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## An Experimental Study of Heavy Metal Extraction, Using Various Concentration of EDTA in a Sandy Loam Soils

<sup>2</sup>D. Naghipoor Khalkhaliani, <sup>1</sup>A.R. Mesdaghinia,

<sup>1</sup>A.H. Mahvi, <sup>2</sup>J. Nouri and <sup>2</sup>F. Vaezi

<sup>1</sup>School of Public Health, Center for Environmental Research,  
Tehran University of Medical Sciences, Tehran, Iran

<sup>2</sup>School of Public Health, Tehran University of Medical Sciences, Tehran, Iran

**Abstract:** This study provides an evaluation of EDTA solution for the removal of lead, zinc and cadmium from a contaminated soil. The field soil contained 68% sand, 12% clay and 20% silt. The performance of EDTA for the treatment of soil contaminated with heavy metals was evaluated in this study. Soil samples containing variable levels of Pb, Zn, Cd were subjected to Ethylene Diamin Tetra-acetic Acid (EDTA) treatment and the extraction of heavy metals was found to vary, ranging from 54.5 to 100%. Thus the feasibility of soil washing for the decontaminated sandy-loam soil with single and several metals were evaluated in laboratory-scale batch experiments. Of the washing reagent test, Na<sub>2</sub>-EDTA 0.1 M solutions were generally more effective for removing heavy metals from soils. Na<sub>2</sub>-EDTA 0.1M preferentially extracted lead over cadmium and zinc. The most efficient washing occurred using the 0.1M EDTA at the lowest pH.

**Key words:** Chelating agents, heavy metals, EDTA, soil remediation, Na<sub>2</sub>

### INTRODUCTION

Soils are effective agents for metal sorption because of their high surface area and the presence of various surface function groups (Gong and Donahoe, 1997). Soil properties, especially the content of Fe, Al and Mn oxides and hydroxides and the content of clay and organic matter, are believed to be the main controls of heavy metal sorption (Gong and Donahoe, 1997; Tessier *et al.*, 1995; Young and Harvey, 1992). Heavy metals in soils are receiving increasing attention due to greater understanding of their toxicology importance in ecosystem and for human health (Ottosen *et al.*, 2001). Heavy metals will continue to be an environmental concern for a long time unless they are taken out from the ecosystem (Chen *et al.*, 2000). Heavy metals can be transferred by the intake of vegetation and human health can be impacted through ingestion of both water and foods that have been contaminated by the soil. Heavy metals are toxic to human as well as to other organisms (Chen *et al.*, 2000). Contaminated sites are also pose a threat to groundwater supplies if the metals are not properly contained and treated. Heavy metals interact with soil matrix and may persist for a long period of time creating long-term hazards to the environment and human health. A promising method for removing heavy metals from soil is chelating extraction (Lo and Yang, 1997). Metals are particularly troublesome because they can

accumulate in soils through adsorption, precipitation and other physico-chemical processes and the presence of very small concentrations can sometime pose a serious health threat to living organisms. Clean up of soils contaminated with trace metals is one of the most difficult and expensive goals in environmental engineering. Soil washing is considered as one of the most suitable on-site techniques for removing trace metals. Recently increased attention has been focused on the use of acidic or chelating agents to dislodge and dissolve trace metals from solids into solution. Citrat acid, Nitrilotree Acetic Acid (NTA) and EDTA are the common alternatives suggested for chelating polluted soils (Zeng *et al.*, 2005; Papassipi *et al.*, 1997). Clearly, action should be taken to control the contamination of soils (i.e. particle size or gravity separation, flotation, etc.) or to employ chemical methods involving the application of appropriate leaching solution to the contaminated soil. Metals can be removed by physical methods. These techniques is to separate the metals from soil by using chelating agents to form soluble metal-chelate complexes. Technologies available for re-mediating metal contaminated soils can be divided mainly into two groups, namely, immobilization methods and separation/concentration methods. In the first type of remediation, contaminants are immobilized thereby preventing the leaching of contaminants into the groundwater. The second type of remediation deals with separating the containment from the soils or reducing the

