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Leaching of Zinc, Cadmium, and Lead in a Sandy Soil Due to Application of Poultry Litter

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Dissolved organic matter in poultry litter could contribute organic ligands to form complexes with heavy metals in soil. The soluble complexes with heavy metals can be transported downward and possibly deteriorate groundwater quality. To better understand metal mobilization by soluble organic ligands in poultry litter, soil columns were employed to investigate the movement of zinc (Zn), cadmium (Cd), and lead (Pb). Uncontaminated soil was amended with Zn, Cd, and Pb at rates of 400, 8, and 200 mg kg^{-1} *soil, respectively. Glass tubes, 4.9-cm-diameter and 40-cm-long, were packed with either natural or metal-amended soil. The resulting 20-cm-long column of soils had bulk density of about 1.58 g cm*$^{-3}$*. Columns repacked with natural or amended soil were leached with distilled water, 0.01 M EDTA, 0.01 M* $CaCl_2$*, or poultry litter extract (PLE) solutions. Low amounts of Zn, Cd, and Pb were leached from natural soil with the solutions. Leaching of Zn, Cd, or Pb was negligible with distilled water. In the metal-amended soil, EDTA solubilized more Zn, Cd, and Pb than* $CaCl_2$ *and PLE. The breakthrough curves of Zn and Pb in the PLE and* $CaCl_2$ *were similar, indicating they have similar ability to displace Zn and Pb from soils. Compared with Zn and Cd the PLE had a small ability to solubilize Pb from metal-amended soil. Thus, the application of poultry litter on metal-contaminated soils might enhance the mobility of Zn and Cd.*

Keywords EDTA, heavy metals, leaching solution, metal-amended soil, poultry litter

Introduction

The poultry industry is one of the largest and fastest growing sectors of animal production in the world (Nicholson *et al.*, 1996). Poultry litter is a valuable source of major plant nutrients and has been used as a soil amendment in agriculture. The increasing poultry industry in Iran, especially in the Hamadan province, produces large amounts of poultry litter, which are used to enhance soil fertility. Besides supplying nutrients, organic litter affects metal solubility (del Castilho *et al.*, 1993). Dissolved organic matter in poultry litter could contribute effectively organic ligands to form complexes with heavy metals in soil (McBride, 1989; Bolton and Evans, 1991). These complexes can be moved downward and may deteriorate groundwater quality (Li and Shuman, 1996). Sewage sludge and forest litter also can release soluble organic ligands, which can complex with trace metal cations. Adding soluble organic ligands have been reported to reduce the sorption of trace metals by soils (Shuman, 1995).

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The mobility of heavy metals in soils depends on total concentration in soil and soil properties and environmental factors (Li and Shuman, 1996). Boyle and Fuller (1987) reported that zinc (Zn) leaching through soil columns was enhanced by elevating the dissolved organic carbon. Lamy *et al.* (1993) concluded that mobility of cadmium (Cd) was enhanced by an increase in soluble organic mater. Camobreco *et al.* (1996) reported that copper (Cu) and lead (Pb) transport in soil was increased in the presence of sphagnum peat. Li and Shuman (1997) reported that poultry litter extract (PLE) removed more Cd and Zn from contaminated soil column than calcium chloride. Dowdy and Volk (1983) suggested that in sandy soils with low organic matter, leaching of heavy metals would occur due to the heavy rainfall or irrigation.

Also in arid and semiarid regions, use of poor quality water has become inevitable for irrigation to compensate rapidly increasing water demands. Substantial leaching losses can occur in this environment when a soluble fertilizer is applied in the soil. Losses of nutrients from soil due to the application of poor water quality also were reported by other researchers (Feigenbaum, 1986; Feignebaum and Meieri, 1988; Jalali, 1997; Jalali and Rowell, 2003), but experimental data on heavy metals leaching from sandy loam under arid and semi arid conditions are few. Thus, to better understand metal mobilization by soluble organic ligands in poultry litter and their contribution to poor water quality, soil columns were employed to investigate the movement of Zn, Cd, and Pb in soil. Because complexation and cation exchange are the two most important mechanisms that affect metal mobility, EDTA and $CaCl_2$ were used as complexing- and exchange-solution, respectively.

