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REMOVAL OF HEAVY METALS (Cu, Cd AND Zn) FROM CONTAMINATED SOILS USING EDTA AND $FeCl_3$

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ABSTRACT

Environmental pollution caused by heavy metals from different industrial activities constitutes a serious risk for the environment. Soils contaminated with metals, such as Cu, Cd and Zn, are often subjected to physical or chemical remediation procedures to purify soils from these metals. Typical chelating agents used for metal extraction and soil washing generally include ethylene-diaminetetraacetic acid nitrilotriacetic acid, diethylenetriaminepentaacetic acid and citric acid. The subject study evaluating the potential of soil washing methods using EDTA and ferric chloride on two types of soils (coarse grained, fine grained). The effects of operating parameters, such as liquid/solid ratio, soil washing chemicals and washing time were examined. In extraction procedure of Cd (266 mg kg^{-1}), Cu (194 mg kg^{-1}) and Zn (497 mg kg^{-1}) from contaminated coarse grained soil with using 0.01 M $FeCl_3$ washing solution (liquid/solid ratio 20) for 2 hours, contaminants were removed 96.66%, 90.02% and 98.25%, respectively. In extraction procedure of Cd (218 mg kg^{-1}), Cu (153 mg kg^{-1}) and Zn (441.6 mg kg^{-1}) from contaminated fine grained soil with using 0.01 M $FeCl_3$ washing solution (liquid/solid ratio 20) for 2 hours, contaminants were removed 98.18%, 97.48% and 98.05%, respectively. Better removal efficiencies have been obtained by using $FeCl_3$. These results confirmed the effectiveness of the soil-washing method with $FeCl_3$ in remediating heavy metals (Cu, Cd and Zn) from different types of soil.

Keywords: EDTA, $FeCl_3$, Heavy Metals, Soil, Soil Washing

1. Introduction

The rapid development of industry and the use of chemical substances in industrial and military activities such as electroplating, metalworking, refinishing, mining, and munitions manufacturing generate metal-contaminated by products that are responsible for a large portion of metal contaminated sites in soils (Tejowulan and Hendershot, 1998). Soil contamination is becoming an important industrial activity in the world (Pociecha and Lestan, 2010). Both surface and subsurface environments have been subjected to heavy metal pollution (Peters, 1999). Cadmium, copper, lead, mercury, arsenic, chromium and zinc are the most hazardous heavy metals (Abumaizar and Smith; Yarlagadda *et al.,* 1995). Soil may be contaminated by heavy metals such as zinc, cadmium, due to the use of sludge or urban composts, pesticides, fertilisers and emissions from municipal waste incinerators, car exhausts, residues from metalliferous mining and the metal smelting industry (Sun *et al.*, 2001). Extremely high contamination is found at industrial areas, where the most frequently identified metals are lead, chromium, cadmium, nickel and zinc (Gusiatin and Klimiuk, 2012). For example, the cadmium pathways to people are soil-plant-animal-man and soil-plant-people. Cadmium-rich soil generally results in cadmium rich food, and geographical differences have been reported in daily cadmium intake and cadmium accumulation in the kidneys (Kawada and Suzuki, 1998). Zn and Cd are not biodegradable and thus tend to accumulate in living organisms, causing diseases and disorder. The elevated level of these heavy metals in water poses a serious threat to human health, animals, and ecological systems (Atar *et al.*, 2012). In Turkey, Cu, Cd and Zn-contaminated soils, which result from the improper disposal of ındustrial waste are an environmental threat in many regions. High concentrations of these metals in soils are confined primarily to certain mineral-rich geological regions and are usually present in stable forms which do not affect living organisms, and do not present risk to drinking water supplies (Abumaizar and Khan, 1996). Heavy metals are highly persistent and tend to be retained in soil matrices over thousands of years. Heavy metals in soil can cause various illnesses affecting human health after entering the human body and may also be toxic to microorganisms, plants and animals in addition deleterious effect on soil fertility (Sun *et al.*, 2001).

