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Review

Phytoremediation: Environmentally sustainable way for reclamation of heavy metal polluted soils

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ABSTRACT

Soil contamination with toxic metals is a widespread environmental issue resulting from global industrialization within the past few years. Therefore, decontamination of heavy metal contaminated soils is very important to reduce the associated risks and for maintenance of environmental health and ecological restoration. Conventional techniques for reclamation of such soils are expensive and environmental non-friendly. Phytoremediation is an emerging technology implementing green plants to clean up the environment from contaminants and has been considered as a cost-effective and non-invasive alternative to the conventional remediation approaches. There are different types of phytoremediation including, phytostabilization, phytostimulation, phytotransformation, phytofiltration and phytoextraction, the latter being most extensively acknowledged for remediation of soils contaminated with toxic heavy metals. Recent literature is gathered to critically review the sources, hazardous effects of toxic heavy metals and environmentally sustainable phytoremediation technique for heavy metal polluted soils to offer widespread applicability of this green technology. Different strategies to enhance the bioavailability of heavy metals in the soil are also discussed shortly. It can be concluded that phytoremediation of heavy metal contaminated soils is a reliable tool and necessary for making the land resource accessible for crop production.

1. Introduction

Heavy metal pollution of soil has become global environmental dilemma due to intensively increasing industrialization and agricultural activities (Chaoua et al., 2018; Woodford, 2019). Heavy metals are highly toxic because, unlike organic matter, they are not biodegradable but can only change their oxidation state and are highly persistent in nature with half-life more than 20 years (Jan et al., 2015; Ayangbenro, and Babalola, 2017; Hadia-e-Fatima and Ahmed, 2018). Fifty-three elements are documented as heavy metals and are considered as universal pollutants with densities greater than 5 g/cm^3 (Sarma, 2011; Saif and Khan, 2017; Prieto et al., 2018). Heavy metals can be grouped into essential and non-essential classes. Essential heavy metals include Co, Cr, Cu, Fe, Mn, Ni and Zn and are considered as essential micronutrients, but become poisonous when taken in excess quantities. Non-essential heavy metals include Pb, Cd and Hg and are highly toxic for living organisms (Monni et al., 2000; ul-Hassan et al., 2017; Sandeep et al., 2019).

Heavy metals are natural components of soil, but human activities have increased their concentration. Sources of heavy metals in soil include excessive application of agrochemicals, sewage sludge, industrial wastewater, biosolids and manure (Bu-Olayan and Thomas, 2009; Alghobar and Suresha, 2017; Haroon et al., 2018; Woodford, 2019). Consequently, heavy metal accumulation in soil causes severe health problems for plants, animals and humans (Kleckerova and Docekalova, 2014; Srivastava et al., 2017). According to the United States Environmental Protection Agency (US EPA), soil heavy metal pollution has caused health issues for about 10 million humans all over the world (U.S. EPA, 2016). Hence, accumulation of heavy metals in crop plants through soil-root interface is a critical hazard (Maimon et al., 2009; Nazir et al., 2015; Sakizadeh and Ghorbani, 2017).

2. Sources of heavy metal pollution in soil

Anthropogenic and geological activities are two different sources through which heavy metals enter the soil (Keshavarzi et al., 2015; Liu et al., 2018). Industrial and domestic wastewater, smelting, mining, fuel manufacturing and agrochemicals are different anthropogenic input points of heavy metals in agricultural and non-agricultural soil (Wei and Yang, 2010; Marrugo-Negrete et al., 2017; Yan et al., 2018).

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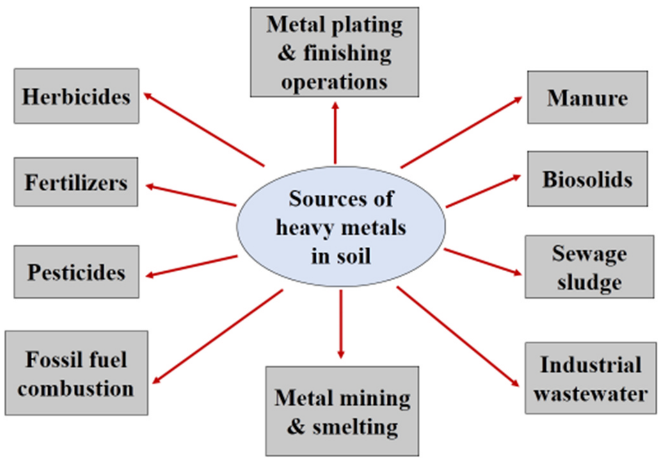

Fig. 1. Sources of heavy metals in soil.

Heavy metals are released into atmosphere through discharge of gases and dust from transport and production of constructed material and energy. In atmosphere, heavy metals exist in the form of aerosols and then through precipitation become deposit to soil (Aksu, 2015; Naderizadeh et al., 2016; Suvarapu and Baek, 2017). Sources of heavy metals in soil are shown in Fig. 1. Excessive application of biosolids, fertilizers, pesticides and herbicides to crops causes buildup of these chemicals in soil and uptake by plant (Atafar et al., 2010; Bitew and Alemayehu, 2017). Application of different biosolids as valuable fertilizers such as composts, sewage sludge and livestock manure to agricultural land leads towards elevating the level of heavy metals in soil (Basta et al., 2005; Bitew and Alemayehu, 2017). Biosolids which are organic material also have different heavy metal contents like Pb, Cd, Hg and Cr. Heavy metals in biosolids applied to soil can also leach to groundwater (Brisolara and Qi, 2013). Concentration of some heavy metals in fertilizers and sewage sludge is shown in Table 1.

Micronutrient (Fe, Zn, Mn, Cu, Co and Ni) deficient soils are provided with these elements for healthy growth of plants and crops (Tripathi et al., 2015). Large amounts of fertilizers are being applied to agricultural soils to supply the crops with adequate quantity of nitrogen, phosphorus and potassium. The compounds which are used to provide these components also possess very minute quantities of toxic heavy metals. Phosphate fertilizers contain considerable amount of toxic heavy metals as compared to potash and nitrogen fertilizers. Due to excessive application of such fertilizers, concentration of heavy metals (Pb and Cd) is going to be increased (Boyd, 2010; Bitew and Alemayehu, 2017). Use of sewage water for irrigation also leads towards contamination of soil and crops (Bridge, 2004; Chaoua et al., 2018; Haroon et al., 2018).

