



**Full Length Article**

# Removal of Lead from Calcareous Contaminated Soils by Organic Acids

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## ABSTRACT

The comparative performance of two organic acids (EDTA, citric acid) for the removal of Pb from two texturally different soils (loamy sand, sandy clay loam soil) was evaluated. The extraction of Pb increased with increasing organic acid concentration and removal from sandy clay soil was lower than from loamy sand soil. Maximum Pb removal (330.6 mg kg<sup>-1</sup>) was achieved in loamy sand soil where highest concentration (32 mmol kg<sup>-1</sup>) of EDTA was applied. However, in sandy clay loam soil, same rate of EDTA application had 1.4 times less Pb extraction (240.0 mg kg<sup>-1</sup>) than that of loamy sand soil. In the case of citric acid relatively poor performance was observed at all the rates of application except at 32 mmol kg<sup>-1</sup>. Calcium extraction efficiency for each soil was different and it was a function of the concentration of organic acids. At lower rates, comparing EDTA performance in different soils, lower application rates (0-16 mmol kg<sup>-1</sup>) resulted in higher extraction of Ca in loamy sand soil as compared to sandy clay loam soil. However at the highest rate (32 mmol kg<sup>-1</sup>), EDTA application resulted in more Ca extraction in sandy clay loam soil. In the case of CA treatments, both the soil responded differently and Ca extraction from soils varied with the concentration of organic acid i.e., increasing CA concentration from 2 to 32 mmol kg<sup>-1</sup> resulted in increased removal of Ca from both the soils. Increasing concentration of CA from 2 to 32 mmol kg<sup>-1</sup> increased Ca extraction from 206.6 to 731.0 mg kg<sup>-1</sup> in loamy sand soils where as for sandy clay loam soil Ca extraction was increased from 92.6 to 747.0 mg kg<sup>-1</sup>.

**Key Words:** Pb-contaminated soil; EDTA; Citric acid; Texture

## INTRODUCTION

Agriculture in urban periphery is partially or completely dependent on sewage water most of which is untreated. Limited data pertaining to heavy metal status of raw sewage receiving soils of Pakistan are available (Ghafoor *et al.*, 2004). Apart from sewage irrigation, it is generally held that soils of arid and semi-arid regions are often rich in total Ni, Zn, Cu and Pb (Han *et al.*, 2002). According to FAO (1992), Pakistani soils contain relatively high available Pb, Co, Se and to some extent Cd compared to most of the other developing countries. Bioavailability of heavy metals is partly governed by physico-chemical characteristics of soils and partly by plant characteristics. It is a major health concern for animals and human. Therefore, soil, plant and water quality monitoring, together with the prevention of entry of metals into food chain via plant uptake, is a prerequisite in order to prevent potential health hazards of metals in agricultural lands.

Soils contaminated with heavy metals need remedial measures. In developing countries like Pakistan, decontamination methods employing physical and chemical

means are not applicable due to high cost associated with these technologies (Salido *et al.*, 2003). Phytoremediation, in general and phytoextraction in particular offers a promising alternative to conventional engineering based technologies (Raskin *et al.*, 1997; Marchiol *et al.*, 2004). Phytoremediation using hyperaccumulator plants have little prospects for Pb contaminated soils, mainly due to low phytoavailability of this metal (McGrath *et al.*, 2002). Therefore, application of chemical amendments (EDTA, elemental sulfur, inorganic acids etc.) to soils containing high levels of total Pb could increase the solubility and phytoavailability of Pb. Because of its strong affinity towards metals as well as little adverse effects on soils properties, EDTA is considered to be the most studied chelating agent (Grčman *et al.*, 2001; Meers *et al.*, 2005). However, due to its long residence time in soils and continued availability for the release of soil-bound metals, leaching of metals like Pb to ground water has been reported (Chen *et al.*, 2004). Moreover, EDTA being a non-selective chelating agent has the ability to complex and reacts with other metals like Ca, Al, Fe, Mg, Pb, Cu, Zn, Cd etc in soil (Kim *et al.*, 2003). Therefore, in order to achieve complete

complexation of target metal (s), extra amount of this chelating agent is applied to soil. Similarly among many factors affecting EDTA enhanced metal solubility, soil texture and calcite contents can influence the performance of this chemical agent (Manouchehri *et al.*, 2006). In calcareous soils, Ca is a major competing cation that can affect the ability of EDTA to extract heavy metals from contaminated soils (Brown & Elliot, 1992; Theodoratos *et al.*, 2000).

