Review

Role of organic amendments on enhanced bioremediation of heavy metal(loid) contaminated soils

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ABSTRACT

As land application becomes one of the important waste utilization and disposal practices, soil is increasingly being seen as a major source of metal(loid)s reaching food chain, mainly through plant uptake and animal transfer. With greater public awareness of the implications of contaminated soils on human and animal health there has been increasing interest in developing technologies to remediate contaminated sites. Bioremediation is a natural process which relies on soil microorganisms and higher plants to alter metal(loid) bioavailability and can be enhanced by addition of organic amendments to soils. Large quantities of organic amendments, such as manure compost, biosolid and municipal solid wastes are used as a source of nutrients and also as a conditioner to improve the physical properties and fertility of soils. These organic amendments that are low in metal(loid)s can be used as a sink for reducing the bioavailability of metal(loid)s in contaminated soils and sediments through their effect on the adsorption, complexation, reduction and volatilization of metal(loid)s. This review examines the mechanisms for the enhanced bioremediation of metal(loid)s by organic amendments and discusses the practical implications in relation to sequestration and bioavailability of metal(loid)s in soils.

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Contents

1. Introduction

The term 'heavy metal(loid)s' in general includes elements (both metals and metalloids) with an atomic density greater than $6\,g\,cm^{-3}$ [with the exception of arsenic (As), boron (B) and selenium (Se)]. This group includes both biologically essential [e.g., cobalt (Co), copper (Cu), chromium (Cr), manganese (Mn) and zinc (Zn)] and non-essential [e.g., cadmium (Cd), lead (Pb) and mercury (Hg)] elements. The essential elements (for plant, animal or human nutrition) are required in low concentrations and hence are known as 'trace elements' or 'micro nutrients'. The non-essential metal(loid)s are phytotoxic and/or zootoxic and are widely known as 'toxic elements'. Both groups are toxic to plants, animals and/or humans at exorbitant concentrations [1].

Health authorities in many parts of the world are becoming increasingly concerned about the effects of heavy metal(loid)s on environmental and human health and their potential implications to international trade. For example, Cd accumulation in the offal (mainly kidney and liver) of grazing animals not only makes it unsuitable for human consumption but also imperils the access of offal products to overseas markets. Similarly, bioaccumulation of Cd in potato, wheat and rice crops has serious implications to local and international commodity marketing [2–7]. For these reasons, there is global urgency to ensure that the heavy metal(loid) content of foodstuffs produced complies with regulatory standards and compare well with those from other countries.

Due to ever-increasing production of livestock and poultry products for human consumption, a large volume of organic wastes from these industries are generated. The large amounts produced must be treated or utilized in a manner that conforms to environmental regulations, including safe disposal onto land. Organic amendments, such as poultry manure compost and biosolid are used as a source of nutrients and also as a soil conditioner to improve the physical properties and fertility of soils. As land treatment becomes one of the important waste management practices, soil is increasingly being seen as a major source of metal(loid)s in the human food chain, introduced mainly through plant uptake and animal transfer [8–10].

With greater public awareness of the implications of contaminated soils on human and ecosystem health, there has been increasing interest amongst the scientific community and regulatory agencies in the development of technologies to remediate contaminated sites. However, unlike organic contaminants, most metal(loid)s do not undergo microbial or chemical degradation and their total concentration in soils persists for a long time after introduction. For diffuse distribution of metal(loid)s (e.g., fertilizer-derived Cd input in agricultural soils), remediation options generally include amelioration of soils to minimize their 'bioavailability'. As a working definition for EPA to use in risk assessment and risk management decision-making, bioavailability of metals is the extent to which bioaccessible metals adsorb onto or absorb into and across biological membranes of organisms [11]. Bioavailability can be minimized through chemical and biological immobilization of metal(loid)s using a range of inorganic compounds, such as lime and phosphate (P) compounds (e.g., apatite rocks), and organic compounds, such as 'exceptional quality' biosolid which meets the highest level of quality in three categories: (1) reduction of pathogens; (2) reduction of vector attraction (odor); and (3) low concentrations of specific metals [12–15]. The more localized metal contamination found in urban environments (e.g., Cr contamination in timber treatment plants) is remediated by metal mobilization processes that include phytoremediation (including phytovolatilization) and chemical washing [16–18].

Bioremediation of metal(loid) contaminated soils includes technologies that involve biological agents including higher plants, microorganisms and organic amendments. Bioremediation is a natural process which relies on bacteria, fungi, and higher plants to alter contaminants and environmental conditions as these organisms carry out their normal life functions and can be enhanced by adding organic amendments to soils (i.e. biostimulation/bioaugmentation). Metabolic processes of these organisms are capable of using chemical contaminants as an energy source, rendering the contaminants harmless by reducing their bioavailability or less toxic products in most cases [19,20].

Conventional methods to remove, reduce, or mitigate toxic substances introduced into soil or ground water via anthropogenic activities and processes include pump and treat systems, soil vapor extraction, incineration and containment. The utility of each of these conventional methods of treatment of contaminated soil and/or water suffers from recognizable drawbacks and may involve some level of risk. The emerging science and technology of bioremediation offers an alternative method to remediate contaminated sites. Bioremediation has been demonstrated and is being used as an effective means of mitigating hydrocarbons, halogenated organic solvents and organic compounds, non-chlorinated pesticides and herbicides, nitrogen compounds, and metal(loid)s and radionuclides.

This review focuses on the potential value of organic amendments in the remediation of metal(loid) contaminated sites. Following a brief overview of the reactions of metal(loid)s in soils and sources of organic amendments, the review describes the mechanisms for the enhanced bioremediation of metal(loid)s by organic amendments. The practical implications of organic amendments on bioremediation are discussed in relation to sequestration and bioavailability of metal(loid)s in soils.

2. Sources of heavy metal(loid)s in soil environment

Heavy metal(loid)s reach the soil environment through both pedogenic (or geogenic) and anthropogenic processes. Most metal(loid)s occur naturally in soil parent materials, chiefly in forms that are not available for plant uptake. Because of their low solubility, the metal(loid)s present in the parent materials are often not available for plant uptake and cause minimum impact to soil organisms. Often the concentrations of metal(loid)s released into the soil system by the natural pedogenic (or weathering) processes are largely related to the origin and nature of the parent material. Apart from Se [21,22] and As [23–25], other

elements (e.g., Cr, Ni, Pb) derived via geogenic processes have limited impact on soil. Unlike pedogenic inputs, metal(loid)s added through anthropogenic activities typically have high bioavailability [26–28]. Anthropogenic activities, primarily associated with industrial processes, manufacturing and the disposal of domestic, agricultural and industrial waste materials are the major source of metal enrichment in soils [1] (Table 1). Atmospheric pollution from Pb-based petrol is a major issue in many developed and developing countries where there has been no constraint on the usage of leaded gasoline. In urban areas, particularly in the US, Pb-based paints are a serious health issue also [29]. Fertilizer, manure and organic amendments addition to agricultural soils is considered to be the major source of most minor elements including metal(loid)s that are essential for plant growth.

2.1. Fertilizer products

Phosphate fertilizers are considered to be the major source of heavy metal(loid) input, especially Cd, in pastoral soils in Australia and New Zealand and paddy soils in Asian countries (Table 1). There have been greater efforts to reduce the accumulation of Cd in soils through the use of low Cd-containing P fertilizers. This is achieved by either selective use of phosphate rocks (PR)s with low Cd or treating the PRs during processing to remove Cd. Superphosphate fertilizer manufacturers in many countries are introducing voluntary controls on the Cd content of P fertilizers. For example, the fertilizer industry in New Zealand has achieved its objective of lowering the Cd content in P fertilizers from 340 mg Cd kg^{-1} P in the 1990s to 280 mg Cd kg^{-1} P by the year 2000. A number of PRs with low Cd contents are available which can be used for the manufacture of P fertilizers, but sources with higher Cd contents continue to be used in many countries for practical and economic reasons [38]. Several chemical processes to remove Cd from phosphoric acid before it is converted to P fertilizers have been examined. These include extraction of wet phosphoric acids with amines and by ion exchange resins. For example, calcinations which refer to heating of PRs, usually in the presence of silica and steam, are aimed at reducing Cd content through its volatilization. However, calcinations may not become a likely option in the fertilizer industry because it is expensive and calcinations decrease the reactivity of PRs, making them less suitable for direct application as a source of P [39].

2.2. Biosolids

Organic amendments such as biosolid (e.g., Cd) and poultry manure (e.g., As) have been regarded as the major sources of metal accumulation in soils, and a large volume of work has been carried to examine the mobilization and bioavailability of metal(loid)s derived from biosolid and manure in soil [40–42]. The heavy metal(loid)s in biosolid most commonly of concern, Pb, Ni, Cd, Cr, Cu and Zn originate primarily from the contamination of these wastes with industrial waste water [42]. Gove et al. [43] reported that biosolid application (250 kg N ha^{-1} year^{-1}) to sand or a sandy loam soil resulted in loadings of approximately Zn: 6 mg kg^{-1}, Cu: 2 mg kg^{-1}, Pb: 5 mg kg^{-1} and Ni: 0.2 mg kg^{-1} of soil on a dry weight basis. Illera et al. [44] demonstrated that biosolid application to soil had little effect on the total concentration of Ni and Cr, but resulted in a considerable increase of Cd, Cu, Pb and Zn as a consequence of the high availability of these metals in biosolid. It is known that they are typically immobilized in soils, but they can be toxic to soil micro flora and can be accumulated in plants and grazing animals [42]. Kao et al. [45] reported that the addition of biosolid enriched in Cu, Pb and Zn reduced the microbial biomasses C and N, indicating that the microbial activities were disrupted by the heavy metal(loid)s.

2.3. Manure

Manure addition is increasingly being recognized as a major source of metal input to soils, with repeated applications having resulted in elevated concentrations of metal(loid)s in soil. For example, the annual metal inputs to agricultural lands in England and Wales from animal manures amounted to 5247 Mg, 1821 Mg and 225 Mg of Zn, Cu and Ni, respectively which represent 25–40% of the total inputs [46]. Similarly, Jinadasa et al. [47] surveyed Cd levels in vegetables and soils of Sydney, Australia and concluded that the increases in Cd and Zn in vegetable soils were due to repeated applications of poultry manure. Xiong et al. [48] investigated the concentrations of Cu in pig, cattle, chicken and sheep manure in China and showed that the mean Cu concentrations in pig, cattle, chicken and sheep manures were 699.6 mg kg^{-1}, 31.8 mg kg^{-1}, 81.8 mg kg^{-1}, and 66.85 mg kg^{-1}, respectively. This can be major input of Cu to agricultural land. Similarly, in New Zealand, land application of dairy pond effluent, based on a N loading of 150 kg N ha^{-1}, is likely to add a maximum of 31.5 kg Cu ha^{-1} and 73.7 kg Cu ha^{-1} through effluent and manure sludge application, respectively [49]. Martinez and Peu [50] estimated that 183 kg and 266 kg Cu and Zn, respectively, were added to soil through 8 years of swine manure application, most of which accumulated in the surface soil.

Metal(loid)s in manure by-products are also derived from ingestion of contaminated soil by the animal, and also during manure collection and handling. A number of metal(loid)s are added to livestock and poultry feedstuff not only as essential nutrients but also as supplement to improve health and feed efficiency. In confined intensive animal production systems, a number of feed additives are used to improve feed efficiency and to reduce outbreaks of diseases [40,51]. Among the many feed additives, the metal(loid)s As, Co, Cu, Fe, Mn, Se, and Zn are added to prevent diseases, improve weight gains and feed conversion, and increase egg production in the case of poultry [52,53]. Similarly, regular use of growth promoters containing metal(loid)s is likely to result in elevated concentrations of these metal(loid)s in manure by-products [54,55]. Since a major portion of the metal(loid)s ingested is excreted in faeces and urine, concentrations in manure by-products depend primarily on their concentrations in the diet [56]. For example, Sutton et al. [57] and Kunkle et al. [58] observed that Cu concentrations in swine and poultry manure by-products were linearly related to Cu added in the diet. Similarly, Mohanna and Nys [59] noticed that by reducing dietary Zn from 190 mg kg^{-1} to 65 mg kg^{-1} in broiler poultry feed resulted in a decrease of Zn concentration in manure by 75%. Introducing highly viscous raw materials such as triticale, rye and barley at high levels in poultry diets has been shown to reduce Zn retention, thereby contributing to increased level of Zn in manure [60]. Li et al. [55] obtained a significant correlation ($R^2 = 0.89$, $p < 0.05$) between Cu in swine feed and feces Cu concentrations. The concentrations of Cu in feed samples ranged between 6.86 mg kg^{-1} and 395.19 mg kg^{-1} and Cu concentrations in pig feces were approximately 5-times greater than in pig feed.

