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

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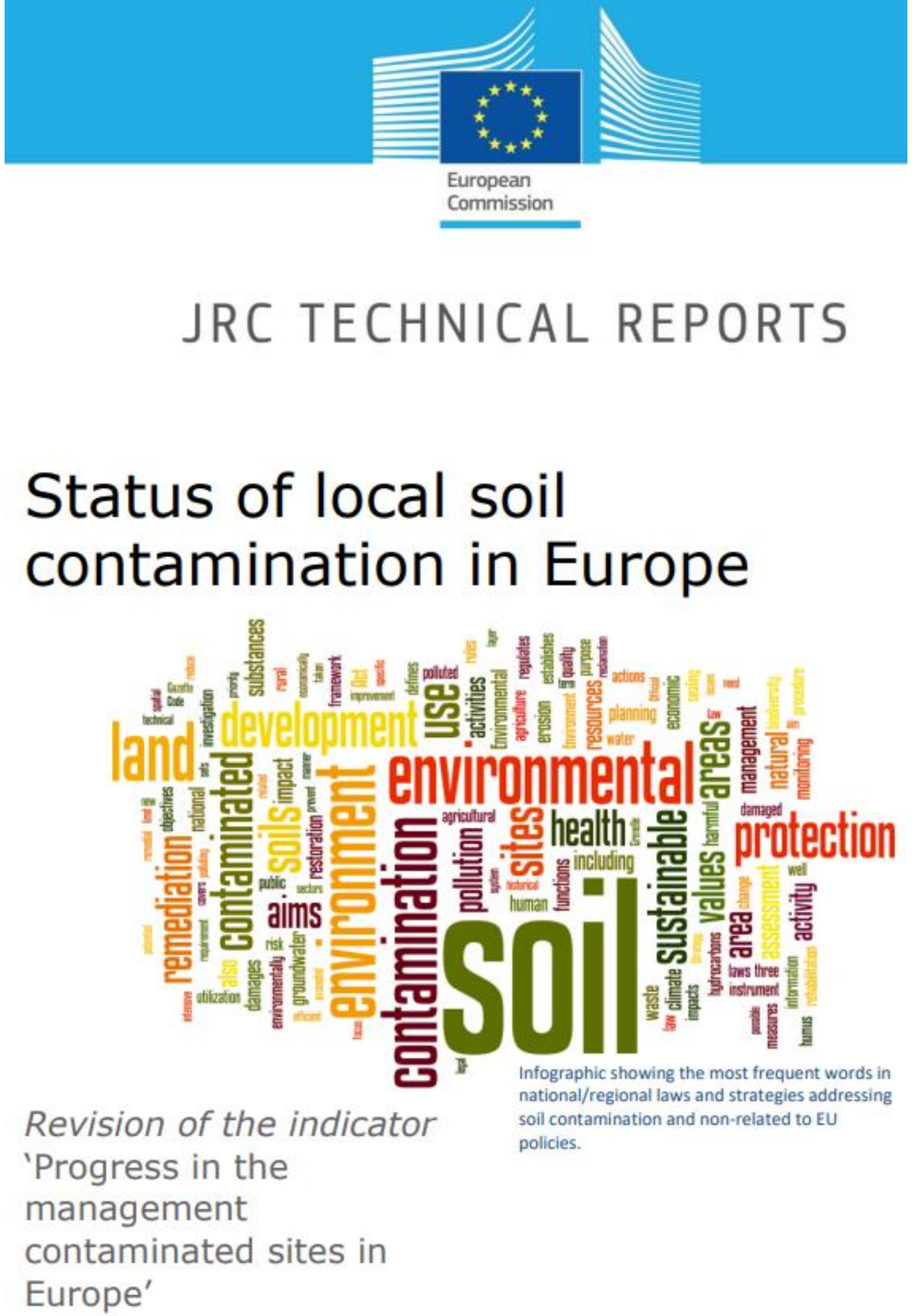
- Soil is a **non-renewable resource**, generated at a rate of a few centimetres per thousand years.

- It plays a critical role in supporting ecosystems and human society.

- However, anthropogenic activities are causing widespread soil contamination and degradation.



Soil pollution and loss:

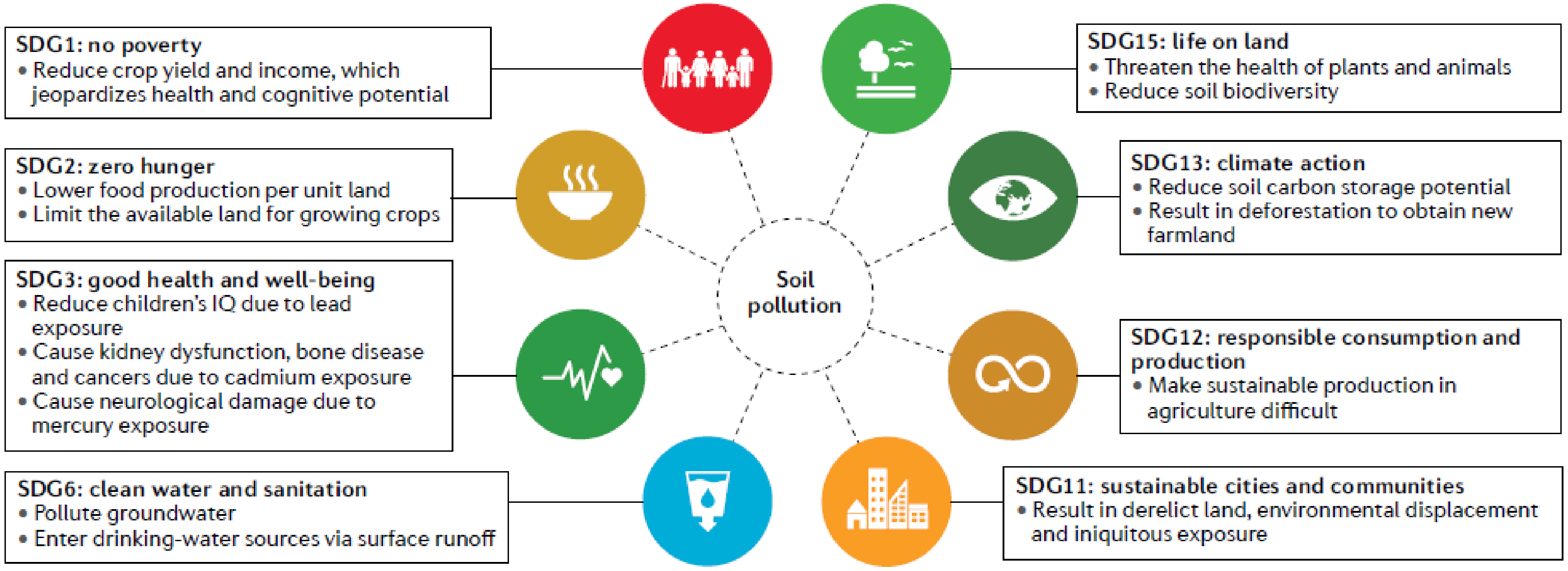


Fig. 1 | **The impact of soil pollution on SDGs.** Soil pollution negatively impacts sustainability, specifically, hindering progress on a number of the Sustainable Development Goals (SDGs) set out by the United Nations.