Technologies currently available for the remediation of heavy metal contaminated soils can be mainly divided into four groups, namely physical, chemical, solidification/stabilization and thermal methods (Dermont *et al.*, 2008). Especially, among of available methods, chemical extraction or soil washing remains one of the more ex situ techniques used by industry. This method is an innovative treatment technology that brings a chelating reagent into contact with the polluting metals. The chelating reagent can increase the aqueous solubility of metals and remove them from the soil particulate fraction and concentrating the contaminants into a smaller volume (Gitipour *et al.*, 2011). Among remediation technologies, soil washing cannot only extra heavy metals or metalloids adsorbed or precipitated into soils, but it can also reduce the volume of contaminated soils (Makino *et al.*, 2008). Soil washing systems incorporating most of the removal techniques offer the greatest promise for application to soils contaminated with a wide variety of organic and inorganic contaminants, but the main target contaminant group is heavy metals (Jang *et al.*, 2007; Khan *et al.*, 2004; Arwidsson *et al.*, 2010). The most widely used aqueous solution is EDTA. The practical value of EDTA is based on their abilities to form stable, water soluble complexes with many metal ions (Voglara and Lestana, 2014; Karwowska, 2014).

EDTA has the ability to chelate or complex with almost all heavy metal ions in 1:1 metal-to-EDTA complexes (Zhou *et al.*, 2011; Dipu *et al.*, 2012). This chelating effect greatly increases the solubility of heavy metals in the washing solution and is, therefore, conducive to remove heavy metals from soils There are two major considerations in the selection of chelating agents for soil washing to remove heavy metals: (i) thermodynamically assisted complexation between metals and EDTA; and (ii) EDTA-promoted dissolution (Shahid *et al.*, 2014; Zhang *et al.*, 2010; Luciano *et al.*, 2013). The principal of chemical extraction is based on the functioning of chelating reagent in the solution as surfactants to bind metal while soil is washed (Abumaizar and Khan, 1996).

In general, the weakly-bound heavy metal is relative easier to be removed by EDTA solution. Besides, heavy metals themselves also affect their removal by EDTA, because of the differences in their chelating ability with EDTA, which can be evaluated with the stability constants of heavy metal-EDTA complexes. However, to remediate the existing heavy metal-contaminated soils, only the operating conditions such as contact time and liquid/solid ratio. In the past, Eliot *et al.,* (1989) and Reed *et al.,* (1996) investigated the removal of various heavy metals from contaminated soils and showed that the complexing agent EDTA can enhance most of the heavy metal removal. Recently, several papers deal with the extraction of heavy metals, and copper in particular, by soil washing but the aim of most of them was to compare the effectiveness of different chelating agents (EDTA, citric acid) towards selected metal extraction or to optimise the solid/liquid ratio to achieve a stated metal extraction yield (Palma and Ferrantelli, 2005).

Heavy metals in soil are present in several different forms. The various metal species can be present in a soluble ionic, exchangeable, organically bound or a residual form. Certain of these forms are more mobile, while other forms are very stable and are not converted readily from one form to another. Different metal fractions within soil require different methods to separate them from the soil matrix. The most common technique employs chemical extraction with different reagents to mobilize analyte metals selectively from soil. The various metal fractions behave differently toward different chemical reagents.

Recently developed a novel cost-effective on-site soil washing technique. Calcium chloride and iron (III) chloride were selected as the soil washing chemicals because of their Cd-extraction efficiency, cost-effectiveness and low environmental impact (Nagai *et al.*, 2012; Makino *et al.*, 2006). Makino *et al.*, (2006) elucidated the mechanisms of soil washing with iron(III)chloride and demonstrated that it is more effective than other metal salts or hydro chloric acid (Nagai *et al.*, 2012). Thus, soil washing necessary to investigate an environmentally friendly and cost effective soil remediation technology (Kim and Kim, 2011).

Soil washing could allow to remove, to control or to prevent a possible pollution of soil by heavy metals. Implementation of this remove plan depends mainly on exchange reactions between heavy metals such as Cu, Cd, Zn, EDTA and $FeCl_3$. Unfortunately, this mechanism is still unclear. In order to fill this lack of knowledge, we used complementary liquid/solid ratio and contact time on the metal removal of different soil approaches. Significant new results and interpretations allowed determining the almost complete removal of heavy metals by EDTA and $FeCl_3$ in aqueous solution. In addition, the object of this study was to obtain a better understanding of the influences of soil washing on physical and chemical properties of different soil.