As in the case of animal diet, the majority of metal(loid)s used in animal health remedies also eventually reach the end-use by-products. Addition of As to feed as an additive to control coccidiosis in poultry has been shown to result in a seven-fold increase in As level in poultry litter [61]. Similarly the excessive use of Cu compounds as a growth promoter in swine and poultry, and as a footbath in milking yards to treat lameness in dairy cattle [62], can result in elevated concentration of Cu in effluent and manure products [49].

Christen [63] obtained a direct correlation between water-extractable As in soils and the amount of poultry litter applied, implicating this material as a major source of As input in soils.

Table 1

Selected references on metal(loid) concentrations in phosphate compounds, cattle manure, poultry manure, swine manure, municipal sewage sludge and municipal solid waste [3,30–37].

Sample	Concentration ($mg\,kg^{-1}$)												
	As	B	Cd	Co	Cr	Cu	Hg	Mn	Mo	Ni	Pb	Se	Zn
Phosphate compounds													
Gafsa phosphate rock	4		38	3		Cu		7					393
Jordan phosphate rock	12		<1	4		4		5					235
North Carolina phosphate rock	23		48	2		8	0.4–2.1	7			<1–51		400
Sechura phosphate rock	5		11	3		9		91					178
Nauru Island phosphate rock	3		100	6		4		122					1010
Arad phosphate rock	7		12	4		8		2					560
Single super phosphate			32	77		32		890			488		165
Triple superphosphate			70	47		15		75			238		418
Diammonium phosphate			10	16		49		307			195		112
Cattle manure													
Dairy manure			–		15	7.2		372		9.0	8.6		67
Dairy liquid and solid manure	1.3	8.1	0.2			139	0.02		2.5	0.8	2.2	3	191
Cow dung						200		700					800
Cow manure	6.8		0.7	2.23	–	17.5	<0.4	172		9.6	7.5	–	–
Cattle manure (composted)	3.0		0.5	3.55	–	–	<0.4	186		6.2	2.6	–	–
Cattle manure (composted)	5.2		0.4	3.57	14.4	–	<0.4	357		8.7	5.4	0.48	164
Feed lot manure						16.5		149					6480
Feed lot lagoon						0.51		0.29					1.8
Dairy cattle FYM	1.63		0.38	5.32		37.5				3.7	3.61		153
Dairy cattle slurry	1.44		0.33	5.64		62.3				5.4	5.87		209
Beef cattle FYM	0.79		0.13	1.41		16.4				2.0	1.65		81
Beef cattle slurry	2.6		0.26	4.69		33.2				6.4	7.07		133
Poultry manure													
Poultry dropping						400		1800					2300
Broiler litter						313		246					327
Poultry litter				313				246					327
Broiler litter	34.6		4.93	9.9		6.1		501		2.46	0	1.23	743
Dried poultry waste from caged hens without any litter	0.57		–	2.0	6	30.7	<0.04	166	5.0		–	0.38	158
Broiler/turkey letter	9.01		0.42		17.17	96.8				5.4	3.62		378

Layer manure	0.46		1.06		4.57	64.8				7.1	8.37		459
Poultry litter	43	51	3	6	–	748		956	6	15	11		718
Deep-pit poultry litter		19	2	8	6	19		271		14	13		252
Poultry manure			0.48		7.3	54.3		465	7.69	7.0	2.3		550
Poultry manure		390											
Swine manure													
Swine manure			0.25		33	1338		869		12.4	14.0		1440
Swine dung						1000		2100					2900
Cu-enriched swine manure		17.8				1279		197					231
Swine FYM	0.86		0.37		1.98	374				7.5	2.94		431
Swine slurry	1.68		0.30		2.82	351				10.4	2.48		575
Sewage sludge													
Sewage sludge–Athens	4.97		11.2		75.1	54.7		1248		53.4	2.47	1.24	294
Urban compost			0.48		71	119		214		15	324		328
Urban compost			0.45		65	89		350	4304	85.3			354
Sewage sludge			11.4		645	870		497		479	226		1788
Denwer sewage sludge	8.1		26	7.1	280	816	7.8	220	84.9		950	4.57	1672
City sewage sludge	14.3		104	9.6	1441	1346	8.6	194	14.3		1832	3.1	2132
Austin sewage sludge (Autinite)	9.4		3.3	4.10	106	300	1.5	430		36.7	86.9	2.57	563
Milwauke sewage sludge (Milorganite)			7.2	4.07	2940	–	1.1	142		31.2	130	1.04	450
Anaerobic sewage sludge			4.4		5.1	709		129		5.3	67		407
Nu-Earth sludge			210										4140
Municipal solid waste													
Fresh municipal solid waste			6.0		16	139		816		25	216		2677
Degraded waste			3.0		53	173		643		21	420		1658
Municipal solid waste			9		55		1.2				330		
Green waste			0.075		20	37					87		214
Mixed refuse compost			5.5		71	274					513		1510
Wet waste from Kitchen	0.5		0.1		1.6	14	0.1			1.2	8.0		

The organic As compounds have been used as feed additives for swine disease control and weight improvement in China. Li and Chen [64] investigated As concentration in pig feeds and manures ranged from 0.15 mg kg^{-1} to 37.8 mg kg^{-1} and 0.42 mg kg^{-1} to 119.0 mg kg^{-1}, respectively. They reported that the potential soil arsenic increase rates resulting from land application of pig manure might range between 11.8 μg kg^{-1} year^{-1} and 78.9 μg kg^{-1} year^{-1} based on the loading rates of pig manure of 2.7–57.2 t ha^{-1} year^{-1}. Similarly, Kornegay et al. [65] examined the distribution of Cu in soils that received swine manure with low (59–88 mg kg^{-1}) and high (1180–2810 mg kg^{-1}) Cu concentrations, the latter was caused by feeding swine with Cu-enriched feed. Expectedly, there was a 3- to 4-fold increase in ethylenediamine tetraacetic acid (EDTA)-extractable Cu concentration in soils treated with Cu-enriched manure, indicating the dominance of Cu-organic matter complexes.

Soil ingestion has been identified as an important source of Cd ingestion by grazing sheep and cattle in New Zealand and Australia [66,67]. For example, it has been estimated that in New Zealand, sheep ingest 11–30 g soil d^{-1} soil in the summer and 264–275 g soil d^{-1} during the winter. The corresponding values for cattle are 220–470 g soil d^{-1} in summer and 900–1600 g soil d^{-1} in winter [68]. Based on these values and the average Cd concentration of 0.1–0.5 mg kg^{-1} in pasture soils, it can be estimated that approximately 15 mg and 90 mg of Cd is ingested annually through soil by sheep and cattle, respectively most of which is excreted in the manure. Similarly, Smith et al. [69] reported that sheep ingest soil at rates varying according to season. Soil ingestion rates in sheep ranged from 0.1% to 44% of dry matter intake and the median winter Pb intake was 783 mg d^{-1}, while in summer the median Pb intake was reduced to 55 mg d^{-1}. The principal source of Pb intake was found to be by direct ingestion of soil which accounted for more than 50% of ingested Pb for most of the year.

Advances in the treatment of sewage water and isolation of industrial wastewater in the sewage treatment plants have resulted in a steady decline in the metal content of biosolid. Furthermore, stabilization using alkaline materials has resulted in the immobilization of metal(loid)s in biosolid. Similarly, diet management in animal and poultry industries have resulted in low levels of metal(loid)s in manures [40,70,71]. A number of studies have shown that alkaline-stabilized biosolid compost that are low in total and/or bioavailable metal content and metal immobilized manures can be used as an effective sink for reducing the bioavailability of metal(loid)s in contaminated soils and sediments [72–74].

3. Reactions of heavy metal(loid)s in soils

3.1. Adsorption and complexation

Sposito [75] defined sorption as the accumulation of a solute at the interface between the aqueous solution phase and solid phase. Adsorption indicates formation of surface complexes which can be either physical attachment or bonding of metal ions and molecules onto the surface of another component. Adsorption of charged solutes by a charged adsorbent can be classified into specific and non-specific retention [38]. In general terms, non-specific adsorption is a process in which the charge on the solutes balances the charge on the sorbent through electrostatic attraction, displacing other like-charged ions from the surface [38]. Specific adsorption refers to chemical bond formation between the solute and the functional groups on the adsorbent [75].

Adsorption of heavy metal(loid)s strongly depends on soil pH [35,76]. For example, the amount of Pb sorption is a function of pH and increases sharply with pH, up to pH 5 and reaches a maximum at pH 8.0 with slight increase between pH 6 and 8; however, this depends on the adsorbent. The increase in Pb sorption with

pH is attributed to an increase in negative charge, precipitation as hydroxides and formation of hydroxyl species that are more strongly retained compared to free metal ion species [77,78]. The formation of multi-nuclear metal-hydroxy species in solution is the precursor to formation of metal-hydroxy precipitates from a homogenous solution [79].

Adsorption of heavy metal(loid)s is also highly dependent on soil components that include silicate clays, organic matter, and iron, aluminium and manganese oxides [76]. Redox potential and cation/anion exchange capacity also control heavy metal(loid)s sorption, but a single factor rarely accounts for their sorption in soil [80]. Metal(loid)s form both inorganic and organic complexes with a range of soil components [38]. Surface complexation indicates semi-covalent bond formation between dissolved ions and surface function groups [81]. Soil organic matter has a high affinity for metal cations due to the presence of ligands or functional groups [82]. When soil pH increases, H$^+$ dissociates from functional groups such as carboxyl, phenolic, hydroxyl, and carbonyl functional groups, thereby increasing the affinity for metal cations [38]. The general order of affinity of heavy metal(loid)s on organic matter is as follows [1,83,84]:

$$Cu^{2+} > Hg^{2+} > Cd^{2+} > Fe^{2+} > Pb^{2+} > Ni^{2+} > Co^{2+}$$
$$> Mn^{2+} > Zn^{2+} > As(V) > As(III)$$
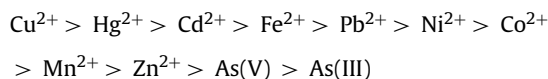
Soil surface possesses various hydroxyl groups with different levels of reactivity concerning the dissociation of H$^+$. The deprotonated aluminol groups interact with heavy metal(loid)s to form stable surface complexes [85]. When the metal(loid) ion is directly bound to surface functional groups of soil particles, the complexes are called inner-sphere complexes. When the water molecules interposed between the surface functional group and metal ions, the complexes are called outer-sphere complexes. Generally, inner-sphere complexes are more stable than outer-sphere complexes due to covalent bonding in inner-sphere complexes [81]. An important aspect of metal–organic matter interactions is the ability of organic functional groups to form multi-dentate complexes, thereby increasing the stability of the bond. Heavy metal(loid)s readily form complexes with natural ligands such as humic and fulvic acids or anthropogenic ligands such as EDTA and nitrilo triacetic acid (NTA) [81]. Multi-dentate complex formation in solution, known as chelation, greatly enhances the metal solubility. Heavy metal(loid)-EDTA or NTA complexes can enhance their mobility. This property can be applied to phytoextraction technology in order to remove heavy metal(loid)s from soil [86]. However, ternary complex formation (complex of three components – metal, ligand and organic acid) in solution increases the risk of metal mobilization to groundwater reserves [87].

Certain factors affect heavy metal(loid)-organic complexes and these variables are temperature, solution and soil pH, ionic strength, dominant cations and soil type [38,88]. Soil pH is the most significant factor influencing metal(loid)–soil chemistry. For example, when pH increases the surface functional group is negatively charged. Consequently, metal cations are almost completely removed from soil solution at high pH [89]. Soil type and components have an important effect on heavy metal(loid)s complexation. In general, fine grained soils have more heavy metal(loid) retention potential than coarse grained soils because the former contain large amounts of active surface sites such as clay minerals, iron and manganese oxyhydroxides, and humic acids [81].

In the case of metalloids, such as As, the effect of soil acidity on adsorption is manifested through two interacting factors–the increasing negative surface potential on the plane of adsorption and the increasing amount of negatively charged As(V) species present in soil solution. While the first factor results in lower As(V) adsorption, the second factor is likely to increase adsorp-

tion. Thus the pH effect on As(V) adsorption is largely influenced by the nature of the mineral surface. For example, in soils with low oxide content, increasing pH had little effect on adsorption, while in highly oxidic soils, adsorption decreased with increasing pH [90]. Soil acidification affects the solubility of Cr through its effect on adsorption/precipitation and oxidation/reduction reactions [91–95]. While the adsorption of Cr(VI) in soil increases with decreasing pH, the adsorption of Cr(III) decreases [93].