Materials and Methods

Soil

Since Azandarian area is the most important agricultural region of Hamadan province and also because of poultry litter usage, the soil sampling was carried out in this region. The sample was taken from the 0-to-30-cm layer of agricultural soil in Hamadan, western Iran. The soil was selected to represent typical, continuously cultivated (for grapes, *Vitis vinifera* L.) and sandy loam. Soil was air dried and passed through a 2-mm-mesh sieve before being stored in polyethylene bags. Soil pH, electrical conductivity (EC), organic matter, cation exchange capacity (CEC), and texture were determined using standard laboratory procedures. Total concentration of Zn, Pb, and Cd in the soils was determined on filtered extracts obtained from 2-g samples, which were digested overnight with 12.5 ml 4 M HNO_3 at 80°C (Sposito *et al.*, 1982). Soil was belonged to Azandarian series (Typic Calcixerepts). Some of the relevant chemical and physical properties of an air-dry <2 mm sample are in Table 1.

Poultry Litter Extract

Poultry litter was collected from the University farm in Hamadan. Poultry litter extraction was made by stirring 50 g of dry poultry litter (sieved to 2 mm) in 1.0 liter deionized water. The suspension was stirred for 2 h, centrifuged for 20 min, and filtered through Watman 42 filter paper. After adjusting the pH from 8.2 to 7.0 with 6 M HCl, this solution was used for only 1 week while being stored at 4°C (Li and Shuman, 1997). The metal concentrations for Zn, Cd, and Pb of the PLE were analyzed by atomic absorption spectrophotometer (AAS)

Table 1

Selected physical and chemical properties of studied soil

Soil type	Classification	Sand	Silt	Clay	pH	CEC	OM	CaCO$_3$	Total (mg kg^{-1})		
			(g kg^{-1})			(cmol$_c$ kg^{-1})	(g kg^{-1})	(g kg^{-1})	Zn	Cd	Pb
Azandarian	Typic Calcixerepts	784	133	83	6.3	11.2	34.8	47	99.67	0.5	73.87

(Varian, spectra 220). The concentrations were as follows: Zn, 0.46 mg l^{-1}; Cd, <0.01 mg l^{-1}; Pb, 0.16 mg l^{-1}.

Metal-Amended Soil

The soil was amended with Zn, Pb, and Cd at 400, 200, and 8 mg kg^{-1}, respectively, using $ZnCl_2$, $CdCl_2 . 2H_2O$, and $PbCl_2$, respectively. The amended soil sample was saturated with distilled water, mixed, air dried, sieved (2 mm), and stored for leaching experiments.

Leaching Experiments

The leaching columns consisted of Pyrex tubes, 40-cm long; with an inner diameter of 4.8 cm. Columns were filled with soil to a height of 20 cm by uniform tapping to achieve a uniform bulk density of 1.58 g cm^{-3}. During packing each soil was funneled into columns, while the walls were being simultaneously tapped with a wood rod to achieve a uniform packing at the same bulk density. A Whatman No. 42 filter paper was placed at the bottom of the leaching column. The bottom of the column was covered with nylon mesh. The solution was pounded (about 5 cm above the soil surface) on the soil column, and maintained during the leaching process so that the effluent flow was kept at an average of 13.8 to 28.8 mm h^{-1}. Water level varied slightly according to the outflow rate. A filter paper was placed on the soil surface to minimize soil disturbance from the addition of leaching solution. Then, soil columns were leached with H_2O, 0.01 M $CaCl_2$, 0.01 M EDTA, or PLE. Leachates were collected in 10-ml increments and after 8 pore volumes in 50-ml increments. Leaching continued until 20 pore volumes were collected. Pore volume (PV) is that part of soil not occupied by the soil matrix. In the field, without the soil saturation, the PV is filled partly with soil air and partly with soil water. In leaching columns, the PV is filled entirely by the soil water. The PV of soil columns was calculated from value for the bulk density and particle density (2.65 g cm^{-3}) of the soil in the column (Rowell, 1994). Pore volume of the columns was 143 ml. The study was conducted in two replicates at room temperature (22–24°C). The concentrations of Zn, Cd, and Pb were determined in the leachate by AAS (Li and Shuman, 1997). The results were reported using the average of two replicate columns. The t-test was used for comparing the means of leaching solutions at $p \leq 0.05$ using Minitab software (version 13.1, Minitab Inc.).