Other than EDTA many naturally occurring biodegradable chelating agents (citric (CA), oxalic, malic acids etc.) have also ability to increase solubility and plant availability of heavy metals including Pb (Nigam *et al.*, 2001; Chen *et al.*, 2003; Evangelou *et al.*, 2006). However, due to low stability binding constants and biodegradation, relatively little leaching occurs due to their application. Among naturally occurring organic acids CA had been extensively used in chemically enhanced phytoextraction studies (Meers *et al.*, 2005). In the current study, two organic acids contrasting in nature (EDTA & CA) were used to assess their impact on metal mobility in two texturally different soils (loamy sand, sandy clay loam). Citric acid was selected as potential alternative to EDTA, as it combines the ability to degrade rapidly in the soil with that ability to complex heavy metals and thus render them more phytoavailable (Meers *et al.*, 2005). Findings of such studies could be helpful in selecting the optimal chemical amendment as well as in determining the optimal application rate for enhanced phytoextraction of specific heavy metal contaminated soil in the field (Wang *et al.*, 2007). The present study was planned to evaluate the ability of EDTA and CA to increase mobility of Pb and Ca and to assess the affect of texture on effectiveness of EDTA and CA to extract Pb from contaminated soils.

## MATERIALS AND METHODS

Soils used in this experiment were collected from two agricultural fields irrigated with untreated city sewage. The sandy clay loam soil was collected from village 217-RB, Kajlianwala, Faisalabad. While the second soil, loamy sand was collected from the Land utilization Farm of University of Agriculture, Faisalabad situated at village Uchkera. These soils have developed in alluvium derived from the Himalayas mountain ranges and transported to the Indus River and its Tributaries during the late Pleistocene age (Riaz-ul-Amin, 1986). Therefore the soils are young, dominated by illite type clay minerals and carry negligible pH dependent negative charge (McNeal, 1966; Ranjha *et al.*, 1993).

After collecting soils from surface (0-20 cm), these were air-dried, ground with wooden roller and passed through 2 mm sieve and stored in plastic jars for designed analyses. Soil paste pH ( $pH_s$ ) was determined with help of WTW pH meter after standardizing it with buffer solutions of pH 7.01 and 9.20. Cation exchange capacity (CEC) was

determined following  $NH_4$ -acetate method. Electrical Conductivity of soil saturated extract ( $EC_e$ ) was determined with help of EC meter (TOA, CM-40V). Soluble cations and anions were determined according to the procedure described by the US salinity lab staff (1954). Soil organic matter (OM) was determined by oxidizing organic carbon in the soil with  $K_2Cr_2O_7$  and back titrating the unutilized  $K_2Cr_2O_7$  against  $FeSO_4$ . Calcium carbonate contents were determined with help of calcimeter (Moodie *et al.*, 1959).

The efficiency of synthetic chelant (EDTA) was compared with a natural organic complexing chelant (citric acid). Self spiked soils were used because these soils had the advantage of good homogeneity in terms of heavy metal concentration and speciation, soil composition, contamination process and contamination period.

The lab scale batch extraction experiment was conducted with a soil to solution ratio of 1:10 ( $g\ mL^{-1}$ ) at room temperature ( $20 \pm 2^\circ C$ ). Deionized water, lab-grade EDTA in the form of  $Na_2$  salt and citric acid was used for preparing solutions of EDTA and citric acid. The extraction of metals was performed in 100 mL clean plastic tubes with 4 g of contaminated soil and 40 mL each of EDTA or citric acid solution prepared in deionized water (DI). No pH adjustment was made during extraction experiment. The experiment was conducted in triplicate. A set of control experiment was also carried out using deionized water (DI) water as the extraction solution to determine the amount of water extractable metals in soils. The tubes were shaken for 2 h at 200 rpm over an end to end shaker. After shaking, the slurries were centrifuged, filtered through  $0.45\ \mu\ m$  Whatman membrane filter and filtrates were analyzed for metals (Pb, Ca) concentrations using Atomic Absorption Spectrometer (Model Thermo S-Series). Description of experiment is given in the Table II. The protocol for the extraction procedure employed in this study was the same employed by Lim *et al.* (2005).