3.2. Precipitation

Precipitation is an important process of metal(loid) immobilization in the presence of anions such as sulfate, carbonate, hydroxide and phosphate when the soil pH and the metal(loid) concentration are high [1]. Co-precipitation as well as precipitation as salts contributes to the immobilization of heavy metal(loid)s [80]. Co-precipitation of metal(loid)s takes place especially in the presence of Fe and aluminum (Al) oxyhydroxides [96]. For example, Contin et al. [97] reported co-precipitation of Cd, Cu, Ni, Pb and Zn onto precipitated Fe (hydr)oxides. Hydroxide precipitation is the most common and effective method of treatment for heavy metal(loid)s [98]. Liming often increases the precipitation of metal(loid)s. For example, Lee et al. [99] used granulated lime and calcium carbonate as coagulants to remove heavy metal(loid)s from contaminated water. They showed that the main removal mechanism of heavy metal(loid)s in their experiments was precipitation. Sulfide precipitation is also considered as an effective process for the precipitation of highly toxic heavy metal(loid)s. Metal sulfides are the least soluble minerals under reducing conditions. The attractive feature of the sulfide precipitation is the efficiency for metal(loid) removal over a broad pH range due to low solubility of metal sulfides and fast reaction [100]. However, to maintain low metal(loid) solubility using sulfides requires the maintenance of reducing conditions, and for most surface soils is not practical.

Lead forms precipitates with carbonates, phosphates, and sulfates present in the soil [101]. Lead precipitation with carbonate is more common in calcareous soil than in non-calcareous soil [102]. Park et al. [103] demonstrated the formation of Pb precipitates such as carbonate ($PbCO_3$), chloride ($PbCl_2$) and hydroxide chloride [Pb(OH)Cl] with the reaction of Pb with Mg/Al layered double hydroxides in aqueous solution. Phosphate compounds are the most common amendment to precipitate heavy metal(loid)s effectively in contaminated soils or water. The usual stability sequence of metal phosphates is Pb > Cu > Zn [104]. Most of the studies on Pb stabilization have used various P-containing amendments, which reduce the Pb mobility by ionic exchange and precipitation of pyromorphite-type minerals [$Pb_5(PO_4)_3X$; X = F, Cl, B or OH] [80]. The common precipitate is hydroxypyromorphite or chloropyromorphite [104,105].

3.3. Oxidation/reduction

Arsenic, Cr, Hg and Se are most commonly subjected to microbial oxidation/reduction reactions, whilst divalent metals such as Zn, Cd, Pb and Ni are not. The oxidation/reaction reactions are grouped into two categories, assimilatory and dissimilatory. In assimiliatory reaction, the metal(loid)s substrate acts as a terminal electron acceptor, thereby promoting bacterial growth. In the dissimilatory reactions the metal(loid) substrate has no specific function in the physiology of the microorganisms, and occurs by fortuitous reductions coupled to microbial oxidations of simple organic acids and alcohols, H_2 or aromatic compounds [106].

Some anaerobic bacteria use Se(VI) as a terminal electron acceptor for their growth. Reduction of Se(VI) to Se(0) is an important process to precipitate Se from contaminated water. Bacteria also enzymatically reduce Cr(VI) to the less mobile and toxic Cr(III),

and reduce Hg(II) to volatile Hg(0), which can be used as a remediation strategy [107]. Lindstrom and Sehlin [108] reported that archaebacterium *Sulfolobus acidocaldarius* has the ability to oxidize As(III) to As(V). The rate of oxidation of As(III) to As(V) in aqueous systems is increased by several orders of magnitude by the presence of dissolved Fe(III) and illumination with near ultraviolet light [109]. Since As(V) is more strongly retained by inorganic soil components and less toxic than As(III), microbial oxidation results in the immobilization and bioremediation of As. Similarly, Cr(III) is strongly retained onto soil particles and less toxic than Cr(VI), the reduction of Cr(VI) to Cr(III) can enhance the immobilization of Cr, thereby rendering it less bioavailable. Lie et al. [110] isolated bacteria from Cr-contaminated landfill and demonstrated that toxic Cr(VI) was reduced effectively into comparatively less toxic Cr(III) by *Bacillus* sp. Sun et al. [111] reported the photochemical reduction of Cr(VI) by organic acids with α-OH in the presence of Fe(III) and indicated that the complex formation between Fe(III) and organic acid is a key step for the photocatalytic reduction of Cr(VI).

3.4. Methylation/demethylation

Methylation is considered to be the major process of volatilization of As, Hg and Se in soils and sediments, resulting in the release of toxic methyl gas [112]. Although methylation of metal(loid)s occurs through both chemical and biological processes, biological methylation is considered to be the dominant process in soil and aquatic environments. Thayer and Brinckman [113] grouped biomethylation into transmethylation and fission. Transmethylation implies transfer of an intact methyl group from methyl donor to another compound. Fission refers to the transfer of a molecule such as formaldehyde or formic acid from a methyl source to another compound. Afterward the resulting group is reduced to a methyl group.

The metabolic methylation of inorganic As is considered to be a process of detoxification in aquatic organisms [114]. Inorganic As is methylated in the organism by alternating reduction of As(V) to As(III) and addition of a methyl group from *S*-adenosylmethionine to form methylarsonic acid and dimethylarsinic acid which are less reactive with tissue constituents and are more readily excreted in urine than inorganic As [115].

Methylation and demethylation are important processes regulating the Hg cycle in the environment [116]. Microorganisms can methylate Hg in water and soils. Methylation of Hg in the environment occurs mainly by biological processes involving sulfate reducing bacteria [117] under anoxic conditions [118]. Pongratz and Heumann [119] reported that bacterial culture released trimethyl Pb and monomethyl Cd as well as dimethyl Hg. Abiotic methylation of Hg can occur by methylcobalamin, methyltin compounds, and humic matter. Among these methyl donors, humic matter is the most promising environmental methylating agent [120]. The reaction between oxidized mercury and small organic molecules such as acetic acid is considered as a potential abiotic methylation process. Oxidized Hg bound to thiols in humic molecules can be abiotically methylated by humic substances [121]. Demethylation of methyl Hg can occur by both reductive and oxidative pathways and result from either cellular detoxification or metabolic processes in microorganisms. End products of reductive demethylation are CH_4 and either Hg(II) or Hg(0) [122].

Methylation of Se occurs through biomethylation in which microorganisms or plants convert inorganic Se into dimethyl selenide or dimethyl diselenide [123]. Methylated species of Se are volatile, and methylation of Se can be resistance mechanism of bacteria [124]. Methylation of inorganic and organic Se to dimethyl selenide and demethylation of trimethylselenomium ion to dimethyl selenide are considered detoxification processes of Se.

Zhang and Frankenberger [125] reported that trimethylselenonium ion is a major urinary Se metabolite of animals.

3.5. Biological modification of local soil environments

Certain microbial processes can enhance metal(loid) solubility, thereby increasing their bioavailability and potential toxicity, whereas other processes result in the immobilization, thereby decreasing their bioavailability. On the one hand, solubilization of metal(loid)s can occur by chemolithotrophic (autotrophic) and chemoorganotrophic (heterotrophic) mobilization mostly by the release of inorganic and organic acids, siderophores and other complexing agents, and thereby accelerating redox, methylation, demethylation and biodegradation [126]. On the other hand, microbially induced metal(loid) immobilization can occur by biosorption, precipitation, reduction, accumulation, intracellular deposition, localization and sequestration [127]. Microbial products derived, induced or excreted by the presence of certain metal(loid)s can result in their removal through adsorption. Metallothioneins which are small cysteine-rich polypeptides that can bind metal(loid)s, phytochelatins, cysteine-containing γ-glutamyl peptides, metal-thiolate clusters and microbial exopolymers composed of polysaccharide are involved in heavy metal(loid) binding and detoxification [128–130].

A number of studies have demonstrated metal immobilization by bacteria. For example, *Azotobacter* sp. and *Micrococcus luteus* immobilized 490 mg Pb g^{-1} and 310 mg Pb g^{-1} whole cells, on a dry weight basis, respectively. This was mostly located in the cell wall and membrane [131] and in lipid extracts of *M. luteus* cells [132]. Similarly, Zn was removed effectively from Zn containing medium by inoculation of sulfate-reducing bacteria (SRB) enriched on phosphogypsum as a sulfate source. The mechanism of Zn removal is precipitation with sulfide in the medium [133]. Di gregorio et al. [134] isolated a bacterial strain, related to the species *Stenotrophomonas maltophilia* and resistant to Se(III), from the rhizosphere of the Se hyperaccumulator legume *Astragalus bisulcatus*. This strain reduced soluble and harmful Se(III) to insoluble and non-bioavailable elemental Se [134].

Plant roots also affect on local soil environments and chemistry of metal(loid)s. The ability of plant roots to remove heavy metal(loid)s from contaminated soils has been used as emerging environmental remediation technology. Changes in bioavailability of metal(loid)s in the soil is often resulted from root-induced changes in soil properties [135]. The factors influencing metal(loid) bioavailability in soil include root-induced pH changes, metal binding by root exudates, detoxification of metal(loid)s by phytochelatins, root-induced microbial activities, and root depletion as a consequence of plant uptake [136]. Plant root exudates include a variety of soluble substances such as organic acids which form complexes with metal(loid)s and dissolve the solid phase metal(loid)s in the soil [137].

4. Sources of organic amendments

Organic wastes have been utilized as beneficial soil amendments for centuries [138,139]. Animal manures have in the past been widely used as a source of essential nutrients and other benefits to soils. The more recent concern about soil contamination has resulted in organic wastes been used as materials for remediation of contaminated sites. In addition, the increase in wastewater generation and intensification of livestock has resulted in large quantities of solid organic wastes from very widely different sources with variable composition [138,139]. Several industries generate significant quantities of organic waste, such as paper mill factories, olive mills, etc. [140].

Municipalities generate several types of organic waste which may be used as a soil amendment for land reclamation [141]. Municipalities generate two major sources of organic waste for soil amendment. The major organically based by-products include biosolids (often referred to as sewage sludge) and municipal solid wastes (MSW). Municipal solid waste is the general waste stream generated in local municipalities, including garden waste. Organic waste is largely separated from other materials (i.e. general waste, glass, aluminum, non-recyclables) for re-use often after composting [142]. Disposal of organic wastes is a significant issue for virtually all governments, with the soil area available for disposal of organic wastes declining over the last few decades [138]. Municipal solid wastes are derived from disposal of the general waste stream that includes food scraps and yard trimmings as well as miscellaneous products which are separated from non-compostable materials [142]. Nevertheless, MSW often contain significant levels of heavy metal(loid)s due to incomplete separation of industrial waste streams. Municipal solid wastes are generated in significant quantities, with approximately 0.1×10^6 Mg year^{-1} and 20×10^6 Mg year^{-1} generated in the US and France, respectively (Table 2). Biosolid

Table 2
Quantities of various organic waste by-products from municipal and industrial wastes produced on an annual basis.

Material	Location Nation, State	Quantity of Organic waste (10^6 Mg year^{-1})	Re-used (%)	References
Biosolid	US	7.5	56	[141,143]
Biosolid	Victoria, Australia	0.7	–	[144]
Biosolid	New Zealand	1–2	–	[145]
Paper mill Sludge	US	5.6	14	[146]
Biosolid	Canada	0.4	49	[147]
Biosolid	United Kingdom	1.1	–	[148]
Biosolid	Netherlands	0.3	–	[148]
Cattle, poultry, swine	US	174	–	[143]
MSW	US	0.1	–	[143]
MSW	France	20.5	7	[149]
Poultry manure	US	44.4	–	[150]
Poultry manure	New Zealand	0.59	–	[150]
Feedlot manure	Australia	>1	–	[151]
Green waste (urban garden waste)	Australia	>3	–	[151]
MSW	Poland	12.3	1.8	[152]
MSW	India	48	–	[153]
Rice husk	India	20	–	[153]
Rice wheat straw	India	12	–	[153]
MSW	UK	28	–	[154]
Pulp and paper	EU	11	55	[155]

generation per annum is generally more significant however. The amount of biosolids generated per annum in the US is approximately 7.5×10^6 Mg year^{-1}, approximately $1.5–2 \times 10^6$ Mg year^{-1} of wet biosolids (varied moisture content) in New Zealand [145], 0.4×10^6 Mg year^{-1} dry weight in Canada and 1.1×10^6 Mg year^{-1} in the United Kingdom, and 0.07×10^6 Mg year^{-1} in Victoria, Australia. Unfortunately, a substantial proportion of biosolids are not re-used for beneficial purposes (i.e. land application, contaminated site reclamation, energy production) due to local public opposition and potential hazards, and are disposed of in landfills [141,145,147,149].

