

# Remediation of soil contaminated with heavy metals-A Review

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**Abstract** - Soil contamination is a major problem arising worldwide as result of speedy urbanization and industrialization. The unorganized discharge of domestic and industrial effluent contaminates soil and water with variety of toxic chemicals. The presence of toxic metals in soil and water are hazardous for ecosystem. Chemically contaminated soils are not suitable for agriculture and construction purposes due to their poor physical and chemical properties. Such soils need remediation before use for any application. Various methods are available for decontamination of such soils for soil improvement. However, methods for removal of toxic heavy metals need more research inputs. The Chemical remediation of soil contaminated with hazardous heavy metals by washing with chelating agents was reported by many researchers. This review presents the different methodology adopted by researchers for remediation of soil contaminated with heavy metals.

**Key Words:** Soil Contamination, Soil Remediation, Toxic Heavy Metals, Soil Washing, Chelation.

## 1. INTRODUCTION

Soil and water get polluted with heavy metals due to industrial processes and improper disposal of garbage [1,2]. Arsenic, cadmium, chromium, copper, lead, nickel and mercury are generally present in polluted soil and water. These heavy metals are toxic to human health and ecosystem [3,4]. Heavy metal may get swept away from contaminated areas to new areas by wind and rain. Prevention of contamination of soil with heavy metal is very essential because cleaning of contaminated soils is very difficult and expensive.

Methods for remediation of contaminated soil for reducing the harmful effects of heavy metal include excavation, in situ remediation, phytoremediation and chemical extraction [5,6].

## 2. METHODOLOGIES

Different methodologies of soil remediation are discussed here based on literature survey. Excavation and removal of the contaminated soil is a rapid ex situ remediation method by which complete removal of the contaminants is possible [10]. Disadvantage of excavation method is that the contaminants are simply moved to a different place, where they will contaminate another site as well as during removal and transport of contaminated soil there is risk of

contaminated soil falling to other places. Excavation is a costly process and cost increase further if large area is to be excavated.

Some in-situ remediation methods are being used which do not remove heavy metal but immobilize them in soil [7]. On increasing pH of soil to 6.5 or higher, solubility of cationic metals get decreased which makes them less available to plants thus blocking their entry in food chain but on increasing pH, solubility of anionic metals is increased which makes them more available to plants. Drained soil have improved soil aeration which allow metals to get oxidized and make them less soluble in water thus less available to plants but chromium is more available to plants in oxidized forms. Availability of chromium to plants is reduced on getting it treated with organic matter.

Availability of cationic metals to plants get reduced on application of phosphate to contaminated soil because it react with cationic heavy metal to form metal-phosphate compounds. Due to extremely low solubility potential values of metal-phosphate compounds, these compounds do not enter the food chain by getting absorbed into plants as well as these compounds does not get leached to the ground water. Phosphate-based metal stabilization is a very rapid and easy to apply process. Phosphate reagent can be applied in a wet or dry form and can be used to stabilize metals in situ or ex situ within 24 to 48 hours of application. Radionuclides such as thorium, uranium, radium and cesium were also converted to safe and stable metal-phosphate compounds. Waste streams toxicity levels (leachability) of metal phosphate compounds of Hg, Se, Zn, Pb, Ba, Cr and Cd are reported to be below Universal Treatment Standard (UTS) and Resource conservation & recovery act (RCRA) regulated levels so these metal phosphate compounds can be left on the site rather than transported off the site to a hazardous landfill. Cost of in- situ fixation of heavy metal is about half the cost of excavation and disposal of soil contaminated with heavy metal because it saves on transportation and disposal cost [8,9].

EPA (Environmental protection agency of USA) approved phosphate-based chemical stabilization process is commercially available non-hazardous method that permanently immobilize a wide range of heavy metals. The treated soils contain extremely stable metal-phosphate compounds which are stable to long-term pH related deterioration. EPA's Toxic Characteristic Leaching Procedure (TCLP) test and Multiple Extraction Procedure (MEP) test

measure long-term stability of metal-phosphate compounds because it simulates the long term leaching effect of environmental exposure of stabilized metals. Ten MEP extractions simulate 1,000 years of metals stability and each TCLP extraction simulates 100 years of stability. MEP, TCLP, Synthetic Precipitate Leaching Procedure (SPLP), redox potential, bioavailability and Germany's DIN (Deutsches Institute fur Normung) leachate test established the effectiveness of this metals stabilization process.

