



Heavy Metal Contamination in Soil: Sources, Accumulation in Vegetables and Remedial Measures: A Review

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Abstract: Industrialisation and mechanisation create a lot of heavy metal pollution around the globe. Both anthropogenic and natural sources are responsible for the discharge of heavy metals into the environment. Anyhow, these toxic metals reach into soil, water bodies, plants and finally to human beings through the food chain. These toxic metals create several problems in plants and living beings after intake from the soil and accumulate in their bodies. Heavy metals also exhibit toxic effects on soil biological activities by affecting key microbial processes and hamper soil microbes' activities. Due to industrial development in urban areas, heavy metal contamination has become a severe threat to peri-urban agriculture prevalent for vegetable production. There has long been a need for decontamination of these agricultural resources and prevent further contamination from averting the adverse effects on living beings. In this article, an attempt has been made to provide an extensive understanding of different sources of heavy metal, such as zinc (Zn), copper (Cu), lead (Pb) and cadmium (Cd) *etc.*, in agro-ecosystem and their possible risks to soil and plants. This review has also made an effort to present brief information on remediation techniques, especially phytoremediation.

Key words: Heavy metal, pollution, soil, food chain, living beings, remediation

Introduction

Agricultural lands face the severe menace of deterioration due to unrelenting population pressure, urbanisation, and excessive utilisation of water resources. Globally, land degradation has several economic, social, ecological and environmental consequences. These adverse effects are directly or indirectly related to soil fertility and agricultural productivity, *i.e.*, soil erosion, waterlogging, reduced plant growth and changes in soil biodiversity (NRSC 2014; Datta and Young 2005). The data revealed that land degradation constituting 75% of the earth's usable landmass affect 4 billion people around the globe

(Barbier *et al.* 2018). Presently, heavy metal contamination is one of the most urgent environmental issues globally (Gupta *et al.* 2019). Heavy metals are omnipresent in the environment. It has crossed the permissible limits due to natural/native and anthropological sources (human-induced) and processes (Ratul *et al.* 2018; Kumar *et al.* 2019). Duffus (2002) reported that the density of a "heavy" metal ranges from 3.5 to 7 g cm⁻³. 'Heavy metal' is a popularly used and widely recognised term for a large group of elements with a density greater than 5 g cm⁻³. Heavy metal pollution of soil, water, and plants (crops, vegetables and trees) is one of the prevailing critical environmental predicaments in developing countries, including India, because of their toxicity, bio-magnification capacity, and non-

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biodegradability (Lu *et al.* 2011). Heavy metals may enter the food chain through plants, and therefore, humans can also be exposed to them (Intawongse and Dean 2006). It can impair the health of living beings through several absorption pathways such as direct ingestion, dermal contact, diet through the soil-food chain, inhalation and oral intake (Yu *et al.* 2018). Heavy metals are widely distributed in the environment and are considered significant chemical food contaminants. Heavy metals include both the elements, *i.e.*, essential for normal metabolic processes, called micronutrients (Fe, Mn, Cu, Zn, Mo), which in excessive quantities are more harmful to plants than to animal bodies, as well as elements such as As, Hg, Pb and Cd, which is harmful in low concentrations. Solubility, distribution and mobility of metal depend on pH, Eh (Redox potential), dissolved oxygen (DO), concentration and type of ligands and chelating agents. The problem is rising severely in industrial towns and rapidly developing cities. In the periphery of cities, cultivated soils are polluted with deposition or discharge of hazardous heavy metals, thereby increasing the levels of hazardous substances in food chains.

As stated earlier, heavy metals that accumulate in soil significantly contribute to the contamination of crops and indirectly become the leading component of

health hazards (Sawut *et al.* 2018). Vegetables are an everyday diet taken by populations worldwide due to their richness in vitamins, minerals, fibres, and anti-oxidative effects. Heavy metal pollution is one of the problems that arise due to the increased use of fertilisers and other chemicals to meet food production demands for human consumption. The tradition of growing vegetables within and at the edges of cities is ancient (Smith 1996). These cultivated lands are contaminated with heavy metals mainly through vehicular emissions, pesticides and fertilisers, industrial effluents, sewage sludge and other anthropogenic activities. The amount of heavy metals contamination differ from one location to another as the application of fertilisers and pesticides, effluents, and other human activities differ at each location. The present review is a compilation of different aspects of toxic heavy metal contamination in soil, its transport through the food chain and remediation techniques. Additionally, the review highlights the maximum allowable limits of potentially toxic metals in soils, water and vegetation.

Sources of heavy metals

Agro-ecosystem receives metals from natural and anthropogenic sources. Weathering of rocks, erosion and volcanic activity are the most important natural

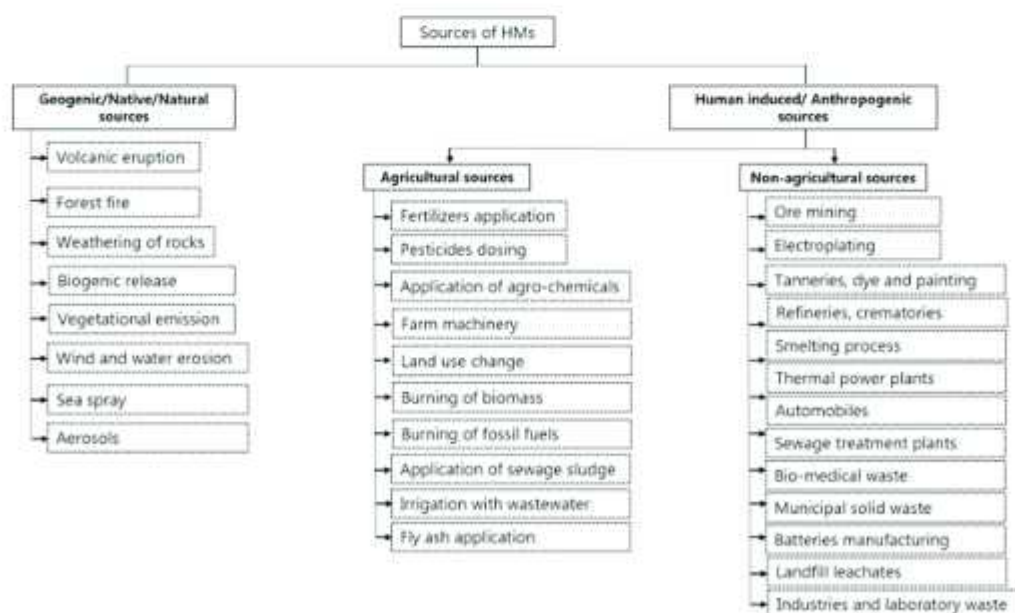


Fig. 1. Natural and anthropogenic sources of heavy metals (Kumar *et al.* 2019)