Animal manure is another major source of organic amendments [139]. The principal sources of manure are derived from chicken, swine and cattle farms, although farm dairy effluent is also significant. Confined animal production methods have increased the efficiency of animal food production, but have also led to a variety of environmental issues. The amount of waste generated from animal farms in the US far exceeds all municipal by-products. The use of metal(loid)s (eg. Cu, Zn, As, Se and Co) in animal diets to supply adequate nutrition or control disease have resulted in metal(loid) enriched organic waste streams. However, the usefulness and properties of animal manures depends on numerous production factors. The chemical, biological and physical properties depend on factors such as animal type, animal attributes (age, size), water use, manure collection (floor type), season, bedding type and storage and handling of manures [156]. Composting of animal manure is often desirable, to reduce the quantity of animal manure needing disposal, and to stabilize the organic material.

There are a range of other organic waste products which have been used as soil amendments, although these are typically produced in smaller quantities. Papermill waste water treatment processes in some countries may be an exception. In the US in 1995 it was estimated that 5.6×10^6 Mg dry weight of paper mill sludge was produced, of which only 14% was re-used for land application [146]. The use of papermill sludge has been shown to produce benefits to crop production on agricultural fields and reclamation of mine sites [157–159]. However, reclamation of mine soils or tailings with paper mill sludge often requires additional nutrient inputs [157,159,160], owing to the high carbon content and low nitrogen levels [146].

Treatment of organic waste streams is increasingly being designed to suit the end-use [161]. Although biosolids are often modified primarily for the control of odor and disease control during the waste water treatment process [138,161,162], they are increasingly mixed with other products. The most common biosolid products are alkaline stabilized biosolids, which is the most common method for generation of Class A biosolids [140,163]. Biosolids have been similarly blended with lime, other alkali and various salts after biosolid production for improving alkalinity and soil productivity. Lime and gypsum treated anaerobically biosolids post production were used by Pietz et al. [164,165] for revegetation of acidic coal refuse material. It was found that the lime + biosolid and gypsum + biosolid treatments reduced soil acidity and dissolved Al and increased plant productivity compared to control and individual amendments. Similarly, Maddocks et al. [166] reported that only the application of biosolid and Bauxsol^{TM}, which is a by-product of alumina processing, concurrently enabled both the grass *Bothrichloa insculpta* and the tree species *Eucalyptus paniculata* to grow well on an acid, metal contaminated mine overburden. Sajwan and Youngblood [167] reported increased growth of Sorgum with mixtures of fly-ash and biosolid in a greenhouse study. In addition, additional materials may be necessary to increase fertility or reduce leaching and runoff losses of environment contaminants, such as P and N [168–171]. It is often the case that a single waste material does not possess all the properties required to ameliorate site contamination, acidity, sodicity, etc. and the use of multiple blended materials is often required. Thus, the production of land amendments for contaminated soil reclamation is likely to require specifically blended waste by-products. Furthermore, some of these amendments may contain potentially toxic elements which require careful consideration when applied to soils.

5. Approaches to bioremediation of heavy metal(loid) contaminated soils

Unlike organic contaminants, most metal(loid)s do not undergo microbial or chemical degradation and two approaches are employed in mitigating their impacts and remediation of contaminated environments. These include (i) immobilization and (ii) mobilization of metal(loid)s, thereby controlling their bioavailability. Bioavailability of metal(loid)s plays a key role in both these two approaches.

The bioavailability of metal(loid)s in the soil environment has been defined as the fraction of the total metal(loid) in the interstitial pore water (i.e., soil solution) and soil particles that is available to the receptor organism [172]. Considerable controversy exists in the literature relating to the definition and the methods used for its measurements. For instance, microbiologists often regard the concentration that can induce a change either in morphology or physiology of the organism as the bioavailable fraction, whereas plant scientists regard the plant available pool as the bioavailable fraction [172]. Recent studies have indicated that the transformation of contaminants in soils is a dynamic process which means bioavailability changes with time [173–178].

A more generic definition of bioavailability is the potential for living organisms to take up metal(loid)s through ingestion or from the abiotic environment (i.e., external) to the extent that the metal(loid)s may become involved in the metabolism of the organism. More specifically, it refers to the biologically available fraction (or pool) that can be taken up by an organism and can react with its metabolic machinery [179]; or it refers to the fraction of the total concentration that can interact with a biological target [180]. In order to be bioavailable, the metal(loid)s have to come in contact with the organism (i.e., physical accessibility). Moreover, metal(loid)s need to be in a particular form (i.e., chemical accessibility) to be able to enter biota. In essence, for a metal(loid) to be bioavailable, it will have to be in an accessible form to the biota concerned.

In the case of immobilization, the bioavailability of metal(loid)s is minimized by allowing them to react with the soil for a longer period (ageing) or by adding soil amendments. A number of studies have documented the effect of ageing on the immobilization of metal(loid)s in soils [175,181,182] and also the potential value of various organic and inorganic soil amendments in reducing the bioavailability of metal(loid)s in soil [183–186]. For example, various phosphate compounds have been found to be very effective in the immobilization of Pb in soils [187–189] and USEPA recommends this technique for risk based remediation of Pb contaminated sites [190]. The primary objective of this technique is to reduce the risk of metal(loid)s reaching the food chain through plant uptake and off site contamination through leaching and erosion. Increasingly, plants with the associated microbial community are used to achieve the stabilization of metal(loid) contaminated soils (i.e. phytostabilization) [191].

In the case of mobilization, the bioavailability of metal(loid)s is increased by transforming the metal(loid)s from the solid phase to the soil solution phase. A number of studies have demonstrated the value of various soil amendments such as EDTA in enhancing the mobilization of metal(loid)s in soils [4,192–194]. However, EDTA is toxic to plant and microorganisms and not easily degradable [195]. Recently, ethylenediamine disuccinate (EDDS) has been identified
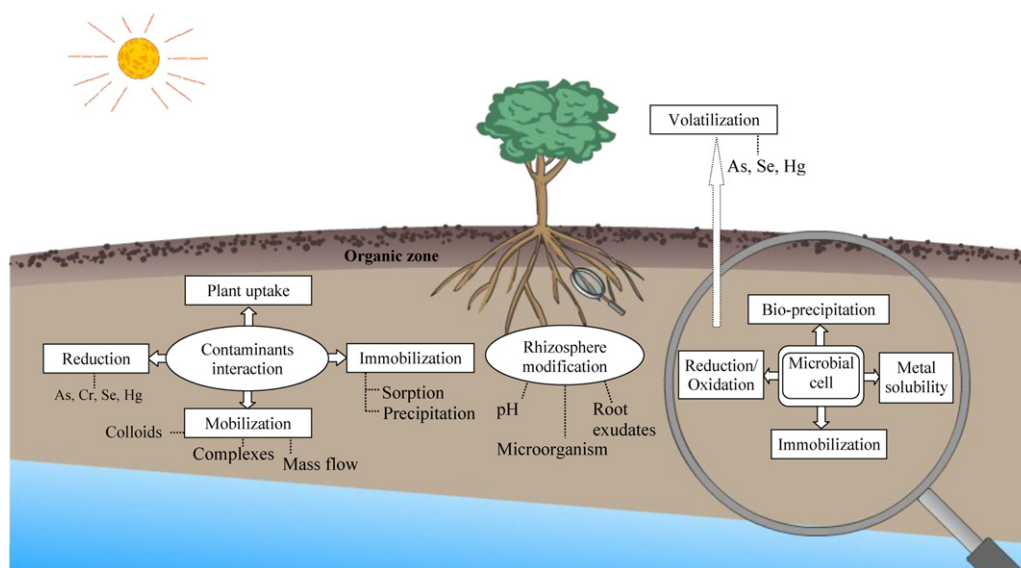
Fig. 1. The role of organic amendments in regulating various bioremediation processes that include immobilization, reduction, volatilization and rhizosphere modification.

as a promising biodegradable alternative for persistent compounds such as EDTA for application in enhancing the mobilization of heavy metals [196]. The mobilized metal(loid)s are subsequently removed either through soil washing [197,198] or plant uptake (i.e. phytoremediation) [199,200]. Removal of metal(loid)s through phytoremediation techniques and their subsequent recovery or safe disposal of plants such as incineration and ashing are attracting research and commercial interests. However, when it is not possible to remove the metal(loid)s from the contaminated sites by phytoremediation, other viable options, such as *in-situ* immobilization should be considered as an integral part of risk management.

6. Mechanisms for enhanced bioremediation of heavy metal(loid)s by organic amendments

Organic amendments enhance bioremediation of heavy metal(loid)s through various processes that include immobilization, reduction, volatilization and rhizosphere modification (Fig. 1).

6.1. Immobilization

It has often been shown that the addition of organic amendments to soils increases the immobilization of metal(loid)s through adsorption reactions (Table 3). The organic amendment-induced retention of metal(loid)s is attributed to an increase in surface charge [218] and the presence of metal(loid) binding compounds [219]. For example, Bolan et al. [220] noticed that the addition of biosolid compost increased the surface charge of the amended soils, which is attributed to the higher pH and surface charge of the biosolid compost. However, Li et al. [221] have shown that in the case of alkaline-stabilized biosolid compost addition to soil, the increase in charge is not in proportion to the amount added, which is often attributed to the enhanced solubilization of organic matter by the alkalinity of material. Similarly, it has been shown that the removal of organic matter altered the surface charge of variable charge soils [222,223]. The presence of phosphates, Al and Fe compounds and other inorganic minerals in municipal sewage sludge is also believed to be responsible for increasing metal(loid) sorption. For example, Li et al. [221] reported evidence for greater affinity for Cd adsorption by the inorganic components of the biosolid compost-amended soils, indicating that the increased adsorption

of Cd is independent of the added organic matter and of a persistent nature. Lozano Cerezo et al. [224] have shown that treatment with sewage sludge increased the pH and the organic matter contents and decreased the availability of heavy metal(loid)s with time in a clay quarry. Organic amendments were effective for immobilization of Cu by increased formation of copper–organic matter complexes, and for Cr by reduction from Cr(VI) higher valency to Cr(III) and subsequent precipitation as chromic hydroxide [16].

Compost treatment in soils from an Italian mining area improved soil physical characteristics such as particle size distribution, cracking pattern and porosity. The development of better soil structural characteristics may be able to prevent the dispersion of metal contaminated particles by formation of water stable aggregates. Compost addition improved the growth of ryegrass and tall fescue (*Festuca arundinacea*), and decreased their content of Zn and Pb in stems and leaves [225]. Similarly, Pichtel and Bradway [226] reported that composted peat had a significant effect on spinach (*Spinacea oleracea*) and cabbage (*Brassica oleracea*) tissue metal concentrations by reducing bioavailable Pb and Zn in soil. Cobalt, Cr, Cu, Ni, Pb and Zn immobilization by various organic amendments are presented in Fig. 2 [227].

Melamed and Boas [228] evaluated the effectiveness and drawbacks of physico-chemical amendments to mitigate Hg pollution, originated from Hg(0) use in gold (Au) recovery. Results indicated that the use of oxides, phosphate, and organic matter may be effective in the immobilization of Hg(II), depending upon Hg speciation. Calcium was effective in counteracting the increased solubility enhancement of Hg(0) due to the presence of humic acid. Tejada et al. [229] investigated the effect of crushed cotton gin compost and poultry manure on the biological properties of a soil contaminated with Ni. The addition of organic amendments increased enzyme activity such as urease, benzeneboronic acid-protease, β-glucosidase, alkaline phosphatase and arylsulfatase. The enzyme activity was highest in crushed cotton gin compost. This may have been due to the adsorption capacity of Ni being higher in the humic acid of crushed cotton gin compost than in the fulvic acid-amended soil. Similarly, Aguilar-Carrillo et al. [230] tested the efficiency of sugar foam rich in calcium carbonate in the immobilization of As, Cd and thallium (Tl) in an acidic soil. The amendment of sugar foam retained the metal(loid)s and the percentage of extraction by the toxicity characteristic leaching procedure (TCLP) was reduced. This indicated that the sugar foam had the potential to immobilize As, Cd

Table 3
Selected references on the potential value of organic amendments in the immobilization and plant uptake of metal(loid)s.