Results and Discussion

Leaching of Zn from Soils

The results of the leaching are presented as breakthrough curves (graphs showing the relationships between concentrations of cations or anions and cumulative water passing out the columns) (Rowell, 1994; Jalali and Rowell, 2003). In all treatments, the two sides of the breakthrough curves have different characteristics (Figures 1–3). The leading edge is sharp and the following edge shows tailing. In soil leached with EDTA, the Zn, Cd, and Pb concentration increased at an average rate of 332, 13, and 415 mg l^{-1} per pore volume, respectively, for the first part of the breakthrough curves. For other leaching solutions, those rates were reduced. Movement of heavy metals in soil is affected markedly by their extent of sorption by the soil. Chemical processes can influence sorption reaction, control the concentration of heavy metals in solution, and their transport through the soil profile. Distribution coefficients are used to relate the amount of heavy metals sorbed by the soil to that dissolved in the water (Alloway, 1995). At the initial breakthrough curves of the heavy metals, the adsorption coefficient is large when the heavy metal concentration in the liquid phase is low. With increasing concentration of heavy metals, it drops considerably to a maximum value at the highest breakthrough concentration of the heavy metals. After the peak breakthrough concentration has passed away, adsorption coefficients increase gradually with decreasing concentration of the heavy metals (Carrillo Gonzalez *et al.*, 2004). Breakthrough curves gave similar shapes with no indication of an extended leading edge that would result from by-pass flow. The repacked sieved soil (2-mm) does not have the structural features that normally give rise to by-pass flow (Wong *et al.*, 1990).

Leaching of Zn with distilled water in the native soils was negligible, and in the presence of $CaCl_2$, EDTA, or PLE was small.

The breakthrough curves for Zn in metal-amended soil when leached with the different leaching solutions are presented in Figure 1. Peak concentration Zn in the leachates was observed in about 1 pore volume followed by a decrease in the subsequent leachate fractions, with each of the treatments (Figure 1). In soil leached with PLE, release of Zn was small, with a steady decrease in Zn concentration as the experiments proceed. The maximum Zn concentration (121 mg l^{-1}) was observed for about 1 pore volume, after which the concentration of Zn decreased rapidly with a long tail from 43 to 30 mg l^{-1} during the rest of the percolation. Concentration of Zn in the PLE leachate was higher than the drinking water standard, which is 5 mg l^{-1} (Adriano, 1986). Table 2 shows the total amounts leached in 5, 10, 15, and 20 pore volumes. The amount leached varied from 16.8 to 25.7% of the added Zn after 5 to 20 pore volumes had passed through the column. This result indicated that some Zn was retained by the soil. Li and Shuman (1996) stated that these results probably were due to preliminary sorption of organic ligands on to the soil with the creation of new sorbing surfaces. Petruzelli *et al.* (1992) also showed that the addition of sewage sludge extract increased the amount of heavy metals retained by soil. Long-term application of wastewater could elevate the trace element contents in soil (Page and Chang, 1985). The leaching of Zn by EDTA is shown in Figure 1. The maximum Zn concentration (350 mg l^{-1}) was observed for about 1 pore volume followed by decrease in the subsequent fractions. In this soil Zn was leached with EDTA due to solubilization by chelate formation (Norvell, 1991). The leaching of Zn by the EDTA was larger than that for the PLE, indicating a high potential of EDTA to solubilize Zn in the soil. The amount leached fraction varied from 31.2 to 44.8% of the added Zn after 5 to 15 pore volumes had passed through the columns (Table 2). Li and Shuman (1996) also reported that EDTA leached more Zn than PLE. These results suggest that EDTA is a potential agent to remove Zn from metal-contaminated soil.

