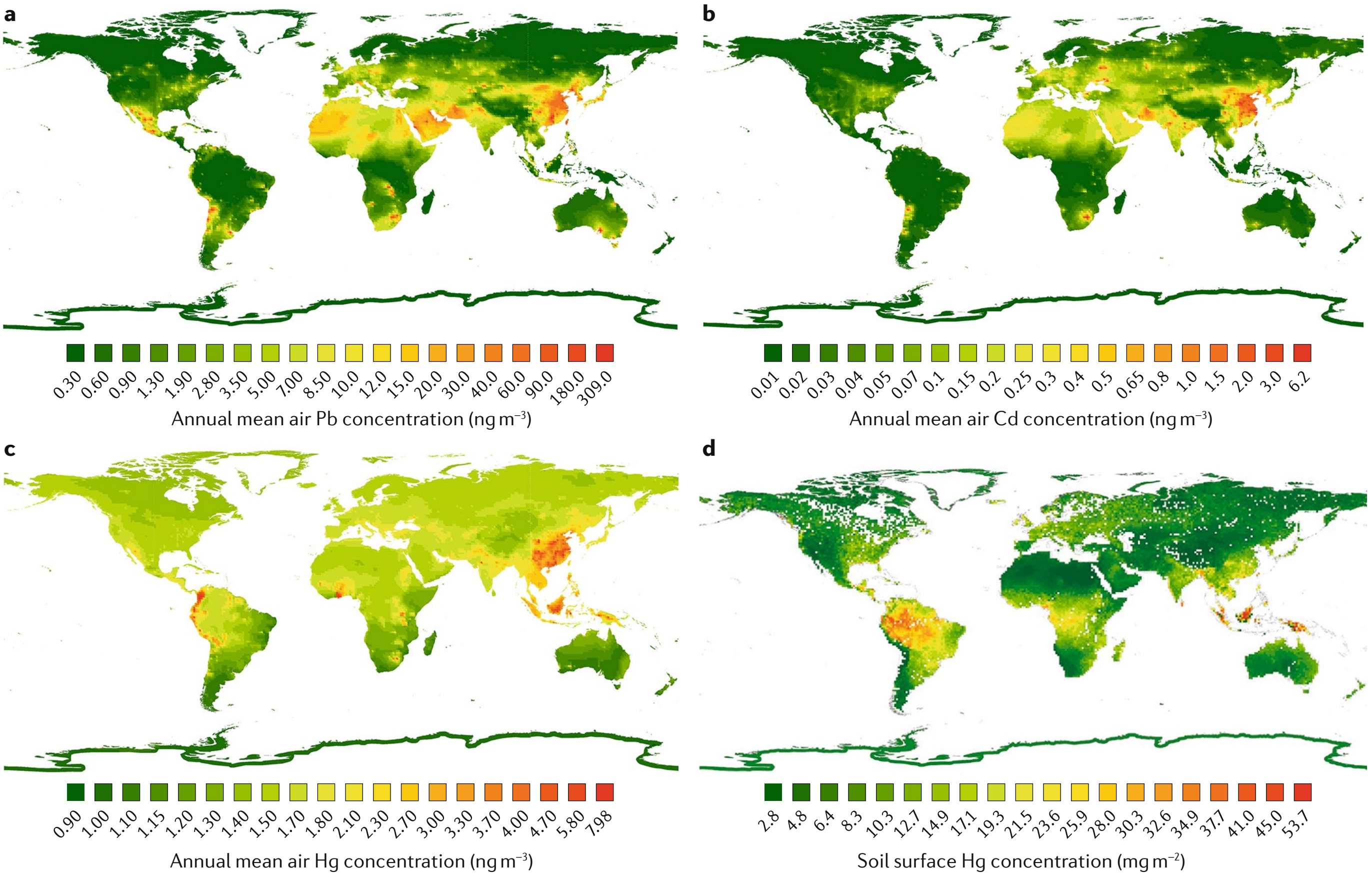

Fig. 3 | **Spatial distribution of heavy metal(loid)s.** The annual mean distribution of heavy metals in the air (panels **a**–**c**) and concentration in the soil (panel **d**). **a** | Lead concentration in air[72]. **b** | Cadmium concentration in air[72]. **c** | Mercury concentration in air[72]. **d** | Mercury concentration in the top 20 cm of soil based on model simulations[75]. Panels **a**–**c** adapted with permission from REF.[72], EMEP. Panel **d** adapted with permission from REF.[75], ACS.

The inherent bioavailability of different heavy metal(loid)s also varies substantially. For instance, the concentration of lead in soil tends to be much higher than that of other heavy metal(loid)s, nearly 40 times higher than cadmium and 100 times higher than mercury, due to the high natural background and high lead-emission levels[23,78]. However, lead has a low inherent bioavailability because it forms insoluble compounds such as pyromorphite and adsorbs strongly to soil minerals such as manganese oxides[81]. In contrast, cadmium has a much higher bioavailability than lead, arsenic and mercury because it exists mainly in exchangeable phases and has a comparatively low adsorption potential[82]. The oxidation state of the heavy metal(loid)s can also change their bioavailability to the plants. For example, the As(III) oxidation state of arsenic causes a decrease in plant growth, stomatal conductance and photosystem II efficiency of *Atriplex atacamensis*, whereas As(V) does not impact these processes[83].

