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EFFECT OF APPLICATION OF EDTA ON THE EFFICIENCY OF PHYTOEXTRACTION OF METALS FROM SOIL NEAR DUMPSITES BY SOME SELECTED PLANTS IN ZARIA NIGERIA

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ABSTRACT

The aim of this study is to investigate the effect of the application of ethylenediamine-tetraacetic acid (EDTA) on the efficiency of phytoextraction of Cd, Co, Cu, Ni, Pb, and Zn by *Vetiverazizanioide*, *Ipomoea asarifolia,Helianthus annus*, *Ricinuscommunis* L. and *Cymbopogoncitratus*harvested from the soil treated with EDTA and that not treated with EDTA (natural). The concentrations of the metals in the plant species and soil were determinedusing atomic absorption spectrophotometry. The results showed that the metal concentrations in the soil (before planting) **at the dumpsites**were higher ($P \leq 0.05$) than the control and were above (except Cu and Zn) the standard limits as recommended by FAO/WHO). **The concentrations of metals in all the plants harvested from soil the treated with EDTA were found to be higher ($P < 0.05$) than those from the soil without EDTA treatment.**The percentages removal of Cd ranged from 36.7 - 52.5 (natural) and 59.7 - 70.3 (with EDTA); Co:- 0.6 - 15.7 (natural) and 1.2 - 19.2 (with EDTA); Cu:- 24.5 - 44.3 (natural) and 32.1 - 70.5 (with EDTA); Ni:- 20.5 - 88.4 (natural) and 35.6 - 90.5 (with EDTA); Pb:- 13.0 - 21.3 (natural) and 27.9 - 42.1 (with EDTA) and Zn:- 17.1 - 28.0 (natural) and 19.9 - 36.0 (with EDTA).This demonstrated that EDTA is an efficient soil amendment additive in enhancing metals desorption from the soil by increasing their accumulation in plants, hence enhancing the efficiency of phytoextraction of the heavy metals from the soil by the plants forthe purpose of detoxification of dump sites.

***Keywords**:* Bioaccumulation, Dumpsites, EDTA, Heavy metals, Phytoextraction, Plants.

1.0 INTRODUCTION

Heavy metal contamination in soil is a major concern because of their toxicities and threat to human life and the environment. The accumulation of these metals is a consequence of several activities like chemical manufacturing, painting and coating, mining, extractive metallurgy, nuclear and other industries. The introduction of industrial and municipal solid wastes into the environment has contributed greatly to the increase in levels of heavy metals in soil, and vegetations grown in dumpsites.

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The soil and plants on these sites constitute a serious threat to the health of people living around such areas (Inuwa*et al*., 2007; Ahmadpour*et al*., 2012).

Phytoextraction process involves plant roots removing metals from contaminated soils/sediments and transporting them to leaves and stems for harvesting and disposal without destroying the soil structure and fertility (Yashim*et al*., 2015). This can be an effective remediation method at a variety of sites and on numerous contaminants (Ahmad *et al*., 2017).Phytoextraction which can effectively remove contaminants from contaminated soils is the most promising for commercial application. It has been accepted widely both in developed and developing nations for its potential to clean up the polluted and phytoextraction is the reduction inthe concentration of metals in contaminated soils to regulatory levels within a reasonable time. The extraction process depends on the ability of selected plants to grow and accumulate metals under the specific climatic and soil conditions of the site being remediated.This technology is currently gaining considerable importance due to its potential for application to real world ecosystems. It is cheaper and does not degrade the physical or chemical profile of the soil. This process uses commonly cultivated plant species producing sufficiently large vegetative matter and they are subsequently stimulated to take-up heavy metals by adding particular agents to the soil or directly onto the plants (Jarosz*et al*., 2013). For more than four decades, synthetic chelates are being widely used to increase heavy metals bioavailability in both soil and hydroponics, which can efficiently enhance phytoextraction (Azhar*et al*., 2006).Chelate-assisted phytoextraction or induced phytoextraction, in which artificial chelates are added to treated soil to increase the mobility and uptake of metal contaminants,seems to be the most promising (Jarosz*et al*., 2013). Several studies documented that chelating agents such as ethylenediamine-tetraacetic acid (EDTA), N-(2-hydroxyethyl)-ethylenediaminetriacetic acid (HEDTA), diethylenetriaminepenta acetate (DTPA), ethylenediaminedisuccinate (EDDS), Nitrilotriacetic acid (NTA) and citric acid (CA) can be used to increase metal mobility, thereby enhancing phytoextraction.