Amendments	Substrate	Metal(loid)s	Observations	References
Poultry compost	Soil (Alfisol) from barren farm land spiked with Cd	Cd	Application of poultry manure compost transformed 47.8–69.8% of soluble/exchangeable Cd to the organic-bound fraction and consequently decreased Cd uptake of plants by 56.2–62.5% compared to the control.	[201]
Green waste, municipal solid waste–derived compost	Highly acidic clay-loam soil contaminated with As, Cu, Pb and Zn from mine site	As, Cu, Pb	Either green waste or municipal solid waste-derived composts reduced plant metal uptake by binding metal to organic matter.	[74]
Coir, green waste compost and wood bark	Metal-contaminated mine waste	Pb, Cu, Zn	Application at the rate 10% and 20% significantly reduced the plant availability of Pb, Cu and Zn because of the ability of the waste to immobilize metals in soil.	[202]
Compost	Soil (Ferralsol) spiked with Cd	Cd	Compost application decreased exchangeable Cd by 70% and was effective in reducing the phytotoxicity of Cd.	[203]
Sewage sludge, municipal solid waste compost, garden waste compost	Metal-contaminated mine soil from a pyrite mine	Zn	Application of organic amendment reduced 0.01 M $CaCl_2$ extractable Cu, Zn and Pb concentration.	[204]
Bio-sludge, bio-fertilizers	Black cotton (calcareous) soil spiked with As, Cr and Zn	As, Cr, Zn	A significant reduction in the metal uptake in plant, when amended with biosludge and bio-fertilizer, which is attributed to the stabilization of heavy metal(loid)s in the soil.	[205]
Green waste compost	Soil from a dismantled zinc smelter	Cd, Zn	The green waste compost reduced the leaching of Cd and Zn up to 48% in soil.	[206]
Organic manures	Soil from metal contaminated sites in Bangladesh	Mn, Cr, Ni	Organic manures reduced Mn, Cr, and Ni uptake.	[207]
Biosolid	Varaible charge soils in New Zealand	Cu, Cr^{6+}	Organic amendments are effective in reducing phyto availability of Cu and Cr(VI) by formation of organic metal complexes and reduction to immobile form, respectively.	[16]
Municipal biosolids, woody debris, wood ash, pulp and paper sludge, compost	High metal waste materials from historic mining site	Zn, Pb, Cd	Surface application of biosolids mixed with wood ash or other amendments resulted in significant decreases in acidity and $Ca(NO_3)_2$ extractable Zn in the subsoil. The amendments were able to restore a vegetative cover to the metal contaminated materials.	[208]
Cyclonic ashes, biosolids, water treatment residuals	Fine fraction of mine tailings	Cd, Pb, Zn	All amendments reduced soil solution and NH_4NO_3 extractable metals	[209]
Vermi compost	Waste water	Zn	Vermicompost adsorbed a maximum of 2.49 mg Zn g^{-1} at pH 2 from kaolin wastewater; The fine soil particle contained in vermicompost reduced the adsorption capacity of the organic matter of vemicompost.	[210]
Coir fibre	Aqueous solution	Ni, Zn	Modification of coir fibre by oxidation with hydrogen peroxide resulted in an increase in metal adsorption (The modified coir fibres adsorbed 4.33, 7.88 and 7.49 mg g^{-1} Ni, Zn and Fe, respectively, against 2.51, 1.83 and 2.84 mg g^{-1} for the unmodified coir fibres).	[211]
Carbon from coconut coirpith	Aqueous solution	Cd	The adsorption capacity of activated carbon (250–500 μm) prepared from coirpith was 93.4 mg Cd g^{-1} at pH 5.0.	[212]
Citric acid treated soya bean straw	Aqueous solution	Cu	The maximum Cu adsorption capacity at pH 6 was 0.64 mmol g^{-1} for the base washed and citric acid modified soybean straw.	[213]
Hemp fibers	Aqueous solution	Pb, Cd, Zn	The maximum adsorption capacity of hemp fibers for Pb, Cd and Zn ions from single solutions was the same (78.0 mmol kg^{-1}) but varied in ternary mixture (74.0, 35.0 and 35.0 mmol kg^{-1}, respectively).	[214]
Coconut shell powder	Aqueous solution	Cd	The maximum adsorption capacity of coconut shell powder was 285.7 mg g^{-1} for Cd.	[215]
Olive stone waste	Aqueous solution	Pb, Ni, Cu, Cd	The maximum adsorption of metals by olive stone waste was 68.8, 44.7, 36.3 and 31.9 mmol kg^{-1} for Cd, Pb, Ni and Cu, respectively.	[216]
Cattle-manure-compost based activated carbons	Aqueous solution	Cu	The maximum adsorbed metals by cattle-manure-compost based activated carbons ranged 44.0–95.0 mmol kg^{-1} for Cu and 20.0–44.0 mmol kg^{-1} for Pb.	[217]

and Tl in acidic soils. In addition, scanning electron microscopy in back-scattered electron mode showed the formation of Al-hydroxy polymers which were associated with these three metal(loid)s, probably through direct coordination or the formation of ternary complexes.

6.2. Reduction

Most metal(loid)s are subject to abiotic and biotic redox reactions, which influence biogeochemical behaviour. However, redox reactions in soils are most important for As, Cr, Hg and Se. Organic amendments play a pivotal role especially in the reduction of these metal(loid)s by providing a source of electron donor and carbon substrate for microorganisms. The value of organic amendments in the reduction of metal(loid)s in relation to remediation of contaminated soil and water sources is presented in Table 4. It has often been noticed that addition of organic amendments such as manures and crop residues enhances the reduction of Cr and Se [231,233,234]. Various organic materials, such as powdered leaves [240] and *Pinus sylvestris* bark [241], seaweed [232], black carbon

Table 4
Selected references on the potential value of organic amendments in the redox reactions of metal(loid)s from soil and water.

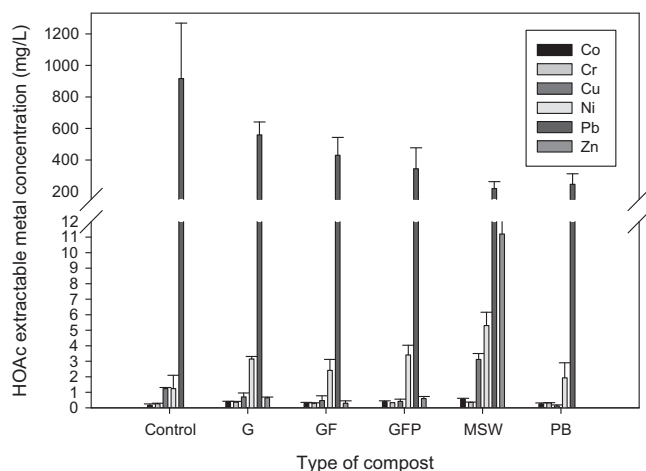| Amendments	Metal(loid)s	Substrate	Observations	References
biosolid compost, farm yard manure, fish manure, horse manure, spent mushroom, pig manure and poultry manure	Cr(VI)	Soil	Organic amendments enhanced the rate of reduction of Cr in the soil. There was a difference in the extent of Cr(VI) reduction among the soils treated with organic amendments, which was attributed to the difference in dissolved organic carbon in the soil.	[231]
Seaweed	Cr(VI)	Water	Protonated brown seaweed was effective in reducing Cr(VI) and reduction increased with a decrease in the pH of seaweed biomass. Electrons required for Cr(VI) reduction also oxidized the organic compounds in the seaweed biomass.	[232]
Black carbon (BC)	Cr(VI)	Water	Phenolic groups on surface and large surface area of BC were dominant drivers of Cr(VI) reduction and the resultant Cr(III) was bound to BC surface by surface complexation and precipitation.	[233]
Soybean meal and rice bran.	Cr(VI)	Soil	Soybean meal and rice bran reduced resin extractable Cr(VI) in soil, which was attributed to enhanced reduction of Cr because of more DOC and easily decomposed matter.	[234]
Composted cow manure	Cr(VI)	soil	Chromate leaching was reduced in soils in the presence of elevated organic matter because of reduction followed by retention on cation exchange sites or precipitation.	[235]
Carbon amendments- lactate, ethanol and acetate	Cr(VI)	Soil	Bacteria reduced most of Cr(VI) in anaerobic conditions and lactate was more effective as a bio stimulant for reduction be acting as an electron source.	[236]
Sewage sludge and poultry litter	As	Soil	The mixture of sewage sludge and poultry litter reduced. As(V) to more mobile and toxic As(III).	[237]
Casein amino acids	Se	Soil	Application of both insoluble (casein) and soluble (Casamino acids) organic amendments to Se contaminated soil enhanced dimethyl selenide production and subsequent removal of Se from soil through volatilization.	[238]
Rice straw	Se	water	Selenate is reduced to selenite in the presence of rice straw, as rice straw carries Se-reducing bacteria.	[239]

[233] have been used to reduce Cr(VI) from industrial effluents. Addition of biological waste materials, such as poultry and livestock manures, and biosolids has often been shown to increase the amount of dissolved organic carbon (DOC) in soils either by acting as a source of DOC or by enhancing the solubilization of the soil organic matter [242,243]. The easily oxidizable organic carbon and DOC fractions provide the energy source for the soil microorganisms involved in the reduction of metal(loid)s, such as Cr [234] and Se [244] and non-metals, such as N [245] and S [246].

While Cr(III) is strongly retained onto soil particles, Cr(VI) is very weakly adsorbed and is readily available for plant uptake and leaching to groundwater [247–249]. Thus, reduction of Cr(VI) to

Fig. 2. Acetic acid extractable Co, Cr, Cu, Pb and Zn concentration of soils treated with various organic amendments (G; green waste-derived compost, GF; green waste and catering waste-derived compost, GFP; green waste, catering waste and paper waste-derived compost, MSW; municipal solid waste-derived compost, PB; commercially available peat-based compost) [227].

Cr(III) can enhance the immobilization of Cr, thereby rendering it less bioavailable. Bolan et al. [231] investigated the effect of 7 organic amendments (biosolid compost, farm yard manure, fish manure, horse manure, spent mushroom, pig manure and poultry manure) on the reduction of Cr(VI) in a mineral soil low in organic matter content. Addition of organic amendments enhanced the rate of reduction of Cr(VI) to Cr(III) in the soil. At the same level of total organic carbon addition, there was a significant difference in the extent of Cr(VI) reduction among the soils treated with organic amendments. There was, however, a significant positive linear relationship between the extent of Cr(VI) reduction and the amount of DOC in the soil. Addition of the biosolid compost was also found to be effective in reducing the phytotoxicity of Cr(VI). The reduction of Cr(VI) by various organic amendments is presented in Fig. 3.

Arsenic exists in environment as As(VI) and As(III). Organic arsenic species (monomethylarsenic acid and dimethylarsenic acid) can also be present in environment, but tend to be found at lower concentrations [250]. Arsenite is of great concern because of its toxicity and high mobility [251]. Arsenate in soils and sediments can be reduced to As(III) by bacteria [252,253]. Since As(III) is less strongly retained than As(V) by inorganic soil components, microbial reduction results in the mobilization of As and becomes more toxic than As(V). Soil organic matter influences the reduction of As, converts As(VI) to As(III) in aerobic environments, thereby increasing the toxicity of As. In copper-chromium-arsenate contaminated soils, As(III) increases with an increase in soil organic matter [254].

Higher oxidation states of Se, Se(IV) and Se(VI) are toxic and the reduction of soluble Se(VI) and Se(IV) to the less toxic Se(0) converts Se into an insoluble mineral form. Selenite is less mobile than Se(VI) because the former is strongly adsorbed onto soil minerals and organic matter under near neutral pH conditions [255]. Soluble Se is not typically found under reducing conditions because less soluble forms such as Se(0) are thermodynamically favoured. When Se(IV) and Se(VI) are introduced into moderately reducing
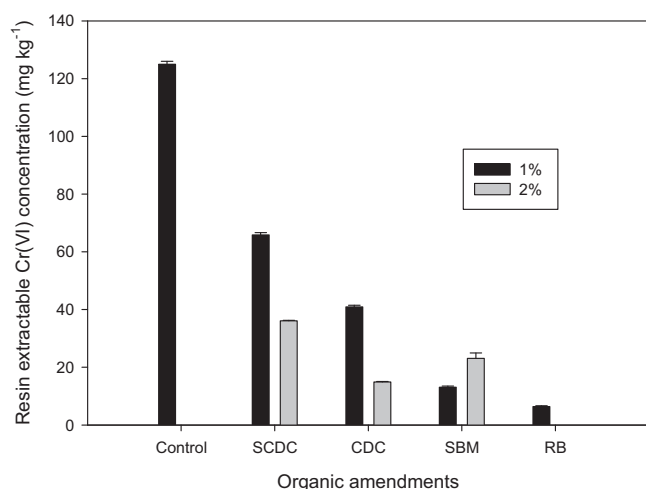
Fig. 3. The amounts of resin-extractable Cr(VI) in Cr(VI)-spiked soils (500 mg Cr(VI) kg^{-1} soil) at 8 days after amending with 1% or 2% organic matter (SCDC; sugarcane dregs compost, CDC; cattle-dung compost, SBM; soybean meal, RB; rice bran) [234].

Fig. 4. Influence of different organic amendments (2% or 5%) on Se volatilization by wheat (FM; farmyard manure, PM; press mud, POM; poultry manure, AL; Arhar leaves) [280].

conditions they are rather quickly transformed through microbial mediation to Se(0) and/or organic Se compounds [239].

