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REVIEW PAPER

# A review on remediation technologies for heavy metals contaminated soil

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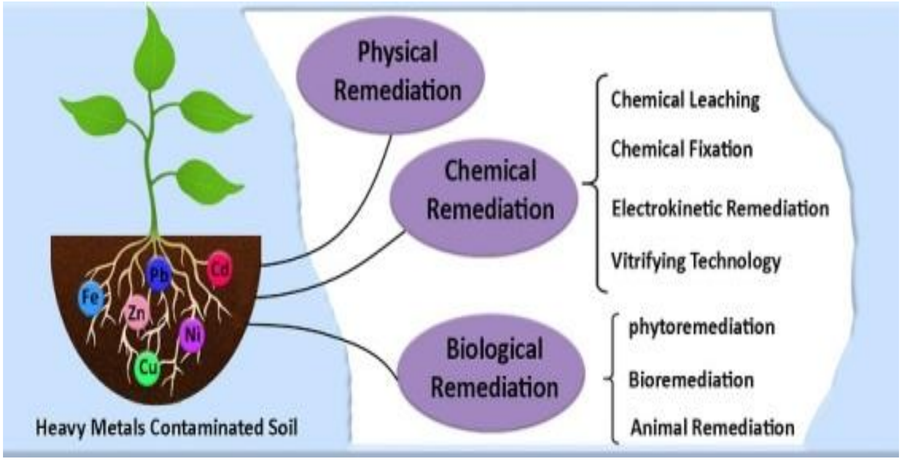
## Highlights

- Due to industrialization and urbanization, environmental safety of soil has become a challenge.
- Biotechnological tools gradually became important technique for the last few decades for removal of metal ions pollution.
- Potassium phosphate is considered more effective in extracting arsenic among various potassium and sodium salts.
- The remediation mechanisms such as extracellular complexation, precipitation, oxidation-reduction reaction

## Graphical Abstract

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## Abstract

The Today, synthetic products such as industrial waste, pesticides, batteries, paints, and industrial or domestic sludge widely applied, as well as and manufacturing can adversely result in heavy metal contamination of urban and agricultural soils. Simultaneously, by growth of industrialization and urbanization, the environmental safety of soil has become of great concerns. Based on investigating the status of soil contamination, the remediation technologies of soil contaminated by heavy metals were focused in the present study. To this aim, physical remediation, chemical remediation and biological remediation were all paid attention. To supply required references to the present study, the mechanisms of remediation, strengths and drawbacks developing trend were discussed. It is proposed that for effective and economic remediation of soil, a better understanding of remediation procedures and the various options available at the different stages of remediation is highly necessary.



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## 1. Introduction

Soil is a natural and dynamic body that formed by soil builder processes and factors, and including minerals and organic materials that covers the outer crust of the earth, that plants are able to grow in it. In ecosystem, soil is considered as basic environmental elements and the important material basis for human being survival and development. However, soil also becomes a way for contaminants entrance into the environment, ignoring contamination derives from air pollution, water pollution or soil pollution itself (Fawzy et al., 2019; Khatun et al., 2019; Li et al., 2019; McBride and Zhou, 2019; Yu et al., 2019). Soil Contamination occurs due to various agricultural and industrial activities including fossil fuel combustion, agricultural use of fertilizers and pesticides, mining waste, and landfill leaching (Almehdi et al., 2019; Azizollahi et al., 2019; Kumari and Dey, 2019). Potential harmful contaminants have been accumulated in the upper soil during thousands of years, starting from the mining for haematite and later for copper (Gong et al., 2019; Kumar et al., 2019; Parlayıcı and Pehlivan, 2019). In terms of their mobility and bio toxicity on living ecosystem, removing metals from soil is an essential task. Soil contamination has become a serious problem with the economy and industry development. Soil contamination by Heavy metal is more serious than other soil contaminations (Demarco et al., 2019; Gómez-Garrido et al., 2018; Guo et al., 2019; Jeelani et al., 2018; Liu et al., 2018). By the accumulation of heavy metals and metalloids, soils contamination may occur. Emissions from the rapidly expanding industrial areas, disposal of high metal wastes, mine tailings, leaded gasoline and paints, land application of fertilizers, animal manures, sewage sludge, pesticides, wastewater irrigation, coal combustion residues, spillage of petrochemicals, and atmospheric deposition are of the principal reasons (Arreghini et al., 2018; Hossain et al., 2018; Khan et al., 2008; Lu et al., 2018; Tampouris et al., 2001; Zhang et al., 2018). Heavy metals are toxic to all organisms if present in high concentrations. The concentrations of heavy metals in soils can vary widely, even in uncontaminated soils. Marked differences in the geochemical composition of the rocks can result in wide ranges of total concentrations of elements in soils, even in soils not contaminated. Nevertheless, concentrations of heavy metals can cause toxicity in soil organisms and susceptible plants dependent to the factors affecting the bioavailability of the elements (Ebadi and Hisoriev, 2017; Galal et al., 2019; Mai et al., 2019; Nan et al., 2019).

