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DEVELOPMENT OF A LOW COST REMEDICATION METHOD FOR HEAVY METAL POLLUTED SOILS

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SUMMARY

High concentrations of heavy metals in the soils raise potential long-term environmental and health concerns because of their persistence and tendency to accumulate in the environment and along the food chain. Heavy metals are not biodegradable and once they are in soil they persist unless removed, hence requiring decontamination.

This study examined the feasibility of heavy metal removal from the soil using plants growing naturally in the surroundings of selected polluted sites in Tanzania and using buried nodules of the sorbent materials zeolite and autoclaved aerated concrete (AAC) in a laboratory experiment.

Locally growing plants proved to have potential for heavy metal removal from polluted soils. For example, *Sporobolus* sp. is a hyperaccumulator of Cu. Four other species, *Launea cornuta* (Oliv & Hiern) O. Jeffrey, *Tagetes minuta* (L.), *Sporobolus* sp. and *Blotiella glabra* (Bory) Tryon showed high potential for phytoextraction of Cu, while *Dioscorea* spp. (yam) and *Stylochaeton natalensis* Schott showed high potential for Cu phytostabilisation. No hyperaccumulators of Pb and Zn were identified in the area, but *Tephrosia candida* and *Tagetes minuta* (L.) were identified as potential plants for phytoextraction of Pb, while *Conyza bonariensis* (L.) Cronquist, *Launea cornuta* (Oliv & Hiern) O. Jeffrey, *Tagetes minuta* (L.), *Blotiella glabra* (Bory) Tryon, *Pteridium aquilinum* (L.) Kuhn and *Polygonum setogulum* A. Rich were identified as potential plants for phytoextraction of Zn. *Sphaeranthus africanus* (L.) and *Pteridium aquilinum* (L.) Kuhn showed potential for phytostabilisation of Pb and *Stylochaeton natalensis* Schott for phytostabilisation of Zn.

The nodules were produced in rectangular shapes with dimensions 2 cm width, 4 cm length and 2.5 cm height. The AAC nodules were cut to that size from larger pre-fabricated blocks. Zeolite grains with a size of 1-2 mm were mixed with cement as binder and formed to the final rectangular shape (density 1.1 g cm⁻³). These nodules were buried at medium depth in rectangular PVC boxes (40 cm length, 21.5 cm width and 9 cm height) filled with soil. The soil used was artificially polluted by adding salts of Pb and Zn. Lead nitrate Pb(NO₃)₂ and zinc chloride (ZnCl₂) were used to spike the soil with Pb and Zn respectively. The sorption experiment demonstrated that both materials tested had potential in decontaminating polluted soil. AAC had a higher concentration of both Zn and Pb bound to the matrix than zeolite when the soil was spiked with one metal. The sorption capacity of zeolite and AAC in a mixed metal scenario (Pb and Zn) showed less difference from that for Pb or Zn separately. The analysis of metal speciation in soil showed that the majority of the metals were firmly bound to components of the soil and was not readily released on irrigation with water in the experimental set-up.

Owing to variations in plant sensitivity and soil conditions, the phytoremediation effect can highly be augmented by combining a hyperaccumulator species with sorbent material. Identification of the best combinations and designs remains the subject of future research.

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LIST OF PAPERS

The thesis is based on the following two papers, which are referred to in the text by their Roman numerals.

- I. Mkumbo, S., Renman, G. & Mwegoha, W. 2012. Assessment of the phytoremediation potential for Pb, Zn and Cu of indigenous plants growing in a gold mining area in Tanzania. *International Journal of Environmental Sciences* 2 (4), 2425-2434.
- II. Mkumbo, S., Renman, G. & Mwegoha, W. 2012. Active removal of lead and zinc from polluted soil by in situ sorption to mineral nodules. Manuscript.

Paper I is reproduced with permission from the journal.