In this study, changes in some physical, chemical, soil properties and heavy metal speciation in a soil before and after washing with ferric chloride ($FeCl_3$) and ethylenediaminetetraacetic acid (EDTA) were assessed. Two different soils, all contaminated with Cu, Cd and Zn, from locations in Aksaray and Çanakkale were used in this study. Soil samples taken from near Aksaray and Municipality of Kepez in Çanakkale, which was contaminated by multi-heavy metals (Cd, Cu and Zn). The goal of our study was to investigate the effects of liquid/solid ratio and contact time on the metal removal of different soil. The results obtained in this study are expected to provide important insight for remediating heavy metals contaminated soils in future engineering applications.

2. Materials and Methods

2.1. Soil Sample

Surface soils (15 cm) were collected from Aksaray and Kepez district Canakkale of Turkey. The analysed soil samples were homogenized and passed through a 2 mm sieve. Rocks and other large material not passing through the sieve were removed. The soil then was left to air dry for three days while periodic hand mixing continued, being careful to break large clumps that formed during drying. Only then soils were then considered as homogenized and stored in a cooler at four degrees Celsius until they were tested.

Table 1. Physical and chemical parameters of analysed soil samples

Parameter	Soil Sample 1*	Soil Sample 2**
pH	8.67	7.80
Moisture Content (%)	1.25	5.50
Appearance	Black-coloured, coarse grained	Light-coloured, fine grained
Sand (%)	70.0	12.0
Silt (%)	23.0	20.0
Clay (%)	7.00	68.0
Total Cd (mg kg^{-1})	266.5	218.1
Total Cu (mg kg^{-1})	194.2	153,3
Total Zn (mg kg^{-1})	497.5	441.6

* Soil sample of Aksaray ** Soil sample of Kepez

Total concentration of Cu, Cd and Zn in the soils were analysed determined by soil digestion with 9 mL HCl and 3 mL HNO_3 inductively coupled plasma optical emission spectrometry (Optima 2100DV ICP, Perkin-Elmer, Boston, MA). Chemical and physical tests were performed to characterize the two bulk soils that were used during this experimental process. These tests were conducted to measure the physiochemical properties of the soils. Each test was performed on homogenized bulk soils and was performed in triplicate. Recommended quality assurance and quality control samples were taken in each step of analysis to increase the accuracy of the results. Table 1 lists individual methods used to characterize soils.

The procedures to measure pH for this research was performed with an aqueous soil suspension 1:2 (m/v) and pH meter (WTW, Weilheim, Germany).

2.2. Soil Contamination

The soil samples were artificially contaminated in the laboratory. About 0.5 kg of oven dry soil samples, surfactant and target contaminants, Cu (II), Cd (II) and Zn (II) in the form of $Cu(NO_3)_2*1/2H_2O$, $CdSO_4*8\ H_2O$ and $Zn\ (NO_3)_2*7\ H_2O$ and for digestion before ICP analysis, formed the experimental materials. Distilled water was used for washing, diluting and for use as a control. The slurry followed by shaking them separately on a wrist action shaker (Zhicheng, ZHWY-200B) at 100 rpm for 48 hours at room temperature (25 ± 2 °C). After centrifugation at 3000 rpm for 15 minutes, the supernatant was removed and the two soil samples were dried in the oven at 100 °C for 48 hours. Following this, the dried contaminated soil samples were kept for two month.

2.3. Extracting Agent

Two soil-washing extracting agents were chosen to test for this experiment. As stated previously, acids and chelating agents are the two most common types of extracting agents used in soil-washing processes. In the common acid extracting agents (nitric, hydrochloric, and oxalic) nitric was chosen to represent acid extracting agents because it is widely employed in full-scale soil-washing processes. Similarly, EDTA was chosen (from EDTA, DTPA, and NTA) to represent chelate extracting agents because of its wide-spread use as a chelating agent in full-scale soil washing processes. Chelating agents were used in this study: Na_2EDTA (Sigma-Aldrich, 99% pure) and ferric chloride $FeCl_3*\ 6H_2O$ (Merck 99% pure).

