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Influence of EDTA and NTA on heavy metal extraction from sandy-loam contaminated soils

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ABSTRACT

In this study, Pb, Cd, and Zn as heavy metal compounds were extracted from the sandy-loam soil; the soil was prepared synthetically inside the laboratory and also taken from the field by using EDTA and NTA as the chelating agents. By using EDTA 0.1 M to extract the contaminants from the synthetically contaminated soil with single metal concentration of 500 mg/kg, the efficiency of extracting Pb, Cd, and Zn in the first pore volume (PV) were found to be respectively 43.84%, 20.6%, and 43.7%; the corresponding amounts after the third PV for the contaminants were 85.25%, 83.2%, and 69.7%, respectively. The efficiency of NTA 0.1 M in extracting Pb, Cd, and Zn in the first PV was 8.3%, 59.6%, and 14.87%, respectively; after the third PV, the corresponding efficiency reached to 21.8%, 83.56%, and 24.49%, respectively, for the contaminants. At pH 4.5, the efficiency of extracting Pb, and Zn using EDTA and NTA were higher than other levels of pH; and also, the maximum Cd extraction was achieved in pH 7. Considering the samples taken from the field, the extraction efficiency of Pb, Cd, and Zn using EDTA 0.1 M after the fifth PV were 89.42%, 64.21%, and 63.5%, respectively.

1. Introduction

Ever increasing growth of population and industrialization along with rapid improvement of quality of life have resulted in the production and disposal of hazardous solid wastes into the environment [1,2]. Soil contamination caused by the release of non-biodegradable pollutants (e.g. heavy metals) into the environment has become one of the main concerns among societies [3–6]. It should be noted that anthropogenic activities have been considered to be the main source of air and soil contamination; in fact, the contamination is mostly related with heavy metals in most populated regions around the globe [7–9]. It should be noted that absorbing high dosage of heavy metals can cause acute toxicity in human beings, microorganisms, animals, and vegetation's [10]. Since heavy metals are mostly adsorbed by soil particles, washing soil with merely water cannot remove these compounds from this media in an efficient and appropriate way [11,12]; however, the chemical soil washing process removes these contaminants selectively and efficiently from the soil media [13].

Over the last decades, different chemical compounds have been used as chelating agents for the process of chemical soil washing. Comparing with acid soil extraction, these agents have higher efficiency in extracting metals from the soil media and comparatively have lower effects on the physical, chemical, and biological characteristics of soil; and also, the solubility of the extracted metals in the solution is of high degree [14,15]. So far, several forms of chelating agents have been studied and applied for extracting and improving these metals, including ethylenediaminetetra acetic acid (EDTA), ethylenediaminetriacetic acid (HEDTA *N*-(hydroxyethyl) [16–18]. In a similar study for the extraction of heavy metals from marine sediments using EDTA, EDDS, NTA and citric acid tested for metal removal, EDTA showed a good removal efficiency for tested heavy metals [19].

In order to decide whether artificial chelating agents can be applied, several important factors should be taken into account. It should be considered that these agents unselectively extract metals from the soil media and at the presence of polyvalent

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cations (e.g. Fe, Mn, Ca, Mg, etc) could result in the formation of specific complexes; hence, the concentration of the agents should be higher than that of the contaminants in order to have acceptable results [20]. In addition, the chemical state of the metals in the soil media affects the efficiency of the agents in extracting these compounds; in other words, recently contaminated soil medias with heavy metals are more liable and accessible [21,22]. The biodegradability of the chelating agents and also their high capacity in binding with metals are of utmost importance. Furthermore, it can be of great advantage if the agents were recoverable and reusable.

In recent years, due to environmental hazards caused by non-biodegradable chelating agent, there is growing interest in using biodegradable and environmental-friendly chelating agents such as Ethylene Diamine Disuccinic acid (EDDS) and Nitrilo Triacetic Acid (NTA) for heavy metals extraction from soil and sediments [23,24]. EDTA has been selected as one of the most effective chelating agents mainly for four reasons:

- EDTA has an effective ability in chelating the metal cations (e.g. Pb, Cd, Cu and Zn);
- EDTA can be applied in washing various types of soils;
- recoverability and resusability of EDTA after the process of soil washing;
- extracting heavy metals from the soil by applying this agent does not result in strong acidification of the soil [25].