ABSTRACT

High concentrations of heavy metals in the soils have potential long-term environmental and health concerns because of their persistence and accumulation tendency in the environment and along the food chain. This study was aimed at studying the feasibility of heavy metals removal from the soil using plants naturally growing in the surroundings of selected polluted sites in Tanzania and soil application of the sorbent materials zeolite and autoclaved aerated concrete (AAC). The results showed that *Sporobolus* sp. is a hyperaccumulator of Cu. Four other species, *Launea cornuta* (Oliv & Hiern) O. Jeffrey, *Tagetes minuta* (L.), *Sporobolus* sp. and *Blotiella glabra* (Bory) Tryon showed high potential for phytoextraction of Cu. No hyperaccumulators of Pb and Zn were identified in the area, but *Tephrosia candida* and *Tagetes minuta* (L.) were identified as potential plants for phytoextraction of Pb, while *Conyza bonariensis* (L.) Cronquist, *Launea cornuta* (Oliv & Hiern) O. Jeffrey, *Tagetes minuta* (L.), *Blotiella glabra* (Bory) Tryon, *Pteridium aquilinum* (L.) Kuhn and *Polygonum setogulum* A. Rich were identified as potential plants for phytoextraction of Zn. The result from sorbent experiments showed that both materials had a potential for remediating metal polluted soils. The AAC had a higher removal capacity for both Zn and Pb than zeolite. The removal capacity of zeolite and AAC in a mixed metal experiment (Pb and Zn) showed a little difference in the sorption capacity of AAC and Zeolite for Pb and Zn respectively. Speciation of the metal in soil shows that the major part of the metal was associated with firmly attached component of the metal in the soil. Compared with the total metal concentration analysed, the available component accounted for 13-39% for Zn and 31-39% for Pb. It can be suggested to co-remediate polluted soils using reactive sorbent nodules and hyper-accumulating plant species. Identification of the best combinations and designs remains the subject of future research.

Key words: autoclaved aerated concrete; decontamination; heavy metal; hyperaccumulator plants; sorbent; zeolite.

1. INTRODUCTION

Heavy metals are naturally occurring elements, which under normal conditions are present at low concentrations in the soil. Weathering of rocks, soils and volcanic eruptions are among the natural sources of heavy metals in the environment. However, nowadays the anthropogenic contributions of heavy metal pollution to water, air and soil far surpass the natural sources. The main anthropogenic sources of heavy metal pollution include: mining, metal smelting, metallurgical industries, and other metal-using industries, waste disposal, corrosion of metals in use, agriculture and forestry, fossil fuel combustion, and sports and leisure activities

(Hutchinson & Meema, 1987; Cunningham *et al.*, 1995; Mdegela *et al.*, 2009).

Like all other developing nations, Tanzania has recently witnessed an increase in mining and industrial activities. These rapid changes have been exacerbated by the radical change that occurred in the mid-1980s from a socialist to a capitalist economy, which is market-orientated and private sector-led, (Ngowi, 2009). The mining sector, being the overall leading sector in the rate of expansion, has emerged in many facets. However these ventures have not made any significant contribution to the economy of the local population or of the nation at large, while at the same time they are giving rise to great

concern in terms of environmental pollution of soil and surface water (Kitula, 2006).

Among the mining sectors, gold mining is currently the leading type, since gold is a highly exploited mineral in Tanzania. Gold mining and subsequent processing of gold ores are known for their potential contamination of the environment and risks to human health (van Straaten, 2010). Although the metal can be naturally weathered from its mother rock, grinding these rocks and exposing them to agents of weathering as dumped waste rocks accelerate their pollution capacity in an area (Bitala, 2008).

Heavy metals are not biodegradable (Sobukola *et al.*, 2010), and therefore once they are present in soil or water they tend to remain there for a long time unless they are actively removed. Through pollution of the environment, the heavy metals can find their way into the human body through food, drinking water and air. As trace elements, some heavy metals (*e.g.* copper and zinc) are essential to maintain the metabolism of the human body (Fraga, 2005; Duruibe *et al.*, 2007). However, at higher concentrations they can lead to poisoning (Cambra *et al.*, 1999). Heavy metal poisoning can arise as a result of, for instance, drinking-water contamination (surface or ground water), high ambient air concentrations near emission sources, or intake via the food chain (Tripath *et al.*, 1999; Zukowska & Biziuk, 2008).

1.1. Effects of metals in the environment

Heavy metals are among the biosphere pollutants of major global concern, due to their ability to bioconcentrate and bioaccumulate up the food chain, their environmental persistence and their chronic toxicity (Mdegela *et al.*, 2009). As such, heavy metals remain one of the best-known environmentally hazardous substances. These substances can accumulate and biomagnify unnoticed in an ecosystem until they reach

toxic levels (Barak & Mason, 1990; Barlas, 1999).