EDTA has been employed for soil remediation due to its strong complexes-forming ability. EDTA is poorly biodegraded in the soils though its effectiveness at complexing metals. Metal–EDTA complexes could increase solubility and phytoavailability of metals in soils. When EDTA is applied to soils, a large fraction of the total metals is dissolved and becomes available for phytoextraction. It is a non-selective extracting agent that can form a strong complex with a variety of metals in soils (Chen *et al*., 2012).Here, the solubilization of heavy metals must be enhanced to increase extraction efficiency.

The aim of this study is to investigate the effect of the application of EDTA on the efficiency ofphytoextraction of heavy metals from soil near metal dumpsites by *Vetiverazizanioide*, *Ipomoea asarifolia*,*Helianthus annus*, *Ricinuscommunis* l. and *Cymbopogoncitrates.*

2.0 MATERIALS AND METHODS

2.1 Study Areas

Three scrap metal dumpsites (at Dakace, Gaskiya and Hanwa) in Zaria, Nigeria (11° 07' 51" N; 7° 43' 43" E) were selected for the research work because of the activities that involve disposal of metal containing materials into the biosphere, which may lead to increase in the amount of toxic metals entering the environment. Another site which is about 2km away, where there were no activities involving disposal of metal containing materials was also selected to serve as a control.

A test area of 6m x 4m was selected at each site and divided into two parts (3m x 2m) where the studied plants (*Vetiverazizanioide*,*Helianthus annus*, *Ricinuscommunis* L., *Ipomoea asarifolia* and *Cymbopogoncitratus*) were grown. The various plant speciewere planted in the designated field plots with spacing of 20cm x 20cm for all the tested plants as described by Zhuang*et al*. (2007). The plants were allowed to grow naturally under natural climatic conditions and exposed to natural day and night temperatures, with neither mineral fertilisation nor optimum irrigation so as to assess the feasibility of the remediation process. Weeds were controlled by mechanical method. After 10 weeks when the plants might have achieved maximum biomass production, a single dose of 5mmol/kg EDTA (Na_2-EDTA) was applied to the soil of one part of the plots at each study site. Seven days after the application of the EDTA solution, the plants were harvested and the associated soil samples were collected (Yashim*et al*.,2016).

The choice of these plants was based on their widespread availability and demonstration of tolerance to conditions not favourable for the growth of other plants in Nigeria. Also, these plant specie have the ability to co-exist on the same field (Okoronkwo*et al*., 2014). This feature prompted their simultaneous study under the similar test conditions.

2.2 Sample Collection and Treatment

Soil samples were randomly collected from the surface to a depth of 20 cm from each dumpsite (before planting) and the control. At harvest, whole plant samples (25of each specie) were randomly collected from each of the study areas and soil samples were also collected from the surface to a depth of 20 cm around each plant root zone and then mixed together.

The collected soil samples were air-dried at room temperature for 3 days, while the plant samples were washed, cut into pieces and air dried. The dried soil and plant samples were ground, sieved (500µm sieve) and kept in clean polythene bags for further analysis.

One gram (1g) of each of the sieved soil and plant samples was digested separately with 20 cm^3 of aqua regia (a mixture of 3 parts concentrated HCl to 1 part concentrated HNO_3) on a hot plate in a fume cupboard, until a clear solution was obtained. To the hot solution, 30 cm^3 of distilled water was then added and filtered through a Whatman No. 42 filter paper into a 50 cm^3 standard volumetric flask and then made up to the mark with distilled water (Yashim*et al*., 2015). Cadmium, cobalt, copper, nickel, lead and zinc were analyzed in the plant and soil samples using atomic absorption spectrometer (AAS), with the analyses being done in triplicate.

