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ORIGINAL PAPER

# Efficiency of chelating agents in retaining sludge-borne heavy metals in intensively applied agricultural soils

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**Abstract** This paper presents an evaluation of different chelating agents for their effectiveness in removing Cu, Co and Zn in three distinctly different types of sludge-amended soils. Soil types (Luvisol, Arenosol and Vertisol) were each mixed with an anaerobically digested sludge at a 1:1 ratio followed by leaching with three types of chelating agents, namely: ethylenediamine tetra acetic acid, nitrilotriacetic acid and acetic acid. Aqua regia method was used to quantify pseudo total metal before and after treatment. Generally, chelating agents can be out competed by soil colloids in attracting cations. The efficiency of chelating agents was found to follow this order ethylenediamine tetra acetic acid $<$ nitrilotriacetic $=$ acetic acid in all the three metals, with ethylenediamine tetra acetic acid being the most effective chelating agent. More heavy metals were removed in Luvisol and Arenosol than in Vertisol implying that soils rich in clay fraction retain more cations than soils with minimal clay fraction. Similarly, copper responded positively to chelation than zinc and cobalt in Luvisols and Arenosols, although the results were not conclusive for Vertisols.

**Keywords** Chelating agents · Heavy metals · Ligands · Sewage sludge

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## Introduction

The use of anaerobically digested sludge as an effective soil conditioner may lead to bio accumulation of toxic heavy metals in the terrestrial environment. In particular, heavy metal contamination of the soil is one of the most important environmental problems throughout the world (Doumett et al. 2008; Nouri et al. 2006; Wuana et al. (2010). For instance, biosolids can significantly increase heavy metal concentration in cultivated soils (Sloan et al. 2001) and/or agricultural soils (Steinnes 1990). Concern of the pollution caused by heavy metal-containing sludge has accentuated the need to carry out numerous investigations as well as to develop various methods/techniques to remove the toxic heavy metals from contaminated sites. Some of the popular methods include ion exchange (Parkpian et al. 2002), Phyto-remediation (Neugschwandtner et al. 2008) and chemistry (Sanchez-Martin et al. 2007; Khodadoust et al. 2005; Ito et al. 2000, Naoum et al. 2001; Marchioretto et al. 2002). Moreover soils are effective agents of heavy metal sorption, particularly soils with high contents of organic matter, clays and Fe, Al, Mn oxides and hydroxides (Tessier et al. 1995; Gong and Donahoe 1997). Although experimental trials in the field of heavy metal removal from sewage sludge are numerous (Lee et al. 2005; Veeken and Hamelers 1999; Naoum et al. 2001), it is evident that knowledge gaps still exist warranting a need to further study the interaction of heavy metal with different types of soils as well as to devise heavy metal removal techniques. Few, if any, studies have attempted to study ways of preventing and discouraging transfer of heavy metals from sludge to soil. Despite its toxicity and heavy metal load, sludge is a valuable by product since it is a source of all essential nutrients and has also been used to improve soil porosity, structure, bulk density, aggregation and water holding capacity (Ngole 2007). Using adequate chemical or biological methods, a selective removal of heavy metals to make sludge non-hazardous is possible and it is even possible to remove and recover heavy metals for possible reuse (Lee et al. 2002). The objectives of this study are twofold: (a) to assess the potential of three chelating agents; Ethylenediamine tetra acetic acid (EDTA), Acetic acid (AA) and Nitrilotriacetic acid (NTA) to prevent transfer of heavy metals from sludge to the soil, and (b) to study the effects of soil type–ligand interactions on the decontamination of sludge. Wuana et al. (2010) reported that the solubilization/exchange/extraction of heavy metals by washing solutions differs considerably for different soil types. Hence the study focused on the extraction of Zn, Cu and Co in three soil types with contrasting soil textures: Luvisol, Vertisol and Arenosol.

Recent study on sewage effluent and composited sludge (Emongor and Ramolemana 2004) as well as past studies (Leeper 1978) among an array of heavy metals have singled out Cu, Co, Zn and Cd as the most noticeable toxic metals in both composited sludge and dammed waste ponds. Hence, the choice of the three heavy metals (Cu, Co and Zn). In particular, Cu and Zn are reported to build-up on subsurface soils following additions or irrigation with consequent accumulation on vegetable crops grown in that media (Dikinya and Areola 2010).