Also, it is shown that NTA has their biodegradability as opposed to EDTA so that is being employed for metals extraction [26,27].

The present study describes a column experiment in which the efficiencies of EDTA and NTA as chelating agents in removing Pb, Cd, and Zn through soil washing process from sandy-loam were assessed. It should be noted that the experiments were conducted on an artificially contaminated sandy-loam and also real ones taken from the field.

2. Materials and methods

2.1. Soil sampling and characterization

In this study, the samples of sandy-loam soil were collected from Rasht, Gillan, Iran at upper horizon of 0–20 cm depth of the soil. The samples were uniformed and air dried; and then, using a sieve with 2 mm diameter pores, the samples were characterized, which can be seen from Tables 1 and 2. To prepare stock solutions of Pb, Cd and Zn, lead nitrate (99.0 w/w%), cadmium nitrate tetrahydrate (99.0%), and Zinc nitrate hexahydrate (99.0 w/w%) were applied respectively. These solutions were then used to prepare synthetically contaminated soils. EDTA (99.0 w/w%) and Nitrilo Triacetic Acid NTA (99.0 w/w%) were used to prepare in the preparation of as soil washing solutions. According to the methods proposed by Reddy and Chinthamreddy [28], the soils were then spiked with a Quinter nary mixture of lead, cadmium and zinc nitrates in order to have higher levels of metal contamination. 130 mL of 5000 mg/L of the quinternary metal stock was added to 1300 g of the air-dried parent soil, and then the suspension samples were placed on a shaker with 180 rpm and room temperature of 25–27 °C for 48 h; after this, the samples were incubated for 4 weeks. The spiking stage, as mentioned above, was intended to furnish about 500 mg/kg as target concentration [15,28].

Table 1
Physical and chemical characteristics of soils used for artificial contaminated soil.

Parameters (Unit)	Values
Sand (%)	68
Silt	20
Clay	12
pH H_2O	6.5
OC (%)	3.12
CEC (meq/100 g)	17.8
Cd (mg/kg Soil)	0.82
Zn (mg/kg Soil)	78.3
Pb (mg/kg Soil)	38.9
Ca (mg/kg Soil)	4194
Mg (mg/kg Soil)	106

Table 2
Physical and chemical characteristics of soils used as real contaminated soil.

Parameters (Unit)	Values
Sand (%)	62
Silt	28
Clay	10
pH H_2O	6.9
OC (%)	2.8
CEC (meq/100 g)	19.2
Cd (mg/kg Soil)	36.88
Zn (mg/kg Soil)	642.5
Pb (mg/kg Soil)	923.5
Ca (mg/kg Soil)	5155.25
Mg (mg/kg Soil)	215.44

In addition, the real contaminated soils were collected from Lead and Zinc industries in Zanjan, Iran. When the samples were collected, it was tried to characterize the soils, which can be seen from Table 2. A column with diameter and height of respectively 7.5 cm and 15 cm was built and filled with the air-dried soil (1.46 kg per column); and also, the soil was pressurized to obtain a uniform bulk density of 1.8 ± 0.05 g/cm^3 using compact machine.

The soil columns were at first saturated with deionized water, flowing upward to prevent the entrapment of air within the soil pore space. It should be noted that leaching of the solution contained with chelating agents through the high density soil column caused by gravity force takes a long period of time. In addition, a gradual decrease in the flow rate occurs which is mainly due to decreased hydraulic conductivity of solution during the leaching; considering this, the pilot scale soil columns which were prepared for single and multi-metals extraction processes were operating under 2.5 PSI pressure. The pilot is shown in Fig. 1.

In order to assess the effect of the concentration of the agents used in this study on the efficiency of the process, three concentrations of 0.1, 0.01 and 0.005 M for each agent were

Fig. 1. Photograph of the pilot scale soil columns.