The bioaccumulation factor (BF) was calculated to determine the degree of metal accumulation in the plants grown at the dumpsite (Sun *et al*., 2011).

$$BF = \frac{\text{Concentration of metal in plant}}{\text{Concentration of metal in soil}}$$

3.0 RESULTS AND DISCUSSION

The mean concentrations of the studied metals in the soil at the three study areas (before planting) and the control are shown in Table 1. The metal concentrations at the dumpsites were higher ($P \leq 0.05$) than the control and were above (except Cu and Zn) the standard limits as recommended by Food and Agricultural Organisation and World Health Organisation (FAO/WHO, 2010)as reported by Akpoveta*et al*., 2010. Using one-way ANOVA, there was no significant difference ($P > 0.05$) in the metal level at the three dumpsites. This implies that the dumping of scrap metals on the soil has contributed significantly to the higher concentrations of heavy metals at the dumpsites.

Table 1: Mean Concentration of Heavy Metals (mg/kg) in Soils at the Three Dumpsites and the Control before Planting

	Cd	Co	Cu	Ni	Pb	Zn
Dakace	8.00±0.03	8.30±0.04	6.50±0.03	51.30±0.04	69.30±0.04	55.93±0.11
Gaskiya	7.00±0.02	9.90±0.05	5.30±0.03	84.93±0.07	75.90±0.04	28.90±0.12
Hanwa	6.00±0.02	8.70±0.05	5.60±0.05	47.90±0.06	76.90±0.05	25.58±0.06
Control	1.87±0.01	7.37±0.04	2.49±0.03	6.50±0.02	18.23±0.03	23.95±0.08
***Standard regulatory limits**	5	8	30	40	10	90

*Standard regulatory limits as reported by Akpoveta *et al*., 2010.

3.1 Concentrations of Metals in Plants

The concentrations of the metals under study in the plants harvested from the three dumpsites are presented in Figures 1 – 3. The concentrations of metals in all the plants harvested from soil treated with **EDTA** were found to be higher ($P < 0.05$) than those from soil without **EDTA** (natural) treatment in the three metal dumpsites. It was observed that concentrations of **Co** and **Pb** were highest in *Cymbopogon citrates than other plants* harvested from soil treated with **EDTA** and without **EDTA** (natural) treatment in the three metal dumpsites. Similarly, **Cu** and **Zn** were highest in *Helianthus annus, while Cd was highest in Ipomoea asarifolia*. These results are similar to those obtained by Awokunmi*et al*. (2015) and Yue-bing*et al*. (2011). The application of EDTA raises the concentration of the heavy metals in the

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the plants from the rhizosphere of the soil, and also facilitated the metal translocation from roots to shoots.

When **EDTA** is applied to soils, a large fraction of the total metals is dissolved and becomes available for phytoextraction without inducing a strong acidification of the growth medium (Yashim*et al*., 2015; Muhammad *et al*., 2017). In general, the total phytoextraction of the metals in the plants increased significantly ($P < 0.05$) with the application of **EDTA** compared with the natural phytoextraction.