Soil pH is among the most important environmental factors controlling the bioavailability of heavy metal(loid)s[84,85] and can drastically influence the solubility of soil metal(loid)s; for example, cadmium forms insoluble compounds under alkaline conditions (above pH 7.5) but is highly soluble in acidic pH while increasing bioavailability[82]. Local pH and, thus, heavy metal(loid)s bioavailability, are impacted by dynamic biological systems, as in the rhizosphere, where local pH is influenced by root activities and soil amendments[86–88].

Soil organic matter also influences the bioavailability of heavy metal(loid)s, but its effects are complicated. For instance, cadmium can adsorb onto carboxylic, phosphoryl, sulfhydryl and phenolic hydroxyl groups present in organic matter, reducing its bioavailability. However, dissolved humic substances can also form soluble complexes with cadmium, increasing its bioavailability[82]. Humic acids can form complexes with mercury that are highly stable and have low mobility[89,90], whereas compounds of mercury and fulvic acids are more labile and, thus, more bioavailable than mercury–humic-acid complexes[78,91]. Roots release low-molecular-weight organic compounds such as oxalic acid that act as metal chelators, which increase the bioavailability of certain heavy metal(loid)s[47]. Binding to non-organic matter can also influence bioavailability. Among different geochemical fractions of heavy metal(loid)s, exchangeable and easily mobilized metal(loid)s species, such as carbonate-bound metal(loid)s, are more bioavailable and toxic than those species that are less easily mobilized, such as Fe and Mn oxide bound, organic matter bound and residual fractions[92].

Rhizosphere
The very narrow region of soil in the vicinity of plant roots.

Humic acids
Soil organic substances that coagulate when strong base extracts are acidified.

Fulvic acids
Soil organic substances that remain soluble when strong base extracts are acidified.

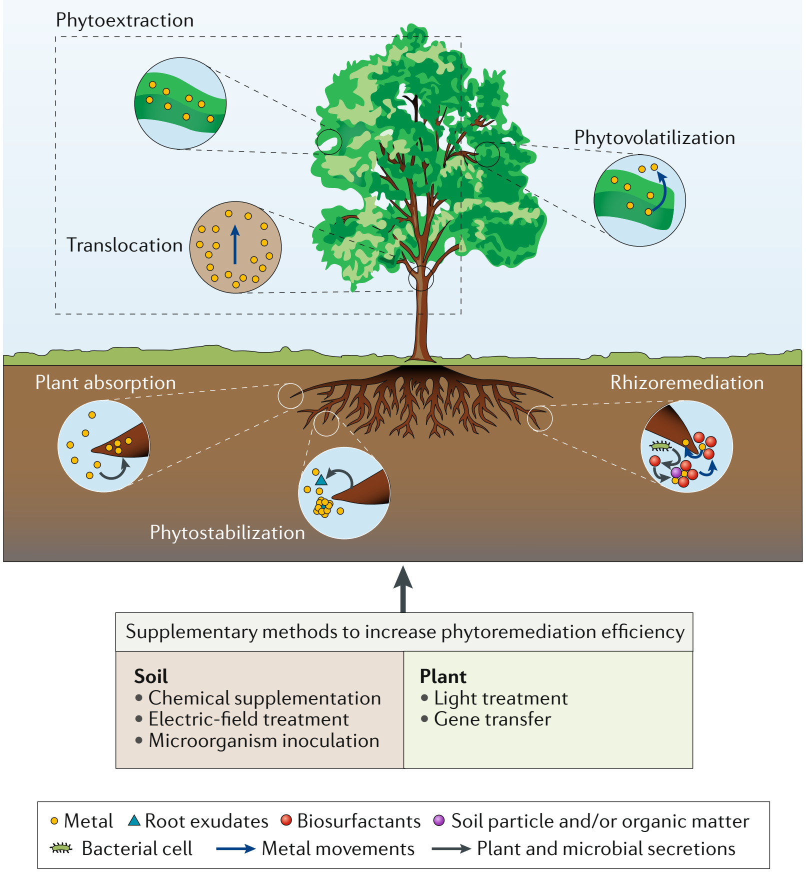

Fig. 4 | **Phytoremediation.** Natural methods of removing or detoxifying soil metal(loid)s, and supplementary methods to increase phytoremediation efficiency[257]. Roots can absorb heavy metal(loid)s from the soil or release factors to stabilize heavy metal(loid)s and prevent bioaccumulation (phytostabilization). Soil microbes can release factors to aid absorption of heavy metal(loid)s (rhizoremediation). Once taken up by roots, heavy metal(loid)s can translocate to the above-ground biomass of the plant, where they can be lost by transpiration (phytovolatilization) or be removed from the field by harvesting the plant. Adapted from REF. 257, CC BY 3.0.

Bioremediation

The interactions between plants, microbes and heavy metal(loid)s are exploited in bioremediation strategies, which use living organisms for soil decontamination[93,94]. These organisms can be plant or microbial species that are resistant to toxic heavy metal(loid)s and capable of thriving in highly contaminated agricultural soil. Some of these species can adsorb heavy metal(loid)s or release compounds that bind with them[95,96], thus, affecting contaminant bioavailability and toxicity. Other species can extract and remove heavy metal(loid)s from the soil environment[97].