The European countries have invested a lot to remediate the contaminated soils (Aresta et al., 2008). In the 1980s, the U.S. Congress has passed the Comprehensive Environmental Response, Compensation and Liability Act (CERCLA), namely superfund program, in order to protect the human health and remediate the environmental pollution. There are many other laws and regulations, such as The Resource Conservation and Recovery Act (RCRA), Superfund Amendments and Reauthorization Act (SARA), emphasize the standard and behavior of soil remediation. From 1982 to 2002 year, the area of remediated land has reached 18.35 million $m^3$. The Britain also passed Environmental Protection Act in the 1990s and in which the second part clearly stated that the principle of polluter responsibility. As compared to the developed countries, the investment and research in the remediation of contaminated soil was not far enough. The prevention of heavy metals contaminated soil is not only needed to control the sources, but also enhance the remediation of contaminated soil (Nicholson et al., 2003).

## 2. Materials and Methods

### *2.1. Physical remediation*

The physical remediation consists of soil replacement method and thermal desorption, as well. The soil replacement is to clean soil to restore or partly restore the contaminated soil in order to dilute the pollutant concentration, increase the soil environmental capacity, and thus remediate the soil (Abumaizar and Smith, 1999; Aresta et al., 2008; Kos and Leštan, 2003). The soil replacement is classified into three classes such as soil replacement, soil spading and importing new soil. Soil replacement is removing the contaminated soil and importing new soil, which is suitable for small contaminated area. Besides, the replaced soil should be treated possibly, or else it will incur the second pollution. Soil spading is deeply digging the contaminated soil, to spread the pollutant into the deep sites and achieving the aim of diluting and naturally degrading. New soil importing is dumping clean soil in place of the contaminated soil, completely from base to surface or mixing to decrease the pollutant concentration. The soil replacement is suitable for soil with small area and polluted severely and which costs a lot (Abumaizar and Smith, 1999; Kos and Leštan, 2003). According to the temperatures, the traditional thermal desorption can be classified into high temperature desorption (320~560 °C) and low temperature desorption (90~320 °C). Some advantages including simple process, mobile devices and reuse ability of the remediated soil are all counted for this technology. A company of mercury collection and service in USA has used this technology for in-situ remediation and developed commercial service. However, the limited factors, such as the expensive devices, long desorption time, limit its application in the soil remediation (Dandan et al., 2007).