volume of contaminated soil (Khodadost *et al.*, 2005). Soil washing is a variable treatment alternative for metal contaminated sites. Chemical extractions are some times introduced in the washing fluid to enhance the efficiency of heavy metal extraction. These extractions include acids, bases, chelating agents, electrolytes and redox reagents. Process parameters in soil washing include the mode of extraction (e.g., batch or column), extractant type and concentration, pH, electrolyte concentration, liquid -to-solid ratio (L/S) and retention time. The soil related parameters are pH, particle size distribution and mineral type of metal to be extracted and their concentration, distribution and physicochemical forms in the soils. The kinetics of metal desorption/dissolution is also a crucial parameter as it can affect the treatment duration and cost (Tandy *et al.*, 2004; Lim *et al.*, 2004; Reed *et al.*, 1996). Using EDTA to remove heavy metals has been proven to be an effective method due to strong complexing ability of EDTA (Samani *et al.*, 1998). Even though EDTA has shown to be a suitable chelating agent for remediation of heavy metal, not much information is available with high heavy metal removal efficiency. The objectives of this research were to further investigate the feasibility of EDTA to achieve high removal efficiency of heavy metals from polluted soils. The performance was evaluated for different soil samples containing variable levels, from 500-1000 mg kg<sup>-1</sup> in Pb, Cd and Zn. The advantages of using chelating agents in soil washing is high efficiency of metal extraction, high thermodynamic stabilities of the metal complexes formed, good solubility of metal complexes and normally low adsorption of the chelating agents and their metals complexes on soils (Abumaizar and Smith, 1999; Peters, 1999; Tandy *et al.*, 2004).

**MATERIALS AND METHODS**

The Sandy-Loam soil, which has been sampled from roadside along agricultural lands of Gilan province in Iran, was selected for this study. This research was conducted for a year in the Department of Environmental Health Engineering and Department of Chemistry of Tehran University in 2004. The surface soils, (0-20 cm) were sampled, air dried, ground, passed by a 2 mm sieve and stored in plastic bags prior to laboratory analysis and use in batch experiments. Soil pH was determined using glass electrodes in a soil: water ratio of 1:1 and the particle size distribution of soils were analyzed by the Pipete method (Gee and Bauder, 1986). Organic carbon was determined by the Walkley-Black wet combustion method (Rhoades, 1982). Exchangeable cations and Cation Exchange Capacity (CEC) were determined using ammonium acetate at pH 7. (Rhoades, 1982).

Table 1: Selected properties and concentration of the Cd, Zn and Pb in unpolluted field soil

Soil properties	S-L
pH H <sub>2</sub> O	6.50
OC (%)	3.12
Particle Size analysis (%)	
Sand	68.00
Silt	20.00
Clay	12.00
CEC	
Meq/100 g	17.80
Total content (mg kg <sup>-1</sup> soils)	
Cd	0.82
Zn	78.30
Pb	38.90
Ca	4194.00
Mg	106.00

**Soil preparation:** Since the concentration of heavy metals in soil samples is usually less than 100 mg kg<sup>-1</sup>, hence the soil samples have been contaminated artificially (Table 1).

Several solutions containing Zn, Cd and Pb were prepared by using acetate of these metals. These solutions were added to a portion of the mentioned soil sample at a rate of 100 mL/10 g (solution/solid) in centrifuge tubes. Therefore, the soil sample received 500 mg kg<sup>-1</sup> or 1000 mg kg<sup>-1</sup> heavy metals. The suspension samples were then placed on a mechanical shaker operated at 180 rpm and at a room temperature (27-30°C) for 48 h.