Bioremediation tends to be more sustainable than traditional thermal or physico-chemical techniques such as soil washing, which can remove or destroy living organisms and soil organic matter, jeopardizing long-term soil health and diminishing post-remediation soil productivity[98]. Bioremediation also brings other sustainability benefits, including decreased cost, increased worker safety and smaller life cycle environmental footprints compared with traditional remediation methods[98], maximizing the economic, social and environmental benefits of soil remediation[99]. These benefits have prompted the remediation industry to move towards such nature-based solutions[100,101]. In this section, we discuss three bioremediation approaches: phytoremediation, microbial bioremediation and integrated methods.

Phytoremediation

Phytoremediation for soil decontamination employs indigenous[102] or imported species[103] of plants, including ones that are genetically modified[85,104]. This approach is adaptable to different plot sizes through planting and cultivating an appropriate number of selected phytoremediation plants, and considering intrinsic biogeochemical processes associated with plant growth, metal(loid)s speciation and changes in soil. Phytoremediation techniques include phytostabilization, in which root exudates reduce metal bioavailability in the rhizosphere, and phytovolatilization, which exploits plant evapotranspiration systems to transfer contaminants from the soil to the atmosphere[105] (FIG. 4). However, the most commonly used and well-studied phytoremediation technique is 'phytoextraction'[105]. In this approach, plant species take up heavy metal(loid)s from the soil through their roots; the heavy metals then accumulate in the plant's above-ground biomass, which is harvested. The biomass is typically incinerated, leaving behind a metal-concentrated bottom ash usually disposed of in landfills[106,107]. Particles that result from biomass combustion can pose a health risk and incineration requires appropriate filtration or scrubbing techniques. However, harvested biomass can be used as a feedstock for bioenergy production[108–110] or pyrolyzed to form biochar[111], with appropriate safety considerations.

Soil–plant–metal interactions. Heavy metal(loid)s enter plant tissue through various pathways (FIG. 4). For example, on a molecular level, plant-root systems are not completely selective and will take up heavy metal(loid)s from interstitial soil water, such as cadmium and arsenic, that have properties similar to nutrients required by the plant, such as zinc and calcium[112–114]. After entering root systems, heavy metal(loid)s are translocated from the roots to shoots and leaves, and then to fruits or seeds. Studies have also shown that heavy metal(loid)s will also enter plants from the atmosphere via foliar transfer[115], although unlike soil–root transfer, the molecular mechanisms involved in atmosphere–leaf transfer are not well understood.

Heavy metal(loid)s concentrations, the presence of chelating compounds, plant characteristics and soil properties all affect soil–plant–metal interactions and plant uptake rates[116–120] and, therefore, the effectiveness of phytoremediation. For instance, metal(loid) ions can form insoluble complexes, causing precipitation on soil-particle surfaces that inhibits their uptake by plants. Complexation of heavy metal(loid)s with larger molecules of soil organic matter can also hinder plant uptake. Plant Fe^{2+} uptake systems are upregulated in iron-deficient soil, allowing more Cd^{2+} to be taken up by root cells through Fe^{2+} channels[121]. Similarly, elevated concentrations of Zn^{2+} and Ca^{2+} can act as competitors to Cd^{2+} for plant uptake[112], mitigating cadmium toxicity towards the plant or enhancing essential mineral-element uptake[113]. Methods to augment biomass generation and heavy metal(loid)s uptake have been developed, including the application of artificial light[122,123], chemical supplementation[124,125], electrical-field treatment[126], microorganism inoculation[127] and gene transfer[128].

Plant selection. Plant selection is a critical step in phytoremediation, as species vary widely in their ability to uptake or immobilize different contaminants. Although indigenous species are preferred because they are adept at surviving in local environmental conditions, the use of introduced species might be necessary to speed up remediation[105]. Regardless of origin, hyperaccumulators[129,130] (plant species that extract large amounts of heavy metal(loid)s) are advantageous to use as they can speed up remediation of sites contaminated with high levels of heavy metal(loid)s. However, as they tend to take up a limited variety of heavy metal(loid)s[129], hyperaccumulators might not be suitable for soils contaminated with many different metal(loid)s species[131]. In these cases, fast-growing and high-biomass phytoremediation plants including willow, eucalyptus and poplar trees can be used to extract a wide range of heavy metal(loid)s from soil, although application of such plants may prevent agricultural production for years or possibly decades due to their relatively slow metal-extraction rates[132–134]. However, it is feasible for farmers to apply intercropping techniques, which enable the growth of phytoremediation plants alongside agricultural crops[135,136].

Hyperaccumulators are deemed most suitable for use on occupied agricultural sites to reduce heavy metal(loid)s contamination in developing countries, where there is great pressure for crop production[137]. To assist in this context, research has focused on the identification of hyperaccumulator species that are able to grow alongside crops[138–140], allowing remediation of the soil and the prevention of contaminants from endangering food crops simultaneously. Other research has focused on the use of crop plants that are able to take up contaminants but do not bioaccumulate them in edible parts; for example, crops may produce a grain that is suitable for animal consumption, while contaminants are enriched in the shoots or roots, which can then be removed as part of a phytoextraction strategy[141]. The main drawback to this approach is that heavy-metal(loid)s-extraction rates can be relatively low and use of these plants can prevent the growth of more valuable crops.