## Materials and methods

### Soils and sample preparation

Samples of three different soil types: Luvisol, Arenosol and Vertisol (FAO 1998) with distinct physical and chemical characteristics were collected from the most agricultural active regions of Botswana. Particle size analysis showed huge variations in their soil textures (Table 1). Samples from these soils were each sieved to a <0.02 mm fraction before analysis. The anaerobically digested sewage sludge were sampled from the Gaborone City Council wastewater treatment plant and later pulverized to fineness (<0.02 mm) using a bench top ScutteBuffalo hammer mill. The sludge sample was subsequently air dried and oven dried at 50°C to ensure complete dryness prior to analysis.

**Table 1** Particle size analysis of the soil samples

| Soil type | Sand (%) | Silt (%) | Clay (%) | Soil texture |
|---|---|---|---|---|
| Luvisol | 21 | 60 | 19 | Silt loam |
| Arenosol | 88 | 5 | 7 | Sand |
| Vertisol | 10 | 70 | 20 | Silt clay loam |

### Experimental design

The experiment was a two-factor Completely Randomized Design where the primary factor was homogenized 1:1 (soil:sludge) mixtures of Luvisol, Arenosol and Vertisol. The secondary factor was three types of chelating agents: ethylene-diamine-triacetic-acid (EDTA), acetic acid (AA) and Nitrilotriacetic acid (NTA) including a control (where no chelating agent was applied). In this study, samples of the three soil types were each leached with three chelating agents (EDTA, NTA and AA). There were a total of 36 samples (12 samples from each soil type mixture), 3 samples within each soil type samples receiving each treatment. A response in this case is denoted by $Y_{ijk}$, where $i$ indicates the soil type, $j$ indicates the chelating agent and $k$ is the observation number. The means denoted by $Y_{average}$ are means for each treatment combination. Table 2 summarizes the experimental design.

### Baseline analysis of sludge and soil mixing

To gather baseline data/information, three replicates of dried sludge samples were analyzed for several variables including pH and concentration of Zn, Cu and Co using standard methods (APHA 1989). Analysis of pure sludge and each of the three types of soils for total concentration of Zn, Cu and Co was intended to gather baseline information on total metal content on soils and sludge before the two were mixed. Aqua regia method of metal extraction (Alloway 1995) and 5th Step of Tessier and Campbell (1979) as described in the methodology section were used to assay the total concentration of Zn, Cu and Co on both sludge and the three types of soils. Meanwhile, the pH measurement in sludge was accomplished by adding 25 mL of $H_2O$ to each 10 g of the three replicates of sludge

**Table 2** Notation for two-way experimental design

| Soil type | Chelating agents | | | |
|---|---|---|---|---|
| | EDTA (1) | NTA (2) | AA (3) | Control (4) |
| Luvisol | $Y_{111}$ | $Y_{121}$ | $Y_{131}$ | $Y_{141}$ |
| | $Y_{112}$ | $Y_{122}$ | $Y_{132}$ | $Y_{142}$ |
| | $Y_{113}$ | $Y_{123}$ | $Y_{133}$ | $Y_{143}$ |
| | $Y_{average}$ | $Y_{average}$ | $Y_{average}$ | $Y_{average}$ |
| Arenosol | $Y_{211}$ | $Y_{221}$ | $Y_{231}$ | $Y_{241}$ |
| | $Y_{212}$ | $Y_{222}$ | $Y_{232}$ | $Y_{242}$ |
| | $Y_{213}$ | $Y_{223}$ | $Y_{233}$ | $Y_{243}$ |
| | $Y_{average}$ | $Y_{average}$ | $Y_{average}$ | $Y_{average}$ |
| Vertisol | $Y_{311}$ | $Y_{321}$ | $Y_{331}$ | $Y_{341}$ |
| | $Y_{312}$ | $Y_{322}$ | $Y_{332}$ | $Y_{342}$ |
| | $Y_{313}$ | $Y_{323}$ | $Y_{333}$ | $Y_{343}$ |
| | $Y_{average}$ | $Y_{average}$ | $Y_{average}$ | $Y_{average}$ |

samples (ratio 2:5) followed by vigorous shaking for 30 min at 120 rpm. The pH was then measured on the supernatant liquid using a pH electrode. Similarly, the sludge samples were mixed with each of the three soil types samples at volume per volume (v/v) percent ratio of 50:50 sludge:soil. In order to enhance sludge–soil interaction, the mixtures of each of the three soil samples were homogenized and incubated for a week at 25 ± 3°C under suitable humidity of approximately 50% achieved by continuous wetting of the sample as and when necessary.