The wet-aging stage was essential to ensure complete and even exposure of every soil particle to contamination. At the end of the 48 h wet-aging period, the suspension was centrifuged to separate the solid phase from solution. The supernatant liquid phase from the centrifuge tubes was filtered and the equilibrium concentration of heavy metals in the liquid was measured using Atomic Absorption Spectrometer (AAS). The pH of the solution was measured and all the contaminated samples pH was about 7. The contaminated soil was then washed with DI water to remove the entrapped water in the soil. So an artificially contaminated soil sample was used in this study. The advantage of deliberately contaminating the soil is that a rather homogeneous test sample, with consist heavy metal concentration and specification, soil composition, contamination process and contamination period can be obtained. This would minimize ambiguity in the extraction of potential results arising from sample heterogeneity (Elliot and Shasteri, 1999).

**Batch extraction with EDTA:** Batch extraction experiments were conducted using an EDTA solution at various concentrations and contact times to determine appropriate range of concentration and dosage of the washing solution to achieve high heavy metal extraction efficiencies. The extraction solution was prepared from

Table 2: Remediation efficiency vs. reaction time of contaminated Sandy -loam soil with 500 mg kg<sup>-1</sup> Conc.

	Time (h)									
	0.5	1	2	3	4	6	12	24	36	48
Pb	99.6	99.8	100.0	100.0	100.0	99.6	100.0	99.8	99.6	100.0
Zn	84.3	84.5	84.2	84.6	84.5	85.1	84.6	84.0	85.2	85.0
Cd	80.3	80.2	80.3	80.7	80.3	81.2	81.1	81.3	82.2	82.3

reagent-grade disodium ethylenediaminetetraacetat (Na<sub>2</sub> EDTA). The EDTA concentration was 0.005, 0.01 and 0.1 M and the pH values of the three solutions were 4.7, 4.53 and 4.46, respectively. The contact time for metal extraction was 2 h. All the experiments were performed at room temperature (27-30°C). A soil sample of Teng used with a 2.5:1 liquid/solid ratio was employed for all soil extraction experiments. Ten gram of soil sample and 25 mL of EDTA solution were added to a polyethylene centrifuge tube. The tube was sealed with a lid and then placed on a mechanical shaker operated at 180 rpm at room temperature for the desired contact period. Two hours reaction time was deemed sufficient based upon batch rate desorption tests conducted over 48 h period. After mixing, the samples were allowed to settle for about 15 min and then centrifuged and filtrated through a 0.45 µm member filter. The pH of the washing solution before contact with the soil and the pH of the filtrate were measured and recorded. Following filtration, the filtrate was acidified to a pH of <2.0 with 1:1 HNO<sub>3</sub> for heavy metals analysis. It was assumed that the metal concentration of the filtrate was released from the contaminated soil. Remediation efficiencies were determined by dividing the heavy metal release quantities to the initial quantity in the soil. All heavy metal analysis performed by using a Perkins-Elmer Atomic Absorption Spectrometer (AAS). DI Water was performed to provide a baseline for the removal obtained by chemical washing.

### RESULTS AND DISCUSSION

Based on this study and the results shown in Table 2 a reaction time of 2 h was considered suitable since the curves illustrate that no appreciable amount of metal was extracted beyond this time. Table 2 represents the remediation efficiency of the contaminated soils in 0.5, 1, 2, 3, 4, 6, 12, 18, 24, 36 and 48 h time periods for three types of metals Cd, Zn and Pb. The vacillation of remediation process results usually occurs in initial hours and then the efficiency will follow a constant rate, therefore the optimum time period, which is used as a bench mark for comparison, was 2 h. Apparently Pb, Zn, and Cd release were very rapid such that reached an equilibrium within 1-2 h of extraction time. Table 3 and 4 represent the remediation efficiency by various rates of EDTA concentrations at pH original (about 7) on a

Table 3: Remediation efficiency (%) of single metal-contaminated soil (Sandy-loam) with Conc. 500 mg kg<sup>-1</sup> using various M EDTA

EDTA	Pb	Cd	Zn	pH
0.1	100.0	79.3	84.7	7
0.01	90.9	77.9	82.4	7
0.005	71.8	73.5	80.1	7

Table 4: Remediation efficiency (%) of multi metal-contaminated soil (Sandy-loam) with Conc. 500 mg kg<sup>-1</sup> using various M EDTA