Ok YS* et al. (2022) *Nature Reviews Earth and Environment* 1:366

O'Connor D, Deyi H., **Ok YS** and Lanphear BP (2021) *Nature Sustainability* 3:77

The effects of iniquitous lead exposure on health

Disadvantaged communities are vulnerable to the impacts of lead exposure risking further worsening of their living standards, an outcome likely to weaken global efforts towards the Sustainable Development Goals. We urge policy makers to adopt protection systems aimed at safeguarding the most threatened populations.

David O'Connor, Deyi Hou, Yong Sik Ok and Bruce P. Lanphear

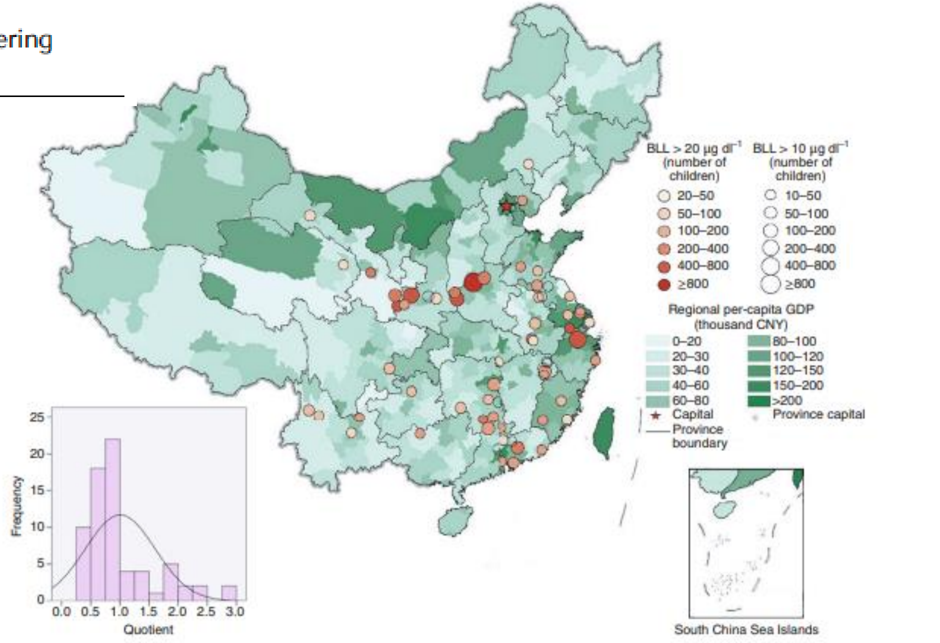


Fig. 1 | **Distribution of 75 lead exposure incidents reported in the Chinese news media since 2000.** These incidents involve >10,000 children with BLLs exceeding the regulatory threshold (10 μg dl^{-1}), and >3,000 children with acute lead poisoning (BLL > 20 μg dl^{-1}). The inset chart shows that where these incidents occurred, the quotient of the regional per-capita GDP and provincial per-capita GDP is typically less than 1. Based on 10,000 bootstrap samples, the median quotient value is 0.85 (95% confidence interval = 0.72 to 0.95). CNY, renminbi.

- The growing issue of soil pollution has caught the attention of national and international bodies, both governmental and non-governmental.

- In 2017, the UNEA adopted a resolution that requested a number of bodies to report on **global soil pollution**. These bodies, including WHO and FAO, are required to assess the extent of the problem, monitor future trends and identify associated risks and impacts by 2021.

Soil pollution – speed up global mapping

Deyi Hou & Yong Sik Ok

Too few countries are investing in national surveys of soil pollution. A global map is urgently needed, not least to prevent international trading of contaminated produce and the migration of persistent organic pollutants across borders. We urge all member states at next month's fourth session of the UN Environment Assembly (UNEA) to speed up their assessments.

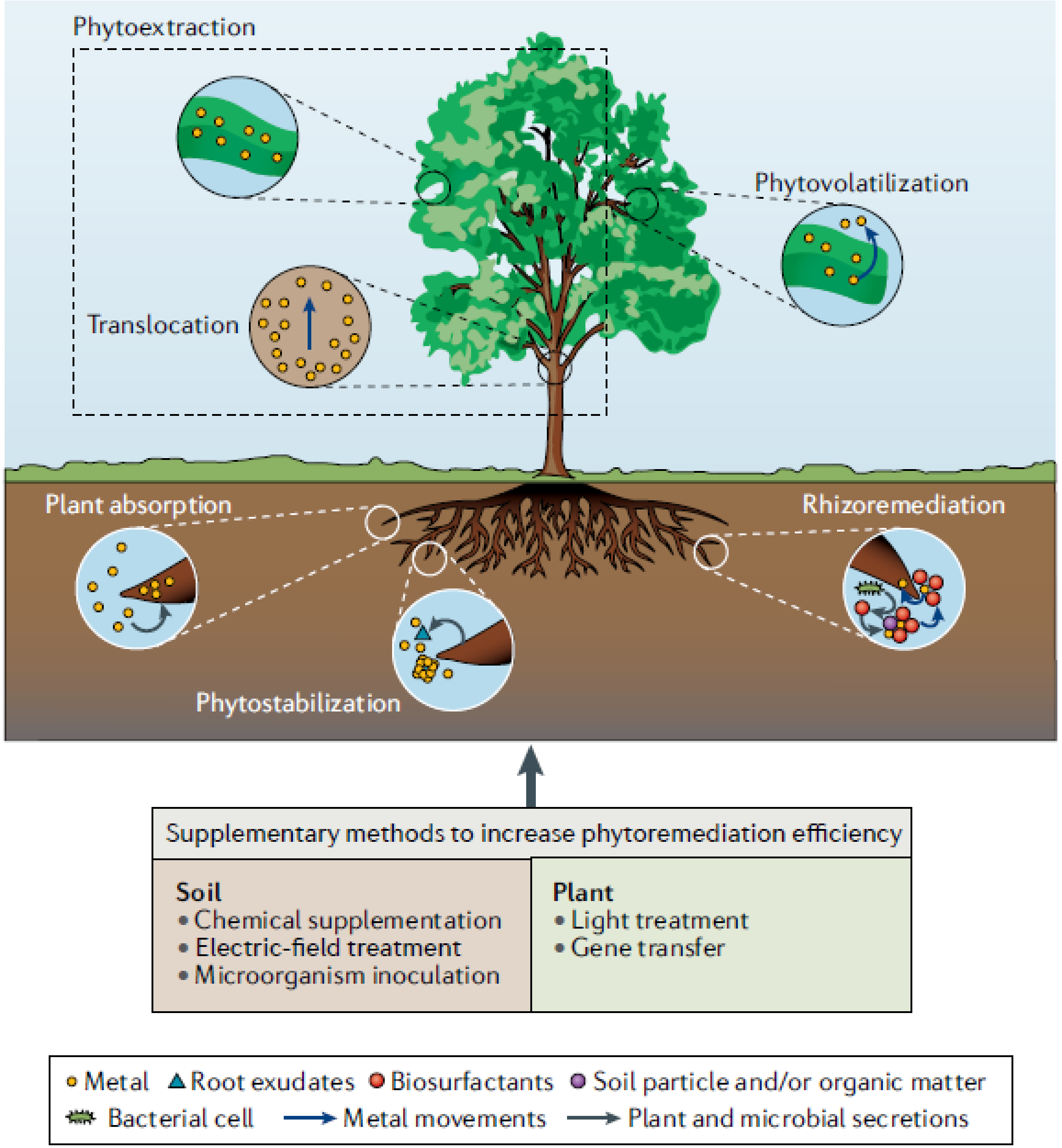
A global map of soil pollution will also guide policymakers on protecting soils; inform chemical and waste management (see Y. Geng *et al. Nature* **565**, 153–155; 2019); prevent further pollution by identifying sources and controlling polluter behaviour; and reduce risks to public health and the environment.