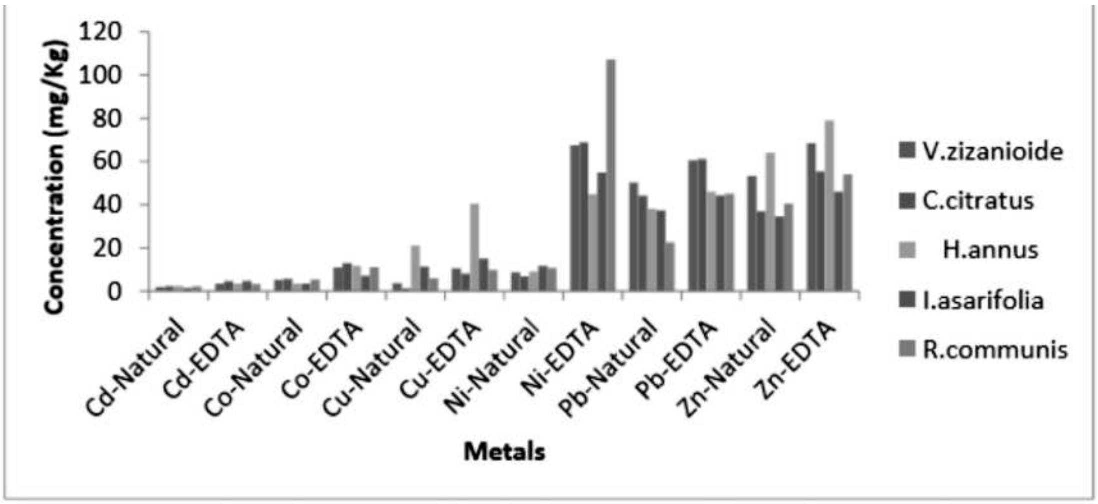

Figure 1: Concentrations of Heavy Metals in the Plants Harvested from Dakace Dumpsite

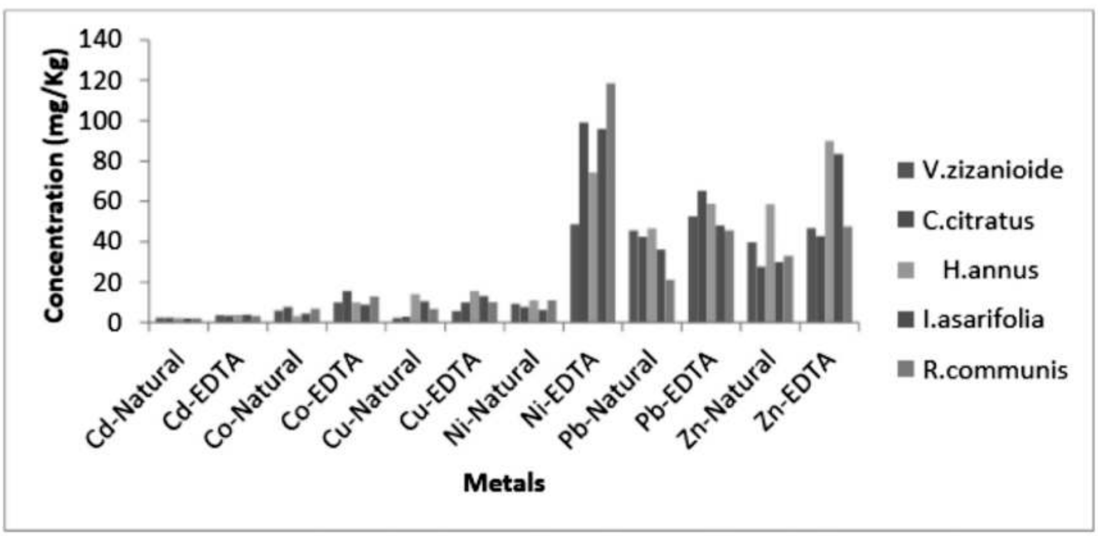

Figure 2: Concentrations of Heavy Metals in the Plants Harvested from Gaskiya Dumpsite