sources whereas, ore mining operations, smelting, electroplating, industrial effluents, landfills, use of pesticides and phosphate fertiliser, and atmospheric deposition are the vital anthropogenic sources of heavy metal contamination in agricultural soil (Herrero *et al.* 2019). Mining operations, crushing, collecting ores, mineral refining and purification, and tailings processing are evident sources of environmental pollution, *i.e.*, soil, water and vegetation, along with the discharge of wastewater from mining (Fig. 1.). Generally, the trace amount of element releases in the environment by weathering rocks over an extended period, but rapid industrialisation also adds a massive amount of these metals in the environment day by day. The composition of parent material and the pedogenic processes are the primary factors responsible for the metal content in soils.

Heavy metal contamination in the soil environment

Soil heavy metal pollution is a universal problem for food production and environmental health (Liu *et al.* 2018). Soils from more than 10 million sites world-wide have been confirmed to be contaminated, with more than 50 per cent of such sites contaminated with toxic metals (USEPA 2014). Extensive studies on heavy metal contamination of urban and semi-urban regions were reported widely. Li *et al.* (2014) identified heavy metal pollution sources of soils primarily from anthropogenic sources such as agriculture, urbanisation, municipal waste and industrial development. In addition, they have found that the leaching of heavy metals led to a degradation of food safety, thus causing hazards to the environmental ecosystem. Long-term application of sewage and industrial wastewater results in the build-up of heavy metals in the environment, reducing soil functionality and nutrient consistency, product toxicity and food chain degradation (Karthikeyan and Singh 2004).

Compared with the reference soils, the concentration of heavy metals such as Cd, Cr, Ni, Pb and Zn in the sewage irrigated soils showed significantly high values ($P < 0.05$), which suggested a substantial build-up of irrigated soils, especially Cr, Ni and Zn. (Chen *et al.* 2010). Shi *et al.* (2020) studied the effect of

long-term utilisation of E-waste dismantling activities on the heavy metals pollution in soils of rice-growing areas of southeastern China. This research determined the concentration of heavy metals such as Cd, Cu, Ni, Pb and Zn to assess the temporal trends over the past decade. From 2006 to 2016, the averages values of Cd, Cu, Ni and Zn in paddy soils were increased by 0.11, 11.81, 1.01 and 6.82 mg kg⁻¹, respectively. The average concentration of Pb in soils decreased by 14.06 mg kg⁻¹.

Al-Wabel *et al.* (2017) studied the accumulation of heavy metals in the agricultural soils of Saudi Arabia. They observed that the application of synthetic fertilisers, pesticides, and manures were the primary sources for its accumulation. The results of principal component analysis combined with correlation matrix suggested that Fe, Mn, Zn, Cu, Cr Ni, Cu, and Co represent natural abundance in soil. However, Cd, Pb, and Cu are man-made inputs, mainly due to pesticides and fertiliser applications. Wang *et al.* (2020a) analysed the influence of industrialisation and urbanisation on agricultural soil quality in Jiangsu Province, China. The level of Cd, Pb, Cr, Cu, Zn, Hg, and As contents in soil were measured. A decreasing trend of heavy metal contamination was observed from south to north, consistent with the economic development gradient. The spatial heterogeneity of heavy metal contamination was mainly influenced by industrialisation development. Cr, Cu, Zn, and As were affected by geogenic and man-made sources, while Cd and Pb were mainly affected by the latter. The Hg was mainly derived from industrial activities such as petrochemical production. Wang *et al.* (2020b) reported that in the vicinity of the petrochemical industrial area, China, the mean contents of Hg, Cd, As, Pb, Ni and Cu were 0.18, 0.69, 16.22, 47.24, 31.62 and 93.06 mg kg⁻¹, respectively. The spatial distribution of heavy metals in the surface samples was influenced mainly due to the industrial activities during the petroleum refining and trans-shipment process. Nickel was the primary pollutant in the petroleum refining process.

Hu *et al.* (2019) stated that the significant increase in heavy metals in the agricultural soils of Jiangxi Province's, China was due to the massive use of chemical fertilisers, manures, and agricultural waste, which was degrading the soil quality and also damaging

the terrestrial ecosystem. They also pointed out that heavy metals had shown mainly a negative impact on people's health due to contaminated food products. They explained that soil Hg level mainly originated from municipal activities, which accounted for 75.3% of the total sources. The primary sources of Ni in soil were municipal activities, agricultural activities, and industrial and mining activities, which account for 38.2, 27.5, and 25.1% of the total sources, respectively. Soil Cu was derived predominantly by agricultural activities (36.6%), followed by municipal activities (29.8%), and manufacturing and mining activities (25.8%). Yang *et al.* (2018) found that Cu, Pb, Cd, and As concentrations in soil and vegetables were higher in the mine-affected area than in the reference area (far away from the mine area). They reported potential safety hazards for the communities close to the mining field.