In soils and sediments, reduction is largely mediated by microorganisms [256,257]. Microorganisms have been reported for the enzymatic reduction of metal(loid)s in the soils during the metabolism of organic matter. Chromium (VI) can be reduced anaerobically to less toxic and less mobile Cr(III) by *Shewanella oneidensis* [258] and *Pantoea agglomerans* [259]. Municipal solid waste increases microbial activity [260], thereby microorganisms can facilitate anaerobic reduction of metal(loid)s.

Anaerobic Se-respiring bacteria can use Se(VI) and Se(IV) as terminal electron acceptors and precipitate as Se(0). High organic matter in sediments promotes the reduction of Se(VI) and Se(IV) to less toxic Se(0) [261]. Siddique et al. [262] reported the reduction of Se(IV) and Se(VI) to insoluble Se(0) in drainage water in the presence of rice straw, which has been attributed to the presence of reducing bacterial communities. Reduction of Se species may be promoted by organic functional groups released from rice straw, which act as electron donors [263]. Waste by-products have been shown to assist in remediation of Se contaminated soils [264–266]. Crop residue application to Se contaminated soils was found to reduce Se accumulation in canola (*Brassica napus*) leaves [264]. Industrial by-products, such as ashes from combustion of biomass and red gypsum, reduced the mobility and availability of Se in soils [265].

Dissolved and natural organic matter associate very strongly with Hg, altering its speciation and bioavailability in aquatic and terrestrial environments [267]. Mercury forms strong bonds with humic substances and appears to stabilize Hg(II) in the environment [268]. Humic matter reduces Hg(II) to Hg(0) in the water [269]. Mercuric ions in the water reduce to Hg(0) mainly by photochemical reduction. In the presence of dissolved humic substances, the reduction rate depends on the intensity and wavelength of entering light [270]. Mercury forms strong covalent bonds with reduced sulfur functional groups in soil and this complexation facilitates the mobility of mercury from natural and contaminated soils and sediments [271,272]. Dissolved organic carbon stimulates microbial growth, thereby promoting formation of methyl mercury in some environments. By contrast, the presence of DOC may also inhibit methylation by reducing the amount of inorganic mercury available for methylation through complexation [273]. In bio-reduction of Hg, glucose is a more favourable substrate than acetate. In acetate culture, Hg forms complexes Hg(CH$_3$COO)$_4^-$ and
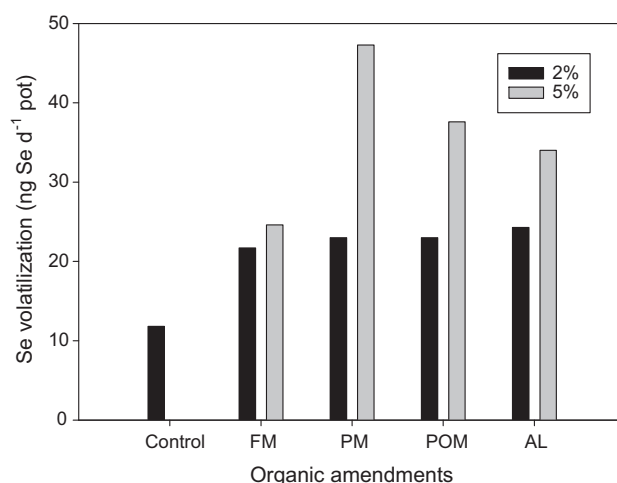
Hg(CH$_3$COO)$_3^{2-}$, so microbial cells are less likely to transport Hg through microbial cell membranes for reduction [274].

6.3. Volatilization

The majority of metal(loid)s cannot be volatilized from soil or waters. However, As, Hg and Se may be volatilized through reduction and methylation reactions. For example, bacterial reduction of Hg(II) is known to be transformed to gaseous Hg(0) and subsequently lost to the atmosphere [275–277]. Similarly, biological methylation is effective in forming volatile compounds of As such as alkylarsines, which could easily be lost to the atmosphere [112]. Volatile derivatives of As include arsines, mono-, di-, trimethylarsine and arsenic oxides. Volatile As compounds are more mobile, bioavailable and often considered more toxic in comparison to pentavalent arsenic species [112], but volatile methylated arsenic oxides are less toxic than As(III) [278] and can easily and rapidly be oxidized and demethylated in air [112]. Therefore, biovolatilization can offer an effective remediation technology for As contaminated soil or water. Similarly, Se undergoes biomethylation in soils and aquatic systems [244], thereby resulting in the release of gaseous methylated compounds. Soil microbes volatilize Se as dimethylselenide and dimethyldiselenide and it is an important part of the Se cycle in nature [279]. Selenium biomethylation is of interest because it represents a potential mechanism for the removal of Se from contaminated environments, and it is believed that methylated compounds, such as dimethyl selenide is less toxic than dissolved Se oxyanions.

Since microorganisms play a vital role both in the reduction and methylation reactions, addition of organic compounds has been shown to enhance the volatilization loss of metal(loid)s (Table 5). For example, Dhillon et al. [280] noticed that addition of organic amendments enhanced the volatilization of Se from soil. Addition of press mud, poultry manure, plant leaves and farm yard manure increased the volatilization of Se, thereby decreasing its uptake by maize (*Zea mays*) and cowpea (*Vigna unguiculata*) (Fig. 4). Similarly, Calderone et al. [266] demonstrated that the addition of organic amendments promoted volatilization of Se. Especially, the application of gluten stimulated volatilization of Se (1.7- to 3.2-fold over the control). Huysmans and Frankenberger [290] isolated *Penicillium* sp. from evaporation pond water which was capable of methylating and subsequently volatilizing organic As. They found that the addition of amino acids, tryptophan, leucine, valine, phenylalanine, isoleucine and glutamine promoted trimethylarsine

Table 5
Selected references on the potential value of organic amendments in the volatilization of metal(loid)s from soil, sediments and water.

Amendments	Metal(loid)s	Substrate	Observations	References
Press mud, poultry manure, plant leaves, farm yard manure	Se	Soil	Organic amendments increased the rate of Se volatilization in seleniferous soils by 1.8-4.0 times compared to control plots; the greatest increase was observed with press mud followed by poultry manure, arhar (*Cajanus cajan*) leaves and farmyard manure.	[280]
Organic C sources (carbohydrates and proteins)	Se	Agricultural evaporation ponds	Proteins are known to stimulate Se volatilization dramatically as compared with carbohydrates as the energy source. Protein amendments increased 10% Se loss after 43 days of incubation. Protein provides methyl groups to form volatile dimethylselenide.	[281]
Citrus (orange) peel, cattle manure, barley straw, grape pomance	Se	Sediment	Organic amendments increased the volatilization of Se. The most effective organic amendment was cattle manure with an average Se emission of 54 μg Se h^{-1} m^{-2} while the background emission of volatile Se averaged 3.0 μg Se h^{-1} m^{-2}. After 22 months, cattle manure treatment removed 57.8% of the total Se content.	[282]
Methionine, casein	Se	Sediment	Se volatilization rate without organic amendments was less than 25 μg m^{-2} d^{-1}. After amending the sediment with 71.4 mg methionine kg^{-1} soil, Se volatilization rates were 434 μg m^{-2} d^{-1} in vegetated plots with *Sporobolus airoides* and 289 μg m^{-2} d^{-1} in irrigated bare plots. With the amendment of 572 mg casein kg^{-1} soil, rates increased to 346 μg m^{-2} d^{-1} in irrigated bare plots and to 114 μg m^{-2} d^{-1} in vegetated plots.	[283]
Pickleweed shoot tissues	Se	Soil	The addition of pickleweed shoot tissues (approximately 1.5 kg m^{-2}) to the soil surface resulted in 2.2-fold increase in biogenic volatile Se (251.6 μg m^{-2} d^{-1}.) over control plot.	[284]
Casein, gluten	Se	Soil	Volatile dimethylselenide release was 2.1 and 2.6 times higher in the casein and gluten-amended soils, respectively compared to the control soil. In columns containing 1% casein or gluten in the top 5 cm of soil, the cumulative loss of dimethlyselenide via volatilization was about 9% higher than in unamended soil.	[285]
Yeast extract	Hg	Freshwater pond	About 30% and 70% of Hg(II) was lost through volatilization in incubations supplemented with 0.1 and 1 mg L^{-1} yeast extract, respectively. The rate of volatilization was related to level of nutrient amendment and consequently, stimulation of heterotrophic activity.	[286]
Cow dung	As	Sludge	Cow dung was used as a substrate for the bacterial growth during As volatilization by methanogenic bacteria. Cow dung addition (25 g L^{-1}) volatilized about 35% of As. The maximum As concentration which can be volatilized was 1.08 mg of As(V) g^{-1} substrate because higher substrate concentration decreased arsenic volatilization rate.	[287]
Compost	As	Soil	Arsenic loss up to 16% from the compost amended soils was attributed to microbially-mediated As volatilization in the chromated–copper–arsenate contaminated soil.	[288]
Cellulose	As	Soil	Arsenicals are subject to microbial reduction and methylation leading to volatilization as arsines. Cellulose addition enhanced arsine evolution.	[289]

production by 10.2 to 11.6-fold over the control without amino acid supplementation.

6.4. Rhizoshpere modification

Organic amendments have been shown to affect the chemistry (e.g., pH, organic acids, soil solution composition) and biology (e.g., microbial community) of soil. Similarly, it is being increasingly recognized that the soil immediately surrounding plant roots (rhizosphere) is a modified microbiologial and chemical environment due to plant–soil–microbe interactions. The changes in soil chemistry due to soil amendment and plant growth can therefore influence the transformation, mobility and bioavailability of metal(loid)s [184,291,292]. The effect of organic amendments on some of the rhizosphere properties in relation to metal(loid)s dynamics is presented in Table 6.

6.4.1. pH

Application of organic amendments such as biosolids and manures often decreases the pH of rhizosphere soils [292,293].

The rhizosphere acidification can be attributed to a combination of mechanisms, including: (i) cation–anion exchange balance; (ii) organic acid release (e.g., citric, malaeic, lactic, oxalic, propanic, butyric acids); (iii) root exudation and respiration and (iv) redox-coupled processes involving changes in the oxidation state of Fe, Mn and N and consuming or producing of H$^+$ [307]. In the rhizosphere, the various origins of H$^+$ released by roots in their immediate vicinity and the underlying physiological mechanisms involved are now well elucidated [136]. However, the contribution of some of these processes such as respiration, exudation of organic anions and redox-coupled processes in the rhizosphere needs to be further clarified. The contribution of organic acid exudation to the pH of soil solutions depends on the soil pH, the assemblage of organic acids, and to some extent the ionic strength of the soil solution [308]. Within the rhizosphere, soil acidification is mainly caused by an imbalance in the carbon and nitrogen cycles caused by uptake of cations-anions and respiration [309]. A major source of H$^+$ fluxes in the rhizosphere is related to the differential uptake of cations and anions by plant roots [307,310,311]. Uptake of solution NH$_4^+$ results in a loss of positive charge in

Table 6
Selected references on organic amendments-induced rhizosphere modifications in relation to heavy metal(loid)s remediation in soils.