Each treatment was replicated thrice following completely randomized design (CRD). Least square means and then standard errors (SE) were reported to differentiate between treatments. The graphs were plotted in Microsoft EXCEL package.

## RESULTS

**Lead extraction by organic acids from soils.** Lead extraction with organic acids at different concentrations is present in Fig. 1 and 2. Extraction of Pb increased with increasing application rates of organic acids. Minimum Pb extraction was found with de-ionized water. Metal removal from sandy clay loam (SCL) soil was reasonably lower than that from loamy sand (LS) soil. Maximum Pb removal ( $330.6\ mg\ kg^{-1}$ ) was achieved from loamy sand soil with the highest concentration ( $32\ mmol\ kg^{-1}$ ) of EDTA. However in SCL soil same rate of EDTA application resulted in Pb extraction ( $240.0\ mg\ kg^{-1}$ ) which was 1.4 time lower than that from loamy sand soil. With citric acid relatively, lower

extraction was observed at all the rates of application except at 32 mmol kg<sup>-1</sup>. However, higher Pb extraction was found from LS soil compared to SCL soil.

**Calcium extraction with organic acids from soils.** The results (Fig. 3 & 4) depict that Ca extraction effectiveness from soils was different and it was a function of the rate of organic acids. At lower rates (0-16 mmol kg<sup>-1</sup>) EDTA extracted higher of Ca in loamy sand soil as compared to sandy clay loam soil. However at the highest rate (32 mmol kg<sup>-1</sup>), EDTA resulted in more Ca extraction from sandy clay loam soil than that from other soil. For CA treatments, both the soils responded differently and Ca extraction remained a function of its concentration i.e., increasing CA concentration from 2 to 32 mmol kg<sup>-1</sup> increased removal of Ca from both the soils. Increasing concentration of CA from 2 to 32 mmol kg<sup>-1</sup> increased Ca extraction from 206.6 to 731.0 mg kg<sup>-1</sup> from LS soil whereas for SCL soil the corresponding Ca extraction increased from 92.6 to 747.0 mg kg<sup>-1</sup>. The CA application up to 16 mmol kg<sup>-1</sup> resulted in higher Ca extraction from loamy sand soil; however, increasing CA application rate from 16 to 32 mmol kg<sup>-1</sup> extracted more Ca from SCL compared to that from LS soil.

## DISCUSSION

We used two organic acids i.e., EDTA (a strong complexor) and citric acid (a weaker complexor). The EDTA could form strong complexes with heavy metals as well as major cation (s) such as Ca (Manouchehri *et al.*, 2006), as indicated by the high value of its metal binding stability constants 18.00 and 10.18, for Pb and Ca, respectively. However, citric acid is relatively a weaker complexant than EDTA with metal binding stability constants of 4.08 and 3.50 for Pb and Ca, respectively. Organic acids were used in two soils differing in texture as well as calcite contents. Loamy sand contained relatively low level of CaCO<sub>3</sub> compared to that of the sandy clay loam soil.

Results of the present study (Fig. 1) show that EDTA was better in releasing soil-bound Pb compared to citric acid (Fig. 2). The ability of EDTA to enhance the release of Pb from insoluble or sparingly soluble compounds compared to other chelating agents has been attributed to its higher binding capacity for Pb as observed in other studies (Blaylock *et al.*, 1997; Huang *et al.*, 1997; Wu *et al.*, 1999). A recent study by Nascimento *et al.* (2006) investigated the release of Pb in soil solution after application of chelating agents *viz.* EDTA, DTPA, oxalic acid, citric acid, vanillic acid and gallic acid. Authors concluded that EDTA was one of the most efficient chelating agents as it significantly increased the concentration of Pb in soil solution after 24 h and 7 d of application. Furthermore, authors noted a 217-fold increase in Pb concentration in soil solution of EDTA-treated pots (10 mmol kg<sup>-1</sup> soil) compared to that for the control pots following 7 d of application. In an earlier study, Shen *et al.* (2002) reported EDTA to be the most efficient