Zheng *et al.* (2016) observed that wetland conversion to the forest had caused noticeable losses of all the heavy metal assessed. Higher concentrations of Cr, Zn, Cu, Ni and As were found in rice cultivation and dryland agriculture with frequent cultivation compared to forest land, minimalist by human activities. Chaoua *et al.* (2019) have been involved in highly toxic metals such as Zn, Cu, Pb and Cd in water, agricultural soils and crops and their possible harm to human health in the Marrakech region. Heavy metals like Zn (112.71 mg kg⁻¹), Cu (17.70 mg kg⁻¹), Pb (57.36 mg kg⁻¹) and Cd (11.22 mg kg⁻¹) were found in irrigated soil. The pattern for heavy metal concentrations was Zn>Pb>Cu>Cd for all the samples. The daily intake for Cadmium and Lead exceeded the permissible limits. To assess the human health risk of heavy metals, it is necessary to calculate the level of human exposure with the help of the Health Risk Index (HRI) to that metal by tracing the route of exposure of pollutants to the human body. The HRI for Zn ranged from 0.054-0.174, for Cu 0.031-0.242, for Pb 2.407-7.973, and Cd 0-5.059. The HRI for Cd and Pb was > 1, which suggests a potential health issue.

In India, the use of industrial effluents and wastewater for irrigation, widespread use of pesticides and fertilisers and the degradation of sewer systems are considered to lead to unacceptable amounts of heavy metal in soils (Mapanda *et al.* 2005). Soils of large areas

are polluted with Pb, Cu, Cr and Ni in Surat, Gujarat and Pali, Rajasthan (Krishna and Govil 2007). According to Paul *et al.* (2015), the agricultural soils of Jajmau, Kanpur, India, were heavily contaminated with heavy metal pollution and revealed that the Cr, Cu, Zn, Pb, and Cd contents were significantly higher than the reference values. Machender *et al.* (2013) study on Chinnaru river basin (CRB), India shows that toxicity of heavy metals such as Ba (370–1710 mg kg⁻¹), Cr (8.7–543 mg kg⁻¹), Cu (7.7–96.6 mg kg⁻¹), Ni (5.4–168 mg kg⁻¹), Zn (49–478 mg kg⁻¹), V (39.8–162.8 mg kg⁻¹), As (3.4–19.6 mg kg⁻¹), and Pb (4–66 mg kg⁻¹) are due to the excessive usage of inorganic inputs. Rattan *et al.* (2005) conducted a case study on the long-term impact of irrigation with sewage effluents on heavy metal content in soils, crops, and groundwater. Results showed an increase in organic carbon content ranging from 38 to 79% in sewage-irrigated soils compared to tube well water-irrigated ones. On average, the soil pH dropped by 0.4 units as a result of sewage irrigation. Sewage irrigation for 20 years resulted in a significant build-up of DTPA extractable Zn (208%), Cu (170%), Fe (170%), Ni (63%) and Pb (29%) in sewage-irrigated soils over adjacent tube-well water irrigated soils, whereas Mn was depleted by 31%.

Meena *et al.* (2016) investigated the risk of sewage-irrigated soils about transferring trace elements to rice and wheat grain. It informed that under wheat crop, sewage irrigation led to a significant increase of zinc (141%), copper (219%), iron (514%), nickel (75.0%), and lead (28.1%) in sewage-irrigated soils over neighbouring tube well water irrigated soils over four decades. Under the rice crop, there was also a significant buildup of phosphorus (339 %), sulphur (130 %), zinc (287 %), copper (352 %), iron (457 %), nickel (258 %), lead (136 %), and cadmium (147 %) in sewage-irrigated soils as compared to that of tube well water-irrigated soils. Soils irrigated with sewage water for more than 20 years showed a significant buildup of Zn (2.1 times), Cu (1.7 times), Fe (1.7 times), Ni (63.1%) and Pb (29%) in sewage irrigated soils over groundwater irrigated soils (Simmons *et al.* 2006). Likewise, Yadav *et al.* (2003) reported that the wastewater discharged from all districts of Haryana contained micronutrients such as Zn, Fe and Co to 30.1, 178.8 and 4.3 mg L⁻¹, respectively. The

excessive accumulation of heavy metals in soil using wastewater irrigation can lead to contamination of soil and increased heavy metal uptake by crops, thus affecting food quality and safety. Satpathy *et al.* (2014) investigated the effects of fertilisers and pesticides, particularly chemical fertilisers, on paddy fields that release potentially toxic heavy metals into the soil. It was found that heavy metals in the paddy soil. Mn concentration ranged from 12.5 to 53.9 $\mu\text{g g}^{-1}$, Zn concentration ranged from 3.8 to 33.8 $\mu\text{g g}^{-1}$, Cu concentration ranged from 0.03 to 2.9 $\mu\text{g g}^{-1}$ in the paddy field soils, and in the concentrations of non-essential toxic metals, Pb ranged from 5.3 to 19.8 $\mu\text{g g}^{-1}$, Cr ranged from 1.3 to 7.8 $\mu\text{g g}^{-1}$, and Cd from 0.02 to 0.6 $\mu\text{g g}^{-1}$. The concentrations of metals in paddy field soils were higher than those of the surrounding reference soil but below permissible limits. The BAF (bioaccumulation factor) the ratio of the concentration of the element in the grain to that in the corresponding soil) was calculated for each rice sample to quantify the bioaccumulation effect of rice on the uptake of heavy metals from the soils. The ranking order of bioaccumulation factor (BAF) for heavy metals was $\text{Zn} > \text{Mn} > \text{Cd} > \text{Cu} > \text{Cr} > \text{Pb}$ indicating that the accumulation of micronutrients was more than that of non-essential toxic heavy metals.

Verma *et al.* (2012) analysed iron mine overburden samples from the Sukri iron ore deposits, Jharkhand's West Singhbhum, and found higher iron concentrations along with Mn, Pb and Zn. The heavy metal content of the overburden of the mine is responsible for their movement in the environment and for soil and water degradation. While working on sediment analysis from Goa's Mandovi estuarine mangrove ecosystem, Veerasingam *et al.* (2015) observed that all sediment cores showed enrichment of heavy metals in the upper part of core sediments decrease in concentration with depth, suggesting an excess of anthropogenic loading including mining activities. Kumar and Chopra (2015) studied heavy metal accumulation in soil and crops irrigated with glass industry effluent. Application of effluent increased the

heavy metal contents of the soil and crops (wheat and barley). The contents of different heavy metals in the soil used for the cultivation of *Brassica juncea*, *Triticum aestivum* and *Hordeum vulgare* were ranged Cd (0.66-0.84 mg kg^{-1}), Cr (0.24-0.28 mg kg^{-1}), Cu (4.37-5.84 mg kg^{-1}), Fe (6.84-7.58 mg kg^{-1}), Mn (1.38-1.56 mg kg^{-1}), Pb (0.23-0.29 mg kg^{-1}), Zn (3.75-4.15 mg kg^{-1}) after irrigation with glass industry effluent.