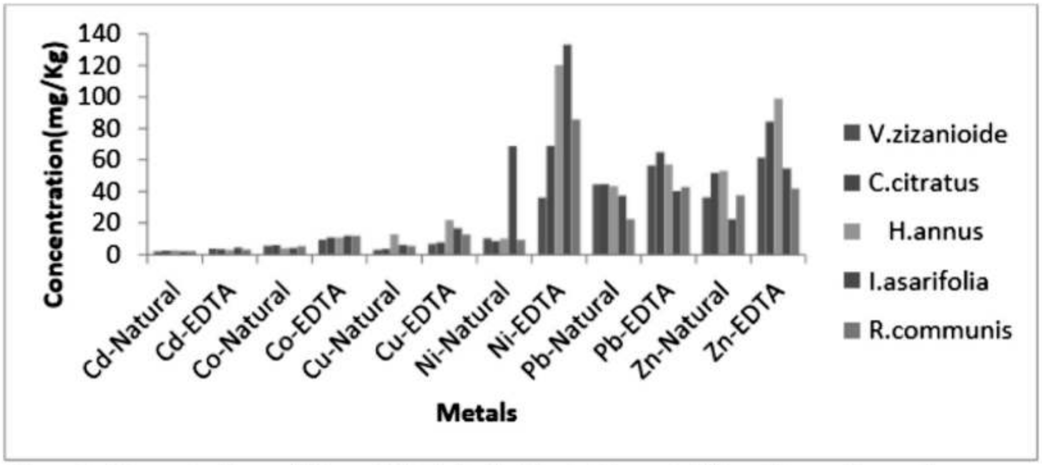

Figure 3: Concentrations of Heavy Metals in the Plants Harvested from Hanwa Dumpsite

3.2 Bioaccumulation factor (BF)

Bioaccumulation factor (BF), defined as the ratio of chemical concentration in a plant to its concentration in the soil, is used to measure the effectiveness of a plant in concentrating pollutant into the plant(*Yashim et al*., 2016).The BF for the studied metals at the three dumpsites in the plants under investigation from soil treated with EDTA and without EDTA (natural) treatment are presented in Figures 4 – 6. *Helianthus annus* harvested from soil treated with EDTA showed highest BF for Co (3.3), Cu (8.2), Ni (9.2) and Zn (6.3); Co (4.3), Cu (7.4) and Pb (2.4); Co (2.2), Cu (5.8), Ni (6.4) and Zn (3.8) at Dakace, Gaskiya and Hanwa dumpsites respectively.*Cymbopogon citrates* harvested from soil treated with EDTA showed highest BF for Cd (1.9) and Pb (1.7) at Dakace dumpsite, while it was Cd (1.8) at Hanwa dumpsite. *Ipomoea asarifolia* harvested from soil treated with EDTA at Gaskiya dumpsite showed highest BF for Ni (8.0) and Cd (1.6). *Ricinuscommunis* only showed highest BF for Pb (2.1) at Hanwa dumpsite.

For phytoextraction to become a viable technology, plants with high tolerance and accumulation rates for several metals are required because polluted soils often contain high levels of several trace metal contaminants (Ghasemi*et al*., 2017). The bioaccumulation factors were determined for each studied metal in the plant specie at the three study areas to ascertain their phytoremediation potentials.

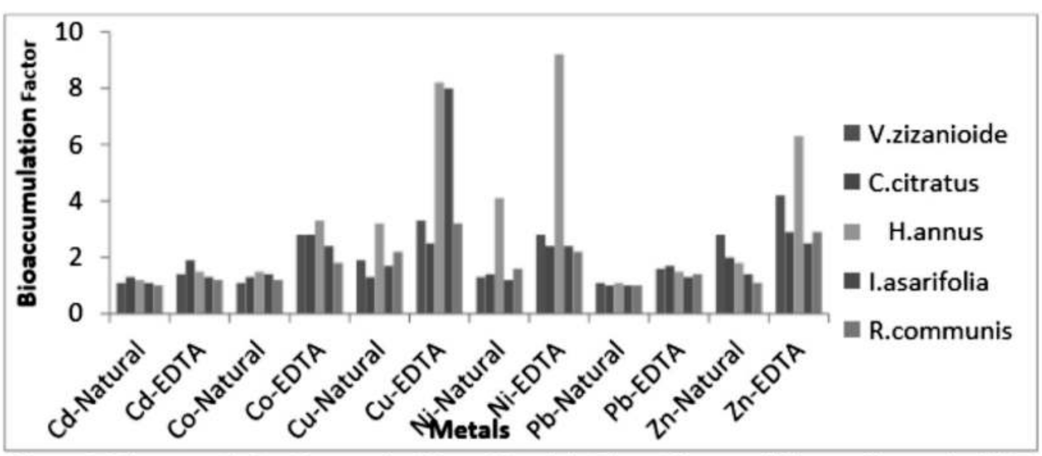

Figure 4: Bioaccumulation Factors for Heavy Metals in Plants Harvested from soil treated with EDTA and without EDTA (natural) treatment at Dakace Dumpsite