Availability of anionic metals such as arsenic to plants get increased on application of phosphate to the soil so anionic metals can not be fixed by phosphate-based chemical stabilization process. As is clear from the discussion, suitable immobilization technique can be applied only if nature of heavy metal present in soil is known.

Phytoremediation of contaminated soil for removal of heavy metals or converting heavy metals to harmless form is done by using vegetation and microbiota. Phytoremediation at a contaminated site can be possible only if plant used for phytoremediation can survive in that climate and is tolerant to presence of heavy metals in soil. Cost of phytoremediation is less than the cost of excavation or in situ fixation but process of phytoremediation take longer time to accomplish than other treatment methods [12]. Phytoremediation is very efficient when heavy metals in low to medium concentrations are spread over a wide area. Phytoremediation can be used in conjunction with other remedial methods.

Chemical extraction or soil washing is ex-situ technique in which an aqueous solution of extracting agent is added to soil contaminated with heavy metals [11]. Soil washing is very cost-effective and ensures rapid cleanup of a contaminated site by completely removing heavy metals [10,13].

Washing of soil with water can remove only small amount of heavy metals because they occur in sorbed state and are sparingly soluble in water [14,15]. For effective removal of

heavy metals, some chemicals are to be added to the washing water which can transfer heavy metals from the soil into solution [16]. Chemical used for soil washing must be capable of breaking strong bonds between the soil and metals and dissolving the metals. Surfactants, cyclodextrins, chelating agents and acids are being used for soil washing [17-23]. Suitability of chemical used for effective soil washing is dependent on type of soil and heavy metal. The identification and quantification of heavy metal in the soil before and after soil washing is necessary to design efficient soil washing procedure [24]. Strong acids (nitric, hydrochloric) used as extracting agents for soil washing dissolve clay mineral and degrade the soil crystalline structure therefore organic acids (oxalic, citric, formic, acetic, succinic, maleic, lactic, fumaric acids) and chelating agents (ethylene-diaminetetraacetic acid (EDTA),

diethylenetriaminepentaacetic acid (DTPA), nitrilotriacetic acid (NTA), citric acid, ferric chloride) are being preferred as alternatives to mineral acid [25-31].

When soil contaminated with heavy metal is washed with aqueous solution of chelating reagent, it bind with metal to form complex. Complex of chelating reagent and heavy metal is also called metal chelate which is soluble in water therefore metal get removed from the contaminated soil [32-37]. EDTA is widely used for washing of contaminated soil because of its abilities to form water soluble complexes with almost all heavy metal [38-47].

Ferric chloride ( $\text{FeCl}_3$ ) and EDTA were used by Bilgin et al. [48] to extract Cu, Cd and Zn from the contaminated soil. After 1 h of soil washing, 90.57% of Cd, 73.22% of Cu and 99.59% of Zn was removed from contaminated soil using 0.01 M EDTA as washing solution with liquid/soil ratio of 20. Udovic and Lestan reported that 73% of Pb, 23% of Zn and 74% Cd was removed from contaminated soil by using EDTA as washing solution [49].

The best removal efficiencies for Cd, Cu and Zn were obtained when soil was washed with 0.01M  $\text{FeCl}_3$  as washing solution with liquid/soil ratio of 20 on rotary shaker at  $200 \pm 5$  rpm for 2 h. On increasing washing time from 0.5 to 2 h percent removal of Cd increased from 83.14 to 98.18. On increasing washing time to 2h percent removal of Cu and Zn also increased to 97.48% and 98%. Lim et al. [50] reported that 95% of Pb, 89% of Cd and 90% of Ni was removed from contaminated soil.

Soil washing with EDTA and  $\text{FeCl}_3$  solutions is environment friendly and due to high liquid/soil ratio of 20, removal efficiency for removal of Cd, Cu and Zn from contaminated soils got improved. Removal efficiency of EDTA for removal of Cd, Cu and Zn from contaminated soil was lower than that of  $\text{FeCl}_3$  but EDTA provided shorter washing time than  $\text{FeCl}_3$  for the removal process.

Extraction of Cd from contaminated soil by soil washing with aqueous solution of Calcium chloride and  $\text{FeCl}_3$  is reported to be very efficient, cost-effective and environment friendly [51-53]. Makino *et al.*, reported that soil washing with  $\text{FeCl}_3$  was more effective than with calcium chloride or hydrochloric acid [52].