A wide variety of hyperaccumulator species specific for a range of metal(loid)s species have been identified, including the Cretan brake fern *Pteris cretica* for arsenic[142], *Sedum plumbizincicola* of the Crassulaceae family for cadmium and zinc[143,144], the grass species *Pogonatherum crinitum* for lead[145], *Celosia argentea* (the plumed cockscomb or silver cock's comb) for manganese[146] and *Pronephrium simplex* of the Thelypteridaceae family for rare-earth elements[147]. The increasing pool of hyperaccumulators, screened and selected from nature, offers new options to tackle difficult-to-treat sites. Meanwhile, researchers are developing transgenic plants to enhance the resistance, volatilization and accumulation of heavy metals in selected plants[148,149]. However, biosafety remains a concern due to potential transfer of conditional lethality and antibiotic-resistance markers to higher levels of the food chain[150,151].

Hyperaccumulators
Plant species that extract and concentrate certain heavy metal(loid)s within their biomass when grown in metal-contaminated soils.

Field successes and challenges. Many laboratory studies on phytoremediation have been conducted on spiked soils containing concentrations of heavy metal(loid)s hundreds or thousands of times higher than those found at contaminated sites[152–154]. The rationale for this method is to identify hyperaccumulator species more easily and better elucidate the molecular mechanisms at work by subjecting plants to high stress levels. However, care must be taken in extrapolating the data from such experiments and applying it to field operations.

In recent years, there has been an increasing number of field trials to verify the effectiveness of phytoremediation strategies at more environmentally relevant concentrations, as well as to determine field-related factors influencing their efficiency[155]. Phytoremediation field trials have been carried out globally, with the majority conducted in China[105] and preliminary large-scale (>500 m^2) phytoremediation field trials have been carried out for various heavy metal(loid)s contaminants in China, Switzerland, Germany, France and so on[105]. These studies provide evidence to evaluate the resilience, stability, suitability and effectiveness of phytoremediation plants under various environmental conditions. The initial large-scale phytoremediation field trials on heavy-metal(loid)s-contaminated soils were conducted in the early 1990s[156], when it was suggested that this approach could reduce metal concentrations to acceptable ranges on otherwise productive land. Since then, many greenhouse pot studies and small-scale outdoor field trials have been conducted, which have confirmed various hyperaccumulator species as being effective in reducing the soil concentration of a range of heavy metal(loid)s and helped identify practices to increase uptake levels. Plant density[157], initial plant size[119], cropping and harvesting strategies such as double cropping[158,159], transplantation and double harvesting[158,159] have been identified as crucial factors affecting success in these studies; however, these small-scale field studies, often conducted at the metre scale, suffer from inconsistency owing to their small size. Phytoremediation efficiency is also affected by soil heterogeneity[160]; hyperaccumulators are often identified and selected for highly contaminated soils, but may be less effective in soils with a lower degree of contamination. In addition, influencing parameters can vary during field treatments, meaning that long-term studies that report annualized treatment efficiencies are preferable to shorter trials[161,162].

More recently, larger, hectare-scale field trials have been performed. Variability in the results from these larger trials tends to be lower than that seen among smaller studies (TABLE 1). An agricultural trial across 11.1 hectares grew the hyperaccumulator species *P. vittata* and *S. alfredii* at a site previously contaminated with lead (351 ppm), cadmium (320 ppb) and arsenic (37 ppm) in the Guangxi Zhuang Autonomous Region in

Table 1 | **Results of large-scale phytoremediation field studies in agricultural soil polluted by heavy metal(loid)s**