Amendments	Mode of action	Observations	References
Sugar beet	pH	Mixed amendment of sugar beet, *Aspergillus niger* and rock phosphate significantly decreased pH in rhizosphere soil of *Cistus albidus*.	[292]
Biosolid	pH	The application of biosolids decreased pH of pore water of rhizosphere soil of *Lolium perenne*.	[293]
Humic acid	Organic acid	Application of humic acid increased bioavailability of various heavy metal(loid)s and enhanced their translocation from root to shoot.	[294]
Biosolids	Organic acid	Biosolids enhanced the organic acid production in the rhizosphere of *Zea mays* L.	[295]
Crop residues	Microbial activity	Soils amended with crop residues increased microbial diversity in wheat rhizosphere.	[296]
Food waste compost	Microbial activity and enzyme activity	Application of food waste compost significantly increased bacterial and fungal populations, soil enzyme activities in the rhizosphere of lettuce plant.	[297]
Soluble organic carbon	Microbial cativity	Structural manipulation of rhizobacterial communities can be mediated by artificial exudates in the form of soluble organic carbon.	[298]
Compost	Microbial activity	Addition of composts to soil increased the incidence in the tomato rhizosphere microbial population.	[299]
Sludge, wheat straw	Microbial activity	Addition of organic amendments significantly enhanced the microbial population in the rhizosphere of wheat plants.	[300]
Sewage sludge	Enzyme activity	The long-term application of sewage sludge significantly increased phosphatase activity in the rhizosphere of rape and wheat plants.	[301]
Municipal solid waste	Enzyme activity	Amendments of municipal solid waste increased the contents of organic C and enzyme activities in *Beta vulgaris* and *Triticum turgidum*.	[302]
Humic acid	Enzyme activity	Application of humic acids increased plasma membrane H^+-ATPase activity in the root of maize (*Zea mays*) seedlings	[303]
Municipal solid waste	Enzyme activity	Application of municipal solid waste compost increased protease hydrolyzing casein, β-glucosidase and dehydrogenase activities in the rhizosphere of plant.	[304]
Sewage sludge	Enzyme activity	Application of sewage sludge increased urease activity (16–43%) in the rhizosphere of Ransom soybean.	[305]
Composted organic residue	Enzyme activity	The nutrient content (NPK) and enzymatic activities (dehydrogenase, urease, protease-BAA, acid phosphatase and β-glucosidase) increased in the rhizosphere soil with the organic amendment.	[306]

solution, which is counter-balanced by a corresponding release of H^+ into the rhizosphere [312]. Apart from this, N transformation and nitrate leaching have been suggested to be major causes of soil acidification [313]. The crop type also plays a major role in rhizosphere acidification. Nitrogen fixing plants tend to decrease soil pH more than non-N fixing plants, since the alkaline release does not occur significantly as NO_3^- is not required. Tang [314] reported that the lupin (*Lupinus angustifolius*) and subterranean clover (*Trifolium subterraneum*) without receiving nitrogen fertilizer produced approximately 1.4 mmol H^+ per kg soil over the experimental period, on average. The impact of plant on the rhizosphere also varies temporally, and at different points along the plant root [312].

In the soil environment, metal uptake by plants increases with decreasing pH, despite it is known to decrease with decreasing pH in nutrient culture [315,316]. For example, higher Cd uptake was obtained for lettuce (*Lactuca sativa*) and Swiss chard (*Beta vulgaris*) grown on acid soils (pH 4.8 to 5.7) than on calcareous soils (pH 7.4 to 7.8) [317]. Khan and Jones [318] showed that the addition of lime resulted in the largest reduction in metal extractability with diethylene triamine pentaacetic acid and $Ca(NO_3)_2$ and phytoavailability of Cu, Fe and Zn.

Acidification affects the leaching and residence time of many metal(loid)s in soil. Tyler [319] observed that the amount of metal(loid)s released from the mor soils of Sweden increased with decreasing pH. Approximately 85% of the total Cd was released from the soil at pH 2.8. The time needed for a 10% decrease in the total concentration of Cd in the mor horizon through leaching was estimated to be 1.7 years at a pH of 2.8, 4-5 years at a pH of 3.2, and 20 years at a pH of 4.2.

6.4.2. Organic acids

'Rhizodeposition' describes the introduction of C compounds and nutrients by plant roots. This generally includes water-soluble exudates, secretions, lysates, gases and mucilage [320]. Organic compounds such as carbohydrates, carboxylic acids and amino acids in rhizodeposits attract microorganisms and therefore are most responsible for additional microbial growth within the rhizosphere [321]. In the rhizosphere, the exudation of C-compounds has been the major research focus for its major impact on soil microbiology and C biogeochemistry [322], particularly the role the rhizosphere may play in global C cycling [307]. Exudation of organic compounds can influence the behavior of trace and toxic metals [323]. Certain low molecular-weight compounds released from plant roots (amino acids, carboxylic acids, sugars, and simple and flavonoid-type phenolics) [324] may form stable complexes with metal cations in the soil solution matrix, and can directly modify metal(loid) availability in the rhizosphere [308]. The role of carboxylates has been examined for their potential impact on the biogeochemistry of metals through aqueous complexation and lig- and exchange processes. Amongst the range of carboxylates exuded in the rhizosphere, malate, citrate and oxalate are expected to have the most dramatic effect due to their ability for complexation of metal(loid)s [325].

Organic exudates are released from the roots when the plant is under stress from mineral deficiency and toxicity [308,312]. Iron deficiency has been known to induce root exudation of a range of organic compounds to improve Fe acquisition in soils [308,326,327]. Exudation of siderophores, another organic exudate, is an additional strategy utilized by plants under Fe deficiency [328]. Phosphorus deficiency is also well known to result in organic anion exudation in a range of plant species [308,312,329,330]. The release of organic acids such as oxalate, malate and piscidate is hypothesized to increase P sequestration from mineral surfaces; however, organic exudates production does not necessarily result in greater P uptake [330]. Root exudate production has also been shown in response to metal(loid) exposure in soil solution [308,331,332]. The exudation of organic acids by plants or associated microbes may complex metal(loid)s and reduce uptake, although increased

uptake is more commonly observed with artificial introduction of organic acids [333].

The promotion of root growth by improvement of soil physical, chemical and biological conditions is also likely to enhance the further modification of metal(loid) behaviour in the rhizosphere [334]. Koo et al. [295] found the total organic acid concentration was highest when biosolid and plant growth was combined, rather than the single treatment. The biosolid provided a baseline of a wide range of organic acids, although the organic acid composition changed considerably with the addition of plants. However, the combined effect was greater than the single roles, possibly due to additional root growth. The interaction of organic amendments with the rhizosphere depends on the nutrient content, heavy metal(loid) availability and the ability of plant species to tolerate heavy metal(loid) exposure, however, limited data exists to date.

Puglisi et al. [335] evaluated the rhizosphere C deposition in maize plants after soil treatment with different organic materials. Available C compounds in the rhizosphere were assessed with the *lux*-marked biosensor *Pseudomonas fluorescens*. Organic amendments (compost, compost water extract, the hydrophobic and hydrophilic fractions of the DOC extracted from the compost) increased available C compounds in the rhizosphere soils amended. Organic amendments enhanced organic C release by maize plant roots. Similar observations were made by Caravaca et al. [336], who reported an increased rhizodeposition in *Olea europaea* subsp. *sylvestris* and *Rhamnus lycioides* treated with composted residue. Caravaca et al. [292] demonstrated that sugar beet-residue amendment increased the total carbohydrates and soluble C-fraction (water-soluble C and water-soluble carbohydrates) in the rhizosphere of *Cistus albidus* L. and *Quercus coccifera* L.

6.4.3. Microbial community

Since rhizosphere microbial communities are strongly influenced by root exudates [337], it has been hypothesized that plants select for beneficial microbial communities in their rhizosphere [338].

The recent scientific evidence indicates that, the higher soluble concentrations of Cu, Zn and Pb in planted compared to non-planted control treatments may be related to the formation of soluble complexes between these elements and organic compounds exuded from growing roots and/or microbial associations. The higher rates of volatile Se produced from the vegetated plots with added methionine compared to bare irrigated plots have been attributed to additional microbial activity associated with plant roots [283]. In this regard, it is well known that plants produce root exudates containing organic acid anions, sugars, vitamins, amino acids, inorganic ions, and some enzymes, which likely have substantial impacts on microbial population development and biological activity. Hence, plant roots and soil microbes and their interaction can enhance metal bioavailability in rhizosphere.

Some prokaryotic (bacteria, archaea) and eukaryotic (algae, fungi) microorganisms can produce or excrete extracellular polymeric substances (EPS), such as polysaccharides, glucoprotein, siderophores, lipopolysaccharide, soluble peptide etc. These substances possess a substantial quantity of functional groups which can coordinate with metal(loid) ions. A number of microbes are involved in EPS production viz., *Bacillus megaterium*, *Acinetobacter*, *Pseudomonas aeruginosa*, SRB and Cyanobateria [339]. Extracellular polymeric substance excretion studies have been limited in cast of soil fungi and algae [340–344]. The cell wall of microbes also plays a major role in metal adsorption/reduction [345]. Metal(loid)s are adsorbed by various functional groups of the cell wall, including phosphate, carboxyl, amine as well as phosphodiester groups. In more recent times, complexation, ion exchange, adsorption (by electrostatic interaction or van der Waals force), inorganic precip-

itation, oxidation and/or reduction have been proposed to explain metal uptake by organism [346,347].

In metal(loid) contaminated soil, the poor performance of plant growth and root development are major limiting factors for phytoaccumulation of metal(loid)s. To overcome these problems, improvement of the microbial activity in rhizosphere in addition of organic amendments is necessary. Plant growth promotion mechanisms by indole acetic acid (IAA), siderophores and 1-aminocyclopropane-1-carboxylate (ACC) deaminase producing microbes in the presence of metal are not well known. Most of recent studies assume that demonstrating the presence of all or even some of these activities is sufficient to elaborate the mechanisms. These findings suggest that (i) IAA promotes plant growth *per se* [348], (ii) ACC deaminase prevents the stress by-product ethylene which inhibits plant growth [349] and (iii) siderophores help plants to acquire sufficient Fe in the presence of overwhelming amounts of other (potentially competing) metal(loid)s [350]. The Ni resistant ACC deaminase-containing plant growth-promoting bacterium could decrease the toxicity of Ni to canola plants [351], there have been a large number of reports of facilitation of metal(loid) phytoextraction through the addition of plant growth-promoting bacteria and fungi. A number of scientific evidences proved that the inoculation of plant growth-promoting bacteria facilitated plant growth and increased the uptake of metal(loid) by the plant [352–355].

In soil, heavy metal(loid)s can have long-term toxic effects within ecosystems [356] and have a negative influence on biologically mediated soil processes [357]. It is generally accepted that accumulation of metal(loids) reduces the amount of soil microbial biomass [358] and various enzyme activities, leading to a decrease in the functional diversity in the soil ecosystem [359] and changes in the microbial community structure [360]. However, metal(loid) exposure may also lead to the development of metal(loid) tolerant microbial populations [361]. For example, many microbes have been reported to reduce Cr(VI) under aerobic and anaerobic condition. Bio-reduction of Cr(VI) can be directly achieved as a result of microbial metabolism [362] or indirectly achieved by a bacterial metabolites such as H_2S [363].

The addition of organic amendments has been reported to enhance proliferation of microorganisms. The application of a range of organic amendments (biosolids, spent mushroom compost, green waste compost and green waste-derived biochar) to bauxite-processing residue sand decreased bulk density and increased mesoporosity, available water holding capacity and water retention. The addition of these amendmens increased soluble organic C, microbial biomass C, basal respiration and the activities of β-glucosidase, L-asparaginase and alkali phosphatase enzymes [364]. The increased porosity may increase oxygen content and diffusion, which enhances microbial activity in soil. The addition of organic amendments, lucerne or dynamic lifter® pellets resulted in an increase of the macroporosity from <10% to >18%, together with reductions in bulk density, and a 50-fold increase in saturated hydraulic conductivity in soil, thereby affecting metal(loid)s dynamics in soils [365].

Chicken litter/leaf compost and dairy cow manure/leaf compost enhanced spore populations of vesicular-arbuscular mycorrhizal fungus relative to those found in crop plots treated with raw dairy cow manure and conventional fertilizer [366]. The beneficial role of organic matter may be related to an improvement of physical properties, such as increased soil aggregate stability, and/or to an increase in microbial activity [367]. Acea and Carballas [368] demonstrated that wheat straw or poultry manure neither inhibited microbial proliferation nor changed the order of the sizes of the various subgroups. However, microbial response was different between treatments. In regard to heterotrophic microbes, amendment with wheat straw increased counts of fungal propag-

ules and hyphae length, while bacteria, particularly actinomycetes and ammonifiers, were lowered. On the contrary, poultry manure treatment favoured bacterial, actinomycetal, ammonifier and fungal mycelium development and propagule generation.

7. Efficacy of organic amendments for bioremediation

Land application of municipal and industrial by-products has been repeatedly shown to increase the vegetative productivity of soils in agricultural and land reclamation settings [161]. Indeed, animal manures have for a very long time been known to improve soil physical, chemical and biological attributes. More recently, plant growth has been shown to be improved by a large diversity of organic wastes, including biosolids, MSW, pulp-mill by-products [146] and in certain cases, industrial wastes [146].

Organic amendments have been particularly beneficial to plant growth and microbial productivity when applied to acidic and nutrient-poor soils [369]. For example, Stephen and Lin [370] reported compost material increased the yield of Chinese white cabbage (*Brassica rapa* L.) only in nutrient-poor soils of Hong-Kong, and not productive soils. Similarly, Sloan and Basta [371] showed that use of alkaline stabilized biosolids increased soil pH of acidic soils, reducing Al toxicity in plants. Biosolids without alkali treatment however do not appear to be an efficient liming agent, particularly in extreme acid conditions found at mine sites and acid sulfate soils [166]. Contaminated soils are difficult to establish plant growth on due to toxicity issues, but also due to unfertile, acidic or saline conditions. Established landfill sites and mine sites are perhaps the best examples. Organic amendments have also been shown to enhance productivity on alkaline, saline and calcareous soils [372–375]. Organic amendments supply micronutrients particularly biosolids and MSW, and often possess moderate to high pH buffering capacity [376–378].