Ke et al. reported that tartaric acid is an environmentally-friendly extractant for remediation of heavy metal contaminated soils. Tartaric acid, in the pH range of 3.5-4.0, was effective for removal of 50%-60% of Cd, 40%-50% of Pb, 40%-50% of Cu and 20%-30% of Zn from the contaminated soil within 24 h. Higher mass transfer coefficients for Cd and Pb than those for Cu and Zn made the removal of Cd and Pb more efficient [54]. Labastida et al reported that 44% lead was removed from soil on remediation with EDTA. Soil after remediation was having a final concentration of  $363.50 \pm 43.50$  mg/kg which is below USEPA standard [55].

Jiang et al. extracted copper and nickel from contaminated soil by washing it with chitosan (biodegradable chelating agent), EDTA and sodium citrate [56]. Extraction efficiency of three chelating agents for removal of copper and nickel from the contaminated soil decrease in the order : chitosan > EDTA > sodium citrate. Extraction efficiency of any of the chelating agents was higher for nickel than for copper which is consistent with the relative stability of chelate complexes of these two metals. Efficiency of metal extraction by soil washing is dependent on pH of solution of chelating agent. Extraction efficiency of chitosan solution for removal of copper and nickel from the contaminated soil increased with decreasing its pH and extraction was most efficient when its pH was 3 - 3.5. Availability of heavy metal is increased in acidic condition. At pH 3-3.5, 43.36% of copper and 37.07 % of nickel were extracted from contaminated soil. Extraction efficiency of heavy metal is increased on increasing concentration of chelating solution. Percentage removal of nickel from contaminated soil increased from 56.22% to 63.34% when concentration of chitosan solution increased from 0.1- 0.4 g/l. The removal of copper from contaminated soil increased from 30.99% to 43.49% on increasing concentration of chitosan solution from 0.1 to 0.3 g/l and percentage of copper removal did not increase on further increasing the concentration of chitosan solution beyond 0.3g/l. After 2 h of washing of contaminated soil with chelating solution, 38.72% of copper and 51.25% of nickel were removed. Extending the reaction time from 2 h to 8 h did not affect the extraction efficiency significantly. A reaction time of 2 h was chosen as the optimal reaction time for extraction of copper and nickel. Extraction efficiencies of copper and nickel increased on increasing liquid/soil ratio. When chelating solution/soil ratio was increased from 5 to 20, the removal of copper increased from 26.34% to 47.22%, while the removal of nickel increased from 41.75% to 66.13%. Liquid/soil ratio was increased by adding same concentration (0.2 g/L) of chelating solution to contaminated soil which means that chitosan amount was increased on increasing liquid/soil ratio. High liquid/soil ratio can prevent clogging of the soil during washing [33] but generate large amount of wastewater, which would increase the cost soil washing. Optimum condition for removal of nickel and copper from contaminated soil were obtained by adding chitosan solution of concentration of 0.3 g/l and liquid/soil ratio of 10 ml/g to 1 g of soil sample and were agitated at a speed of 120 rpm at room temperature for 2 hours.

The removal of toxic metals such as Pb, Zn, Cu and Cd by soil washing with chelators is a viable remediation option. Ethylenediamine tetraacetate (EDTA) is by far the most tested, strong chelator that effectively removes toxic metals from the soil. There is concern that EDTA-toxic metal chelates could leach from remediated soil and contaminate the groundwater [57]. Readily biodegradable chelators such as N,N-bis (carboxymethyl) -l- glutamate (GLDA), iminodisuccinate (IDS), 3-hydroxy-2,2'-iminodisuccinic acid (HIDS), S,S ethylenediamine-disuccinate (EDDS),

methylglycine diacetate (MGDA), and nitrilotriacetate (NTA) have been introduced into soil washing and are being tested as possible substitutes [58-60].

### 3. CONCLUSIONS

Different approaches have been proposed in the literature to assess the sequestering ability of ligands toward metal cations. This review aims to give a methodological contribution in the choice of chelating agents for *soil washing remediation* based on literature survey. The methods here proposed allows determining both the suitability of the ligand for scavenging the target metal ions, and the minimal concentration of ligand to be used, minimizing the adverse effects on essential metal ions. This is particularly relevant when studying the real application of a washing ligand to a specific soil, knowing the total concentrations of metals to be removed before the experimental determination of its behavior. The remediation of metal-contaminated soils using synthetic chelates for soil washing and for enhancing phytoextraction by plants has become one of a number of well studied clean-up techniques in the last two decades.

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