Plant species	Plot size (m^2)	Soil texture	Soil pH	Initial soil OM (g kg^{-1})	Initial soil HM (mg kg^{-1})	BCF[a]		TF[b]	Metal(loid)s removal[c]	Key findings	Ref.
Morus alba	600	–	6.9	–	Cd (3.2)	1st year	<0.09 (s)	<0.3 (s)	3–7 g ha^{-1} year^{-1}	Cd and Pb mostly accumulate in root tissue, but not in fruits, indicating the trees could be used as a crop substitute	251
						2nd year	<0.08 (s)	<0.3 (s)	2–8 g ha^{-1} year^{-1}		
					Pb (181.2)	1st year	<0.02 (s)	<0.6 (s)	40–85 g ha^{-1} year^{-1}		
						2nd year	<0.008 (s)	<0.2 (s)	10–42 g ha^{-1} year^{-1}		
Zea mays	675	Silt loam	5.8	53	Pb (5,844.2)	0.06 (r)		–	7,181 g ha^{-1} year^{-1}	Each hectare can produce ~25 tonnes of corn grain for animal feed; biomass can generate bioenergy fuel equivalent to 1,545 GJ	141
						0.01 (s)		0.25 (s)			
						0.04 (l)		0.69 (l)			
Solanum nigrum	1,500	Sandy loam	6.2	138	Cd (1.91)	5.2 (ap)		–	<233 g ha^{-1}	The plants accumulated Cd in their biomass, enhanced by double cropping and sequential harvesting	158
Averrhoa carambola	1,500	Loam	6.1	43	Cd (1.6)	–		–	213 g ha^{-1}	High-density *A. carambola* removed 5.3% of the total Cd within one season; this decreased Cd bioavailability and uptake (63–69%) by vegetables grown afterwards	252
Salix sp.	1,710	Sandy loam	4.0	30	Cd (2.8)	3.61 (ap)		0.60 (ap)	95 g ha^{-1}	Repeated harvesting of the woody plants prior to leaf fall ensured effective soil decontamination	253
					Pb (283)	0.02 (ap)		0.38 (ap)	55 g ha^{-1}		
					Zn (295)	1.16 (ap)		0.29 (ap)	3,320 g ha^{-1}		
Salix sp.	2,100	Sand	6.6	–	Cd (6.5)	4.3 (s)		–	88 g ha^{-1} year^{-1}	Certain *Salix* species produced up to 12.5 tonnes of dry biomass per hectare per year; Cd and Zn removal increased by 40% with leaf harvest	254
						9.2 (l)					
					Zn (377)	1.8 (s)			3,497 g ha^{-1} year^{-1}		
						10.8 (l)					
Zea mays and *Pteris vittata*	400	–	6.4	–	As (93.6)	5.51 (l)		8.1 (l)	113 g ha^{-1}	Phytoaccumulators grown with maize, limiting As accumulation in maize grains; planting crops in different angular directions improved soil nutrient availability and As uptake	255
Zea mays	4,050	Sand	6.0	50	Cd (67)	0.01 (s)		–	6.4–10.4 g ha^{-1}	Produced biomass for generating 33,000–46,000 kWh of renewable energy per hectare per year	256
					Pb (184)	0.02 (s)			28–46 g ha^{-1}		
					Zn (355)	0.41(s)			1,447–2,826 g ha^{-1}		
Salix sp.	10,000	–	5.6	19	Cd (5.7)	9.82 (l)		–	82–113 g ha^{-1} year^{-1}	Several decades of phytoremediation with *Salix* required to reduce the Cd content of the soil from 5 to 2 mg kg^{-1}, but could be used for bioenergy feedstock	164
Pteris vittata and *Sedum alfredii*	111,000	–	–	–	Cd (0.32)	–		–	85.8% (re)	Phytoremediation decreased soil HM concentrations below national standards at a cost of US$75,375.20 ha^{-1} or US$37.70 m^{-3} of soil, lower than traditional remediation technologies	163
					Pb (350.5)				30.4% (re)		
					As (36.66)				55.3% (re)		

–, data not available; (ap), above-ground part; As, arsenic; BCF, bioaccumulation factor; Cd, cadmium; HM, heavy metal(loid); (l), leaf; OM, organic matter; Pb, lead; (r), root; (re), removal efficiency; (s), stem; TF, translocation factor; Zn, zinc. [a]The BCF represents the ratio of pollutant concentration in the organism to the soil. [b]TF is the ratio of HMs in the shoots and roots of a plant. It represents the ability of a plant to translocate the metal(loid)s from roots to shoots and/or leaves. Only trials with plot sizes larger than 500 m^2 are shown. Heavy-metal concentrations represent the mean total concentration for the whole plant, unless stated otherwise. [c]Removal represents grams of HMs removed per hectare, unless stated otherwise.

Siderophores
Chelating compounds secreted by microorganisms that bind with iron and other metals, increasing their bioavailability.

south-western China[163]. After two years, soluble concentrations of lead, cadmium and arsenic were reduced by 30.4%, 85.8% and 55.3%, respectively. Other large field trials have generally demonstrated that phytoremediation is a promising remedial approach, which can be far more cost-effective than traditional alternatives[163].

Recent studies have aimed to evaluate and enhance the sustainability of phytoremediation. The assessment of a short-rotation willow coppice phytoremediation trial at a heavy metal(loid)s-contaminated agricultural site in Belgium showed this approach to be the most sustainable alternative among various remediation options, due to its capacity to capture atmospheric carbon in plant biomass, while simultaneously treating soil contamination[164]. The use of organic amendments derived from biological waste (such as compost[165], sewage sludge[166] and manure[167]) and industrial waste (such as fly ash and red mud) to enhance phytoremediation have been explored, but some debate remains as to whether these amendments contain harmful levels of contaminants themselves[168–170] and if they actually improve phytoremediation performance[105].

Overall, field trials have been somewhat inconsistent in effectiveness, even among plants of the same species grown in the same plot[171,172]. There are several plausible reasons for this inconsistency. First, phytoremediation efficiency is highly influenced by environmental conditions such as contamination level, soil clay content, weather patterns, soil moisture, organic matter content, pH and salinity[173,174]. Although these parameters are generally homogenous and set at favourable levels in indoor pot experiments, field environments can be highly heterogeneous. Extrapolation of laboratory results might have caused overly optimistic predictions of uptake levels of heavy metal(loid)s in the field[175]. Second, the success of phytoremediation depends on the location of field sampling points, but field heterogeneity causes large differences from one sampling point to the next, often making the interpretation of results challenging.

Microbial bioremediation

Microorganisms exist at high concentrations in agricultural soils[176–178] and possess genes enabling their survival in contaminated soil environments. Many microorganisms are genetically resistant to heavy metal(loid)s[179] and some can survive even under extreme heavy metal(loid)s stress. Native microbes can facilitate the reduction of soil pollution levels or microbes (sometimes, ones that have been genetically engineered) can be introduced to polluted sites to reduce soil metal(loid)s concentrations in a process known as microbial biomediation[180]. Microbe–heavy-metal(loid)s interactions have been studied intensively[181] and a diverse range of microbial species and mechanisms that transform metal(loid)s to chemical species of lower solubility for immobilization, or species of higher solubility for removal[181,182], have now been identified[180,183,184] (FIG. 5). Here, we discuss soil–microorganism–metal interactions and two microbial bioremediation approaches: monitored natural attenuation and engineered microbial bioremediation.