**Table I. Characteristics of soils used in studies**

Characteristic	Unit	Value		
		Loamy (LS)	sand Sandy loam (SCL)	clay
Textural class	-			
Sand	%	85.80	49.10	
Silt	"	10.36	24.50	
Clay	"	3.84	26.40	
<sup>p</sup> pH <sub>s</sub>	-	7.95	8.16	
EC <sub>e</sub>	dS m <sup>-1</sup>	2.05	3.5	
SAR	(mmol L <sup>-1</sup> ) <sup>1/2</sup>	1.60	16.00	
CEC	cmol <sub>c</sub> kg <sup>-1</sup>	4.30	6.15	
CaCO <sub>3</sub>	%	0.92	1.78	
OM	%	0.58	0.82	
Pb (before incubation)	mg kg <sup>-1</sup>	<sup>a</sup> 16.20 (1.78)	32.29 (3.3)	
Pb (after incubation)	mg kg <sup>-1</sup>	<sup>a</sup> 456.20 (325.2)	435.20 (322.3)	

<sup>p</sup>pH by saturated soil paste extract, <sup>a</sup>Total metals while values in parenthesis represent AB-DTPA extractable metals

**Table II. Experimental set up used in heavy metal extraction**

Treatments	Organic acid concentration (M)	Liquid/solid ratio (mL g <sup>-1</sup> )	Organic acid Loading (mmol kg <sup>-1</sup> )
E-2	0.0002	1:10	2
E-4	0.0004	1:10	4
E-8	0.0008	1:10	8
E-16	0.0016	1:10	16
E-32	0.0032	1:10	32
CA-2	0.0002	1:10	2
CA-4	0.0004	1:10	4
CA-8	0.0008	1:10	8
CA-16	0.0016	1:10	16
CA-32	0.0032	1:10	32

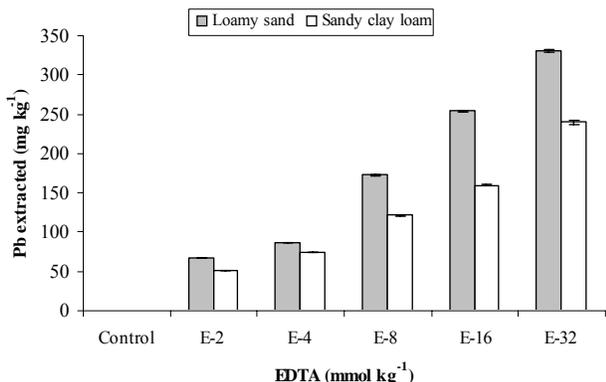
E and CA are abbreviations for EDTA and citric acid, respectively and figures next to E and CA are concentration of organic acids in mmol kg<sup>-1</sup>

chelating agent in the release of soil-bound Pb. Authors reported a 42-fold increase in Pb concentration in soil solution for the EDTA-treated soil (1.5 mmol kg<sup>-1</sup> soil) compared to that for the control 3 d following application.

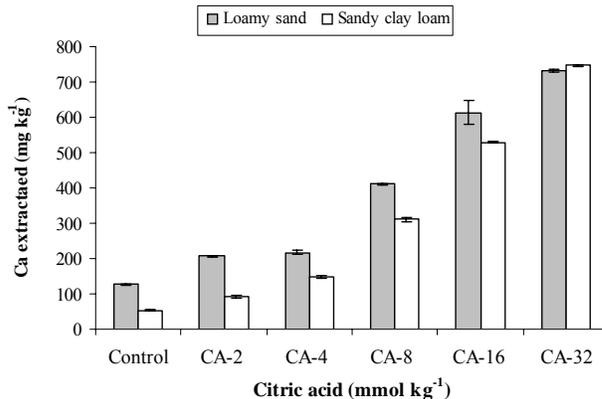
The data (Fig. 1) depict that EDTA efficiency to extract Pb increased with concentration. Several scientists have reported a linear relationship between EDTA concentration and metal removal from soils (Wenzel *et al.*, 2003; Hong & Jiang, 2005). For instance, Kim *et al.* (2003) studied the effects of solution to soil ratio, major cations present in soils and the EDTA: Pb stoichiometric ratio on the extraction of Pb using different Superfund site soils. It was reported that extraction of Pb from Pb-contaminated soils was not affected by a solution: soil ratio as low as 3:1 but instead was dependent on the quantity of EDTA used. Application of EDTA in sufficiently large amount (EDTA: Pb stoichiometric ratio greater than 10) resulted in extraction of most of the Pb from soil.