Pesticides which are being used in agriculture and horticulture sectors are composed of considerable amount of heavy metals such as copper sulfate and lead arsenate compounds are used in pesticides to control pests (McLaughlin et al., 2000; Missimer et al., 2018). Application of domestic and industrial wastewater in agriculture is a common way to solve irrigation problem in arable land. The level of heavy metals in sewage water is low but excessive and long-term irrigation of wastewater effluents has caused the buildup of heavy metals in soil where they fixed in different ways (Gupta et al., 2012; Chaoua et al., 2018). Extent of heavy metals in soil can range from traces to 100,000 mg/kg depending upon type and location of compound (Alloway, 2013; Liu et al., 2018). Heavy metals are main cause of unhealthy environmental condition because they can leach and are nondegradable (Martley et al., 2004; Alloway, 2013; Sidhu, 2016).

3. Environmentally toxic heavy metals and their classification

Heavy metals can be classified into essential and non-essential groups. Group of essential heavy metals comprises of Co, Cr, Cu, Fe, Mn, Ni and Zn and other includes Pb, Cd and Hg. Heavy metals can also be classified as extremely poisonous, moderately poisonous and relatively less poisonous according to their level of toxicity as shown in Table 2. Among non-essential heavy metals, Cd is more bioavailable and bioaccumulate due to its high solubility. Cd has no biological function and in compound form it is present as divalent ion (Volland et al., 2014; Oves et al., 2016). In heavy metal contaminated soil, Cd is mostly present with Zn that is an essential micronutrient. At high concentration Zn causes toxicity in plants, while Cd rarely causes phytotoxicity (Ryzhenko et al., 2017). Vetiver grass can be utilized for the reclamation of Cd contaminated soil because it can uptake and accumulate large concentration of Cd (Banerjee et al., 2016, 2019). In rhizosphere, microorganisms cause sequestration of Cd and thus influence the uptake of Cd by plants (Chen et al., 2019).

In roots of plants that are used for remediation of Cd contaminated soil, high concentration of glutathione and glutathione-S-transferase has found to be associated with detoxification processes (Gill et al., 2013; Wu et al., 2017). Among edible crops, highest level of Cd and Zn was found in maize and that of Pb was found in wheat. Use of Cd contaminated soil for wheat cultivation can cause food contamination because Cd uptake in wheat grains is high as compared to maize (Perrier et al., 2016). Cd has the potential to suppress the activities of antioxidative enzymes mainly glutathione reductase (Wu et al., 2017).

It is very difficult to remove Pb from contaminated soil. Soil's capacity to bind Pb improves with rising level of pH, cation exchange

Table 1
Concentration of heavy metals in fertilizer (Phosphate rock) and sewage sludge.
Source: (Edgell, 1988; Kongshaug et al., 1992; Nishimune, 1993; Wallace and Wallace, 1993; Benckiser and Simmarmata, 1994; Pantelica and Salagean, 1997; European Commission, 2001; Ogunleye et al., 2002; Conceicao and Bonotio, 2006; Jamali et al., 2007; Sabiha et al., 2009).

Country	Metal mg/kg dry material						
	As	Cd	Cr	Cu	Ni	Pb	Zn
Phosphate rock							
Brazil	–	4	70.5	–	116	44.5	299
Morrocco	–	30	291	–	26	7	345
Nigeria	–	–	28	–	–	–	59
North Africa	–	60	105	–	33	6	420
Pakistan	–	7.5	17	–	28	89	67.2
Russia	–	0.1	23.3	–	2	3	19
USA	–	11	142	–	37	12	403
Sewage Sludge							
Germany	–	20–40	1000–1750	1000–1750	300–400	750–1200	2500–4000
Japan	75	85	3000	4300	420	840	7500
Pakistan	4.62	27.3	75.9	177.8	83.0	103.3	406.4
USA	–	10	900	800	100	900	2500

Table 2
Classification of heavy metals according to their level of toxicity.
Source: (GOST, 1983; Mukesh et al., 2008).

Heavy metal	Level of toxicity
Cadmium (Cd), Arsenic (As), Mercury (Hg), Selenium (Se), Lead (Pb), Zinc (Zn)	Extremely poisonous
Nickel (Ni), Cobalt (Co), Chromium (Cr), Copper (Cu), Molybdenum (Mo)	Moderately poisonous
Barium (Ba), Manganese (Mn), Strontium (Sr)	Relatively less poisonous

capacity, organic matter content and phosphate (Tang et al., 2015; Wijayawardena et al., 2015; Zeng et al., 2017). In soil, Pb becomes precipitated as Pb-phosphate and thus becomes unavailable to plants (Zeng et al., 2017). *Brassica juncea* can be utilized to extract Pb form contaminated soil because it transports Pb form root to shoot (Bouquet et al., 2017; Yahaghi et al., 2018). In fruit crops, Pb does not accumulate in their fruiting parts. High concentration of Pb has been found on the surface layer of leafy vegetables and root crops. Pb contaminated soil at a level more than 300 ppm affects the plant survival (Rosen, 2002; Chandrasekhar, and Ray, 2019).

Mercury (Hg) is the liquid metal and byproduct of ore processing. In environment, Hg exists in various forms. Mercuric and mercurous are oxidized forms, while organic and inorganic Hg become reduced to elemental Hg under reducing conditions that can be converted to alkylated form. Alkylated mercury is most hazardous, volatile and soluble in water (Smith et al., 1995; Sun et al., 2016; Beckers, and Rinklebe, 2017). Alkylated mercury is most toxic because due to its strong affinity for sulfur bounded compounds like enzymes and proteins.

Due to rising anthropogenic emissions, Zn has built up in the soil and water systems. Zn is phytotoxic and only some plants have ability to survive under Zn contaminated soil (Pence et al., 2000; Ryzhenko et al., 2017). Zn is a micronutrient and necessary for human health. Its deficiency can cause birth defects (Wang et al., 2015; Calderón Guzmán et al., 2019). Among heavy metals, cobalt (Co) is least toxic and forms different inorganic and organic compounds. In biological systems, Co works as a cofactor for different enzymes. However, at higher concentration it is hazardous for human health (Tchounwou et al., 2012; Leyssens et al., 2017). Nickel (Ni) is an essential element but at higher concentration it causes health effects. Nickel contaminated soil affects microbial growth and microorganisms also develop resistance against Ni (Wuana and Okieimen, 2011; Paulo et al., 2017; X. Xia et al., 2018).

Chromium (Cr) is an ore product of mining industry and in environment present as trivalent (Cr^{3+}) and hexavalent (Cr^{6+}) chromium (Smith et al., 1995; Mandal et al., 2017; Coetzee et al., 2018). Trivalent chromium is an essential trace element and cofactor for many enzymes (Davis and Vincent, 1997; Lewicki et al., 2014). Under aerobic conditions chromium exists as Cr^{6+} and under anaerobic conditions, soil organic matter converts Cr^{6+} into Cr^{3+}. In soils at low pH, Cr^{3+} is dominant that form complexes with Cl^-, CN^-, SO_4^{2-}, NH_3, OH^- (Chrostowski et al., 1991; Gautam et al., 2016).