Determination of heavy metals in soils

A 100 mL of deionised water was added to a 50 g of each soil sample and saturated to field capacity, shaken (120 rpm) and allowed to stand in order to allow solvation of metal ions held in sludge particles followed by leaching with a 200 mL of 0.05 M of each chelating agent. In case of control, a 200 mL of deionised water was used to leach the sample. The leaching was achieved by filtering a sample through a Whatman No. 42 filter paper. The leachate was discarded and the resultant soil was oven dried to constant weight at 50°C analyzed for the total concentration of Zn, Cu and Co. Merck analytical reagents and demineralized water from a Millipore Milli Q system were used for analytical preparations. All extraction procedures were performed using laboratory glassware and polythene bottles cleaned by HCl and rinsed with double distilled water.

Though modified, the aqua regia method for extraction of heavy metal was based on the one described by Alloway (1995) and the 5th step of Tessier and Campbell (1979). Aqua regia extraction is commonly used to determine the total metal content (pseudo total content) in samples of sludge, soil and sediment (Fuentes et al. 2004). In order to assay the total concentration of Zn, Cu and Co; 20 $cm^3$ of concentrated Nitric acid ($HNO_3$) and 10 $cm^3$ of Hydrochloric acid (HCl) was added to 2 g of each of the sample types. The mixture was then gently heated on a hotplate at 70–80°C to dryness. A modified concentration of $HNO_3$ approximately 3.3 mol $dm^{-3}$ was added to the dry residue and then heated for 1 h, filtered using Whatman, hardened low ash filter paper (Grade 54; 22 µm) and subsequently transferred quantitatively into a 50 mL volumetric flask and filled to volume with ultra pure water. The metals of concern were then assayed in the solution obtained using Flame Atomic Absorption Spectrometer model Varian FS 240.

Statistical procedures

Analysis of Variance (ANOVA) was used to evaluate the effects of different treatments. Standard deviation was used to indicate variability, and the least significant difference (LSD) test at a confidence level of 95% and Duncan's multiple range system was used to separate means. Statistical program SAS was used for analysis.

## Results and discussion

Characterization of soil samples and sludge

Physico-chemical characterization of soils is presented in Table 3. Generally, the soil types exist/occur at circumneutral pH of around pH ~ 7 and were significantly different in their organic carbon (% OC) content (Table 3). Vertisols had the highest mean organic carbon of 7.4% whereas Arenosols had the lowest organic carbon content of 3.6%.

Table 4 shows the concentrations of heavy metals in soils before treatment. The results show that Luvisols and Vertisols which are rich in the clay and silt fractions had a high concentration of the heavy metals (Zn, Cu and Co) than Arenosols. Higher heavy metal concentration is directly proportional to % OC and % OM in addition to clay fractions. Humic soils such as Vertisols which are rich in organic matter content have a high nutrient retention and water holding capacities and are consistent with the results in Table 3. It is worth noting that Luvisols and Vertisols are composed of a significant amount of clay and silt fractions. It is the clay fraction of the soil which is colloidal in nature and as such has high affinity to positively charged ions.

Similarly, the initial heavy metal load of sludge ($S_{initial}$) was 40, 26 and 17 mg/kg for Zn, Cu and Co, respectively. Total concentration of heavy metals in sewage sludge indicates the extent of contamination, but gives little insight into the forms in which the metals are present, or their potential for mobility or bioavailability after dispersal

**Table 3** Baseline data on selected characteristics of soil

| Soil types | Selected characteristics of the soil | | | | | |
|---|---|---|---|---|---|---|
| | pH ($H_2O$) | OC (%) | OM | Sand (%) | Silt (%) | Clay (%) |
| Luvisol | 7.26[a] | 3.99[a] | 6.86 | 21 | 60 | 19 |
| Arenosol | 7.40[c] | 3.60[c] | 6.19 | 88 | 5 | 7 |
| Vertisol | 7.19[b] | 7.40[b] | 12.73 | 10 | 70 | 20 |