2.4. Batch Experiments

Batch experiments were then conducted to investigate different parameters involved in the surfactant-enhanced removal of heavy metal contaminated soils. In the batch experiments, 5 grams of soil and air-dried soil were placed in glass bottles containing 50 ml. All soil samples were dried at 105 °C for a minimum of 24 hours before usage. Two different agent solution ($FeCl_3$ and EDTA) at different concentrations were used to determine their effects on the extraction of Cu, Cd and Zn from the contaminated soil. The detailed operating parameters are listed in Table 2. and Table 3. Batch extraction experiments were conducted at a room temperature of 25 ± 2 °C and rotary shaker of 200±5rpm.

Table 2. Washing combinations of EDTA and $FeCl_3$-enhanced soil washing for soil sample of Aksaray

Experiment	Liquid/solid ratio	EDTA Concentration (M)	Washing time (hour)
Exp. 1	10	0.01	0.5
Exp. 2	10	0.01	1
Exp. 3	10	0.01	2
Exp. 4	20	0.01	0.5
Exp. 5	20	0.01	1
Exp. 6	20	0.01	2
Experiment	**Liquid/solid ratio**	**$FeCl_3$ Concentration (M)**	**Washing time (hour)**
Exp. 7	10	0.01	0.5
Exp. 8	10	0.01	1
Exp. 9	10	0.01	2
Exp. 10	20	0.01	0.5
Exp. 11	20	0.01	1
Exp. 12	20	0.01	2

Table 3. Washing combinations of EDTA and $FeCl_3$-enhanced soil washing for soil sample of Kepez

Experiment	Liquid/solid ratio	EDTA Concentration (M)	Washing time (hour)
Exp. 13	10	0.01	0.5
Exp. 14	10	0.01	1
Exp. 15	10	0.01	2
Exp. 16	20	0.01	0.5
Exp. 17	20	0.01	1
Exp. 18	20	0.01	2
Experiment	**Liquid/solid ratio**	**$FeCl_3$ Concentration (M)**	**Washing time (hour)**
Exp. 19	10	0.01	0.5
Exp. 20	10	0.01	1
Exp. 21	10	0.01	2
Exp. 22	20	0.01	0.5
Exp. 23	20	0.01	1
Exp. 24	20	0.01	2

For each tests involving a washing solution, the volume of the solution chosen was 40 ml, because the glass bottles size was 50 ml. 40 ml of solutions were added at varying concentrations to the reactors (glass).

The samples were equilibrated in a wrist action shaker at 2000 rpm for 15 min, and later centrifuged for about 5 minutes, and the supernatant was taken for subsequent ICP analysis of metal concentration. All the batch experiments except those for tests related to contaminant aging were done in triplicate and the reported values denote average metal concentrations. Blank control was performed throughout the experiment.

3. Results and Discussion

3.1. Soil Test

Heavy metals-contaminated soils were chosen for testing based upon their soil classification. A soil most representative of a sand and a clay were chosen so that it could be determined. The sand material selected was from the Aksaray. The clay was collected was from the Kepez, Çanakkale, in Turkey. Aksaray soil has been classified as sand. The colour of the Aksaray soil is a black and coarse grained and soil pH was 8.67. The ground of copper, cadmium and zinc in Aksaray soil was 194.2 mg kg^{-1}, 266.5 mg kg^{-1} and 497.5 mg kg^{-1} respectively. The soil had relatively low level of copper. Kepez soil has been classified as clay. The colour of the Kepez soil is a light and fine grained soil pH was 6.80. The total concentrations of copper, cadmium and zinc were 153.3, 218.1 and 441.6 mg kg^{-1}, respectively. The soil had relatively high level of zinc.

3.2. Effect of EDTA

EDTA is the complexing agent selected for soil washing. To know the effect of Different washing times and liquid/solid ratio to EDTA in removing heavy metal Cu (II) and Zn (II), a few tests were performed. To this end, the different conditions individually to EDTA and the metal removal efficiency was determined. Ethylenediaminetetraacetic acid, a member of the polyamino carboxylic acid family, is a hexahydric acid which act in this way arc called chelating agents and the complexes they form are metal chelates (Kumar, 2011). Typically, the more donor atoms involved in the chelation of a metal cation by a single ligand, the higher the stability of the resulting complex (Kim *et al.*, 2003) The molar concentration of EDTA bigger than the molar concentration of all the heavy metal species presented in the soil. A lot of work used the molar ratio of chelate/metal ([EDTA]/ [Me]) to represent the stoichiometric dose of EDTA.