EDTA	Pb	Cd	Zn	pH
0.1	99.3	79.2	83.1	7
0.01	88.1	78.0	83.0	7
0.005	61.2	69.3	70.7	7

Sandy-Loam soil samples with the contamination rate of 500 mg kg<sup>-1</sup>. The results represent the cardinal effect of EDTA on Pb; meanwhile the increase of the EDTA concentration in Zn and Cd had not a considerable effect on the remediation efficiency (Table 3 and 4). According to Table 3 the remediation efficiency for Pb with the concentration of 0.1 M EDTA was 100% and for the EDTA with the concentration of 0.005 M the efficiency was just 71.8%. A 0.1 M EDTA exhibited complete removal of Cadmium and Lead for a soil to solution ratio of 1:12.5 (Abumaizar and Smith, 1999). Soil flushing tests performed (Palma *et al.*, 2003) on a sandy loam showed that complete Pb removal can be achieved by using four pore volume (4PV) of a solution containing 0.01 M EDTA. Other research results showed that a higher concentration of EDTA and PV were necessary because of the high organic content of soil used in the experimentation. Heavy metal retention by soil is in fact, strongly affected by its natural organic matter concentration (Palma *et al.*, 2003).

Also the effect of EDTA concentration on remediation process of mixed soil (multimetal-soil) was investigated and the results showed that the maximum and minimum efficiencies for the Pb were with EDTA 0.1 M and 0.005 M, respectively, (Table 4). It means that EDTA in single-metal contaminated soils is more efficient than in a multi-metal contaminated soil which this is possibly because of the greater ratio of the EDTA to the metal. Kim *et al.* (2003) reported that lead extraction efficiency was a function of the stoichiometric ratio of the applied EDTA concentration to the total lead concentration in the soil sample.

The excess dosage would ensure that there were always sufficient molecules of chelating agents available to these heavy metals, even though some molecules of

Table 5: Remediation efficiency (%) of single metal-contaminated soil (Sandy-loam) with Conc. 500 mg kg<sup>-1</sup> using 0.1 M EDTA

EDTA	Pb	Cd	Zn	pH
0.1	100	82.4	84.1	3
0.1	100	81.3	84.0	5
0.1	100	80.6	83.5	7
0.1	99.1	79.3	84.6	9

the chelating agent might be precipitated, adsorbed by the soil, or might form complexes with other cations (Ca, Mg, Fe, Al and other trace heavy metals) that originally present in the soil. It shows that Pb, Cd and Zn could be easily extracted by 0.1 M EDTA. It was believed that the high Pb, Cd and Zn extraction efficiencies demonstrated by the chelating agent could be attributed to the formation of strong and stable metal complexes which could overcome the adsorption interaction between the heavy metals and soil solids (Lim *et al.*, 2004). The organic matter in the soil and sludge also might play some part in retaining the metal by complexation. Many other studies have reported a significant influence of soil properties such as pH, organic matter and cation exchange capacities on metal retention by the soil (Surampalli *et al.*, 1998). Tyler and McBride (1992) indicated that the organic matter can retard heavy metal migration in soils. Elliot *et al.* (1986) also reported that the addition of organic matter would enhance retention of most heavy metals. In the next stage of this study the effect of pH on soil remediation was investigated. The remediation was not considerably affected by pH. The remediation efficiency in pH 3 for the Pb was approximately 10% more than in pH 9 for a Clay-Sandy-Loam soil but this increase for Sandy-Loam was not significant (Table 5).

Masky and Calver (1990) concluded that pH was a very important factor for determination the adsorption behavior of metals on soils. This finding is supported by Ottosen *et al.* (2001) who found that Zn is desorbed at a higher pH than Cu which again it desorbed at higher pH than Pb. An important observation from recent soil washing research is that removal efficiencies of metals from artificially contaminated soils via metal adsorption were significantly greater than those for soils from actual waste sites. As observed in Table 1 the initial Zn contaminated soil is higher than Pb and Cd and as a result the removal efficiency of Zn is lower than those of Pb and Cd. Tuin and Tels (1990) found that metal removals from actual waste-site soils were not as high as from an artificially contaminated soil when both soils were washed under similar conditions.