As expected, metal removal from sandy clay loam (clay 26.4%) was slightly lower than that from loamy sand soil (clay 3.8%). It was interesting to note that at lower application rates (2 to 16 mmol kg<sup>-1</sup>), EDTA extracted more Pb from sandy loam soil, however, at 32 mmol kg<sup>-1</sup> slightly higher Pb extraction was found from sandy clay loam soil compared to that of loamy sand soil. This may be due to low permeability and high fixing capacity of sandy clay loam

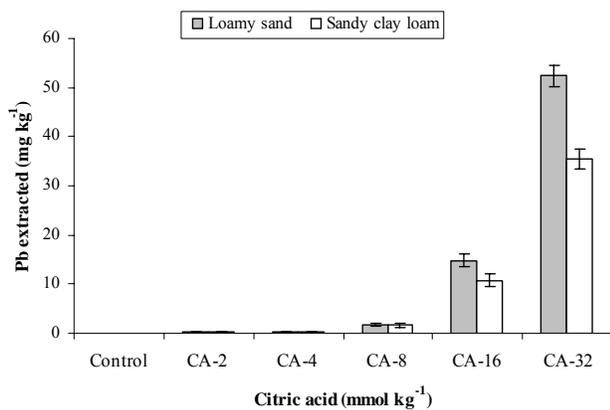
**Fig. 1. Concentration of extracted Pb as a function of EDTA concentration**



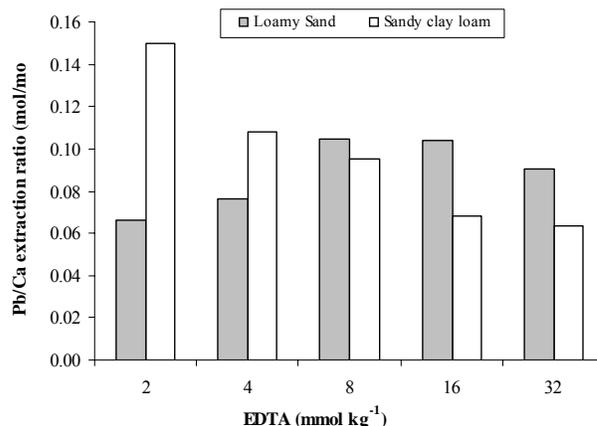
**Fig. 4. Concentration of extracted Ca as a function of citric acid concentration**



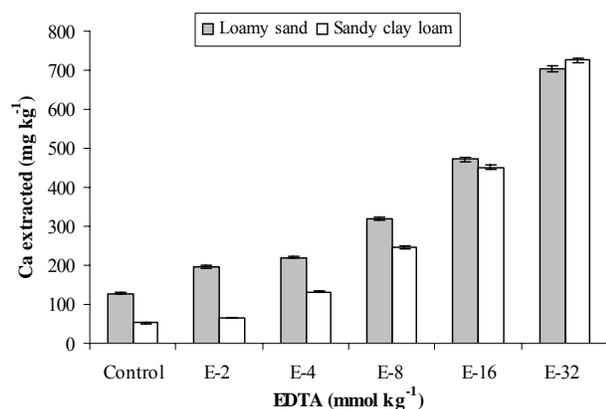
**Fig. 2. Concentration of extracted Pb as a function of citric acid concentration**



**Fig. 5. Extracted Pb/Ca ratio (by mol) by EDTA at different concentrations**



**Fig. 3. Concentration of extracted Ca as a function of EDTA concentration**



soil than those of loamy sand soil. Similar results have been reported by other researchers (Peters & Shem, 1992; Lo & Yang, 1999; Pichtel *et al.*, 2001; Lai & Chen, 2005). Lo and Yang, (1999) reported a 3-10% less metal removal from clayey soil as compared to that from sandy soils.