Worldly, copper (Cu) is mostly used metal because it is an essential micronutrient for both plants and animals. In humans, Cu is important for hemoglobin production while in plants it is important for water regulation and seed production. Copper contaminated soil can cause direct and indirect threats on plants and humans, respectively (Bjuhr, 2007; Jaiswal et al., 2018). In soil, Cu forms complexes with organic matter. Thus, concentration of ionic Cu in soil solution is very low (Wuana and Okieimen, 2011; Lockwood et al., 2015). Copper is toxic due to its radical property that forms hyperoxide radicals which interact with thiol compounds in cell membrane (Nies, 2003).

Arsenic (As) is the most abundant metalloid in nature and found as arsenate (As^{5+}) and arsenite (As^{3+}) (Mukhopadhyay et al., 2002; Watanabe and Hirano, 2013). Arsenate is dominant under aerobic conditions while arsenite is dominant under anaerobic conditions. Arsenic forms methylated compounds that are volatile (Scragg, 2006; Schwartz et al., 2016). Arsenate behaves like phosphate and can enter the microbial cells and prevent the phosphate dependent energy production processes and thus impede oxidative phosphorylation (Lloyd and Oremland, 2006; Urrialde et al., 2017).

4. Heavy metal pollution and toxic effects on environment

Heavy metals are main source of environmental pollution because they can leach and are nondegradable (Martley et al., 2004; Brisolara and Qi, 2013; Sidhu, 2016; Chaoua et al., 2018; Woodford, 2019). Released heavy metals into soil have detrimental effects on living organisms through bioaccumulation and biomagnification processes (Szynkowska et al., 2018; Mortensen et al., 2018). Most heavy metals are toxic for both micro and macro organisms by direct impacts on their physiological and biochemical pathways (Rai et al., 2016; Luo et al., 2019; Ghori et al., 2019) and their concentration in soil ranges from 1 to 100,000 mg/kg (Blaylock and Huang, 2000; Herawati et al., 2000; Long et al., 2002; Alloway, 2013). The highest concentration of heavy metals occurs normally in the topsoil because organic horizons are characterized by the greatest ability to bind heavy metals (Acosta et al., 2015; Gu et al., 2016). Hence, high concentrations of Pb and Cd in upper layer of soil irrigated with contaminated water are taken up by plants (in rhizosphere, root exudates and microorganisms increase mobility of heavy metals) and thus negatively affect the absorption process of essential nutrients (Hédiji et al., 2015; Khan et al., 2016). Lead and cadmium can influence the absorption of Cu^{2+}, Fe^{2+}, Zn^{2+} and Mn^{2+} through competition for sites or processes shared by these cations (Xu and Shi, 2000). Hence, metal contaminated crops mainly vegetables are not safe for human edible purpose due to their network of roots in upper layer of soil (Smolen et al., 2010; Qureshi et al., 2016; Gupta et al., 2019). The maximum permissible limits of toxic heavy metals in irrigation water, soil and plants are presented in Table 3.

Heavy metal intoxicated plants in humans cause carcinogenic and chronic diseases. Human consumption of Pb, Cd and Zn causes respiratory diseases and heart, brain and kidney damage. Heavy metals cause toxicity in biological molecules in three different ways by producing reactive species, blocking functional groups and replacement of basic metal ions (Schutzendubel and Polle, 2002; Morcillo et al., 2016; Wijayawardena et al., 2016; Mandal, 2017). Heavy metals toxicity in plants leads towards stunted growth, low yield and aberrations in metabolic functions (Garbisu and Alkorta, 2001; Schwartz et al., 2003;

Table 3
Maximum permissible limits for toxic heavy metals concentration in irrigation water, soil and plants.
Source: (European Union, 2002; WHO/FAO, 2007).

Metal	Irrigation water (μg/mL)	Soil (μg/g)	Plant (μg/g)
Lead (Pb)	0.015	300	0.30
Cadmium (Cd)	0.01	3	0.2
Chromium (Cr)	0.10	150	5
Arsenic (As)	0.01	20	0.1
Nickel (Ni)	1.40	50	67
Mercury (Hg)	0.01	30	0.03
Copper (Cu)	0.20	140	40
Iron (Fe)	0.50	50,000	450
Zinc (Zn)	2.0	300	60
Manganese (Mn)	0.20	80	500

Asati et al., 2016; Kalaivanan and Ganeshamurthy, 2016). Increased production rate of reactive oxygen species (ROS) is the most obvious outcome of heavy metal toxicity in plants that causes oxidative stress leading towards membrane dismantling (Chen et al., 2012; Pandey and Dubey, 2019). Soil heavy metal contamination can also change native microbial community that ultimately destroys its biochemical properties (Kurek and Bollag, 2004; Paulo et al., 2017; X. Xia et al., 2018; B. Jiang et al., 2019; M. Jiang et al., 2019).

5. Remediation techniques for heavy metal-polluted soil

Decontamination of heavy metal contaminated soils is a main concern in environmental legislation. In-situ and ex-situ are the two approaches that are being practiced for decontamination of heavy metal affected soils. The ex-situ decontamination of polluted soils carried by conventional physico-chemical techniques is expensive. However, in situ remediation of heavy metal affected soils by plants is economical and environment friendly and depends upon the bioavailability of heavy metals (Van Gestel et al., 1992; Khalid et al., 2017; Emenike et al., 2018; Xia et al., 2019). Reclamation processes for heavy metal contaminated soils are classified into physical, chemical and biological processes (Zhou and Song, 2004; Khalid et al., 2017; Emenike et al., 2018). Different remediation techniques for heavy metal-polluted soil are shown in Fig. 2. Remediation mechanisms are based on two basic principles. First is the complete removal of contaminants and second is the transformation of contaminants into less harmful forms by using engineering technologies (Zhou and Song, 2004; Suthersan et al., 2016). Conventional physico-chemical soil remediation methods can be applied at massively heavy metal affected soils but only to small regions (Bio-Wise, 2003; Dada et al., 2015). Application of these methods is limited due to high energy requirement and adverse effects on soil structure and productivity (Leumann et al., 1995; Schnoor, 1997; Danh et al., 2009; Kuppusamy et al., 2017). Microorganisms can be applied for restoration of heavy metal affected soil (Garbisu and Alkorta, 1997; Ayangbenro, and Babalola, 2017). But use of plants for soil reclamation is more beneficial because they extract heavy metals form soil and restore the soil to healthy level (Chirakkara et al., 2016; Sarwar et al., 2017; Emenike et al., 2018; Lajayer et al., 2019).