Accumulation of heavy metals in vegetables through food chain pathway

The heavy metals available for plant uptake are present as soluble components in the soil solution or those that are easily solubilised by root exudates (Blaylock *et al.* 2000; Malav *et al.* 2020). Once hazardous heavy metal enters the food chain, it cannot be removed and thereby becomes circulated into the whole food web. Many hyper accumulated plants serve as food for human beings and animals. Hence, the cycle from soil to humans *via* plants and again into the soil after the death of top consumers provides a path for hazardous heavy metals to remain sustained within the environment for long periods, inducing many harmful effects (Fig. 2). The toxic metal bioaccumulation in vegetables is explained in table 1.

Recent research and its summary have been highlighted in this section. Chandran *et al.* (2012) analysed the Cd, Cr, and Pb concentrations in *Solanum melongena* collected from a sewage irrigated farm. The concentrations of analysed metals were 1.70 mg kg^{-1} , 0.45 mg kg^{-1} , and 0.4 mg kg^{-1} for Cd, Cr, and Pb, respectively. The more significant translocation of Cd was reported due to the active transport or lack of metal absorption to fixed or soluble chelators in the root, or perhaps due to exchange with Ca, Mn and Zn moving through the roots in both vegetables. Zhou *et al.* (2016) investigated the toxic elements (Pb, Cd, Cu, Zn and As) concentrations in 22 vegetable species of six types of vegetables collected from contaminated farmland. The six vegetable types consisted of leafy vegetables, legume vegetables, root vegetables, stalk vegetables, solanaceous vegetables and melon vegetables. It was found that leafy vegetables appear to have the highest propensity to toxic elements accumulation. In contrast, melon vegetables accumulated the lowest concentrations of heavy metals.

Table 1. Bioaccumulation of hazardous heavy metals in different organs of vegetables

Region	HM Species	Vegetable	Concentration of HM (mg/kg)	Reference
Narayanganj, Bangladesh	Cu, Ni, Cd, Cr, Pb and Zn	<i>Basella alba</i> <i>Cucurbita moschata</i> <i>Anolis vidua</i> <i>Trichosanthes cucumerina</i> <i>Spinacia oleracea</i>	Zn-19.762 Cu-9.373 Pb-3.699 Ni-2.92 Cr-1.127 Cd-0.168	Ratul <i>et al.</i> (2018)
Marrakech, Morocco	Zn, Cu, Pb and Cd	<i>Vicia faba</i>	Zn-33.8-77.2 Cu-2.9-7.0 Pb-18.3-45.7 Cd-6.0-12.0	Chaoua <i>et al.</i> (2019)
Nevşehir Province, Turkey	Cd, Pb, Zn, Cr, Cu, Ni and Fe	<i>Solanum lycopersicum</i> <i>Solanum cepa</i> <i>Capiscum annuum</i>	Cd-0.001-0.004 Pb-0.02-0.03 Zn-0.50-1.16 Cr-0.001-0.02 Cu-0.14-0.25 Ni-0.02-0.48 Fe-0.74-4.37	Leblebici and Kar (2018)
Dave, China	Cu, Pb, As and Cd	<i>Vigna unguiculata</i> <i>Ipomoea aquatica</i> <i>Agelais tricolor</i>	Cu-0.46 to 6.67 Pb-0.00 to 1.47 As-0.00 to 0.94 Cd-0.00 to 0.71	Yang <i>et al.</i> (2018)
Korba, Chhattisgarh, India	Cr, Mn, Fe, Ni, Cu, Zn, Cu, Pb and Hg	<i>Solanum lycopersicum</i> <i>Solanumme longena</i> <i>Agelais tricolor</i> <i>Polygonia album</i> <i>Spinacia oleracea</i> <i>Coriandrum sativum</i>	As-0.56-2.08 Fe-192-2255 Cr-1-17 Mn-17-676 Cu-25-71 Zn-24-12	Ramteke <i>et al.</i> (2016)
Sri Ganganagar, Rajasthan, India	Fe, Mn, Cu and Zn	<i>Spinacia oleracea</i> <i>Brassica rapa</i> <i>Solanumme longena</i> <i>Brassica oleracea</i>	Fe-111-333 Mn-20.7-50.7 Cu-10-73.8 Zn-4.8-22.5	Arora <i>et al.</i> (2008)

^a Represents the average value of metals in all vegetable samples collected in particulate study.

However, most of these peri-urban lands (lands in the periphery of the city) are contaminated with pollutants, including heavy metals such as Cu, Zn, Pb, Cd, Ni, and Hg. This loading of heavy metals often leads to degradation of soil health and contamination of the food chain, mainly through the vegetables grown on such soils (Rattan *et al.* 2002). Heavy metals in soils reduce the yield of vegetables by disturbing the metabolic processes of plants (Salakar *et al.* 2011). Singh and Kumar (2006) concluded that soil, irrigation water and some vegetables from peri-urban sites are significantly contaminated by heavy metals, *i.e.* Cu, Cd, Pb and Zn. Tiwari *et al.* (2011) measured metal concentration in ten vegetable crops grown with mixed industrial effluent irrigation near Vadodara, Gujarat, India. Differential accumulation and translocation of

various metals in selected vegetable plant species were observed. A higher concentration of metals was found in order of Fe > Mn > Zn > Cd > Cu > Pb > Cr > As in soils irrigated with industrial effluent than soil irrigated with tube well water; however, the concentration of As, Cr and Pb were below detection limit in tube well water irrigated soils. Contamination assessment of five heavy metals (As, Cd, Cr, Pb and Zn) in five different types of green leafy vegetables *viz.*, mustard (*Brassica campestris*), garden cress (*Lepidium sativum*), fennel (*Foeniculum vulgare*), coriander (*Coriandrum sativum*), and spinach (*Spinacia oleracea*) collected from different market sites of Kathmandu showed a substantial accumulation of heavy metals in roots and leafy shoots of the vegetables (Shakya *et al.* 2013).