Different washing times and liquid/solid ratio were used and the extraction results are shown in Fig. 1 and Fig. 2 for samples of Aksaray soil.

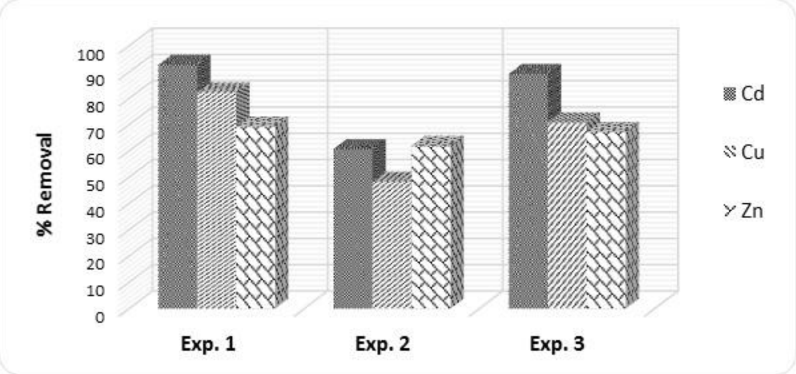

Figure 1. Metal removal among different washing combinations (Exp.1-3)

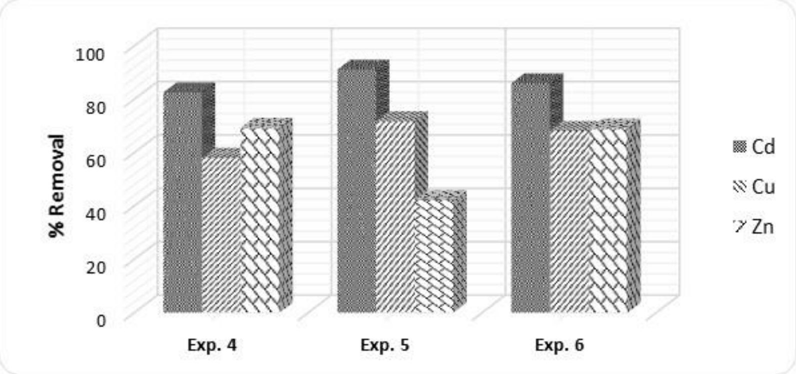

Figure 2. Metal removal among different washing combinations (Exp.4-6)

The obtained removal efficiency of three metals at Exp. 1 The removal of Cd, Cu and Zn were 91.75, 82.27, and 69.24 % respectively. The removal of Cd, Cu, Zn by washing solution using EDTA was rapid during the initial period of washing time. The removal of three heavy metals by washing solution using EDTA was rapid during the initial period of washing time and then on 0.5th and 1st hours heavy metals removal decrease finally 1st and 2nd hours the removal percentage of three heavy metals increased. The same conditions were applied to soil samples taken from Kepez. Extraction results are shown in Fig. 3 and Fig. 4.

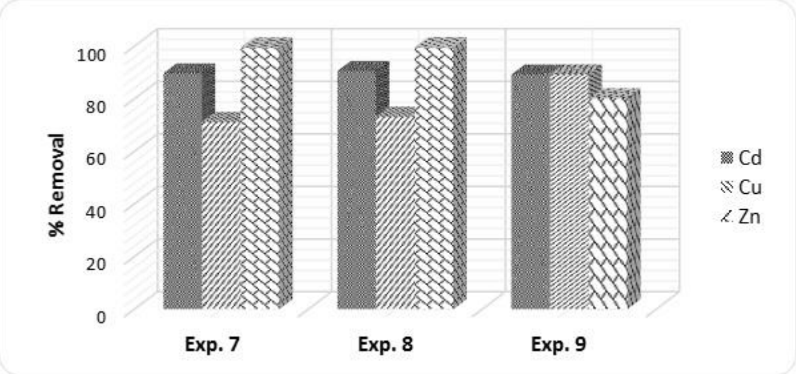

Figure 3. Metal removal among different washing combinations (Exp.7-9)