The World Health Organization and the United Nations Food and Agriculture Organization are among those required by the UNEA since December 2017 to report on the extent of global soil pollution, monitor future trends and identify associated risks and impacts. The results will be presented at the UNEA's fifth session in 2021. Many hurdles must be overcome before a global assessment can be made. Collaboration between developing and developed nations in allocating technical and financial resources is a priority.

Nature **566**, 455 (2019)

doi: https://doi.org/10.1038/d41586-019-00669-x

Contents

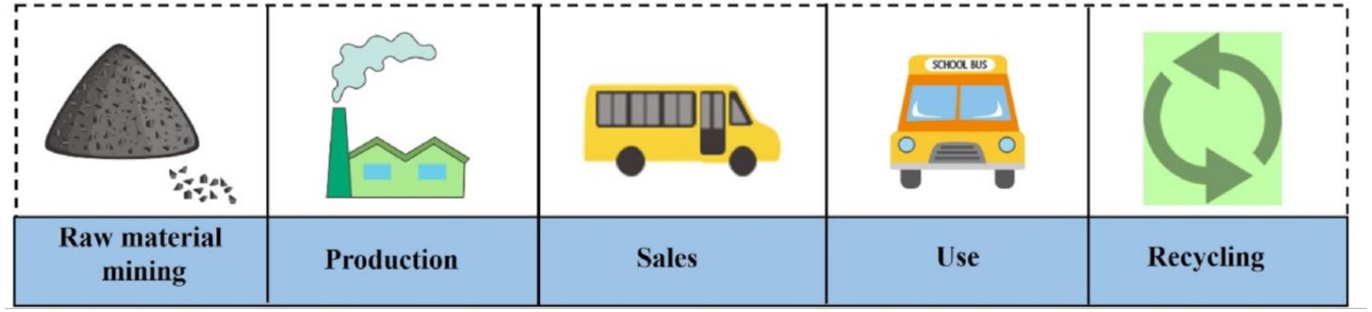


Ok YS* et al. (2022) *Nature Reviews Earth and Environment* 1:366

Occurrence of soil heavy metal(loid)s

- 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.
- Heavy metal(loid)s are even required for renewable technologies in some cases; cadmium and lead, for example, are used in lead–acid and nickel–cadmium battery cells, lead is used in perovskite solar cells and nickel is used in electric-car batteries.

Wang M*, Liu K, Dutta S, Alessi DS, Rinklebe J, **Ok YS** and Tsang CW (2022) *Renewable and Sustainable Energy Reviews* 163:112515



Sources of soil pollution

- Anthropogenic sources of heavy metal(loid)s pollution are associated with agriculture, industry and mining.

- Heavy metal(loid)s present in dusts and aerosols released during mining and smelting activities, fossil-fuel burning, vehicle use, cement manufacture and electronic-waste processing can also enter the soil through atmospheric deposition.

Environmental Science & Technology
Article
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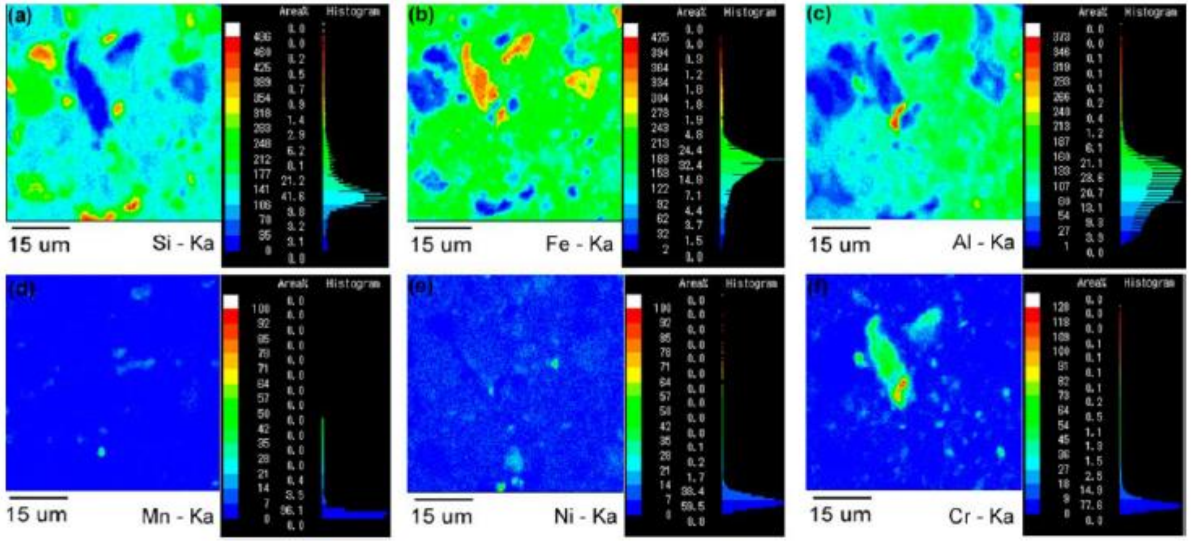
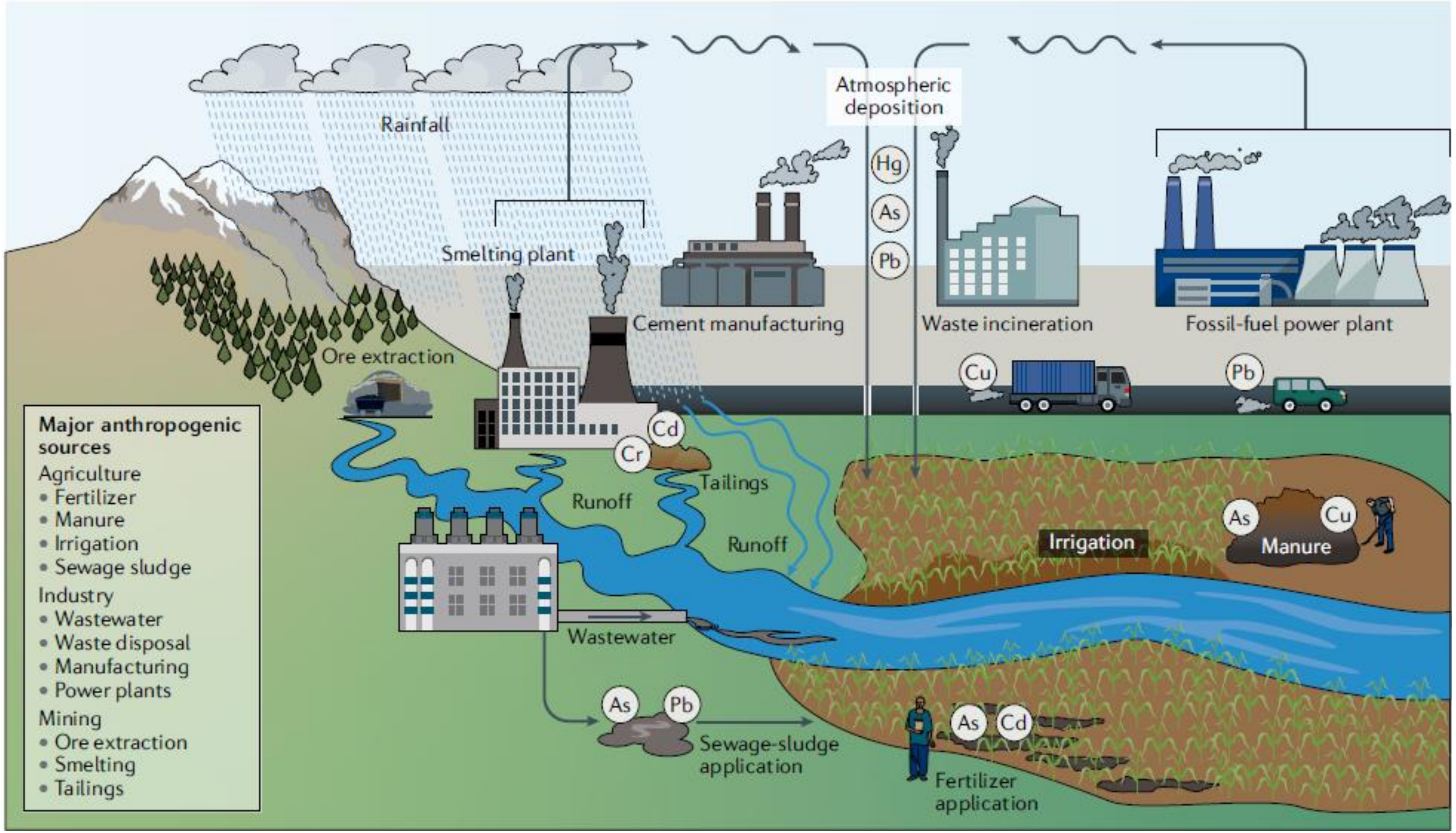
Cr(VI) Formation Related to Cr(III)-Muscovite and Birnessite Interactions in Ultramafic Environments