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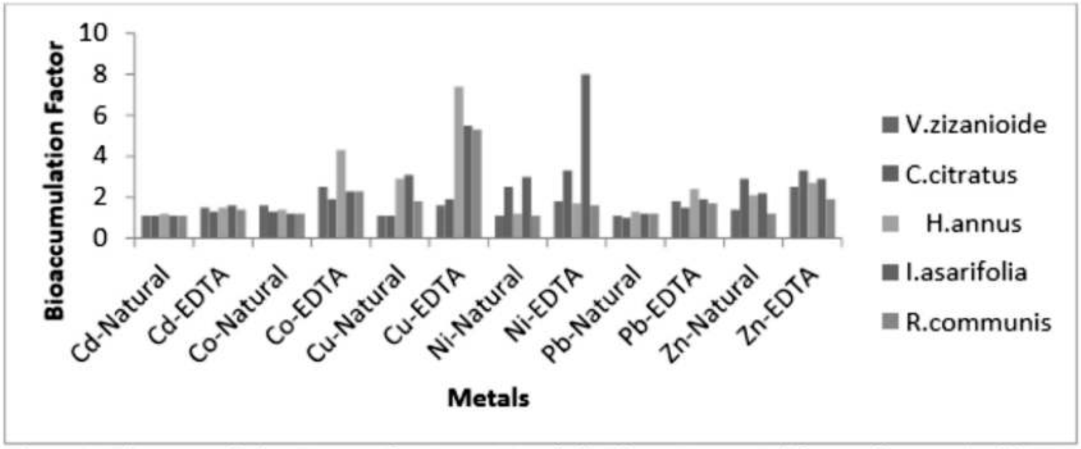

Figure 5: Bioaccumulation Factors for Heavy Metals in Plants Harvested from soil treated with EDTA and without EDTA (natural) treatment at Gaskiya Dumpsite

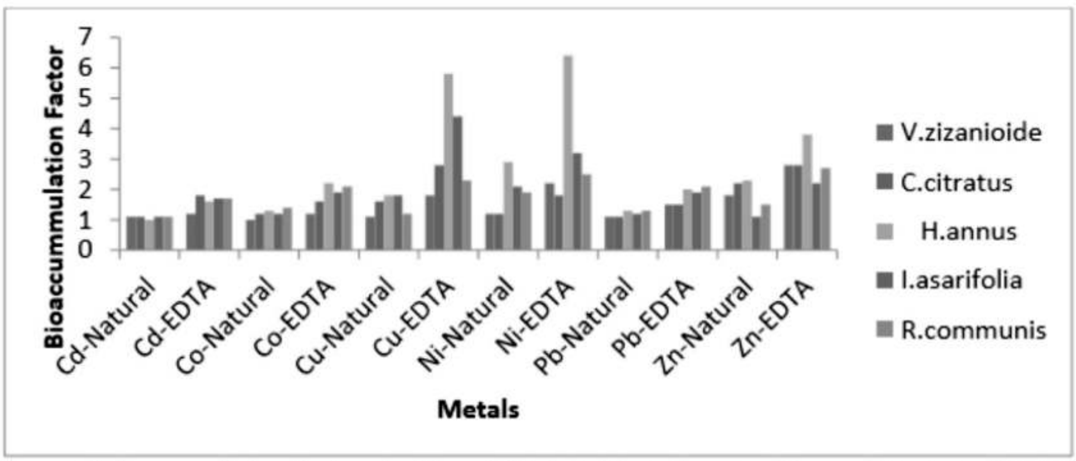

Figure 6: Bioaccumulation Factors for Heavy Metals in Plants Harvested from soil treated with EDTA and without EDTA (natural) treatment at Hanwa Dumpsite

Where the BF is greater than 1, the plant specie is a good accumulator. Plant specievary in their response to take-up of heavy metals during EDTA-assisted phytoextraction (Sun *et al*., 2011). The efficiency of phytoextraction could be enhanced by the right combination of plant specie and chelators.Complexation of heavy metals by chelates could play an important role in controlling heavy metal solubility and concentration in soil, hence their phytoextraction (Yeh*et al.*, 2012).