5.1. *Physico-chemical remediation techniques for heavy metal-contaminated soil*

The physical methods of soil reclamation include soil replacement and thermal desorption processes. Soil replacement includes the use of contaminant-free soil to replace the polluted soil in order to dilute the level of pollutants (Qian and Liu, 2000; Zhang et al., 2004; Fenyvesi et al., 2019). There are three types of soil replacement:

i) Contaminated soil is removed and new soil is introduced
ii) Contaminated soil is digged up and pollutants are spread into deep layers
iii) Imported new soil is added into contaminated soil (Sidhu, 2016; Khalid et al., 2017).

In thermal desorption contaminated soil is heated in order to volatile the pollutants. Contaminated soil is excavated and treated in thermal desorber. Contaminated vapors are captured by gas collection equipment. Thermal desorption is divided into low temperature and high temperature desorption (Aresta et al., 2008; Zhang et al., 2017). Chemical remediation technology of heavy metal contaminated soil includes immobilization techniques and soil washing. Immobilization techniques involve the application of organic and inorganic binding agents to contaminated soil for stabilization of contaminants by the natural processes of sorption, precipitation and complexation. In-situ immobilization processes are preferred due to low inputs of labor and energy (Hashimoto et al., 2009; Sidhu, 2016; Khalid et al., 2017).

5.1.1. *Solidification and stabilization*

Solidification and stabilization are clean up technologies that avoid or slow the release of heavy metals or other contaminants. These methods prevent heavy metals from leaching by binding. Solidification involves the mixing of contaminated material with binding agent that causes the contaminated material to fix together (Evanko and Dzombak, 1997; Xia et al., 2017; W.Y. Xia et al., 2018). Inorganic binding agents include clay, zeolite, charcoal, Fe/Mn oxides, calcium carbonate, fly ash and cement (Fawzy, 2008; Gunatilake, 2015; Zarschler et al., 2016) and organic binding agents include manure, compost and bitumen (Farrell et al., 2010; Lim et al., 2016). Organic and inorganic binding agents can be used in a combination. The dominant mechanism of metal immobilization includes precipitation of hydroxides within solid medium. Solidification involves the mixing of contaminated material with binding agents but those binding agents also have chemical reactions with pollutants to prevent them from leaching into environment (Xia et al., 2017; W.Y. Xia et al., 2018).

5.1.2. *Vitrification*

In vitrification, metal contaminated soil is heated at high temperature to reduce the mobility of heavy metals. During this process high temperature may cause volatilization of organic compounds and some metals such as Hg that should be collected and remaining are converted

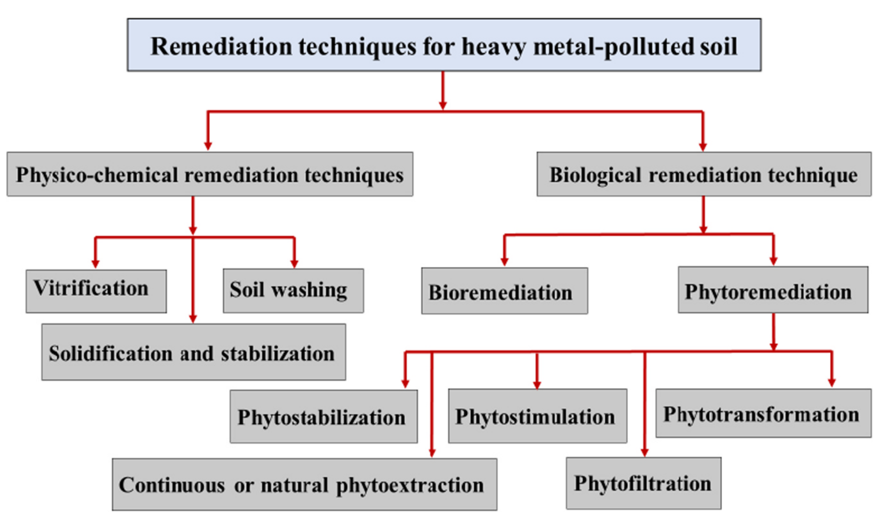

Fig. 2. Different remediation techniques for heavy metal-polluted soil.

into inert glass like product (Asante-Duah, 1996; Kołacinski et al., 2017; RoyChowdhury et al., 2018). There are three major classes of vitrification process which include electrical, thermal and plasma methods (Acar and Alshawabkeh, 1993; Suthersan, 1997; Wait and Thomas, 2003). In-situ vitrification is preferred over ex-situ process and different energy sources can be applied for this technique (USEPA, 1992). In this process an electric current is passed through contaminated soil by using electrodes inserted into the soil. An array of four electrodes is called as a melt and a single melt can decontaminate up to 1000 t of polluted soil (Buelt and Thompson, 1992; Kołacinski et al., 2017).

5.1.3. Soil washing

Soil washing includes the implementation of solvents and mechanical methods to clean up heavy metal contaminated soil (Urum et al., 2003; Dermont et al., 2008; Song et al., 2017). In this technique, fine soil portion is separated from coarse soil that causes reduction in volume of contaminated soil (Riser-Roberts, 1998; Liao et al., 2016). The reduced contaminated soil volume can be treated by other clean up technologies while larger clean soil volume can be managed by utilizing as backfill (RAAG, 2000; Chu and Chan, 2003; Kumpiene et al., 2017). A range of contaminants in soil can be treated by soil washing including heavy metals (Park et al., 2002; Juhasz et al., 2003; Liao et al., 2016). Soil washing includes an in-situ flushing process with washing solvents followed by extraction of heavy metals by ex-situ process from soil slurry (Reed et al., 1995; Dermont et al., 2008). The effectiveness of this process depends upon the potential of washing solvents to dissolve the heavy metals present in the soil (Gombert, 1994; Dermont et al., 2008). Solvents are used according to the nature of contaminants present in the soil. A range of chemicals are applied as solvents for soil washing tht includes organic acids, chelating compounds, surfactants and cosolvents (Gao et al., 2003; Maturi and Reddy, 2008; Zhang et al., 2009).

5.2. Biological remediation technique of heavy metal-contaminated soil: an environmentally sustainable way

Biological remediation approach that is an environmentally sustainable way to reclaim heavy metal contaminated soil includes bioremediation, phytoremediation and combination of both techniques.