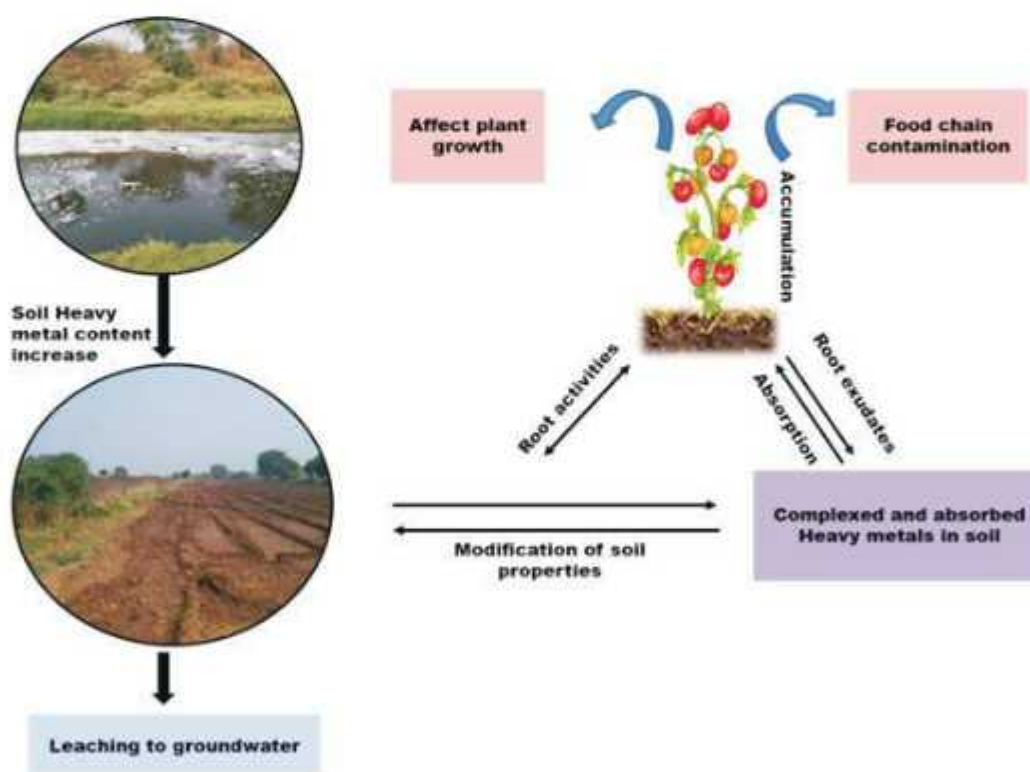


Fig.2. Heavy metal contamination in the agro-ecosystem (Modified from Khalid *et al.* 2018)

Risk assessment of heavy metal toxicity through edible vegetables from the industrial area of Bhilai, Chhattisgarh, showed concentrations of heavy metal, *i.e.* Mn, Fe, Cu, Zn and Pb in leafy vegetables like

spinach (Jena *et al.* 2012). Heavy metal accumulation and distribution pattern in different vegetable crops grown on heavy metal contaminated soil showed a marked difference in metal accumulation, their uptake

and distribution pattern. Crop species also showed a remarkable difference in metal concentration in various plant parts (Singh *et al.* 2012). Heavy metal load in soil and water in peri-urban areas of Delhi are likely to contaminate vegetables with heavy metals and render them unsafe for consumption. Samples of vegetables, *i.e.* spinach (*Spinacia oleracea L.*) and okra (*Abelmoschus esculentus L.*); soil and irrigation water were collected from 5 peri-urban sites of New Delhi to monitor their heavy metal loads. Loads of Cu, Zn, Pb and Cd in okra collected from different sites ranged between 15.3–25.0, 79.4–150.0, 1.1–5.6 and 1.1–7.0 $\mu\text{g g}^{-1}$, respectively. Most (90–100%) of the okra samples registered Cu contamination level below its safe limit (30 $\mu\text{g g}^{-1}$) while 100, 33 and 60% samples crossed the safe limits of Zn (50 $\mu\text{g g}^{-1}$), Pb (2.5 $\mu\text{g g}^{-1}$) and Cd (1.5 $\mu\text{g g}^{-1}$) (Singh *et al.* 2006).

Factors affecting heavy metal content in soils

Accumulation of heavy metals by plants dependson different plant species, translocation and transpiration rate, the concentration of heavy metals in soil, source and form of metals, soil properties such as pH, soil temperature, organic carbon and soil texture *etc.* (Gupta *et al.* 2019). Some important soil-related factors are described in this section.

Soil pH

Solubility and availability of toxic metals are mainly affected by pH of the soil solution. The solubility of metals decreases at high pH and increases at a low value of pH (Sheoran *et al.* 2016).

Redox potential (Eh)

Generally, soil solution is enriched by the ionic form of several metals. Thus, the movement of such metals from soil to plants depends on their oxidation state. For example, Cr exists in two oxidation states *i.e.* Cr^{3+} , which is relatively insoluble in water, while Cr^{6+} is highly soluble and readily available in the soil solution to the plants (NRC 2003).

Soil temperature

A rise in soil temperature may increase the availability of metals in the soil due to an increase in soil organic matter decomposition (Silveira *et al.* 2003).

Soil texture

The texture of soil also influences the solubility and bioavailability of metals in the soil. The availability of toxic elements is highest in loam and sand followed by clay loam, and fine-textured clay soils.