Soil–microorganism–metal interactions. Biogeochemical processes facilitated by microbial activities form the basis of microbial bioremediation[185,186]. A crucial mediator of remediation[187] is the bacterial secretion of siderophores, which primarily transport iron from low-iron soils to cells through specific receptor and transport systems[188]. Fortuitously, siderophores also bind to heavy metal(loid)s[187,189–191]; for example, the bacteria *Alcaligenes eutrophus* secretes siderophores that bind with cadmium, zinc and lead[192]. Bacteria then protect themselves from siderophore-bound heavy metals by producing outer-membrane proteins that facilitate the formation of bioprecipitates, which have low environmental risk

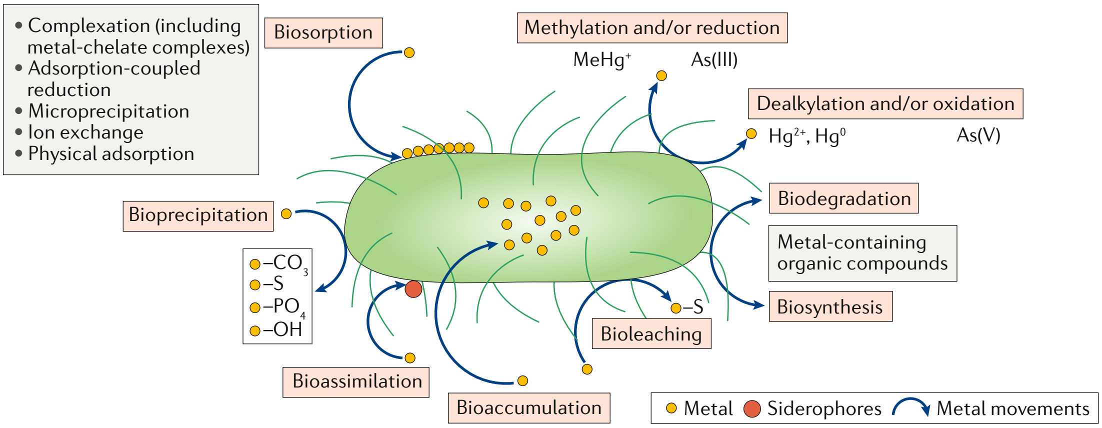

Fig. 5 | **Microbial bioremediation.** Processes by which bacteria can mediate the removal or detoxification of heavy metal(loid)s from agricultural soil. Bacteria can interact with heavy metal(loid)s directly, accumulating them on the cell surface (biosorption). They can also reduce or oxidize metal(loid) species and synthesize or degrade metal-containing organic compounds via catalytic reactions (biosynthesis or biodegradation). Sulfur-oxidizing bacteria can release acids and dissolve metal-containing compounds for leaching of metals (bioleaching). Sulfate-reducing bacteria can precipitate metals by formation of low-mobility sulfides (bioprecipitation). Bacteria can also accumulate metals in the intracellular space by using proteins in their cellular processes (bioaccumulation). Bacteria assimilate metals via iron-assimilation pathways using siderophores (bioassimilation). CO_3, carbonate CO_3^{2-}; OH, hydroxyl OH^-; PO_4, phosphate PO_4^{3-}; S, sulfide S^{2-}. Adapted with permission from REF.[258], Elsevier.

Lime
Calcium-rich alkaline-soil amendments, including marl, chalk, limestone or hydrated lime.

due to low bioavailability[192]. As siderophore production is boosted when iron levels are low, applying lime to contaminated soils can enhance bioremediation efficiency by reducing iron availability to the microbes[193].

Bioleaching and bioprecipitation are mechanisms of microbial bioremediation that rely on the presence of sulfur-oxidizing bacteria (SOB) and sulfate-reducing bacteria, respectively, and play a crucial role in determining the relative abundance of the common oxidation states of sulfur in nature[194]. Bioleaching involves the release of metal ions through the mineral dissolution, for example, by sulfuric acid produced by SOB. Dissolution of metals by sulfuric acid subsequently increases sulfur bioavailability to sulfate-reducing bacteria, which facilitates the bioprecipitation of metals as low-mobility sulfides, effectively removing them from the reactive-metal pool. Laboratory experiments on a multi-metal-contaminated soil by inoculating an SOB showed that metal bioleaching levels were as high as 74% for cobalt, 69% for copper, 69% for manganese and 68% for nickel after six months, the majority of which (80–98%) bioprecipitated as stable sulfide species[195]. Subsequent studies exploited sulfur-cycle bacteria to reduce risks posed by zinc, copper, chromium, lead and nickel in the soil. In this case, the study found that introduced microbial species were able to outperform indigenous species due to their higher sulfur-oxidizing activity[196]. Bioleaching of heavy metal(loid)s from exchangeable, carbonate-bound, Fe-oxide-bound and/or Mn-oxide-bound, organic-matter-bound and residual fractions were observed after only one month because of SOB activity[196]. Several bacterial species associated with the natural sulfur cycle, such as *Acidithiobacillus* spp., *Acetobacter* spp., *Arthrobacter* spp. and *Pseudomonas* spp., can be exploited for bioleaching of heavy metal(loid)s in soils and some fungi species, including *Penicillium* spp., *Aspergillus* spp. and *Fusarium* spp.[197–200]. The activity of *Penicillium chrysogenum*, for example, can mobilize cadmium, copper, lead and zinc in contaminated soils, leading to enhanced bioleaching[201,202].