Anushka Upamali Rajapaksha,[†,‡] Meththika Vithanage,[*,†] Yong Sik Ok,[‡] and Christopher Oze[§]



EPMA analysis of serpentine soil in Sri Lanka. Signal amplitudes are given in the scale.

Sources of heavy metal(loid)s pollution in agricultural soil

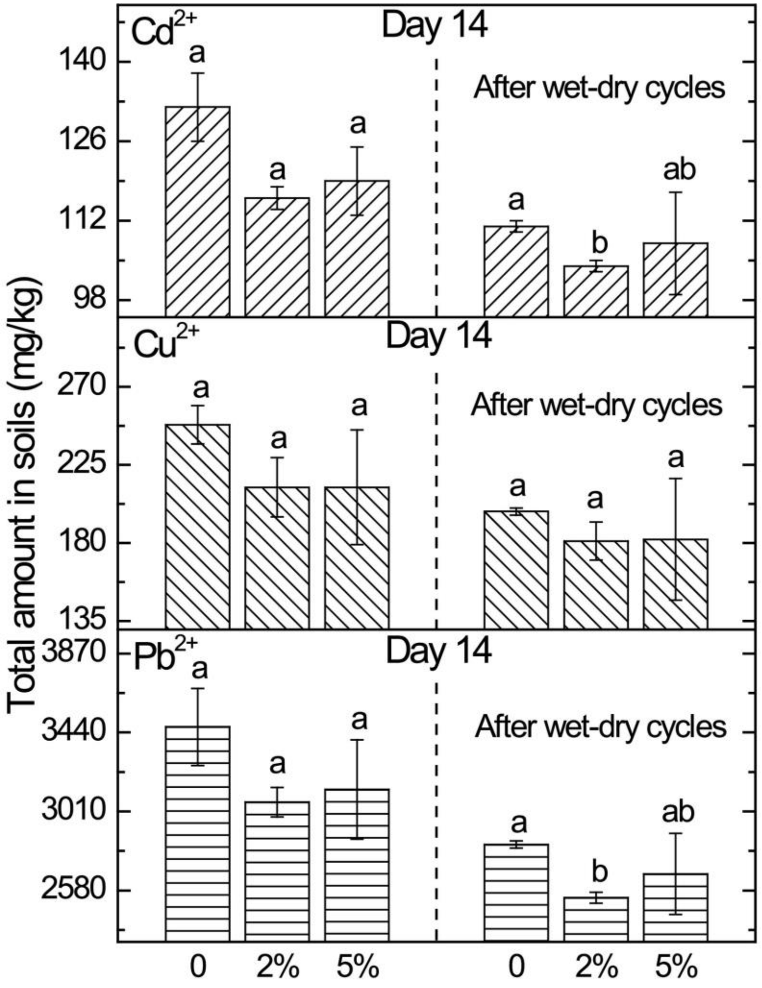


Ok YS* et al. (2022) *Nature Reviews Earth and Environment* 1:366

Heavy metal(loid)s distribution

- Both geogenic and anthropogenic contaminants can accumulate over large spatial areas.

- 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.



Shen Z, Hou D, Zhao B, Xu W, **Ok YS** et al. (2018) *Science of the Total Environment* 619-620:185-193

Heavy metal(loid)s bioavailability

- 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 often reach a dynamic equilibrium with the bulk of heavy metal(loid)s existing in the solid phase of the soil.

- The distribution equilibrium is affected by **soil pH**, moisture, **organic-carbon content**, **redox conditions**, carbonate content, **sulfide content**, **clay minerals** and **metal-oxide content**, factors that can be modified by anthropogenic pollution.

Prediction of Soil Heavy Metal Immobilization by Biochar Using Machine Learning

Kumuduni N. Palansooriya, Jie Li, Pavani D. Dissanayake, Manu Suvarna, Lanyu Li, Xiangzhou Yuan, Binoy Sarkar, Daniel C. W. Tsang, Jörg Rinklebe, Xiaonan Wang,* and Yong Sik Ok*