### *2.2. Chemical remediation*

#### *2.2.1. Chemical leaching*

Chemical leaching is washing the contaminated soil by using fresh water, reagents, and others fluids or gas (Tokunaga and Hakuta, 2002) that can leach the pollutant from the soil. It was found that phosphoric acid proved to be the most promising as an extractant, attaining 99.9% arsenic extraction at 9.4% acid concentration in 6 h. Sulfuric acid also attained high percentage extraction. For the removal of arsenic from contaminated soil, an environment-friendly and cost-effective extraction method has been studied (Alam et al., 2001; Lee et al., 2007). As a sampled soil, a yellow-brown forest soil was contaminated with arsenic (V) and used. Potassium phosphate was most effective in extracting arsenic among various potassium and sodium salts. Arsenic was proficiently extracted by phosphate solution of pH 6.0 at 300 mM phosphate concentration and at 40°C. Among the extractants, in the wide pH valve range, the EDTA can form stable composite with most heavy metals. A soil washing process was applied to remediate arsenic (As)-contaminated stream sediments around an abandoned mine in Goro, Korea (Ehsan et al., 2007). After 1 h of washing with 0.2 M citric acid, removal efficiencies for fine sediments were >95%. When 0.2 M citric acid was mixed with 0.1 M potassium phosphate, the as removal efficiency increased to 100%. It is well-known that, the effect is almost substandard when single extractant is used as the many different pollutants in soil. This let us join or successively use many different extractants. According to results, in removal of heavy metals from the soil samples, Na2EDTA solutions were highly more efficient than $Na_2S_2O_5$. Na2EDTA preferentially exhibited low impact on chromium removal and extracted lead over zinc and cadmium. Removal of Cadmium and, particularly, zinc, by a 0.01 M Na2EDTA solution was increased remarkably by inclusion of 0.1 M $Na_2S_2O_5$. Hence, mixture of the two reagents may provide an economically optimum solution for certain contaminated soils (Ehsan et al., 2007).

Evaluated the efficiency of a washing process with cyclodextrin together with EDTA for the simultaneous mobilization of heavy metals and PCBs from a field-contaminated soil. These studies confirmed that PCB compounds and selected heavy metals can be coextracted efficiently from soil with three successive washes with the same washing suspension having EDTA and cyclodextrin. However, the chelation agents like EDTA is expensive and the biological degradability is bad. Thus, in order to promote the biological degradability of extractants and reduce the risk of second pollution, biological reagent was used to leach the heavy metals in soil. Biodegradable, synthetic organic chelate ethylenediaminedisuccinic acid (EDDS) was used for washing of soil contaminated with 1350 mg/kg of Pb (Li et al., 2009). Also studied the efficiency of tea saponin on metal removal. The results showed that, the removal of Pb, Cd, Zn and Cu were 6.74, 42.38, 13.07, and 8.75%, respectively when using 7 wt % tea saponin as the extractant. The tea saponin can effectively remove acid soluble and reductive metals, which will greatly reduce the environmental risk.

#### *2.2.2. Chemical fixation*

Chemical fixation is to add reagents or materials into the contaminated soil and to use them with heavy metals to form insoluble or hardly movable, low toxic matters, thus decreasing the migration of heavy metals to water, plant and other environmental media and to achieve the remediation of soil (Abumaizar and Smith, 1999; Kos and Leštan, 2003). Clays, metallic oxides, biomaterials, etc. were used as soil conditioning materials. Hodson et al evaluated the ability of bone meal additions (finely ground, poorly crystalline apatite, Ca10(PO4)6(OH)2 to immobilize pollutant metals in soils and reduce metal bioavailability through the formation of metal phosphates has been evaluated (Hodson et al., 2000). Batch experiments and subsequent extraction of metals from controls and bone meal amended soils using 0.01 M CaCl2 and DTPA indicated that bone meal additions decreased the availability of the metals in the soils. Li et al., 2009 studied the efficiency of sodium bentonite, bentonite and diatomaceous earth on remediation of Cd contaminated soil (Li et al., 2009). The results showed that, the concentration of Cd reduced 21.40, 27.63, 27.24, and 32.30% as compared with the control when the additive amount was 20, 30, 50, and 40 g/kg, respectively. There was also report on the remediation of contaminated soil by attapulgite clay (Hong et al., 2002). Results demonstrates that by adding moderate attapulgite clay, Cd concentration reduce 46% in soil, while the soil productivity and quality was not affected. Zhang et al found that the chemical fixation efficiency of phosphate rock, furfural dreg and weathered coal on the contaminated soil (Zhang et al., 2010). The results showed that three conditioning agents could reduce the concentration of Cu, Zn, Pb, and Cd at some degrees. The chemical fixation could remediate the soil with low concentration contaminant; however, the bioavailability of fixed heavy metals may be changed with the changing environmental condition (Bolan et al., 2003). In addition, the use of conditioning agents could change the soil structure at some degrees and have effects on the microbes in soil.