The curved pattern for leached Zn from $CaCl_2$ was similar to the PLE, whereas the leaching with $CaCl_2$ was relatively higher at the first pore volume (Figure 1). The amount leached varied from 20 to 32.9% of the total added Zn after 5 to 20 pore volumes had passed through the columns. Barak and Helmke (1993) stated that a large amount of Ca^{2+} in the $CaCl_2$ can displace Zn from adsorption sites on the soil solids. The Cl^- may also form $ZnCl^+$, which has a low affinity for the reacting surfaces in the soil (Barrow and Ellis, 1986). Li and Shuman (1996) stated that the Ca^{2+} and Cl^- can solubilize only the exchangeable or outersphere associated Zn from soil surfaces. Added Zn may mainly reside in the exchangeable form, which could be released by $CaCl_2$.

The total amount leached by $CaCl_2$ was greater than that leached by PLE (Table 2). The PLE probably released exchangeable forms of Zn, but the retention of the dissolved

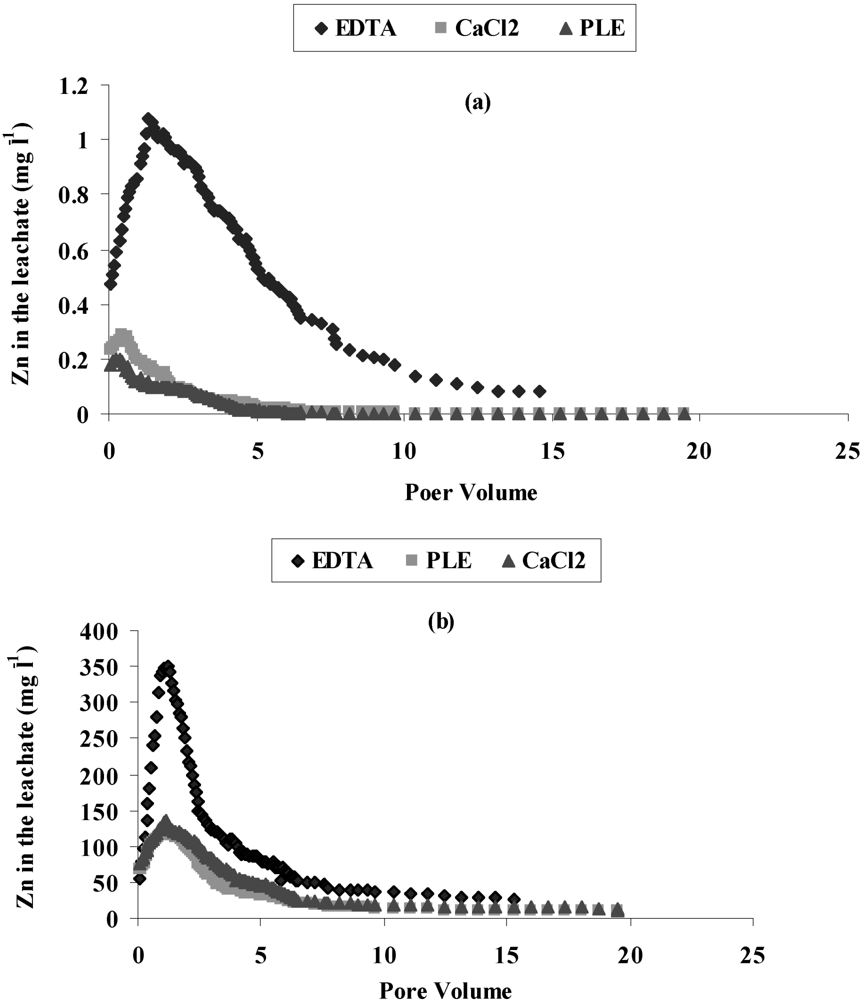

Figure 1. Breakthrough curves for Zn leached with EDTA, $CaCl_2$, or PLE in (a) the Azandarian uncontaminated soil and (b) the Azandarian metal-amended soil.

organic matter by the soil may have provided more active sites to adsorb Zn^{2+} or Zn-organic complexes on soil surfaces (Li and Shuman, 1996). The concentration of Zn in the leachate of distilled water was high, especially at the beginning of the leaching period (82.7 mg l^{-1}) and then decreased and leveled off at about 0.01 mg l^{-1} after 20 pore volumes.