Phytoextraction seems to have considerable potential for decontamination of soils contaminated with heavy metals. The goal of the phytoextraction process is to reduce heavy metal concentrations in contaminated soil to acceptable levels within a reasonable time frame. Figure 7 shows the mean concentrations of the metals in the soil treated with EDTA and without EDTA (natural) treatment in the three metal dumpsites after the phytoextraction process by the five plants (*Vetiverazizanioide*, *Helianthus annus*, *Ricinuscommunis* L., *Ipomoea asarifolia* and *Cymbopogon citrates).* This study has shown that there was a decrease in the concentrations for all the studied metals in the soil after the plants were harvested from Dakace, Gaskiya, Hanwa sites. At the Dakace and Hanwa study areas, it was observed that the decrease in Co concentration was not significant *(*$P > 0.05$) as compared to other studied metals. The percentages removal (Figure 8) of the studied metals from the soils by the five plant specie planted at the study areas treated with EDTA were higher than those by the plant specie not treated with EDTA (natural). The results indicated that percentages removal of

Cd ranged from 36.7 - 52.5 (natural) and 59.7 - 70.3 (with EDTA); Co: 0.6 - 15.7 (natural) and 1.2 - 19.2 (with EDTA); Cu: 24.5 - 44.3 (natural) and 32.1 - 70.5 (withEDTA); Ni: 20.5 - 88.4 (natural) and 35.6 - 90.5 (with EDTA); Pb: 13.0 - 21.3 (natural) and 27.9 - 42.1 (with EDTA) and Zn: 17.1 - 28.0 (natural) and 19.9 - 36.0 (with EDTA).

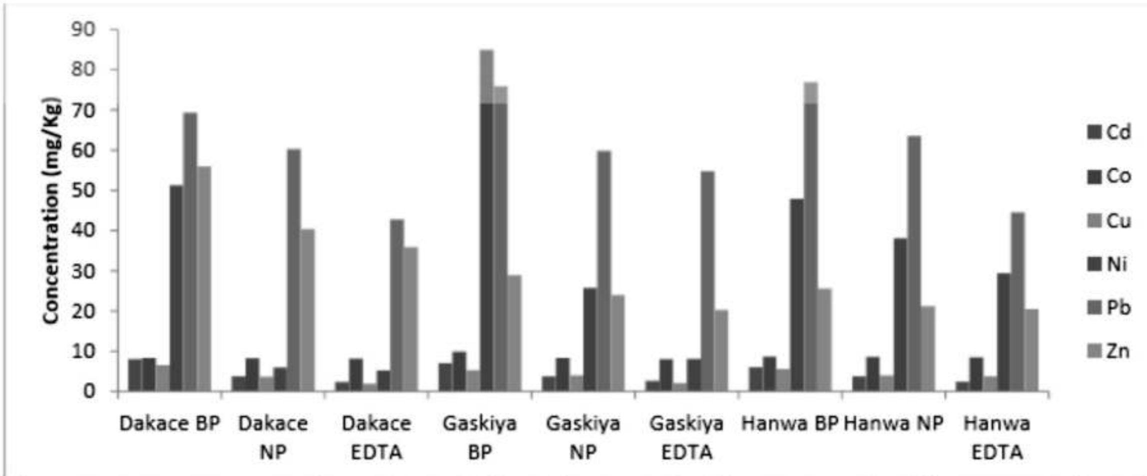

Figure 7: Mean Concentrations of Metals in Soils Treated with EDTA and Without EDTA (natural) Treatment in the Three Metal Dumpsites after the Phytoextraction Process