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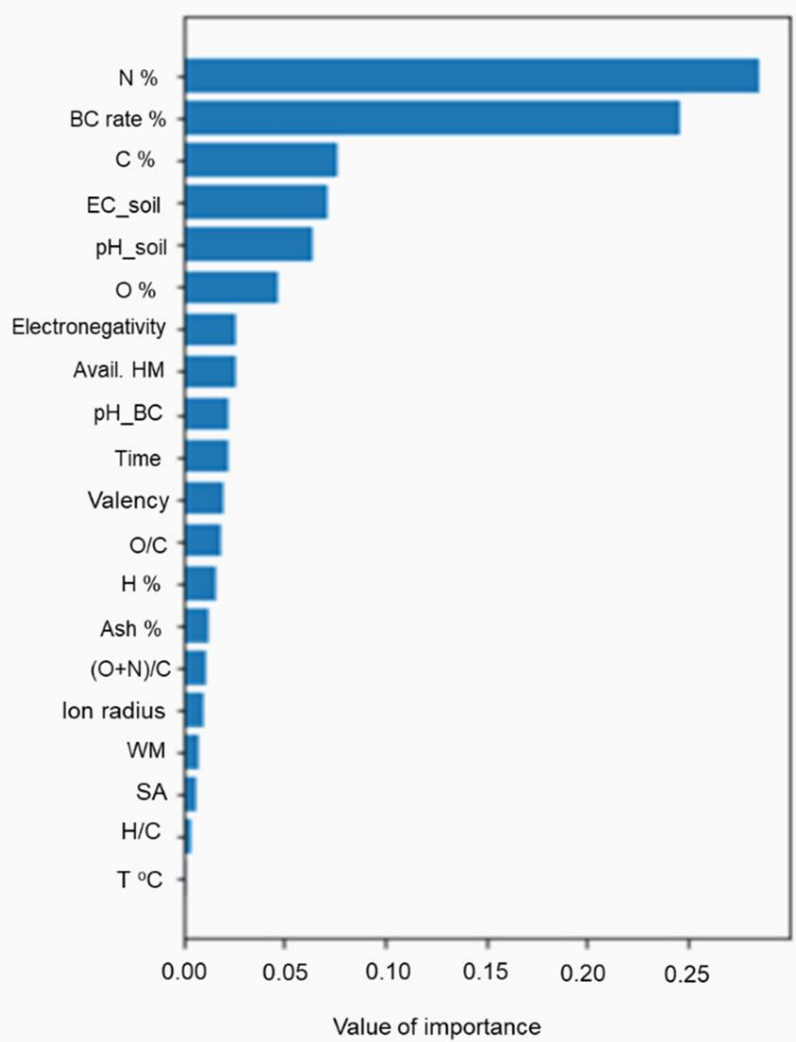
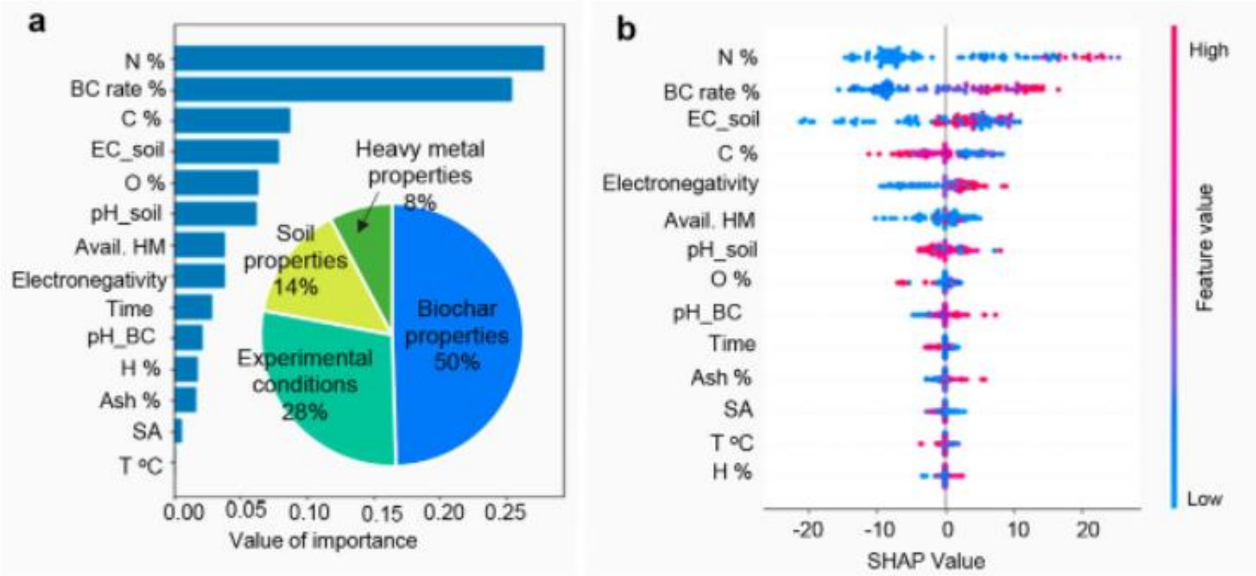


Heavy metal(loid)s bioavailability

- The inherent bioavailability of different heavy metal(loid)s also varies substantially.
- The oxidation state of the heavy metal(loid)s can also change their bioavailability to the plants.
- Soil pH
- Soil organic matter (SOM)

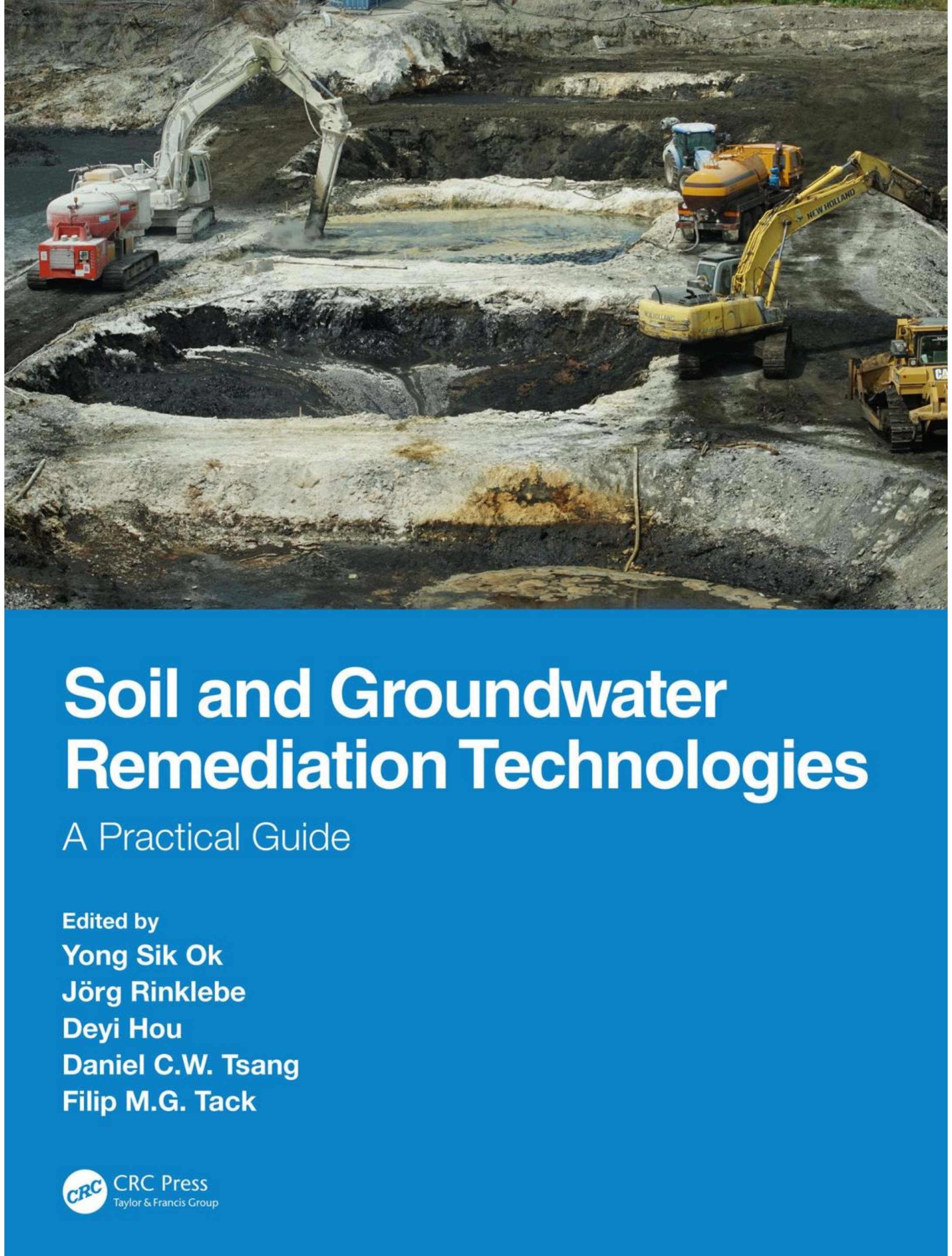


Ok YS* et al. (2022) *Environmental Science and Technology* 56(7):4187-4198

Bioremediation

- The interactions between plants, microbes and heavy metal(loid)s are exploited in bioremediation strategies, which use living organisms for soil decontamination.