Cation exchange capacity (CEC)

This factor plays a vital role in the availability of metal in soil. The soil with low CEC, such as sand, has less binding power to metal and other cations than the soil with high CEC, such as clay (Bhargava *et al.* 2012).

Remediation techniques of heavy metal contamination in soil

Land and water are the two critical components of natural resources on which agriculture's sustainability and civilisation's continued survival depend. Unfortunately, both have been drastically contaminated/degraded by toxic elements (heavy metal). The application of various remediation approaches, such as microbial, physical and chemical approaches, can reduce pollution up to an appropriate level for a healthy environment (Jadia and Fulekar 2009). Throughout the years, various *in-situ* and *ex-situ* remediation methods have been developed to clean or recover heavy metal polluted soils, such as surface capping, soil flushing, electro-kinetic extraction, solidification, and vitrification and phytoremediation (Table 2). These strategies can be divided into five remediation groups: physical, chemical, electrical, thermal and biological. These soil remediation approaches typically use various operating processes and illustrate particular use (Khalid *et al.* 2017).

Classical soil remediation methods are costly, and some require extracting large amounts of soil. Heavy metal cannot be biologically eliminated but can be converted from one oxidising state or nuclear complex to

Table 2. Remediation techniques for metal polluted soil (Liu *et al.* 2018)

	Working mechanisms			Merits		Demerits
	Remediation techniques	Physical containment	Physical containment and isolation	Installation is easy, cheaper technique	Particular area specific technique	
Physical	Surface capping			Installation is fast and secure method	Particular area specific technique, loss of land cropping function	
	Encapsulation					
Electrical	Vitrification	Contaminant deactivation by thermally vitrifying soil		Very high efficiency	Costly, Particular area specific technique, treated land and soil losing environmental functions	
	Electrokinetics	Contaminant removal by electricity		Higher contaminant removal capacity, very less soil disturbance	Time consuming, less efficient	
Chemical	Soil flushing	Contaminant removal by chemical solutions		Higher contaminant removal capacity, very less soil disturbance, easy to install	Particular area specific technique such as best for coarse-textured soils with high permeability	
	Immobilization/stabilization	Contaminant deactivation by physiochemical transformation		Cheaper, easy to install	Metal-specific technique, temporary effectiveness	
Biological	Phytoremediation	Contaminant removal and stabilisation by plants		High public acceptance, cheaper, easy to install, very suitable for large, low contamination locations	Suitable only for shallow contamination, time-consuming, less efficient	
	Bioremediation	Contaminant transformation by microorganisms		Cheaper, easy to install	Low efficiency, merely supplemental to principal remediation techniques	
Physical	Landfilling	Physical containment and isolation		Secure and fast technique	Costly, requiring additional land for waste storage	
Chemical	Solidification	Contaminant deactivation by physically solidifying soil		Secure and fast technique	High cost, requiring additional land for waste storage	
	Soil washing	Contaminant removal by mechanical separation and chemical extraction		Secure and fast technique	Extreme soil disturbance	

another (Kumar *et al.* 2017). Plants and microbes hold the immense ability for doing the same. Heavy metal remediation by plants (phytoremediation) has lowered costs, added an aesthetic benefit, and has long-term practical field applications (Placek *et al.* 2016). Owing to its drawbacks (what are those); however, decontamination of heavy metal from a given location is difficult due to a single solution. However, the application of various phytoremediation methods will be more effective in extracting heavy metals from large fields and eventually will improve agricultural land. Biological processes are less sensitive than conventional approaches to environmental immoderations; they have a clear advantage of being more cost-effective (Cunningham *et al.* 1997). With the example of metal-polluted atmosphere remediation, researchers find that the bioremediation cycle has a restricted outlook mainly because of the non-degradable presence of heavy metals (Marschner 1995).

The use of different plant species in bioremediation of polluted soil environment is an acceptable choice (Table 3), with minimum environmental effects, without damaging the soil, which also provides the ability to recover the heavy metal (Salt *et al.* 1995). Phytoremediation is a cheaper method relative to other bioremediation approaches by 50-80 per cent. The downside of this approach is that it can be a remediation process even more slowly, involving several plant growing seasons. The contaminants may limit plant growth, and the resulting biomass, enriched with heavy metals, may be dangerous in the food chain (Elekes 2014). This biomass is considered waste and requires controlled and responsible disposal because of the risk of toxicity for the environment. The phytoremediation provides economic benefits by turning this tool into a financially self-supporting environmental remediation solution.

Table 3. Different plant species used for phytoremediation in the soil medium

Heavy metal	Process	Plant species	References
Arsenic	Phytoextraction	<i>Pteris vittata</i>	Yang <i>et al.</i> (2017 a,b)
Cadmium	Phytoextraction	<i>Ricinus communis</i>	Yang <i>et al.</i> (2017 a,b)
Chromium	Phytostabilisation	Rose plant	Ramana <i>et al.</i> (2013)
Lead	Phytostabilisation	<i>Hordeum vulgare</i>	Katoh <i>et al.</i> (2017)
Cobalt	Phytoextraction	<i>Pennisetum annuus</i>	Lotfy and Mostafa (2014)
Mercury	Phytostabilisation	<i>Brassica juncea</i>	Shiyab <i>et al.</i> (2009)
Nickel	Phytoextraction	<i>Brassica juncea</i>	Kathal <i>et al.</i> (2016a,b)

Based on processes and applicability, phytoremediation technology is further sub-divided into categories such as (1) Rhizosphere bioremediation: It enhances biodegradation of pollutants by root-associated bacteria and fungi. (2) Rhizofiltration: This process is based on the rhizospheric accumulation of heavy metals such as Pb, Cd, Zn, Ni, Cu and Radionuclides – Cs, Sr, U. (3) Phytostabilisation: It is the transformation of toxic compounds into non-toxic/less toxic forms fixed in the soil thereby reducing the bioavailability of pollutants (Pb, Cd, Zn, As, Cu, Cr,

Se, U, polycyclic aromatic hydrocarbons (PAHs), polychlorinatedbiphenyl (PCBs), dioxins and furans) in the environment. (4) Phytoextraction: It involves plant roots for the uptake of contaminants (preferably heavy metals such as Pb, Cd, Zn, Ni, Cu) that gets accumulated in aerial parts of the plant followed by harvesting the plant biomass for safe disposal (Hyperaccumulation). (5) Phytovolatilisation: It involves volatilisation of Volatile organic compounds (trichloroethylene (TCE), methylene chloride (MC) Tetrachloroethylene (PCE), Carbon Tetrachloride (CT)) through plant leaves or soil surface).