Citric acid was inefficient in extracting Pb from both

the soils at concentrations used in our experiment. Citric acid is relatively a weaker chelating agent than EDTA and does not preferentially extract heavy metals from the solid phase (Kirpichtchikova *et al.*, 2006). Increasing concentration of CA from 2 to 32 mmol kg<sup>-1</sup> increased the Pb concentration into soil solution from 0.2 to 52.4 mg kg<sup>-1</sup> in loamy sand whereas in sandy clay loam it increased from 0.2 to 35.0. Comparing citric acid efficiency for both the soils, higher Pb extraction (52.4 mg kg<sup>-1</sup>) was found from loamy sand soil as compared to that for sandy clay soil where it extracted only 35.0 mg Pb kg<sup>-1</sup> soil. This may be due to high clay as well as carbonate content of sandy clay loam soil which resulted in decreased Pb extraction. It had been reported that citric acid interact preferentially with Ca compared to Pb. Combined with low affinity for Pb as well as high interaction of citric acid with Ca (by decreasing pH & complexation) might have resulted in low efficiency of this chelating agent to extract Pb from sandy clay loam soil (Kirpichtchikova *et al.*, 2006).

Calcium is generally present in soils at high concentration and could greatly reduce the mobilization of target elements (Pb in this study). Results of current study

(Fig. 3 & 4) showed that Ca extraction from both the soils increased with increasing concentration of organic acids. However, at lower levels of application from 2-16 mmol kg<sup>-1</sup>, both the organic acids extracted more Ca from loamy sand compared to that from sandy clay loam soil. At higher level of application (32 mmol kg<sup>-1</sup>), the trend was different. At this level of application, organic acids extracted more Ca from sandy clay loam compared to that from loamy sand soil. The EDTA is a non-selective extractant that can form a strong complex with a variety of metals in soils including alkaline-earth cations such as Al<sup>3+</sup>, Ca<sup>2+</sup>, Fe<sup>2+</sup> and Mg<sup>2+</sup> and target heavy metal (s) such as Pb, Cd, Ni, Zn, Mn (Skoog *et al.*, 1996; Zeng *et al.*, 2005). The presence of competing cations (Ca<sup>2+</sup>, Mg<sup>2+</sup>, Al<sup>3+</sup>, Fe<sup>2+</sup>, Cu<sup>2+</sup>, Cd<sup>2+</sup>, Zn<sup>2+</sup>) and/or the nature of soil matrix can substantially alter the rate of complex formation; predicted solely on the basis of the ratio of EDTA to the target heavy metal (s) (Manouchehri *et al.*, 2006). It is interesting to note that EDTA extracted more Pb than CA in this study, however for Ca extraction, CA performed slightly better than EDTA. A survey of literature reveals that many other workers found results similar to this study (Gramss *et al.*, 2004; Meers *et al.*, 2005; Kirpichtchikova *et al.*, 2006).

The main objective of this study was to assess the role of organic acids in enhancing mobilization of Pb. Both the soils used in this study were calcareous in nature (Table I). At lower application levels, EDTA resulted in more extraction of Pb than its higher levels of application in sandy clay loam soil (Fig. 5). It seems that at lower EDTA levels, competition from Ca to complex EDTA was low which resulted in better extraction of target element (Pb). However, for loamy sand soil, contrasting results were noted. In this soil, more extraction of Pb compared to Ca was recorded at higher levels of application (Fig. 5). These results are contrary to earlier studies where significantly higher extraction of Pb with increasing concentration of EDTA even in the presence of Ca (Kim *et al.*, 2003; Manouchehri *et al.*, 2006) has been reported.

## CONCLUSION

The results from these studies indicated that extraction of Pb from both the soils was dependent upon the rate of organic acids applied. Compared with citric acid, EDTA caused more extraction of Pb at all the levels of application. More extraction resulted from coarse textured soils compared to that from fine textured soils. Overall the results from this study demonstrated that EDTA could potentially be used as an amendment to enhance Pb mobility (extraction) in soils and thus its availability to plants for removal from contaminated soils.

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