5.2.1. Bioremediation

The principal objective of microbial reclamation of heavy metal polluted soil is to immobilize and reduce bioavailability of metals. Heavy metals cannot be degraded by microorganisms but can be converted to another form due to their altered physical and chemical properties. Efficiency of microbial remediation varies with the type of heavy metal and microorganism. Extracellular complexation, intracellular accumulation, oxidation-reduction reactions and precipitation are microbial remediation mechanisms. Microbial leaching is also a remediation tool for heavy metal contaminated soil (Q. Yang et al., 2016; Z. Yang et al., 2016; Yang et al., 2018). Biosorption is the most important mechanism in microbial remediation. Extracellular materials cause immobilization of heavy metals by binding with anionic functional groups of cell surface (Ayangbenro, and Babalola, 2017; Etesami, 2018; Gupta, and Diwan, 2017). Binding forces between metal ions and cell surfaces include covalent bonding, Vander Waals forces and electrostatic interactions (Blanco et al., 2000).

Siderophores that are chelating agents produced by plants, bacteria and fungi play a significant role in microbial remediation. Siderophores bind heavy metals and thus reduce their bioavailability (Schalk et al., 2011; Saha et al., 2016). Biosurfactants produced by microorganisms have the potential to complex heavy metals (Sarubbo et al., 2015; Ma et al., 2016). Complexation increases the dissolution of heavy metals that results in the reduction of metals and production of less soluble salts of metals. Microorganisms can remove heavy metals from environment through enzymatic or non-enzymatic processes (Nealson et al., 1992; Sarubbo et al., 2015; Ma et al., 2016).

5.2.2. Phytoremediation

The objective of soil reclamation methods is to find a solution that is ecofriendly (Martin and Ruby, 2004). Phytoremediation is also called as agro-remediation, botano-remediation or green remediation. Phytoremediation is a green approach and sustainable way for soil reclamation as compared to conventional soil remediation techniques (Mahar et al., 2016). The concept of phytoremediation was first introduced in 1983 and still this technique is at testing stage. Phytoremediation is a best approach to deal with low to average metal contaminated soils and can be applied in combination with other traditional soil remediation approaches for efficient removal of contaminants (Cunningham and Ow, 1996; Helmisaari et al., 2007; Khalid et al., 2017).

Phytoremediation is an environment friendly mechanism in which fast growing plants are used to eliminate, hold or provide nontoxic contaminants in soil or water (Mahar et al., 2016). It is an aesthetical, economical and environment friendly way to detoxify contaminants (Zhang et al., 2010). Phytoremediation can be successful when used plant species can uptake and store high concentration of metal contaminants in their shoot parts. Phytoremediation technology is grouped into five sub classes including phytostabilization, phytostimulation, phytotransformation, phytofiltration and phytoextraction (Parmar, and Singh, 2015; Ramanjaneyulu et al., 2017).

5.2.2.1. Phytostabilization. Phytostabilization, phytoimmobilization or phytorestoration is plant-based inactivation approach to deal with metal contaminated soil (Singh, 2012; Ramanjaneyulu et al., 2017). The objective of this technique is reduction in the mobility and bioavailability of heavy metals and consequently limits their leaching and entry into groundwater and food chain, respectively (Erakhrumen, 2007; Khalid et al., 2017). In phytostabilization plants play secondary role as compared to soil amendments. This technique involves physical and chemical immobilization of metal contaminants by their sorption onto roots and fixation with different soil amendments (Schnoor, 2000; Wuana and Okieimen, 2011). Organic matter, biosolids, clay minerals and phosphate fertilizers are most efficient soil amendments for immobilization of heavy metals (Flathman and Lanza, 1998). The main objectives of plants are to reduce water percolation, limit contact with contaminants, reduce soil erosion and decrease migration of contaminants (Raskin and Ensley, 2000; Akhtar et al., 2013). Phytostabilization is a management strategy and not a long-lasting way because at the end metal contaminants persist in the soil (Vangronsveld et al., 2009).

5.2.2.2. Phytostimulation. Phytostimulation also referred to as rhizodegradation is the disintegration of organic pollutants in rhizosphere with enhanced microbial activity (Mukhopadhyay and Maiti, 2010). Rhizosphere is soil volume about 1 mm nearby root and is influenced by root activity (Pilon-Smits, 2005). In rhizosphere, microbial activity is enhanced in different ways:

i) Root exudates that contain amino acids and carbohydrates enrich indigenous microorganisms;
ii) Roots ensure the supply of oxygen in rhizosphere for aerobic transformations;
iii) Root biomass enhanced the availability of organic carbon;
iv) Mycorrhizae fungi cause the degradation of compounds that cannot be breakdown by bacteria;
v) Plants provide habitat for enhanced microbial population (Yadav et al., 2010).

5.2.2.3. Phytotransformation. Phytotransformation also termed as phytodegradation is the breakdown of organic compounds either by metabolic processes of plants or enzymes produced by plants and does not depend on microbial community (Vishnoi and Srivastava, 2008;

Kumar et al., 2018). Thus, plants can be considered as green liver of biosphere. Phytodegradation is only beneficial for breakdown of organic compounds. Release of volatile compounds to atmosphere through plant transpiration is termed as phytovolatilization. In this technique plants uptake contaminants from soil, transform them to volatile compounds and then discharge those volatile compounds into atmosphere. This technique is limited to organic compounds and Hg and Se heavy metals (Padmavathiamma and Li, 2007; Kumar and Gunasundari, 2018).

5.2.2.4. Phytofiltration. Phytofiltration is the use of plan roots for reclamation of surface and groundwater and wastewater with low level of contaminants (Mukhopadhyay and Maiti, 2010). Firstly, plants are supplied with contaminated water to acclimate the plants and then these plants are transferred to contaminated site for remediation. When roots become saturated, they are harvested (Zhu et al., 1999). Phytofiltration can be rhizofiltration (using plant roots), blastofiltration (seedlings) or caulofiltration (plant shoots) (Mesjasz-Przybylowicz et al., 2004; Da Conceiçao Gomes et al., 2016). In this technique contaminants become absorbed, adsorbed or precipitated and their leaching to groundwater becomes reduced. Root exudates change rhizosphere pH and can cause precipitation of metals on plant roots (Flathman and Lanza, 1998; Javed et al., 2019). In rhizofiltration both terrestrial and aquatic fast-growing plants can be implemented for the extraction of Cd, Cr Cu, Ni, Pb and Zn. Mostly terrestrial plants are used due to their fibrous and longer root system (Dhanwal et al., 2017).