Leaching of Cd from the Soil

Leaching of Cd with distilled water and PLE in native soils was negligible, and was small with EDTA and $CaCl_2$, because of the low Cd content in these soils. The leaching of Cd in the metal-amended soils is shown in Figure 2. Leaching of Cd with $CaCl_2$ was relatively higher than that with PLE, indicating that there was a large portion of Cd in the water-soluble and exchangeable forms than in other forms. The lower leaching of Cd with PLE was possibly due to adsorption of Cd onto the soil surfaces, a process that is similar to

Table 2

Amounts of Zn leached from columns of soil

	Amount leached in 5 PV (381 mm)[‡]		Amount leached in 10 PV (762 mm)		Amount leached in 15 PV (1143 mm)		Amount leached in 20 PV (1524 mm)	
Treatments	kg ha^{-1}	% of applied Zn	kg ha^{-1}	% of applied Zn	kg ha^{-1}	% of applied Zn	kg ha^{-1}	% of applied Zn
PLE	282.8^{a}[†]	16.8	353.8^{a}	21.1	397.3^{a}	23.6	431.2^{a}	25.7
EDTA	596.5^{c}	31.2	750.1^{c}	39.3	855.0^{c}	44.8	—	—
$CaCl_2$	307.0^{a}	20.2	409.3^{a}	25.6	460.5^{a}	29.2	527.4^{a}	33.0

[†] Values followed by same letters within a column indicate no significant difference at $p \leq 0.05$.

[‡] Equivalent to mm was calculated by the following equation: [PV (cm^3)/column surface area (cm^2)] × 10.