Brown et al. [379] combined biosolids with different combinations and amounts of Fe, in the form of Fe oxides, triple superphosphate (TSP) and phosphoric acid to immobilize soil Pb from a smelter contaminated soil in the US. For lettuce, the greatest reductions in Pb uptake were observed in the 3.2% TSP and 1% phosphoric acid. Combinations of biosolid, Fe and P-sources did not show a significant role. However, the gastric bioavailability and bioaccessibility showed good reductions from all treatments. In the rat feeding study component of Brown et al. [379], it was reported that 10% composted biosolid, Fe + 1%TSP, and 1% phosphoric acid addition reduced gastrointestinal bioavailability of Pb by 26, 39 and 26%, respectively.

Reductions in Pb bioavailability have shown particular promise with soil amendments. Ryan et al. [380] reported successful reductions in Pb bioavailability in a swine model with phosphoric acid in a field trial. Farfel et al. [381] amended urban backyards contaminated by Pb-based paint chips with biosolids. Results importantly showed that grass coverage was significantly improved by biosolid addition, thereby reducing exposure and child accessibility to soil-Pb. Secondly, it was also shown that *in vitro* bioaccessibility (pH 2.2) was also reduced. Thus field and laboratory trials have shown that organic and inorganic amendments (see also [382,383]) can reduce gastrointestinal bioavailability of Pb. Similar reductions in gastrointestinal bioavailability for other metal(loid) contaminants such as Cd and As are still lacking.

Despite the successes reported, the long-term potential of organic by-products for metal(oid) immobilizing material, however, is often questioned. Organic by-products from municipal and industrial waste often contains environmental contaminants such as N, P, organic contaminants and metal(loid)s. An additional problem is the ability of organic by-products to immobilize metal(loid) contaminants in the long-term, since over time the organic material

will decompose [384]. It has been proposed repeatedly in the literature that upon degradation of organic material the immobilized inorganic contaminants will be remobilized to other environmental compartments [163,384–386].

The rate of organic amendment degradation depends on the source and treatment of material prior to application to land [387,388]. Composting is a common method used to reduce the easily degradable fractions, such as carbohydrates, proteineceous materials, and increase the extent of humification [387,389]. The extent of humification has been shown to influence the long-term stability of organic wastes [387,389,390], and in some cases, metal sorption [391,392].

Mobilization of heavy metal(loid)s by chelation with organic ligands and subsequent leaching to groundwater supplies is of major concern. Additions of organic amendments have repeatedly been shown to increase metal leaching losses from agricultural settings with application of biosolids [393,394]. Colloidal transport down profile may be a significant transport mechanism in organic amended soils; however, this is also a concern with some inorganic immobilization agents [395].

Similarly, on metal(loid) contaminated soils organic amendments have increased leaching compared to the non-amended soil [227,396]. Schwab et al. [396] studied the effect of aged cattle manure, composted cattle manure and composted garden waste on Zn, Cd and Pb leaching from mine tailings (initial pH 7.4). Results differed between metal(loid)s. In the case of Cd, there was no difference observed between control and any organic amendments after 20 d leaching. For Pb, the aged cattle manure increased Pb leachate concentrations by approximately 6-fold. Whilst for Zn, all organic amendments increased leachate Zn concentrations relative to the control, especially over the initial 10 d. Similarly, Businelli et al. [397] reported that addition of biosolids to an Italian landfill enhanced Cu, Zn and Pb concentrations in leachate from the liner, and Farrell et al. [227] found enhanced initial leaching losses when an acidic contaminated soil was amended with both MSW and garden waste compost. Other examples of enhanced leaching losses of metal(loid)s from organic amendments have been reported [393,394]. Increased DOC concentrations associated with organic amendment are likely to be associated with leaching losses [28,384] due to complexation and chelation of heavy metal(loid)s, especially Cu (see Fig. 5). Nutrient leaching losses from organic soil amendments are also an issue, particularly in the case of N and P from animal manures [168,169,398], and less so for biosolids [393].

Thus numerous studies have shown that despite a reduction in metal(loid) uptake of biota, leaching of contaminants off site is a serious issue with organic amendments. However, enhancement of leaching of metal(loid)s from contaminated soils is often observed in the short term [227], or may not cause an increase of metal contaminants above accepted concentrations for subsurface and surface waters [394,396]. There is limited evidence that supports the 'time bomb' hypothesis for biosolids and other organic amendments. In fact, increasing evidence suggests that whilst decomposition of organic matter in biosolids occurs over long-time frames, limited metal(loid) appears to be released [399–403]. For example, Li et al. [221] obtained evidence for enhanced affinity for Cd adsorption by the inorganic components of the biosolid-amended soils, indicating that the biosolid-induced increase in Cd adsorption is independent of the added organic matter and persists for a long period after biosolid application. The lack of mobilization of metal(loid)s with organic matter decomposition appears to be related to inorganic minerals imbedded in the substrate. Hettiarachchi et al. [382] similarly found that Fe and Al oxide fractions contributed significantly to the sorption of Cd in biosolid-amended soils. Merrington and Smernik [404] reported increased Cd sorption at 2 year old biosolid amended soil, compared to 1 yr. In addition, Hettiarachchi et al. [402] used micro X-ray fluorescence (XRF) and
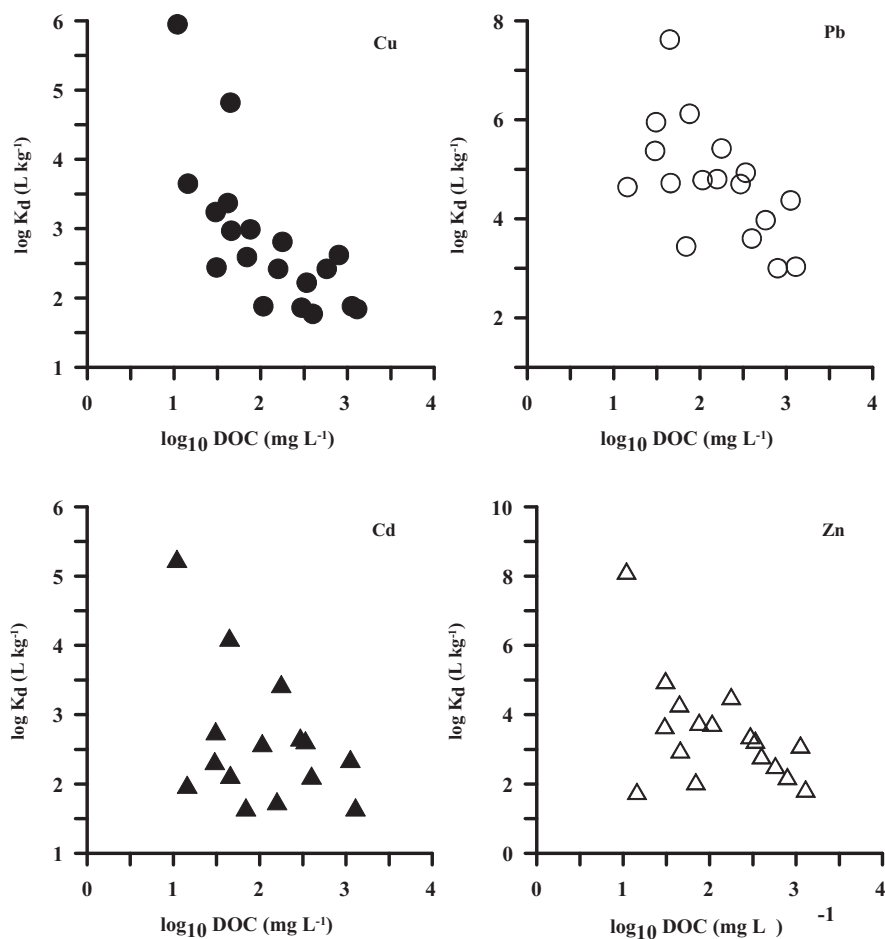
Fig. 5. Regression relationships between dissolved organic carbon (DOC) and partitioning coefficient (K_d L kg^{-1}).

micro X-ray Absorption Near Edge Structure (XANES) spectroscopy to investigate associations of Cd, Cr, Cu, Pb and Zn. These authors found that Cd, Cr, Pb and Zn were closely correlated to Fe, suggesting an association with Fe-rich minerals. The exception was for Cu, which was found to change once organic matter was removed, indicating Cu was predominantly associated with organic matter. By contrast, McBride et al. [405] suggested that biosolids high in Fe and Al rich mineral phases was not effective in increasing Cd sorption in 20 year old biosolid amended soils. Although there is evidence to suggest long-term stability of biosolids, the importance of 'tailor-made' organic amendments with other materials such as P or Fe rich materials is therefore likely to improve their environmental impact in the field.

After application of organic amendments, soils are often tested for toxicity to demonstrate that soils do not pose risk to human or animals. The most commonly used toxicity test methods are earthworm ecotoxicity test, plant growth and bacterial toxicity test. Herwijnen et al. [406] amended metal contaminated soil with composts and a liming product containing organic matter and tested for toxicity reduction capability. They conducted a plant growth test with Greek cress (*Lepidium sativum*), an earthworm (*Eisenia fetida*) survival and condition test and a bacterial toxicity test using *Vibrio fischeri*. Toxicity tests showed significant reduction of metal bioavailability and toxicity for Greek cress, earthworms and bacteria. Alvarenga et al. [407] amended soil with sewage sludge, municipal solid waste compost, and garden waste compost as immobilizing agents in aided phytostabilization of an acid metal-contaminated soil affected by mining activities. They tested the effects of the treatments on soil phytotoxicity and enzymatic

activities. Application of sewage sludge led to the greatest values of dehydrogenase, acid phosphatase, β-glucosidase, protease and urease activities, corresponding to the greatest overall microbial and biochemical activity in amended soils.

8. Conclusions and future research needs

Regular application of organic amendments such as biosolid and manure composts to agricultural soils improves the physical, chemical and biological fertility of soils. However, traditionally these organic waste products have been considered as a major source of metal(loid) input to agricultural soils. With the introduction of advanced wastewater treatment technologies and improvements in feed utilization in animal and poultry industries, the metal(loid) content of these waste products continues to decrease. Hence organic amendments that are low in metal(loid)s can be effectively utilized to remediate soils contaminated with toxic metal(loid)s. Application of organic amendments reduces the bioavailability of metal(loid)s through adsorption and complexation reactions, thereby reducing their transfer through plant uptake and leaching. These amendments also enhance the reduction of metal(loid)s such as Se and Hg, thereby resulting in the release of volatile compounds.

Because of the ever-increasing number of highly metal(loid)-contaminated sites worldwide, concomitant with their economically-prohibitive remediation utilizing modern engineering-type techniques (e.g., soil washing/flushing, electrokinetics, etc.), other more ecologically friendly approaches are now technologically viable. The use of green plants has now been

demonstrated under field scale to offer some promise in phytoremediation. Crucial to an effective phytoremediation strategy is the role of the rhizosphere and their associated microbial assemblages. Root exudates influence the structure and functions of microbial assemblages, which in turn mediate the various biochemical transformations in the root zone, including redox reactions and chemical speciation. In essence, rhizosphere processes play a key role in the transformation, mobility and eventual uptake of nutrients and metal(loid)s contaminants. These rhizosphere processes can be enhanced through organic amendments, thereby achieving natural remediation of metal(loid)s.

However, a major inherent problem associated with the use of organic amendments for the immobilization of metal(loid)s is that although they become less bioavailable, their total concentration in soils remains unchanged. The immobilized metal(loid)s may become plant available with time through natural weathering process or breakdown of organic matter–metal(loid) complexes.

Given the current knowledge on the value of organic amendments in the remediation of metal(loid) contaminated soils, the following research areas could be pursued:

– Impact of organic amendments, especially in the form of nano-material on the chemodynamics of metal(loid)s and their subsequent bioavailability.
– Effect of organic amendments on rhizosphere biochemistry in relation to metal(loid)s dynamics.
– Long-term stability and biogeochemistry of metal(loid)s immobilized by organic amendments.
– Nature and extent of soil mineral-organic matter-microbe interactions as influenced by environmental and edaphic factors.
– Nature of organic matter nano particles and colloids in metal(loid) interactions in terrestrial and aquatic ecosystems.
– Nature of microbial communities as affected by organic amendments dynamics and their role on the remediation of metal(loid)s.
– improved analytical methods to characterize specific compounds or functional groups in organic matter in relation to metal(loid) interactions.
– Mechanistic understanding of the interactions between organic amendments and soil components (e.g., metal oxide surfaces) as a means to predict co-transport of metal(loid)s associated with organic amendment components such as dissolved organic matter.

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