5.2.2.5. Phytoextraction. In phytoextraction, fast growing plants are implemented to remove heavy metals from the environment (soil and water) (Yanai et al., 2006; Van Nevel et al., 2007; Pajevic et al., 2016). Phytoextraction involves two approaches that include continuous or natural and chemically induced phytoextraction (Ghosh and Singh, 2005). Continuous phytoextraction is an approach which includes the removal of heavy metals by network of roots and then directed to upper plant tissues above the ground (Jadia and Fulekar, 2008) as shown in Fig. 3. Harvested plant biomass can be used for biogas production and can also be combusted. Combusted plant biomass can be used for metal recovery, fixed in bricks or disposed in deserted lands. This technique is also known as phytomining or biomining (Singh and Bhargava, 2017a, 2017b). This approach is best to reduce concentration of metal contaminants from soil by plant roots and shoots without affecting soil properties. Metals can be restored from harvestable plant parts (Garbisu and Alkorta, 2001; Singh and Bhargava, 2017a, 2017b). Continuous cropping and harvesting system can cut down level of contaminants in soil (Vandenhove et al., 2001). In contrast to prevailing remediation techniques, this emerging green technology would be ten times more economical (Wan et al., 2016).

Plants use for phytoextraction should have rapid growth, high biomass and extended roots network and should bear and store high levels of heavy metals (Romkens et al., 2002; Sytar et al., 2016). In continuous phytoextraction natural hyperaccumulators are used. Hyperaccumulator plants are used at metalliferous sites (Lasat, 2002; Ghosh and Singh, 2005; Usman et al., 2018). Phytoextraction involves different processes such as

(i) Some metal fraction becomes sorbed at root surface
(ii) Bioavailable metals enter the roots through cellular membrane
(iii) Small fraction of metals taken up bay roots becomes immobilized in vacuole
(iv) Mobile metals in roots enter the xylem
(v) Metals are translocated from roots to tissues of stems and leaves (Lasat, 2000; Adrees et al., 2015).

6. Use of metallophytes for phytoextraction

Plants that can survive in heavy metal contaminated soil are called as metallophytes (Sheoran et al., 2011). Metallophytes are mostly belonged to Brassicaceae plant family and are categorized into three classes; excluders, indicators and hyperaccumulators (McGrath et al., 2002; Bothe, 2011). Metal excluders uptake heavy metals from environment and accumulate into roots but limit their transport towards above ground plant tissues (Malik and Biswas, 2012). Metal indicators uptake contaminants from contaminated soil and accumulate into their upper parts (Sheoran et al., 2011). Metal hyper accumulator should accumulate at the minimum 100 mg/kg As and Cd, 1000 mg/kg Co, Cu, Cr, Ni and Pb and 1000 mg/kg Mn and Ni (Watanebe, 1997; Reeves and Baker, 2000). Heavy metals hyperaccumulation behaves as a defense action in response to plant infective agents (Boyd et al., 1994). Above 400 plant varieties have been recognized as hyperaccumulators with sluggish growth and less biomass production (McGrath and Zhao, 2003; Kramer, 2018). Field implementation of natural hyperaccumulators can be carried out for both phytoremediation and phytomining of heavy metals and precious metals, respectively (Brooks et al., 1998). Some important hyperaccumulators for heavy metal remediation are presented Table 4.

7. Impediments in the success of phytoextraction

The success of phytoextraction depends upon the accessibility of heavy metals to selected plant roots that is directly related to solubility of metal contaminants in soil solution (Felix, 1997; Antoniadis et al., 2017). Hence, bioavailability is the limiting factor for phytoextraction. The dissolution of heavy metals in soil solution is primarily regulated

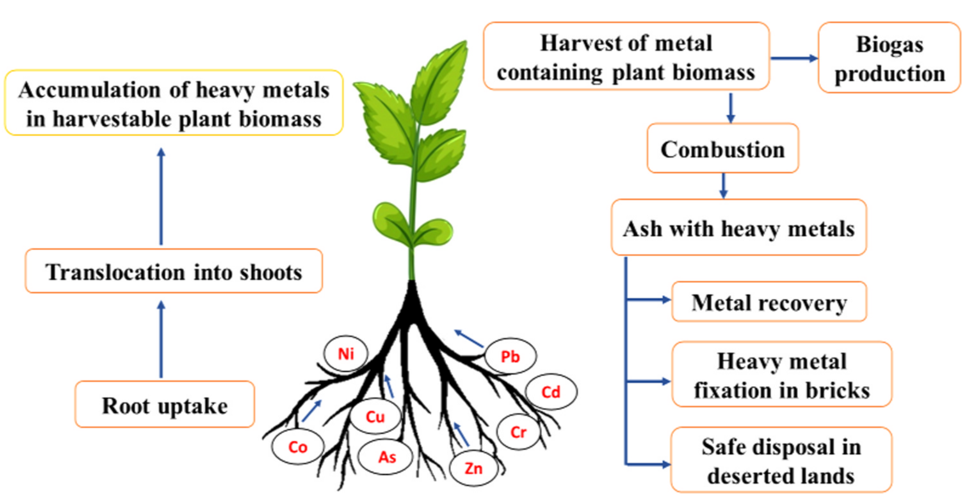

Fig. 3. Phytoextraction process including different management options of end product.

Table 4
Important hyperaccumulators for heavy metals remediation.

Heavy metal	Plant species	Maximum reported concentration (mg/kg)	Reference
Cd	*Azollapinnata, Arabispaniculata*	740, 1127	(Rai, 2008; Zeng et al., 2009)
Cu	*Ipmoea alpine, Eleocharis acicularis*	12,300, 20,200	(Baker and Walker, 1989; Sakakibara et al., 2011)
Pb	*Euphorbia cheiradenia*	1138	(Chehregani and Malayeri, 2007)
Ni	*Alyssum corsicum, Alyssum markgrafii*	18,100, 19,100	(Li et al., 2003; Bani et al., 2010)
Zn	*Thlapsicaerulescens, Potentilla griffithii*	19,410, 19,600	(Banasova and Horak, 2008; Hu et al., 2009)
As	*Pteris vittaia, Pteris ryukyuensis*	23,000, 3647	(Dong, 2005; Srivastava et al., 2006)
Cr	*Phragmites australis, Pteris vittaia*	4825, 20,675	(Calheiros et al., 2008; Kalve et al., 2011)
Mn	*Schimasuperba*	62,412.3	(Yang et al., 2008)

by pH (Baker and Walker, 1989; McNeil and Waring, 1992; Henry, 2000; Javed et al., 2019). High soil pH leads towards greater retention and low dissolution of metals in soil solution, cation exchange capacity and organic matter content (Bliefert, 1994; Li and Shuman, 1996; Tang et al., 2015; Wijayawardena et al., 2015; Zeng et al., 2017). Heavy metals in soil can be categorized into available, exchangeable and unavailable fractions depending upon their uptake by plants (Zhou and Song, 2004; Q. Wei et al., 2008). The successful phytoextraction requires the application of plant species having excessive biomass production, higher growth rate and intense potential to accumulate poisonous heavy metals in their above ground parts (Clemens et al., 2002; Odoemelam and Ukpe, 2008; Kramer, 2018).