BP = Before Phytoextraction, NP = Natural Phytoextraction

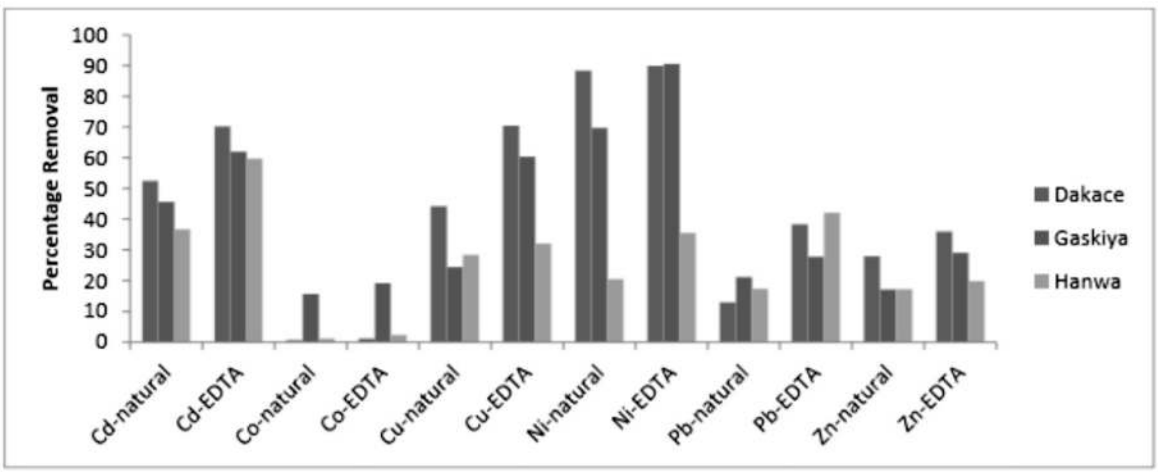

Figure 8: Percentages Removal of Heavy Metal from Soil Treated with EDTA and Without EDTA (natural) Treatment in the Three Metal Dumpsites after the Phytoextraction Process

Phytoextraction seems to have considerable potential for decontamination of soils polluted with heavy metals. The application of EDTA to soil containing heavy metals (Cu, Co, Cd, Ni, Pb and Zn) enhanced the efficiency of phytoextraction of heavy metals from the soil by *Vetiverazizanioide*, *Helianthusannus*, *Ricinuscommunis* L., *Ipomoea asarifolia* and *Cymbopogon citrates*.

The results of this study showed that EDTA is an efficient soil amendment agent in enhancing Cu, Co, Cd, Ni, Pb, and Zn desorption from soil and in increasing their accumulation in plants. The data obtained in the experiment confirmed that all the plants were tolerant to heavy metals. It is generally noted in the literature that EDTA has taken a predominant place in increasing metal removal efficiency (Avtukhovich and Postnikov, 2017).

Combining cropping or co-cropping system has enhanced the phytoextraction rates and the leaching risk caused by the application of the EDTA (Wei *et al*., 2011). Fuksova*et al*. (2009) reported that the remediation efficiency of the individual plant species in the co-cropping system did not differ from those obtained in separate cropping mode. It improved the survival rate of the individual plant/crop and reduced the time period to accomplish the decontamination of a polluted soil (Jenna, 2012; Almas *et al*., 2012).

4.0 CONCLUSION

The percentage removal of the studied metals from the soils by the five plant species planted at the study areas treated with EDTA were higher than those of the plant species not treated with EDTA (natural). Phytoextraction seems to have considerable potential for decontamination of soils polluted with heavy metals. The application of EDTA to soil containing heavy metals (Cu, Co, Cd, Ni, Pb and Zn) enhanced the efficiency of phytoextraction of heavy metals from the soil by *Vetiverazizanioide*, *Helianthusannus*, *Ricinuscommunis* L., *Ipomoea asarifolia* and *Cymbopogon citrates* for remediation purposes.

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