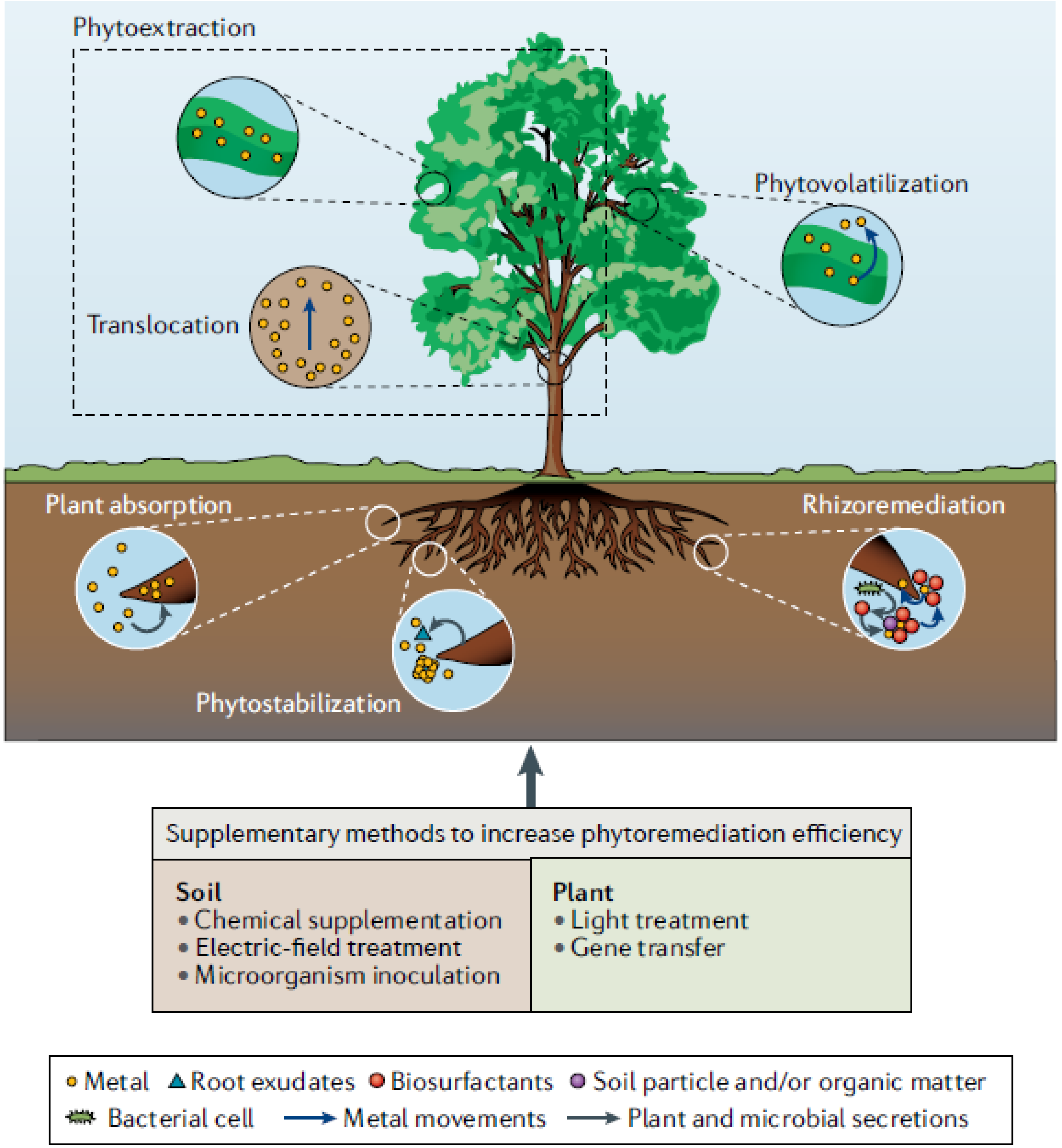
- **Bioremediation** tends to be more **sustainable** than traditional thermal or physico-chemical techniques such as soil washing, which can remove or destroy living organisms and soil organic matter, jeopardizing long-term soil health and diminishing post-remediation soil productivity.



Phytoremediation

- Phytoremediation for soil decontamination employs indigenous or imported species of plants, including ones that are genetically modified.

- 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.



Soil–plant–metal interactions

- Heavy metal(loid)s enter plant tissue through various pathways.

- 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** and, therefore, the **effectiveness of phytoremediation**.