8. Enhancement of phytoextraction efficiency

Phytoextraction efficiency can be enhanced by using the following approaches:

- Increasing heavy metal bioavailability in soil
- Increasing plant biomass and decreasing phytoextraction cycle

8.1. Common approaches to increase heavy metal bioavailability

Two common practices which are being implemented to enhance the phytoavailability of heavy metals include the use of synthetic chelates (Tahmasbian and Sinegani, 2016; Chen et al., 2017; Chhajro et al., 2018) and reduction in soil pH (Zhu et al., 2016a, 2016b; Beiyuan et al., 2017; Agrawal and Singh, 2018). The reduction in soil pH can be accomplished by utilization of acids or acid-producing fertilizers (Murtaza et al., 2015; Zhu et al., 2016a, 2016b; Zhao et al., 2016; Agrawal and Singh, 2018). Synthetic chelates such as EDTA enters the plant roots and form soluble complexes with metals which leads towards enhancing the accessibility of heavy metals (Wenzel et al., 2003; B. Jiang et al., 2019; M. Jiang et al., 2019). But these methods have hazardous effects on physical and bio-chemical features of soil (Singh Brar et al., 2015) and can also cause groundwater pollution (Aziz et al., 2015; Q. Yang et al., 2016; Z. Yang et al., 2016; Postigo et al., 2017). A sustainable way to enhance heavy metal bioavailability by lowering soil pH is the application of acidified manure (Ashraf, 2017).

8.1.1. Chelate-assisted or induced phytoextraction

Due to slow growth rate and low biomass of plants, chemically induced phytoextraction has been proposed. In this approach high biomass and fast-growing crops are used to extract huge concentration of heavy metals whose mobility in soil is raised by chelating agents (Smolinska, 2015; Mani et al., 2015; Patra et al., 2018). Efficiency of phytoextraction is restricted by phytoavailability of heavy metals (Felix, 1997). Accumulation of heavy metals can be enhanced in huge biomass production plants for the development of phytoremediation. Heavy metal accumulation potential of non-hyperaccumulating plants can be enhanced by using chelating substances. Chelating agents have been isolated from plants that are used for extraction and detoxification of heavy metals (Saifullah et al., 2009; Wiszniewska et al., 2016).

Enhanced availability and extraction of metal contaminants from soil by plants treated with chelating agents can be linked to different aspects such as (i) an crease in concentration of heavy metals in soil solution (ii) enhanced movement of metal-EDTA complexes towards roots (iii) less binding of formed complexes with negatively charged cell components of plant cell wall (iv) destruction of physiological barriers in roots due to high level of resultant complexes (v) increased mobility of complexes as compared to free ions results in more translocation of metals from roots to shoots (Evangelou et al., 2007). To enhance the potential of phytoextraction process, EDTA (ethylene diamine tetra acetic acid) was used as a chelating compound in early 1990s. EDTA can enhance the accumulation of heavy metals up to 100-fold (Grcman et al., 2001; Ali and Chaudhury, 2016). Both EDTA and EDTA-heavy metal complexes are highly hazardous for plants and soil microbial community (Chen and Cutright, 2001; Vassilev et al., 2004). Biodegradability of EDTA is very low and it also increases the leaching of heavy metals and thus cause ground water pollution (Lu et al., 2017; Qiao et al., 2017).

Ethylene diamine disuccinate (EDDS) is naturally produced by microorganisms and is effective for enhancing uptake of heavy metals (Sidhu et al., 2018). Bioavailability of Cu, Ni and Zn is enhanced more by the application of EDDS as compared to EDTA while that of Cd and Pb is enhanced more by EDTA than EDDS (Meers et al., 2005; Song et al., 2016). Heavy metal-EDDS complexes enter the roots and then transported to shoots (Luo et al., 2005). EDDS is toxic to some plants but not to soil microorganisms. Nitrilotriacetic acid (NTA) is a biodegradable chelating substance and has not phytotoxic effects (Wenger et al., 2003; De Souza Freitas and Do Nascimento, 2009). NTA has been used to enhance the proficiency of phytoextraction by enhancing the bioavailability of heavy metals. After phytoextraction, harvested biomass can be managed by using for phytomining to produce bio-ore (McGrath and Zhao, 2003; Singh and Bhargava, 2017a, 2017b).

8.1.2. Biological sulfur oxidation to reduce pH and enhance metal bioavailability in soil

Positive effect of elemental sulphur on soil metal solubility because of soil pH reduction has been well documented (Ye et al., 2010), as it possesses slow release acidifying characteristic and is readily available (Chien et al., 2011). Application of elemental sulfur in soil has been recommended to reduce pH and enhance dissolution and bioavailability of heavy metals in soil (Tichy et al., 1997; Seidel et al., 1998; Kayser et al., 2000; Li et al., 2017; Sun et al., 2018). *Thiobacillus* bacteria are the most active sulfur oxidizing bacteria in soil (Besharati, 2003; Zhao et al., 2017). Sulfur is the most common and cost effective natural acidifying element. In soil, *Thiobacillus* bacteria oxidize one mole of elemental sulfur and produce two moles of hydrogen ions in soil and thus cause acidification of soil (Kaplan and Orman, 1998). Karimizarchi et al (2016) incubated the soil for 40 days with elemental sulphur at the rates of 0.5, 1 and 2 $g\,kg^{-1}$ soil and observed that pH decreased from 7.51 to 6.66, 5.45 and 4.8, respectively. Soil temperature and moisture level are two important factors that influence the rate of microbial sulfur oxidation in soil. Production of acid in soil by *Thiobacillus* bacteria is also dependent on sulfur to soil total solids ratio (Tsai et al., 2003).