Mean separation was via Duncan's Multiple Range Test. Means in the same column with different letters are significantly different at the 5% level

**Table 4** Pseudo total heavy metal content of the soils before treatment (control) and sludge

| Soil types | Heavy metal content in mg $kg^{-1}$ | | |
|---|---|---|---|
| | Zn | Cu | Co |
| Luvisol | 0.37[a] | 6.53[a] | 1.52[a] |
| Arenosol | 0.02[b] | 0.98[b] | 0.01[b] |
| Vertisol | 5.02[c] | 11.12[c] | 2.03[c] |
| Sludge | 40 | 26 | 17 |

Means separation was via Duncan's Multiple Range Test. Means in the same column with different letters are significantly different at the 5% level

**Table 5** Comparison of heavy metal content in previous studies and FAO (1992)

| Previous studies | Heavy metal content in mg/kg | | |
|---|---|---|---|
| | Zn | Cu | Co |
| This study | 40 | 26 | 17 |
| Emongor and Ramolemana (2004) | 0.04–0.08 | 0.4–1.1 | – |
| FAO (1992)[a] | 500 | 225 | – |

[a] Permitted concentrations for soil of pH 6.0–7.0 (Directive 86/278/EEC)

in the environment (Lombardi and Garcia 1999). In contrast, Emongor and Ramolemana (2004) reported relatively lower concentrations: 2.0–4.7 mg/kg Cu, 0.4–1.1 mg/kg Ni, 10.2–23.1 mg/kg Zn and 0.04–0.08 mg/kg Cd in sludge. When viewed against the recommended heavy metal concentrations (FAO 1992; see Table 5), the results observed in this experiment study as well as results from earlier studies show that the heavy metal content in Botswana sludge is generally low. Comparison between the results obtained by Emongor and Ramolemana (2004) and the results show a significant increase (Table 4) in heavy metal content in sludge, indicative of contamination attributable to industrialization.

## Effects of soil type on metal retention

Soil type and composition plays an important role for heavy metal retention. In general, coarse-grained soils exhibit lower tendency for heavy metal adsorption than fine-grained soils (Bradl 2004). The adsorption behavior of heavy metals varies among soil types and is a function of one or more soil properties such as clay and organic matter content. Perez-Novo et al. (2008) demonstrated that copper possess higher affinity for organic matter than zinc. Gomes et al. (2001) also found copper to be preferentially adsorbed in a competitive manner over zinc in Utisols, Oxisols and Alfisols. In this study, the same trend was observed (Table 4) in Arenosols and Vertisols. Of the three soil types studied (Table 4), Luvisols and Vertisols retained more copper than zinc and cobalt. Arenosols had the least concentration of all the heavy metals. This can be attributed to the textural differences of the three soil types (Table 3). Since Vertisols are generally fine textured, resulting in a soil body with a high specific surface area and high absorptive capacity at exchange sites and this have led to retention of heavy metals despite efforts to remove the metals by chelating agents (Table 5). Soils rich in clays are known for their ability to effectively remove heavy metals by specific adsorption and cation exchange (Bradl 2004). In contrast, Arenosols had the least load of heavy metals prior to and after treatment due to their textural composition, which is porous and has low water and nutrient retention capacities. It was unlikely for Arenosols to retain a significant load of heavy metals because it lacks colloids which are found on clays.

## Efficiency of chelating agents

Heavy metals are usually complexed with natural ligands such as humic or fulvic acids or anthropogenic complexants such as EDTA or NTA (Bradl 2004). Chelating agents are compounds that form metal complexes, however the efficiency of a chelating compound is influenced by a number of factors such as selectivity and specificity, stability of complexes, and the type of ligand. Complexation will alter metal reactivity affecting properties such as catalytic activity, toxicity and mobility. Heavy metal concentrations after treatment with chelating agents are presented in Table 6. Metals remaining after treatment with chelating agents can only be attributed to metals held tightly by the OH groups and clay colloids. Liu et al. (2007) and Perez-Novo et al. (2008) have found that the important interfaces involved in heavy metal adsorption in soils are predominantly inorganic colloids such as clays, metal oxides and hydroxides, metal carbonates and phosphates. However, no information is advanced on the reversibility of bonds formed in those instances. Efficiency of chelating agents assessed according to amount of heavy metal removed was in the following order: EDTA < NTA = AA (Table 6). This trend was maintained across soil types implying that EDTA out competes colloidal attraction to metals and formed strong complexes with all metals under consideration. In contrast, NTA and AA did not remove as much metals and their efficiency did not vary except when metal type is considered. More Cu was removed than Zn and Co in all the three soil types.