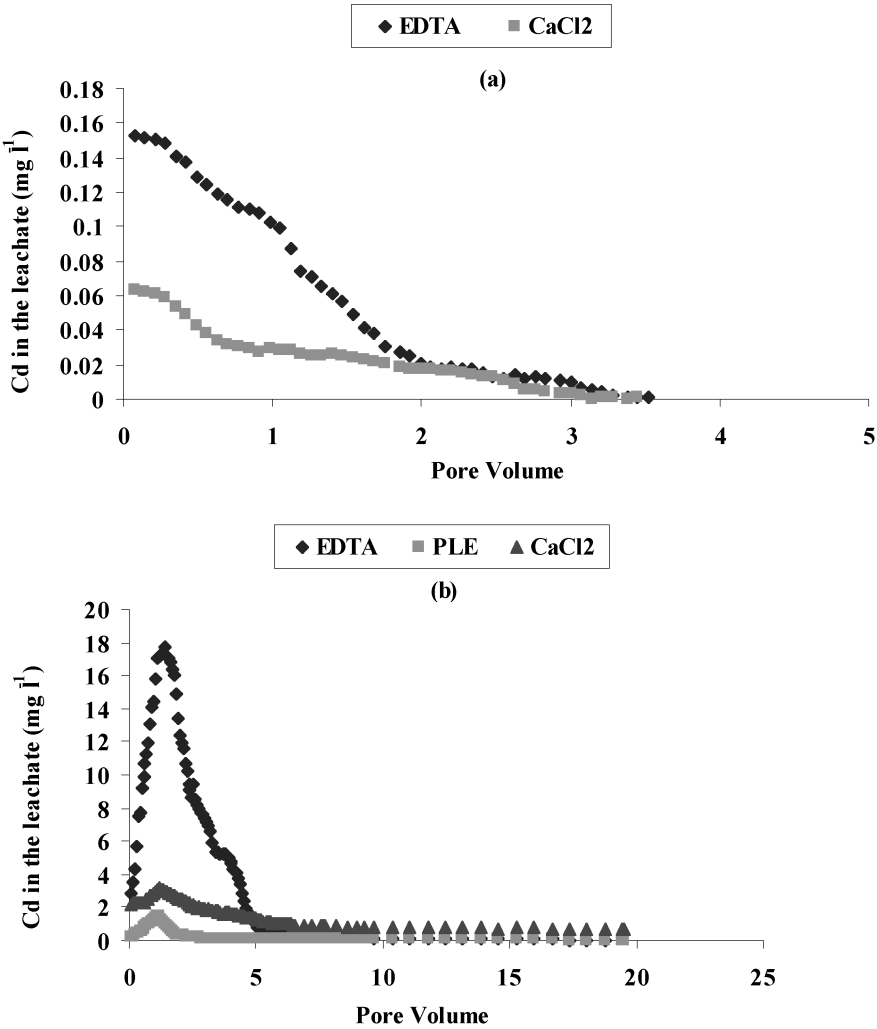

Figure 2. Breakthrough curves for Cd leached with EDTA, $CaCl_2$, or PLE in (a) the Azandarian uncontaminated soil and (b) the Azandarian metal-amended soil.

the mechanism for Zn retention. Some researchers indicated that in metal-amended soil, greater percentage of soil Cd was in the exchangeable fraction than in other fraction (Basta *et al.*, 1993; Berti and Jacobs, 1996). Li and Shuman (1996) stated that cation exchange and inorganic ligands such as Cl^- affectively could release Cd from the soils with a large portion of Cd in the exchange fraction. Leaching of Cd with the EDTA in the metal-amended soils was greater than that with either PLE or $CaCl_2$. The EDTA solubilized Cd from some specifically adsorbed or inner-sphere forms in soil (Yu and Klarup, 1994). Table 3 shows the total amounts of Cd leached in about 20 pore volumes for different leaching solutions. The EDTA solubilized 56.5% of the total Cd after 15 pore volumes had passed through the column. But PLE solubilized 18% of the total Cd, which is less than the amounts of Cd leached by the $CaCl_2$ (43.4%). This result indicates that the addition of soluble organic matter increased the sorption of Cd in soils (Lamy *et al.*, 1993). Petruzzelli *et al.* (1992)

Table 3
Amounts of Cd leached from columns of soil

	Amount leached in 5 PV (381 mm)‡		Amount leached in 10 PV (762 mm)		Amount leached in 15 PV (1143 mm)		Amount leached in 20 PV (1524 mm)	
Treatments	kg ha^{-1}	% of applied Cd	kg ha^{-1}	% of applied Cd	kg ha^{-1}	% of applied Cd	kg ha^{-1}	% of applied Cd
PLE	$4.2^{a\dagger}$	14.4	4.6^{a}	16.0	5.0^{a}	17.0	5.2^{a}	18.0
EDTA	13.3^{b}	50.0	14.8^{b}	55.0	15.3^{b}	56.5	—	—
$CaCl_2$	7.7^{a}	28.5	9.6^{a}	35.4	11.1^{a}	41.0	11.8^{a}	43.4

†Values followed by same letters within a column indicate no significant difference at $p \leq 0.05$.

‡Equivalent to mm was calculated by the following equation: [PV (cm^3)/column surface area (cm^2)] × 10.

demonstrated that the preliminary sorption of organic ligands onto soil might create new sorbing surface to retain Cd and Ni.

Leaching of Pb from the Soil

The leaching of Pb from the native soil (uncontaminated soil) with EDTA, $CaCl_2$, or PLE was negligible, due to the low Pb content in this soil. In the metal-amended soil leaching with EDTA was much larger than with PLE or $CaCl_2$ (Figure 3). Table 4 shows the total amounts leached in 5, 10, 15, and 20 pore volumes. The amount leached varied from 52.3% to 55.8% for EDTA and from 1.7% to 1.9% of the total Pb for PLE, after 5 to 20 pore volumes were passed through the columns. Compared with Zn and Cd the PLE had little ability to solubilize Pb from the soil. The small amount of leaching of Pb with $CaCl_2$ and PLE was probably related to the low amount of Pb in the exchangeable form. Li and Shuman