8.1.3. Novel approach to enhance metal bioavailability by reducing soil pH through the application of microbially augmented acidified cow dung

A novel approach to enhance the bioavailability of the heavy metals has been recently reported by Ashraf (2017) and Ashraf et al. (2018). Authors have reported an increase of 114% and 126% in Pb and Cd concentration, respectively, in the shoots of rye grass that has been achieved by application of the acidified product. The bioaugmented acidified cow dung has been applied to soil to enhance the bioavailability of Pb and Cd to grasses used for phytoextraction in pot trials. Cow dung was acidified by adding elemental sulfur (S°) and molasses solution and also bioaugmented with isolated heavy metal resistant sulfur oxidizing bacteria (SOB). Mechanism of cow dung acidification is shown by equation below (Ashraf, 2017; Ashraf et al., 2018).

$$\begin{array}{ccccccc} 2S^{o} & + & 3O_2 & + & 2H_2O & \xrightarrow{SOB} & 2H_2SO_4 \\ Sulfur & + & Oxygen & + & Water & Molasses & Sulfuric\ Acid \end{array}$$

Minimum inhibitory concentration (MIC) of added SOB for Pb and Cd was 1000 mg/L and 180 mg/L, respectively (Ashraf et al., 2018). SOB oxidized (S°) and produced sulfuric acid (H_2SO_4). Acidified cow dung was applied in slurry form at field capacity level for multiple times to grasses during phytoextraction period. Acidified cow dung enhanced the bioavailability of Pb and Cd by reducing the pH of soil. Diluted acidified cow dung reduced the soil pH by 0.92 point and enhanced the concentration of Pb and Cd in shoot of rye grass from 44 mg/kg to 94.32 mg/kg and 34 mg/kg to 77 mg/kg, respectively (Ashraf, 2017). So, (S°), molasses and SOB can be applied to cow dung to acidify the manure that can be used for phytoextraction of heavy metals. Further, composts prepared with a wide range of organic wastes have many useful applications such as soil restoration (Ashraf, 2017).

8.1.3.1. Mechanism how acidified cow dung enhance the bioavailability of heavy metals (Pb and Cd). Acidified cow dung is enriched with nutrients and microorganisms and also of low pH. Both nutrients and microorganisms enhanced plant growth and promoted phytoextraction because for phytoextraction, plant biomass should be high. Acidified cow dung reduced the soil pH for short time period and enhanced the bioavailability of Pb and Cd. Most of the Pakistani soil is calcareous in nature having pH 8 and above. At this pH metals become immobilized due to their absorption on $CaCO_3$ particles. However, metals become available at pH 7. Sulfate ions present in the acidified cow dung react with water and form sulfuric acid (H_2SO_4). When acidulated cow dung is applied, H_2SO_4 reacted with $CaCO_3$ and dissolve it. As a result, $CaSO_4$ was formed that is a fertilizer as shown in equation below. Heavy metal cations released into soil solution taken up by plants. Further, acidified cow dung is a nutrient rich material. So that, cations present in acidified cow dung exchanged the metal cations present on soil colloidal particles such as clay minerals (Ashraf, 2017).

$$H_2SO_4 + CaCO_3 \rightarrow CaSO_4 + H_2O + CO_2$$

8.2. Increasing plant biomass and decreasing phytoextraction cycle

Plant biomass plays very significant function in the removal of heavy metals through phytoextraction. So, the potential of phytoremediation can be improved by the application of fertilizers and proper irrigation system (Jankong et al., 2007; S.H. Wei et al., 2008; Nie et al., 2010). Efficiency of phytoremediation can also be improved by decreasing phytoremediation cycle. Phytoremediation cycle can be decreased by providing specific requirements of used plant species. This objective can also be obtained by shifting the seedlings of specific plant species to the field in order to limit the time period of phytoremediation. This approach can be beneficial because accumulation of heavy metals in shoots of plants is maximum at flowering stage (Wu et al., 2009).

9. Quantification of phytoextraction efficiency

Potential of phytoextraction can be quantitatively assessed by using bioconcentration factor, translocation factor and time of phytoremediation (Zhuang et al., 2005). Bioconcentration factor is a measure of plant's efficiency for concentrating metals into its tissues from the surrounding environment (Ladislas et al., 2012). Translocation factor is a measure of plants's efficiency for translocating the concentrated metals from its roots to shoots (Padmavathiamma and Li, 2007).

Thus, translocation factor is the ratio of concentration of heavy metals in above ground plant tissues to that in its roots (Mattina et al., 2003; Liu et al., 2010). For the selection of natural hyperaccumulators for phytoextraction of metals, both bioconcentration and translocation factors are important (Wu et al., 2011). Translocation factor for natural hyperaccumulators should be > 1 to show that the level of metals is greater in above ground tissues than below ground tissues. Thus, translocation factor is most important for phytoextraction which involves the harvesting of aerial parts of plants (Wei and Zhou, 2004; Karami and Shamsuddin, 2010).

10. Advantages, disadvantages and future trends in phytoremediation

Phytoremediation is the cheapest technology to treat heavy metal contaminated soil (Rakhshaee et al., 2009). This emerging technology is about 60–80% inexpensive as compared to conventional physico-chemical techniques (Mwegoha, 2008). The harvested plant biomass after phytoextraction can be used for bioenergy production (Van Nevel et al., 2007; Tian and Zhang, 2016; Tripathi et al., 2016; Dhiman et al., 2016). Phytoremediation is a promising clean up technology for heavy metal polluted soil. But, this technique also has some disadvantages like slow growth rate of metal accumulating plant species, low bioavailability of heavy metals and long time period of phytoremediation (Naees et al., 2011; Ramamurthy and Memarian, 2012).

Phytoremediation is an emerging field of research and is limited to pot level with few field studies. Results can be different under greenhouse and field conditions (Ji et al., 2011). Phytoremediation process at field level is affected by different factors such as uneven distribution of pollutants, soil pH, pathogens, nutrients, moisture and temperature. For commercialization this technology must be tested at field level (Vangronsveld et al., 2009). For the success of this technology, identification of plants species having high biomass production and heavy metals accumulation capacity is required (Rodriguez et al., 2005).

11. Conclusion

This review revealed that heavy metals are amongst the most critical threats to the soil and human health. These metals are released into the environment through different sources. Conventional remediation techniques are costly and environmentally devastating. Therefore, it is unavoidable to implement low cost and eco-friendly technologies to remediate heavy metal polluted soils. Phytoremediation of metals is the most effective plant-based approach to remove pollutants from contaminated areas without posing any destructive effects to soil structure. Some plants have potential to absorb toxic metals up to several percent of their dried shoot biomass, these plants are known as hyperaccumulators. New plant species suitable for removing heavy metals from contaminated soils should be fined. Metal bioavailability enhancing strategy such as use of bioaugmented acidified manure can be applied to polluted soil to decrease the soil pH and thus to enhance the availability of heavy metals to plants.

Conflicts of interest

The authors declare no conflict of interest.

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