The metal(loid)s As, Hg, and Se may be discharged by accumulator plants (*e.g.* *Astragalus racemosus*) in gaseous species into the atmosphere with the help of the process phytovolatilisation. So far 721 species of plants have been identified as metal hyperaccumulators and (6) Phytotransformation/phytodegradation: In this process,

the degradation of organic pollutants (Herbicides - atrazine, alachlor, mixtures benzene toluene ethylbenzene and xylene) is carried out by plants utilising enzymes such as dehalogenase and oxygenase; it is not dependent upon rhizospheric microorganisms (Yadav *et al.* 2018) (Fig 3.).

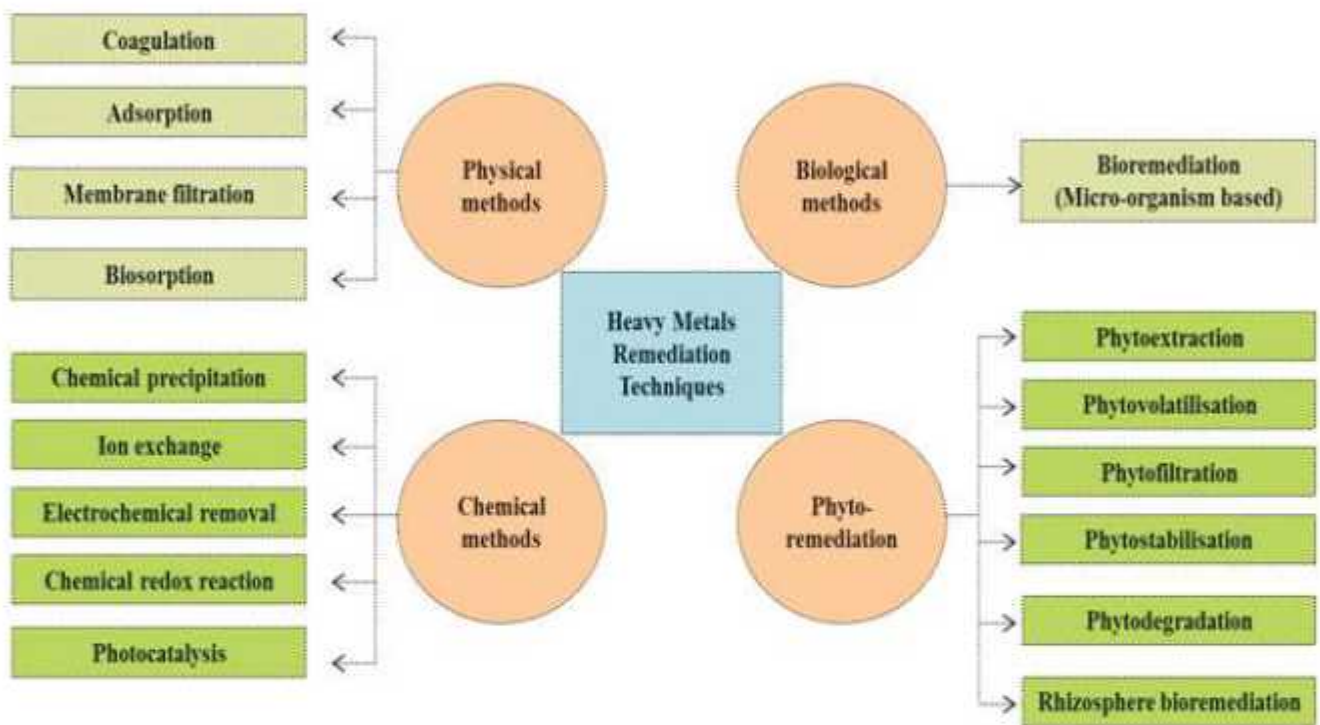


Fig. 3. Schematic representation of overall decontamination methods of heavy metals

Maximum allowable limits of heavy metals in different components of ecosystem

To know the toxic effect or degree of hazardousness of heavy metals in soil, water and vegetation, a maximum allowable concentration is fixed as standards by different organisations/nations. Tables 4, 5 and 6 show the limits of potentially toxic heavy metals in different ecosystem components given by different countries/organisations. The standard adopted by

different countries is helpful to assess the quality of natural resources and check the effectiveness of remediation techniques. The intervention values reflect the quality of soil functionality seriously impaired for human, animal, and plant life. Concentrations that surpass the intervention values are equivalent to significant pollution. Target values indicate the soil concentration required for sustainability or expressed in terms of remedial policy, the soil quality required to fully restore the soil's functionality for human, animal, and plant life.

Table 4. Maximum allowable limits (mg kg^{-1}) of potential toxic metals in soils (Total heavy metal)