Biological reduction provides another important route for microbially assisted soil remediation because the toxicity of heavy metal(loid)s depends on their oxidation state. For example, hexavalent chromium (Cr(VI)) is toxic and carcinogenic, whereas trivalent chromium (Cr(III)) is considered non-hazardous, which enables the remediation of Cr(VI)-contaminated soils by reduction. Bacterial strains resistant to and able to reduce elevated Cr(VI) concentrations include *Pseudomonas fluorescens*, *P. aeruginosa* and *Enterobacter cloacae*[203]. This biological reduction process can be achieved in inundated soils (such as in rice paddies), where oxygen levels are low, or in artificially induced reductive environments, for example, those in which an electron donor is added to induce microbial growth[204,205].

Monitored natural attenuation. The risk posed by heavy metal(loid)s in soil environments can naturally attenuate over time without specific remedial treatment[206]. This phenomenon has been observed at abandoned historic mining sites and adjacent agricultural fields throughout the world[207]. Natural attenuation processes comprise biological, physical and chemical mechanisms[208–210], but the activities of indigenous microbes often drive attenuation; these activities include metal(loid)s sequestration, ion efflux (which can lead to metal precipitation as carbonates near and around cells)[211,212] and extracellular chelation. Indigenous microbes can also mediate biogeochemical reactions that convert mobile heavy metal(loid)s into stable compounds of low bioavailability[208] through adsorption of metal(loid)s to organic matter[213], the formation of carbonates and sulfides (facilitated by *Kocuria flava*, *Sporosarcina pasteurii* and *Terrabacter tumescens*)[214,215], binding to iron and manganese oxides[216], reduction of metal(loid)s to aid the formation of stable compounds (by *Escherichia coli*, *Staphylococcus aureus* and *Staphylococcus xylosus*, for example)[208,217] and the oxidation and hydrolysis of aluminium, iron and manganese species (FIG. 5).

Natural attenuation often takes years or decades to reduce risk levels, although it remains a viable option for remediation when coupled with an appropriate and robust monitoring plan[218]. In some cases, bioremediation based on monitored natural attenuation may be the only practicable option to lower risk, given the difficulties and high costs inherent in treating some agricultural sites, particularly in developing countries[208]. At these sites, agricultural soil-management approaches, such as no-till farming and the use of cover crops, can influence microbial respiration and plant growth[219,220], thus, influencing natural-attenuation rates. For instance, no-till farming increases microbial biomass, soil carbon content and the activity of microbial enzyme activities (such as dehydrogenases, cellulases, xylanases, β-glucosidases, phenol oxidases and peroxidases) in agricultural soil[221], which can accelerate the formation of stable fractions of heavy metal(loid)s.

Engineered microbial bioremediation. Two types of engineered microbial bioremediation exist: biostimulation and bioaugmentation. Biostimulation involves providing indigenous soil microbes with additional nutrients, electron donors or electron acceptors in order to increase their capacity for immobilizing or degrading contaminants in the soil. This approach has been used in remediating heavy metal(loid)s[222], gasoline additives[223], broad-range hydrocarbons[224] and radionuclides[225]. Although indigenous microbes are often excellent candidates for bioremediation because they are acclimated to site conditions[226], laboratory-grown microbial strains can be added to soil, a process known as bioaugmentation[227]. Current commercial bioaugmentation applications use microorganisms collected from environmental samples and enriched in laboratories. There are many successful biofertilizers produced from plant-growth-promoting microorganisms and applied safely in the field[228,229], but, often, microorganisms cultivated under controlled conditions do not survive once placed in competition with indigenous microorganisms in field conditions[230]. Further research is needed to improve the performance of these cultivated microorganisms under field conditions for heavy metal(loid)s immobilization.

Genetic-engineering techniques can be used to improve microbial mechanisms for heavy metal(loid)s resistance[231]. For example, the introduction of a gene encoding phytochelatin synthase from *Schizosaccharomyces pombe*, *SpPCS*, into *E. coli* can enhance the cadmium uptake of *E. coli* by 7.5-fold[232]. Phytochelatins, the protein products of phytochelatin synthase activity, bind strongly to toxic elements such as cadmium, arsenic, lead and mercury[232–234], and render them non-toxic. Insertion of a gene encoding arsenite S-adenosyl methionine methyltransferase (*arsM*) into *Sphingomonas desiccabilis* and *Bacillus idriensis* similarly enables a tenfold increase in the production of methylated arsenic gas[235], allowing arsenic to volatize from the soil. Transferring genes from heavy-metal(loid)s-resistant microbes to other microbial species suitable for microbial bioremediation has potential to increase bioremediation effectiveness, subject to regulatory approval and oversight[212]. However, biosafety issues, including the possibility of horizontal gene transfer, must be taken into account before introducing these organisms into the environment[230].