As can be seen in these figures, the best removal efficiency were obtained in Exp.11 for Cu and Cd. The removal of Cd and Cu were 90.57 and 73.22 % respectively. The obtained best removal efficiency of Zn at Exp. 8. The highest removal efficiency of 99.59% was achieved for zinc using 0.01 M EDTA washing solution with liquid/solid ratio 20 for 1 hour. Udovic and Lestan (2009) reported that EDTA can effectively remove %73 of Pb, %23 of Zn and %74 Cd from soil.

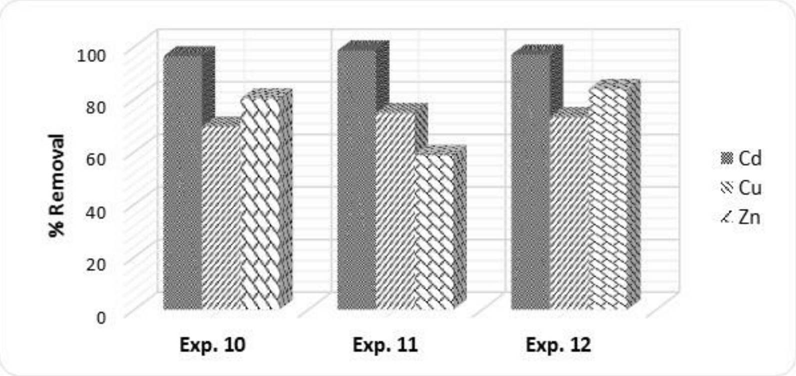

Figure 4. Metal removal among different washing combinations (Exp.10-12)

3.3. Effect of $FeCl_3$

The effect of FeCl3 on the removal of heavy metals contaminated soils was studied. Its show that in Fig. 5 and 6 for samples of Aksaray soil metal removal among different washing combinations.

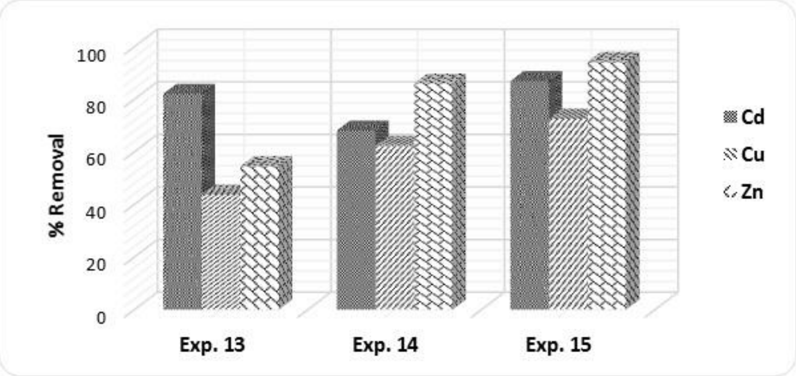

Figure 5. Metal removal among different washing combinations (Exp.13-15)

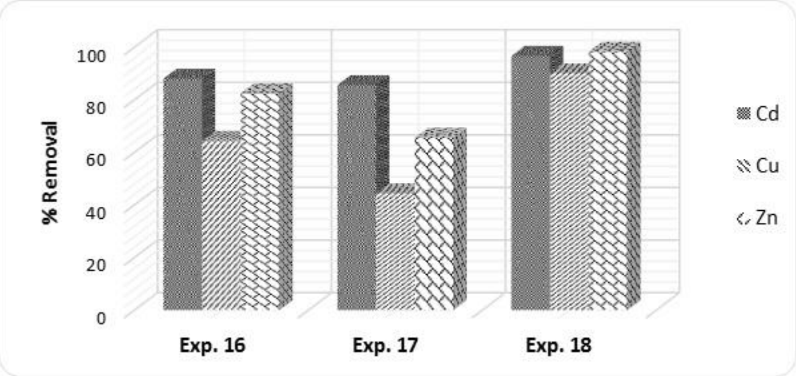

Figure 6. Metal removal among different washing combinations (Exp.16-18)

High L/S ratio improved removal efficiency. The removal of Cd, Cu and Zn were 96.66, 90.02, and 98.25 % respectively. Different washing times and liquid/solid ratio were used and the extraction results are shown in Fig. 7 and Fig. 8. for samples of Kepez soil.