Country/ Organizations	As	Cd	Cr	Hg	Pb	Cu	Ni	Zn	Mo	Reference
Austria	20	1-2	100	1-2	100	60-100	50-70	150-300	10	Visser 1993
Belgium ¹	-	2	100	1	100	50	50	200	-	Visser 1993
Denmark	-	0.5	30	0.5	40	40	15	100	-	Visser 1993
Germany ²	-	1	60	0.5	70	40	50	150	-	Visser 1993
Greece	-	1-3	-	1-1.5	50-300	50-140	30-75	150-300	-	Visser 1993
Finland	-	0.5	200	0.2	60	100	60	150	-	Visser 1993
France	-	2	150	1	100	100	50	300	-	Visser 1993
Ireland	-	1	-	1	50	50	30	150	-	Visser 1993
Italy	-	1.5	-	1	100	100	75	300	-	Visser 1993
Luxembourg	-	1-3	100-200	1-1.5	50-300	50-140	30-75	150-300	-	Visser 1993
Netherlands	-	0.5	30	0.5	40	40	15	100	-	Visser 1993
Portugal ³	-	3	200	1.5	300	100	75	300	-	Visser 1993
Sweden	-	0.4	60	0.3	40	40	30	100-150	-	Visser 1993
Spain ⁴	-	3	150	1.5	300	210	112	450	-	Visser 1993
UK ⁵	50	3	400	1	300	135	75	300	4	Visser 1993
EU	-	1-3	-	1-1.5	50-300	50-140	30-75	150-300	-	EC/DGE 2010
USA ⁶	-	20	1500	-	150	750	210	1400	-	EC/DGE 2010
India	-	3-6	-	-	250-500	135-270	75-150	300-600	-	Awasthi 2000
China	30	0.3	150	0.3	250	50	40	200	-	CNEMC 1990
WHO	-	0.8	100	-	85	36	35	50	-	WHO 1996

¹ Represents the heavy metals value in Wallonia region of Belgium² Represents the heavy metals value in loamy soil of Germany³ Represents the heavy metals value of soil having pH between 5.5 to <7.0 in Portugal⁴ Represents the heavy metals value of soil having pH<7.0 in Spain⁵ Represents the heavy metals value of soil having pH between ≥ 6.0 to ≤ 7.0 in United Kingdom⁶ Represents the heavy metals value in soil on which sewage sludge is applied of USA

Table 5. Maximum allowable limits (mg kg⁻¹) of potential toxic metals in plants and vegetables

HM	Vegetable	FSSAI ¹	WHO ²	India ³	FAO/WHO ⁴	EU ⁵	China ⁶	Germany ⁷
As	Fruit veg	0.10						
	Leafy veg	0.10						
	Root and stem	0.10	-	-	1.0	-	0.5	-
	Brassica	0.10						
Cd	Fruit veg	0.05				0.05		0.1
	Leafy veg	0.20				0.20		0.5
	Root and stem	0.10	0.02	1.0	0.20	0.10	0.05	0.1
	Brassica	0.05				0.20		0.1
Cr	Fruit veg							
	Leafy veg							
	Root and stem	-	1.3	25	-	-	0.50	-
	Brassica							
Hg	Fruit veg							0.05
	Leafy veg							0.05
	Root and stem	-	-	-	-	-	0.01	0.05
	Brassica							
Pb	Fruit veg	0.10				0.10		0.50
	Leafy veg	0.30				0.30		0.80
	Root and stem	0.10	2.0	5.0	3.0	0.10	0.20	0.25
	Brassica	0.30				0.30		
Ni	Vegetables	-	10	1.5	-	-	0.30	-
Zn	Vegetables	50	0.60	50	60	-	20	-
Cu	Vegetables	3.0	10	20	40	-	10	-
Mn	Vegetables	-	-	-	0.20	-	-	-

¹FSSAI - Food Safety and Standards Authority of India (FSSAI 2011)²WHO - World Health Organization, Geneva, Switzerland (WHO 1996)³India - Awasthi 2000⁴FAO/WHO - The Joint FAO-WHO Expert Committee Report on Food Additives (FAO/WHO 2001)⁵EU - European Union commission (EU 2006)⁶China - Hu *et al.* 2017⁷Germany - Merian 1991

Table 6. Maximum allowable limits (mg L⁻¹) of potential toxic metals in drinking water

HM	ICMR ¹	BIS ²	WHO ³	US EPA ⁴	Canada ⁵	EU ⁶	Russia ⁷	Japan ⁸	China ⁹	Pakistan ¹⁰
As	0.05	0.01	0.01	0.19	0.05	0.01	0.01	0.01	0.01	0.05
Cd	0.01	0.003	0.003	0.005	0.005	0.003	0.003	0.01	-	0.01
Cr	0.05	0.05	0.05	0.1	0.05	0.05	0.05	0.05	0.05	0.05
Hg	0.001	0.001	0.001	0.002	0.001	0.001	0.001	0.0005	0.001	0.001
Pb	0.01	0.01	0.01	0.005	0.05	0.01	0.01	0.05	0.01	0.05
Ni	-	0.02	0.02	0.1	-	0.02	0.02	0.01	0.02	0.02
Mn	-	0.07	0.07	-	-	0.07	-	0.07	0.07	-
Fe	0.10	0.30	0.30	0.30	-	0.30	0.30	0.30	0.10	-
Zn	5.0	5.0	3.0	5.0	5.0	3.0	1.0	-	-	5.0
Cu	0.05	-	1-2	1.3	-	1-2	2.0	1.0	1.0	2.0

¹ICMR- Indian Council of Medical Research, India (ICMR 1975)

²BIS- Bureau of Indian Standards, India (BIS 2012)

³WHO- World Health Organization, Geneva, Switzerland (WHO 1993)

⁴US EPA- United States Environmental Protection Agency, Washington, D.C., United States (US EPA 1991)

⁵Canada – (CCME 1991)

⁶EU- *European Union* commission (ECDGE 2010)

⁷Russia – (Radojevic and Bashkin 2006)

⁸Japan – (Radojevic and Bashkin 2006)

⁹China – (SAC MHC 2006)

¹⁰Pakistan – (PEPA 2008)

Conclusion

Heavy metal contamination in agro-ecosystem (soil-water-plant) is a serious concern, especially for peri-urban agriculture. These heavy metals are toxic whenever their concentration goes beyond the permissible limit. Currently, anthropogenic sources boost up the pollution level over geogenic sources. Once the contaminants enter into the soil-water-plant continuum, it spreads to the entire food chain through bio-accumulation and bio-magnification and affects the plant growth, yield and also disrupts the soil quality. There is an urgent need for new developments and strengthen existing environmentally and economically viable remediation techniques to reduce the hazardous effects of heavy metals and restore the ecosystem functions of the contaminated soils. Heavy metal contamination requires review and research for its management and sustainable development of natural resources.

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