MECHANISMS OF TOLERANCE TO CADMIUM TOXICITY

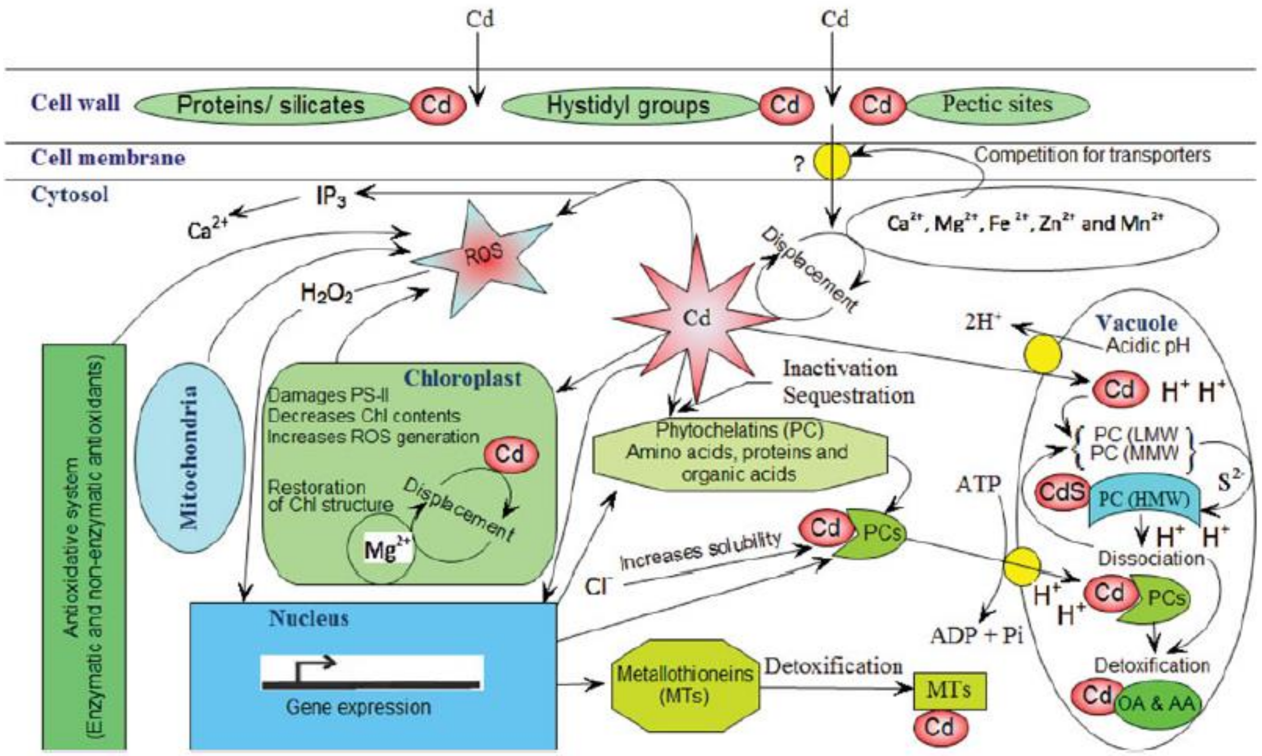
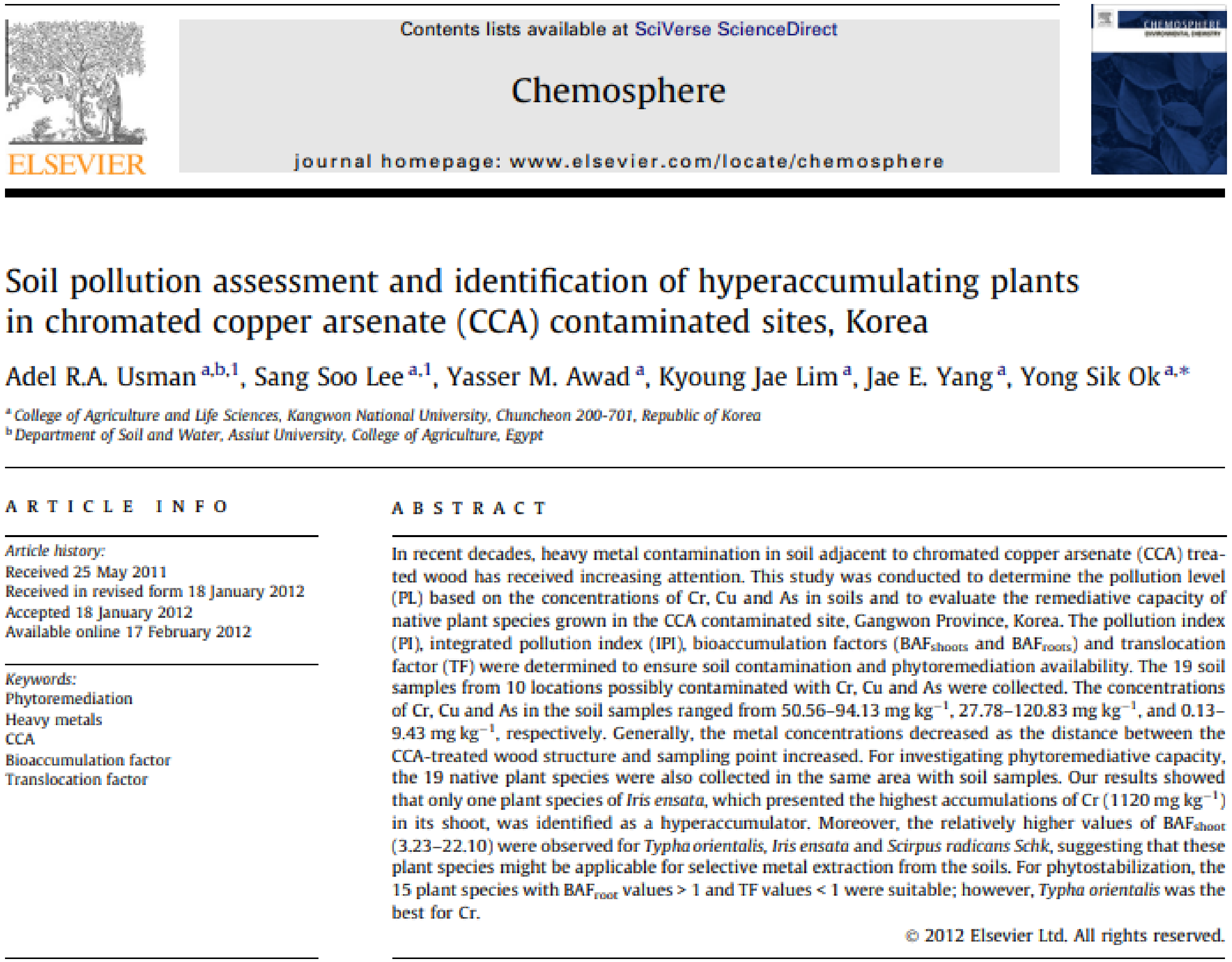


FIG. 4. Schematic representation of various cellular mechanisms involved in sequestration of Cd in plants.

Plant selection

- Plant selection is a critical step in phytoremediation, as species vary widely in their ability to uptake or immobilize different contaminants.
- Regardless of origin, **hyperaccumulators** (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.
- 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**, *Sedum plumbizincicola* of the Crassulaceae family for **cadmium and zinc**, the grass species *Pogonatherum crinitum* for **lead**, *Celosia argentea* (the plumed cockscomb or silver cock's comb) for **manganese** and *Pronephrium simplex* of the Thelypteridaceae family for **rare-earth elements**.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Soil pollution assessment and identification of hyperaccumulating plants in chromated copper arsenate (CCA) contaminated sites, Korea

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ABSTRACT

In recent decades, heavy metal contamination in soil adjacent to chromated copper arsenate (CCA) treated wood has received increasing attention. This study was conducted to determine the pollution level (PL) based on the concentrations of Cr, Cu and As in soils and to evaluate the remediative capacity of native plant species grown in the CCA contaminated site, Gangwon Province, Korea. The pollution index (PI), integrated pollution index (IPI), bioaccumulation factors (BAF_{shoots} and BAF_{roots}) and translocation factor (TF) were determined to ensure soil contamination and phytoremediation availability. The 19 soil samples from 10 locations possibly contaminated with Cr, Cu and As were collected. The concentrations of Cr, Cu and As in the soil samples ranged from 50.56–94.13 mg kg^{-1}, 27.78–120.83 mg kg^{-1}, and 0.13–9.43 mg kg^{-1}, respectively. Generally, the metal concentrations decreased as the distance between the CCA-treated wood structure and sampling point increased. For investigating phytoremediative capacity, the 19 native plant species were also collected in the same area with soil samples. Our results showed that only one plant species of *Iris ensata*, which presented the highest accumulations of Cr (1120 mg kg^{-1}) in its shoot, was identified as a hyperaccumulator. Moreover, the relatively higher values of BAF_{shoot} (3.23–22.10) were observed for *Typha orientalis*, *Iris ensata* and *Scirpus radicans Schk*, suggesting that these plant species might be applicable for selective metal extraction from the soils. For phytostabilization, the 15 plant species with BAF_{root} values > 1 and TF values < 1 were suitable; however, *Typha orientalis* was the best for Cr.

Field successes and challenges

- 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.
- The initial large-scale phytoremediation field trials on heavy-metal(loid)s-contaminated soils were conducted in the early 1990s, when it was suggested that this approach could reduce metal concentrations to acceptable ranges on otherwise productive land.
- **Plant density, initial plant size, cropping and harvesting strategies** such as double cropping, transplantation and double harvesting have been identified as crucial factors affecting success in these studies.