#### *2.2.3. Electrokinetics remediation*

Electrokinetics remediation is a new remediation technology (Cabrera-Guzmán et al., 1990), in which voltage is applied at the two sides of soil and then electric field gradient is formed. The pollutant was carried to two poles treatment room via electro migration, electro osmotic flow or electrophoresis and then further treated (Virkutyte et al., 2002). It is suitable for low permeable soil, and has advantages of easily install and operate, low cost (Cox et al., 1996; Virkutyte et al., 2002) and not destroy the original nature environment (Cabrera-Guzmán et al., 1990; Hodson et al., 2000; Page and Page, 2002). so can attain the environmental remediation and protect the original ecotope (Cox et al., 1996). However, the direct electrokinetics remediation cannot control the pH value of soil system well, and the treatment efficiency was almost low. The main improved methods include adding buffer solution in cathode and anode to control pH value by using ion exchange membrane to control pH value, to add complexant to improve migration, etc. (Hong et al., 2002). Besides, there is combing other methods to remove the heavy metals, such as electrokinetics remediation combined with iron PRB (Hong et al., 2002). Electrokinetic oxidation/reduction combined remediation, and electrokinetic microbe combined remediation (Zhang et al., 2010).

#### *2.2.4. Vitrify technology*

Vitrify technology is to heat the soil at temperature of 1400~2000°C, the process in which organic matters volatilize or decompose. The steam produced and pyrolysis product was collected by off-gas treatment system. After cooling, the melt forms rock shape vitreous, and make heavy metals lose migration. It was reported that the strength of the vitreous is 10 times higher than concrete. Fossil fuel burning or electrode directly heating supplied required energy for ex-situ remediation and then by arc, plasma and microwave energy is transferred. For in-situ remediation, the heat can be through electrodes inserted into the contaminated soil. In short, this technology can remove the heavy metals and the efficiency was high. However, it is complicated and need lots of energy in the melting, which makes it cost a lot and limited in application (Zhang et al., 2010).

### *2.3. Biological remediation*

The biological remediation includes phytoremediation, bioremediation and the combining remediation.

#### *2.3.1. Phytoremediation*

From a global perspective, after the weather, the soil is considered to be the major component of the human environment. Soil not only is the major location for many of land creaturesIn, especially human societies, but also is a unique environment for the life of all species, especially plants. Unlike climate, soil contamination by chemical composition is not easily measurable and there is not a certain definition for pure soil, so we should study potential soil contamination issues in the context of predicting of hazards and potential damages in the soil function.phytoremediation is to use living green plants to fix or adsorb contaminants, and cleaning the contaminants or making their risk reduction or loss. The phytostabilization, phytovolatilization, and phytoextraction are the main three types of phytoremediation (Hong et al., 2002). Phytostabilization stabilizes heavy metals by plants thru adsorption, precipitation and reduction of root. So that, root bioavailability reduce and prevent its migrating into the groundwater and food chain (Hong et al., 2002). Phytovolatilization is transferring the heavy metals into volatile state or adsorbing the metals and transferring into gaseous matter, using special matters secreted by root (Watanabe, 1997). Mercury is the most studied heavy metals. To explore the potential of plants to extract and detoxify mercury, (Bizily et al., 1999) engineered a model plant, Arabidopsis thaliana, to express a modified bacterial gene (merBpe) encoding organomercurial lyase (MerB) under control of a plant promoter. Transgenic plants expressing merBpe grew dynamically on an extensive range of concentrations of monomethylmercuric chloride and phenylmercuric acetate. Plants lacking the merBpe gene were strictly reserved or died at the same organomercurial concentrations. This work suggested that native macrophytes (e.g. trees, shrubs, grasses) engineered to express merBpe may be used to degrade methylmercury at polluted sites and sequester Hg (II) for later removal. However, this technology is only suitable for volatile contaminants, and the application is limited (Hodson et al., 2000). Phytoextraction is adsorbing the heavy metals using tolerant and accumulating plants, and then transferring, storing at the overground parts. Studying the adsorption characterization of different plants and screening high uptake plants is the key of this technology (Table 1).