Integrated methods and phytomanagement

Microbially mediated processes can enhance the efficiency of phytoremediation[236] by transforming heavy metal(loid)s, rendering metabolic nutrients and minerals more bioavailable to aid plant growth, stimulating systems that regulate plant heavy metal(loid)s stress responses or aiding the production of plant hormones that increase plant growth[237] (FIG. 4). The bacterial species *Pseudomonas aeruginosa, Pseudomonas fluorescens* and *Ralstonia metallidurans* produce siderophores that increase contaminant bioavailability to roots, leading to enhanced phytoextraction efficiency[238]. For example, augmentation of soil with these strains can increase chromium and lead uptake by plants by as much as 5.4-fold[238]. Moreover, microorganisms such as species of *Bacillus*, *Achromobacter*, *Stenotrophomonas*, *Brevundimonas*, *Ochrobactrum*, *Pseudomonas*, *Microbacterium*, *Comamonas* and *Sinorhizobium* can lower the toxicity and increase bioavailability of arsenic to plants by increasing arsenic mobilization[239]. A bacterial consortium of *Bacillus methylotrophicus*, *Bacillus aryabhattai* and *Bacillus licheniformis* applied to phytoremediation sites promotes plant growth through enhancing nitrogen fixation and phosphate solubilization, and producing siderophores and other molecules that affect plant hormonal processes. This consortium, when applied to *Spartina maritima*, effectively improved root growth by approximately 60% and bioaccumulation of cadmium, arsenic, copper, lead and zinc by between 17% and 65%[240].

The most significant drawback to bioremediation is the time required to complete treatment, which is sometimes overcome through its coupling with other remediation technologies to shorten treatment length. For example, the production of H^+ and OH^- ions during electrokinetic treatment of soil can produce potential gradients that cause unwanted bands of high residual metal concentration[241]; these issues are mitigated by phytoremediation techniques, as plant roots can extract H^+ and OH^- and residual heavy metal(loid)s[242]. Moreover, electrical fields induced by electrokinetic treatment can transport pollutants from deep in the soil up to the rhizosphere, enhancing phytoremediation effectiveness[126]. *Solanum tuberosum* showed higher zinc and copper accumulation in plant root under supplement of alternating current compared with the control in a laboratory study[243].

The integration of remediation technologies provides a scenario where ecosystem services such as nutrient cycling, carbon sequestration and water storage are restored[244]. Moreover, plants grown in contaminated agricultural fields undergoing bioremediation can be sold as bioenergy products or other profitable products[245]. Moreover, in comparison with traditional remediation strategies, phytomanagement focuses on both risk mitigation and commercial viability by using plants to control contamination while producing marketable biomass, and has been suggested as a viable strategy that can be carried out in large-scale applications[246]. Phytomanagement is considered as either a low cost or a profitable strategy for producing valuable plant biomass such as bioenergy or timber crops, or it can be used to prevent decreased food production on contaminated lands[246,247].

Summary and future perspectives

The accumulation of heavy metal(loid)s in agricultural soils is an obstacle to achieving global food safety and security. Bioremediation is a promising nature-based solution for treating heavy metal(loid)s contamination; however, several issues must be addressed before it can be more broadly implemented.

First, it will be beneficial to accelerate global soil mapping and establish regional models that can adequately predict contaminant distributions and identify pollution sources[248]. Second, the measured effectiveness of bioremediation in the field has been somewhat inconsistent, attributed to heterogeneity in field conditions and artefacts caused by evaluating treatments on a spot-by-spot basis, rather than employing field-wide assessment. Importantly, variability tends to decrease with increasing plot size[249], showing the importance of large-scale field trials. Third, field stations are needed to provide valuable insights into the mechanisms that render heavy-metal(loid)s-contaminated sites resistant to treatment. We suggest there is a need for improved monitoring instrumentation to measure trends in microbial dynamics, metal speciation and fractions, and soil environmental conditions (pH, temperature, redox potential and soil gases), as all these factors can mediate bioremediation effectiveness. Fourth, further research is required in order to decrease clean-up time and expand the applicability of bioremediation techniques to include more sites. Seeking out new natural species for this purpose and developing new genetic technologies that can modify and design the functionality of plant species and microbial strains could play a leading role in future development.

Global agricultural soil pollution by heavy metal(loid)s represents one of the biggest challenges for sustainable development, and developing countries are particularly vulnerable to this threat to food, health and livelihoods. By the 5th session of the United Nations Environment Assembly in 2021, institutions including the WHO and the FAO will elaborate on guidelines for the prevention and minimization of soil contamination, specifically including the use of nature-based solutions[250]. In this context, it is imperative that the international community realizes the seriousness of the threat, takes actions to prevent further pollution and instigates the remediation of contaminated sites with environmentally friendly techniques. Policymakers should foster a bioremediation-enabling environment through policy instruments and increased field-based research funding.

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Author contributions

Y.S.O., D.H., D.O.C., A.D.I., J.L. and D.C.W.T. researched data for the article. Y.S.O., D.H. and D.C.W.T. made a substantial contribution to the discussion of content. Y.S.O., D.H., D.O.C., A.D.I., D.S.A. and J.L. contributed to the writing of the review. Y.S.O., D.H., D.O.C., A.D.I., D.S.A., D.C.W.T., D.L.S., Y.Y. and J.R. reviewed and edited the manuscript before submission.

Competing interests

The authors declare no competing interests.

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