Environmental Pollution 294 (2022) 118627

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Cu phytoextraction and biomass utilization as essential trace element feed supplements for livestock☆

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Table 3
Cu concentration (in mg kg^{-1} DM) in selected feed materials according to different feed composition tables.

	CVB (2016)	Sauvant et al. (2004)	(Rodehutscord et al. (2016))
Feeding materials	Mean values	Mean values	Range
Wheat (grains)	3	7	3.59–5.42
Maize (grains)	1	2	1.04–4.11
Oat (grains)	3	3	3.15–4.25
Rye (grains)	4	5	3.74–4.87
Barley (grains)	4	9	4.27–6.20
Grass, dehydrated	–	7	–

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.

Microbial bioremediation

- Microorganisms exist at high concentrations in agricultural soils and possess genes enabling their survival in contaminated soil environments.

- 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 bioremediation**.

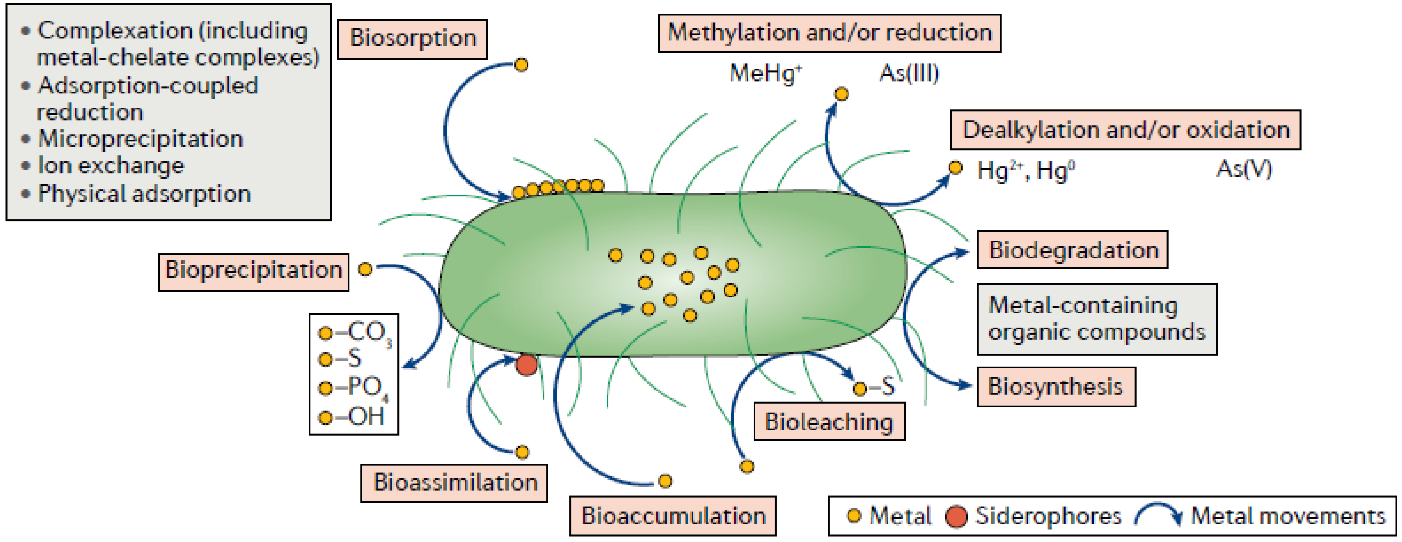

Fig. 5 | **Microbial bioremediation.** Processes by which bacteria can mediate the removal or detoxification of heavy metal(loid)s from agricultural soil. Bacteria can interact with heavy metal(loid)s directly, accumulating them on the cell surface (biosorption). They can also reduce or oxidize metal(loid) 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.

Ok YS* et al. (2022) *Nature Reviews Earth and Environment* 1:366

Soil–microorganism–metal interactions

- Biogeochemical processes facilitated by microbial activities form the basis of microbial bioremediation.
- A crucial mediator of remediation is the bacterial secretion of **siderophores**, which primarily transport iron from low-iron soils to cells through specific receptor and transport systems.
- **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.
- Biological reduction provides another important route for microbially assisted soil remediation because the toxicity of heavy metal(loid)s depends on their oxidation state.

Monitored natural attenuation

- The risk posed by heavy metal(loid)s in soil environments can naturally attenuate over time without specific remedial treatment.
- Natural attenuation processes comprise biological, physical and chemical mechanisms, but the activities of indigenous microbes often drive attenuation.

- 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.
- 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.

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Journal of Controlled Release

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Review article

Sustainable in situ remediation of recalcitrant organic pollutants in groundwater with controlled release materials: A review

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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.
- Although indigenous microbes are often excellent candidates for bioremediation because they are acclimated to site conditions, laboratory-grown microbial strains can be added to soil, a process known as bioaugmentation.

CRITICAL REVIEWS IN ENVIRONMENTAL SCIENCE AND TECHNOLOGY
2019, VOL. 49, NO. 12, 1027–1078
https://doi.org/10.1080/10643389.2018.1564526

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A critical review on bioremediation technologies for Cr(VI)-contaminated soils and wastewater

Shaopan Xia[a], Zhaoliang Song[a], Paramsothy Jeyakumar[b], Sabry M. Shaheen[c,d], Jörg Rinklebe[d,e], Yong Sik Ok[f], Nanthi Bolan[g], and Hailong Wang[h,i]

Biodegradation (2018) 29:311–312
https://doi.org/10.1007/s10532-018-9842-0

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EDITORIAL

Special issue: bioremediation of contaminated soil and water: GeoTrop 2017

Yiu Fai Tsang · Yong Sik Ok · Ajit K. Sarmah · Bin Cao · Ming Hung Wong

Integrated methods and phytomanagement

- Microbially mediated processes can enhance the efficiency of phytoremediation 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.
- The bacterial species *Pseudomonas aeruginosa, Pseudomonas fluorescens* and *Ralstonia metallidurans* produce siderophores that increase contaminant bioavailability to roots, leading to enhanced phytoextraction efficiency.
- 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.

CRITICAL REVIEWS IN ENVIRONMENTAL SCIENCE AND TECHNOLOGY
2016, VOL. 0, NO. 0, 1–31
http://dx.doi.org/10.1080/10643389.2016.1248199

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Phytomanagement of heavy metals in contaminated soils using sunflower: A review

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ORIGINAL PAPER

Use of Maize (*Zea mays* L.) for phytomanagement of Cd-contaminated soils: a critical review

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Integrated methods and phytomanagement

- The integration of remediation technologies provides a scenario where **ecosystem services** such as nutrient cycling, carbon sequestration and water storage are restored.
- Moreover, plants grown in contaminated agricultural fields undergoing bioremediation can be sold as **bioenergy products** or other profitable products.
- 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**.

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.
- 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**.

Summary and future perspectives

- Third, **field stations** are needed to provide valuable insights into the mechanisms that render heavy-metal(loid)s-contaminated sites resistant to treatment.
- Fourth, further research is required in order to decrease clean-up time and expand the applicability of bioremediation techniques to include more sites.
- 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.
- Policymakers should foster a bioremediation-enabling environment through policy instruments and **increased field-based research funding**.

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

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