**Table 1.** Some plant species that used in the phytoremediation process with the ability to absorb some heavy metals**.**

| Plant Family | Plant Species | Metal |
|---|---|---|
| Araceae | *Pistia stratiotes* | Ag, Cd, Cr, Cu, Hg, Ni, Pb, and Zn |
| Asteraceae | *Helianthus indicus* | Pb |
| Fabaceae | *Sesbania drummondii* | Pb |
| Araceae | *Lemna gibba* | As |
| Solanaceae | *Solanum nigrum* | Cd |
| Brassicaceae | *Thlaspi caerulescens* | Cd |

### 2.3.2 Biological remediation

The microorganisms cannot degrade and destroy the heavy metals, but can affect the migration and transformation through changing their physical and chemical characterizations. The remediation mechanisms include extracellular complexation, precipitation, oxidation-reduction reaction and intracellular accumulation. An applied simple technology for extracting precious metals from low-grade ores and mineral concentrates is Microbial leaching. Moreover, microbial leaching can be potentially applied in remediation of mining sites, detoxification of sewage sludge, treatment of mineral industrial waste products, and remediation of soils and sediments contaminated with heavy metals (Bosecker, 2001). Lambert et al., studied the effects of sludge on mycorrhizal (MR) uptake of P, Cu, and Zn, and confirm MR supression of Cu and Zn uptake by P. Sludge reduced uptake at 150 mg/kg P or higher in nonmycorrhizal (NMR) plants with little difference in plant growth among sludges (Lambert and Weidensaul, 1991).

In MR treatments, growth and P-uptake responses to sludge ranged from very beneficial with two sludges to a complete inhibition of the MR response with another sludge. Mycorrhizae substantially increased shoot Cu and Zn uptake only at low soil-P levels. The study of Jones et al indicated that, uptake of Cd from hyphal compartments was higher in mycorrhizal than in non-mycorrhizal plants, corresponding to 96, 127, and 131% of that in non-mycorrhizal plants when 1, 10, and 100 mg Cd $kg^{-1}$ was added, respectively. A large proportion of the increased Cd content of mycorrhizal plants was sequestered in the roots. Consequently, Cd can be transferred from soil to plants by extra radical hyphae of AM fungi, but a reverse transfer is restricted due to fungal immobilization (Joner and Leyval, 1997). However, the biological remediation is susceptible to affected by different kinds of factors, such as temperatures, oxygen, moisture, pH value. It is also limited in applications; such as some microorganisms can only degrade special contaminants, microbes/zymin maybe incur secondary pollution.

### *2.3.3. Animal remediation*

Indeed, animal remediation is distinguishing of some lower animals adsorbing heavy metals, and then degrading and migrating the heavy metals and thus removing and inhibiting their toxicity. The studies showed that, the treatment of the earthworm-straw mulching combinations enhanced plant Cu concentration, and the amount increased by it was lower than that of the earthworm treatment but higher than that of straw mulching treatment (Lambert and Weidensaul, 1991). Zhang et al., studied the use of single extraction methods to predict bioavailability of heavy metals in polluted soils to rice. The results showed that, heavy metals in soil can be measured (Zhang et al., 2010). The accumulation amount increased with the Pb concentrations increasing.

## 3. Conclusion

The research of remediation technologies is still in individual and experimental stage. The development strategy of future remediation technologies is researching green, environmental-friendly biological remediation, combining remediation, in-situ remediation. To this aim, completely quick remediation, and supplying technical supporting for agricultural soil contamination, industrial enterprises Brownfield, mining sites, etc. are to be applied. Soil pollution could affect crop productivity and human health. Investigating the sources, fate and occurrence of soil pollution, and the risks posed to human health has thus been an important area of research.

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