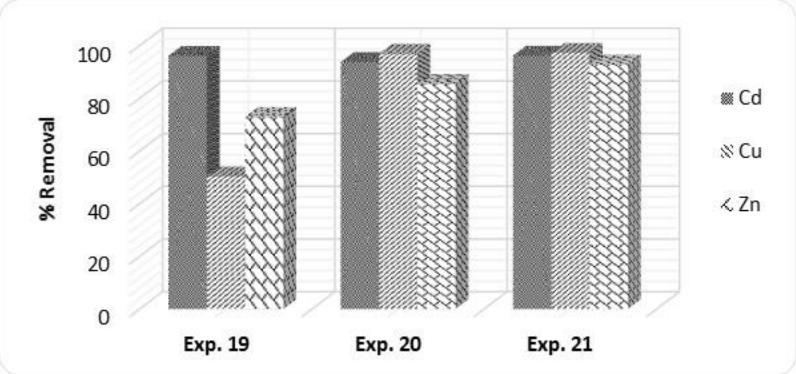

Figure 7. Metal removal among different washing combinations (Exp.19-21)

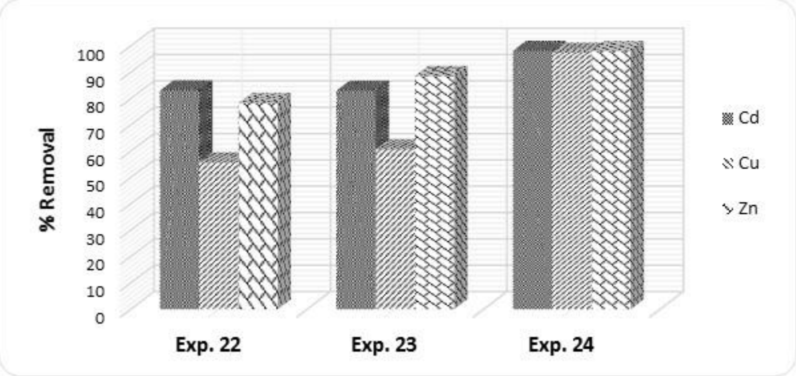

Figure 8. Metal removal among different washing combinations (Exp.22-24)

The best removal efficiencies were obtained in Exp.24 for all metals (L/S:20, $FeCl_3$ conc.:0.01 M, Washing time: 2h, and 200 rpm). The removal of Cd was 83.14, 83.05, and 98.18% when the time was 0.5, 1, and 2 h, respectively. For Cu, the removal increased from 0.5 to 2 h (from 55.81% to 97.48%). Zn had similar trends to Cd and Zn reaching a constant level at around 98 % when the washing time was above 2h. Lim *et al.* (2005) demonstrated that the optimum condition established through this approach could precipitate 95%, 89% and 90% of the extracted Pb, Cd and Ni, respectively.

4. Conclusions

The results of our laboratory scale study indicated that the soil washing is viable for treatment of EDTA and $FeCl_3$ soil extractants, obtained during the leaching of soils contaminated with Cd, Zn, Cu. EDTA and $FeCl_3$ washing were effective for heavy metal removal from soils a significant extent, both of which depended on the operating conditions and contaminant behaviours in soils. The use of EDTA and $FeCl_3$ soil extractant could lead to environmentally dangerless, friendly, feasible new soil heap leaching technology. However, much more work with different soils, heavy metals, sorbents, and operational conditions is needed to fully evaluate its sufficiency. Currently, most studies on EDTA-enhanced soil washing focus on the metal removal effectiveness but a few studies are done by $FeCl_3$. The following specific conclusions can be drawn:

- The higher removal efficiency are taken by $FeCl_3$ both soil types.
- The EDTA provided shorter washing time than $FeCl_3$ at the removal process.

- Soil washing process depends on physical and chemical structure of soil
- Both and $FeCl_3$ are used individually at soil treatment.

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