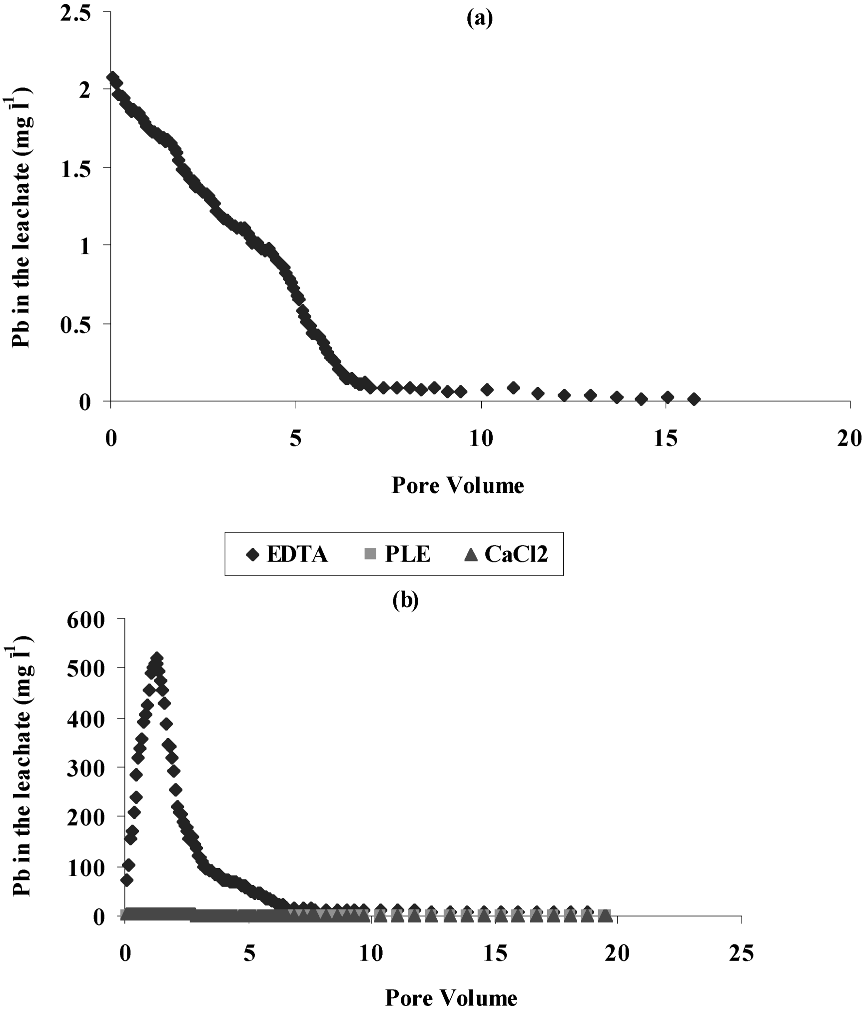

Figure 3. Breakthrough curves for Pb leached with EDTA, $CaCl_2$, or PLE in (a) the Azandarian uncontaminated soil and (b) the Azandarian metal-amended soil.

Table 4
Amounts of Pb leached from columns of soil

	Amount leached in 5 PV (381 mm)[‡]		Amount leached in 10 PV (762 mm)		Amount leached in 15 PV (1143 mm)		Amount leached in 20 PV (1524 mm)	
Treatments	kg ha^{-1}	% of applied Pb	kg ha^{-1}	% of applied Pb	kg ha^{-1}	% of applied Pb	kg ha^{-1}	% of applied Pb
PLE	15.6^{a}[†]	1.7	16.4^{a}	1.8	17.0^{a}	1.9	17.3^{a}	1.9
EDTA	456.1^{c}	52.3	480.5^{c}	55.1	487.0^{c}	55.8	—	—
$CaCl_2$	5.8^{a}	0.7	7.5^{a}	0.8	9.0^{a}	1.0	9.7^{a}	1.1

[†] Values followed by same letters within a column indicate no significant difference at $p \leq 0.05$.

[‡] Equivalent to mm was calculated by the following equation: [PV (cm^3)/column surface area (cm^2)] × 10.