With progressively alkaline conditions, the ability of chelates to enhance solubility of oxides and other solid phase decreases because hydrolysis (the attachment of -OH as ligand) is favoured over completion by EDTA (Ottosen *et al.*, 2001). Such metal ions can be immobilized

Table 6: Remediation efficiency (%) of single metal-contaminated soil (Sandy-loam) with Conc. 1000 mg kg<sup>-1</sup> using various M EDTA

EDTA	Pb	Cd	Zn	pH
0.1	91.3	78.7	80.4	7
0.01	77.8	75.1	80.7	7
0.005	54.5	70.6	76.7	7

Table 7: Remediation efficiency (%) of single metal-contaminated soil (Sandy-loam) with Conc. 500 mg kg<sup>-1</sup> using DI water

Heavy metals	DI water
Pb	8.4
Cd	17.3
Zn	21.8
pH	7.0

in the soil by the formation of insoluble precipitates, incorporation into the crystalline structure of clay particles and metal oxides and/or by physical entrapment in the immobile water surrounding the micro and macropores of soils (Peters, 1999).

Moreover the effect of initial metal concentration in soil for the remediation rate has been investigated by Na<sub>2</sub> EDTA. When the initial metal concentration was increased two times (1000 mg kg<sup>-1</sup>) remediation efficiency by 0.1 M EDTA decreased approximately 9% for Pb and 1% for Cd and 4% for Zn, but when the contamination rate was twice and the 0.005 M EDTA was used, a 45% decrease was observed for lead remediation efficiency which is completely considerable, whereas this decrease for Cd and Zn were about 108% (Table 3 and 6). As another part of the study the remediation process has been done on some samples with the contamination rate of 500 mg kg<sup>-1</sup> by DI. The results represented poor remediation efficiency especially for the lead contaminated soil. An insignificant amount of metals removal by this method resulted in a poor removal efficiency that was less than 22% for Zn, 18% for Cd and 9% for Pb, indicating that the adsorbed HMs could not be readily removed by rising along even though the soil were artificially contaminated in the laboratory. Table 7 represents the removal efficiency with DI water in contaminated soil.

As observed, efficiency is different in the mentioned soil with the different concentration of EDTA especially for Pb, however, the remediation efficiency is maximum for lead. Table 5 provides the remediation efficiency at various pH values that it is nearly independent to pH. The influence of pH and concentration of EDTA on the solubilisation of certain metals in contaminated sediments and found that, in all cases, the lower the pH and the higher the EDTA concentration the greater the extraction recovery. The vacillation of remediation process results usually occurs in initial hours. A rapid uptake of lead from contaminated soil by Na<sub>2</sub> EDTA was observed by Evangelista and

Zownir (Fisher *et al.*, 1998). The possible disadvantages of chelating agent include: 1- EDTA is not easily biodegradable and pose a potential environmental hazard if they remain in the treated soil and 2- chelating agents are relatively expensive chemical compound. The results of batch washing experiments completed in this study indicated that Pb, Cd and Zn can be extracted from artificially contaminated soil using a chelating agent solution. There are significant decrease in the extraction of Pb with low concentration of EDTA (0.005 M). By using a washing solution at a concentration of 0.1 M of EDTA, extraction yields of 100% for Pb, 79.3% for Cd and 84.7% for Zn were obtained. Metal extraction kinetic was found to be fast, a reaction time of 2 h was deemed sufficient contact time between the contaminated soil and the washing solution based upon batch desorption tests conducted over a 48 h period. Result presented in Table 5 are in agreement with Lim *et al.* (2004) who found at the pH 4.7, the amounts of Pb and Cd extracted were slightly higher than those achieved at higher pH values, which were believed to be partly due to dissolution of soil minerals that caused simultaneous release of the adsorbed metals.

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