To evaluate the effectiveness of chelating agents in removing heavy metals, an index of % metal removed was

**Table 6** Heavy metals concentrations (mg/kg) after treatment with chelating agents

| Soil type | Chelating agents | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | Nitrilotriacetic acid | | | Acetic acid | | | EDTA | | |
| | Zn | Cu | Co | Zn | Cu | Co | Zn | Cu | Co |
| 1 | 19.67 | 8.10 | 10.11 | 18.77 | 10.09 | 9.09 | 9.09 | 4.28 | 5.97 |
| 2 | 27.27 | 13.33 | 12.52 | 33.83 | 15.68 | 11.13 | 16.49 | 9.28 | 6.09 |
| 3 | 34.33 | 8.59 | 5.57 | 28.31 | 7.98 | 4.77 | 25.70 | 3.66 | 2.95 |

Soil types 1, 2, 3 denote Luvisol, Arenosol, Vertisol in that order

**Table 7** Efficiency of each chelating agent expressed as a percentage of metal removed

| Soil type | Chelating agents | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | NTA | | | AA | | | EDTA | | |
| | Zn | Cu | Co | Zn | Cu | Co | Zn | Cu | Co |
| 1 | 50.8 | 68.8 | 40.5 | 53.1 | 61.2 | 46.5 | 77.3 | 83.5 | 64.9 |
| 2 | 31.8 | 12.7 | 26.4 | 15.4 | 39.7 | 35.5 | 58.8 | 64.3 | 61.2 |
| 3 | 14.2 | 66.96 | 97.2 | 29.2 | 69.3 | 71.9 | 35.8 | 85.9 | 82.6 |

Soil types 1, 2, 3 denote Luvisol, Arenosol, Vertisol in that order

used as defined in Eq. 1 below. The efficiency of the three chelating agents to remove heavy metals in a homogeneous mixture of soil–sludge varied significantly (Table 7)

$$\text{Percent metal removed}\,(\%) = \left[\frac{(S_{\text{initial}}) - (S_{\text{final}})}{(S_{\text{initial}})}\right] \times 100, \quad (1)$$

where ($S_{\text{initial}}$) and ($S_{\text{final}}$) is the metal load (mg kg$^{-1}$) in soil:sludge mixture before and after treatment, respectively, to give results summarized in Table 7.

Soils treated EDTA had the lowest concentration of the metals in the three soil type mixtures under consideration whereas soils treated with nitriloacetic acid and acetic acid had almost a similar load of heavy metals (Tables 6, 7). EDTA just like NTA is a poly dentate ligand and has multiple binding sites that hold the metal ion strongly into a complex. Whereas AA is mono dentate low molecular weight ligand. Though, EDTA removed a lot more metals than other chelates, it is worth noting that a lot more copper was removed than other metals under observation (Table 7). This might be attributed to the affinity and competitive behavior of copper ions to binding sites either of chelating agents or surface colloids. Bradl (2004) reported the preference series for divalent ions for organic matter as follows Cu > Pb > Fe > Ni = Co = Zn > Mn = Ca. The preference series mentioned above implies that copper exists more in a complexed form than ionic form which is consistent with results in Table 4. Other ligands (NTA and AA) may have formed complexes with the metal ions but the strength of the complexes might have been weak or readily soluble such that they only retained approximately half the total concentration of metals (Table 6). Availability of a metal ion depends on whether is tightly or weakly bound to a ligand–metal complex, however, AA which is a monodentate retained as much metal as NTA which is a multi dentate ligand.

## Conclusion

Vertisols, due to its textural characteristics, imbibed much of the heavy metals compared to Luvisols and Arenosol. Furthermore, metals were removed with ease in Arenosols and Luvisols than in Vertisols. EDTA was the most effective as compared to other two chelating agents which did not differ in their prowess to scavenge for metals. Due to its competitive behavior for either colloidal surfaces or binding sites of chelates copper was the most removed metal than Zn and Co.

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