(1997) stated that Pb bound to the soil matrix has low mobility. Compared with Zn and Cd, the EDTA solubilized more Pb, indicating the potential for leaching of Pb from metal-amended soils. Lead forms chelate complexes with EDTA more than with Zn and Cd. The EDTA formation constant with Pb (Log K = 19.0) is larger than those for Zn (Log K = 17.5) and for Cd (Log K = 17.4) (Norvell, 1991). The $CaCl_2$, leached less Pb than Zn and Cd. This result indicates that compared with Zn and Cd, less Pb is associated with exchangeable fraction. Sposito *et al.* (1982) and Walter and Cuevas (1999) reported that Pb was present mostly in inorganic and residual fractions. It has been reported that pH of the soil plays an important role in metal mobility (Fernandez-Turiel *et al.*, 2001). Thus, Pb would have low mobility in neutral or alkaline soils due to the formation of insoluble salts, whereas Zn would have greater mobility.

Even if distilled water was used as a leaching solution, 0.1%, 5.2%, and 9.7% of the added Pb, Cd, and Zn were leached from metal-amended soils after 20 pore volumes, respectively. This experiment simulates the condition in the field when soil is irrigated with water of good quality or winter rain. The soil had some lime (47 g kg^{-1} $CaCO_3$) that can be solubilized to supply $Ca^{2+}$to replace heavy metals. Because of the low solubility (0.0131 g l^{-1}), this native Ca^{2+} source does not make a significant contribution to replacement of heavy metals from exchange sites (Qadir *et al.*, 1996). In the field, release of CO_2 in the root zone as a result of root and microbial respiration and CO_2 dissolution in water produces H_2CO_3, the acidity of which increases $CaCO_3$ dissolution. Therefore, in the field, where the CO_2 concentration in the soil air is higher than in the atmosphere, dissolution of calcite may be enhanced. Poor quality ground waters are common feature of arid and semi-arid regions. Leaching of nutrients from soil due to application of poor quality water was also reported. Jalali and Rowell (2003) reported losses equivalent to 63 to 170 kg K^+ha^{-1} (12–33% of the initial exchangeable K^+) when 196 mm of solution containing 1 to 15 $mMCaCl_2$ were applied to soil columns in the laboratory. In the studied area, Ca^{2+} is the dominating ion in water wells, and concentrations of Ca^{2+} (in meq l^{-1}) represent an averaged 43.6% of all cations (Jalali, 2005). Its concentration in irrigation water varies from 0.01 to 11.3 mM (Jalali, 2002). Thus, leaching of heavy metals with 10 mM $CaCl_2$ simulates the effects of poor quality irrigation water in arid and semi-arid regions.

Conclusions

Due to their low concentrations in uncontaminated soils, Zn, Cd, and Pb in the natural soil were not mobilized by leaching solutions. In metal-amended soils, large amounts of Zn and Cd were leached, whereas Pb was leached only by the EDTA. The EDTA leached more Zn, Cd, and Pb than $CaCl_2$, and $CaCl_2$ solution leached more than PLE in the metal-amended soils. The amount of Cd and Zn leached with the PLE solution were 18% and 26% of the total added Cd and Zn when 20 pore volumes had passed through the columns, respectively. Thus, the application of poultry litter to the metal-amended soils may accelerate the leaching of Zn and Cd in the soil profile. It would appear that Zn was more affected than Cd. Also these losses by leaching would be more severe on soils having a low CEC, and if uncorrected could eventually result in deficiencies of Zn for plant growth and pollution of groundwater. It can also be concluded that the use of sources of water for irrigation that have high Ca^{2+} concentration can lead to leaching of heavy metals from soil. The leaching columns experiment was not a perfect simulation of the field situation because the leaching rate was relatively fast (13.8–28.8 mm h^{-1}). In the leaching column only about 4 h were required for 5 pore volumes to pass through the column. This rate of movement was not sufficiently slow to ensure that equilibration occurred (the soil is sandy loam and the same flow rate can be expected to occur in the field). If heavy metals are applied in the field, some applied metals may be transformed to highly stable forms.

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