Thus heavy metal polluted soils present a potential public health risk associated with dietary intake of heavy metals. Elevated levels of heavy metals in the soil enhance plant uptake, causing accumulation in plant tissues and eventual phytotoxicity and changes in the plant community (Ernst, 1996; Gimmler *et al.*, 2002). Short-term and long-term effects of pollution differ depending on the characteristics of the metal and soil in question (Kádár, 1995; Németh & Kádár, 2005).

Intake of heavy metals via the soil–crop system is considered to be the dominant pathway of human exposure. Among the heavy metals, copper (Cu), cadmium (Cd), chromium (Cr), lead (Pb), mercury (Hg), zinc (Zn) and arsenic (As) are commonly considered toxic to both plants and humans (Trichopoulos *et al.*, 1997).

Consequently, decontamination of heavy metal polluted soils requires effective approaches in order to safeguard public health. This study therefore examined the feasibility of heavy metal removal from soil using plants naturally growing in the surroundings of selected polluted sites in Tanzania and soil application of sorbent materials such as zeolite and autoclaved aerated concrete (AAC). The feasibility of plant use in this application was examined through identifying accumulator plants growing at contaminated sites. The feasibility of using sorbent materials was examined in a laboratory experiment in which the materials were buried in polluted soils that were irrigated to allow movements of metals in the soil. The chemistry of soils collected from plant sampling sites and those used in experiments was also investigated, with the focus on parameters affecting the movement of heavy metals in soils, such as pH, electroconductivity, particle size distribution and organic matter content of the respective soils.

1.2. Remediation practices

The current practice of remediation of heavy metal contamination in many parts of the world relies heavily on dig-and-dump or encapsulation, neither of which addresses the issue of decontamination of the soil (Pulford & Watson, 2003; Mwegoha, 2008). Immobilisation or extraction by physicochemical techniques can be expensive and is applicable only for small areas where rapid, complete decontamination is required. This practice may not be feasible for Tanzania for economic reasons (Mwegoha, 2008). Recent developments in the field of environmental restoration have led to invention of the phytoremediation technique (Marques *et al.*, 2009).

Phytoremediation is a low-cost biotechnology, with almost no maintenance costs and eventually even an investment return in the form of wood, biomass or fibres (Trapp *et al.*, 2003; Ana *et al.*, 2009). The challenge with this method is that no study has been conducted to identify native hyperaccumulator plants that can be used in phytoremediation. According to Mwegoha (2008), introduction of exotic species has resulted in problems in many parts of the world. Such problems include:

- i) Introduction of invasive species, which ultimately affects the local biodiversity.
- ii) Introduced plants may fail to cope with the climatic and soil conditions at the site.

The success of this method requires plants which can tolerate high concentration of metal in the soil and be able to accumulate the metal. The major mechanism in tolerant plant species appears to be compartmentalization of metal ions (i.e., sequestration in the vacuolar compartment or cell walls), which excludes them from cellular sites where processes such as cell division and respiration occur, thus providing an effective protective mechanism (Chaney *et al.*, 1997; Ana *et al.*, 2009).

Reduction of the influx across plasma membrane as well as binding to cell wall and

root exudates can also be possible avoidance strategies (Hall, 2002).

Metal in the soil exist in different fractions (speciation). Some of which are available for uptake while some fractions are not available for plants uptake. The metals considered available for plant uptake are those that exist as soluble components in the soil solution or are easily desorbed or solubilized by root exudates or other components of the soil solution, these portions representing often only a small part of the total metal content of the soil (Blaylock & Huang, 2000).

The limited bioavailability of various metallic ions, due to their low solubility in water and strong binding to soil particles, restricts their uptake/accumulation by plants (Ana *et al.*, 2009).

Besides the bio-availability of metal in the soil plant species vary significantly in the ability of accumulating metals from contaminated soils. According to Ana *et al.*, (2009) plants with potential for phytoextraction should have the following characteristics:

- i) be tolerant to high levels of the metal;
- ii) have a profuse root system;
- iii) have a rapid growth rate;
- iv) have the potential to produce a high biomass in the field; and
- v) accumulate high levels of the metal in the harvestable parts, as generally the harvestable portion of most plants is limited to the aboveground parts (although the roots of some crops may also be harvestable).

Adsorption of heavy metal in water by Zeolite and AAC has been reported by several researchers (Kietlińska & Renman, 2005; Paola *et al.*, 2008; Renman & Renman, 2012).

The capacity of these materials to adsorb metal in the soil is explored through a laboratory scale experiment.

The general aim of this thesis was to develop a low cost method for remediation of heavy metals polluted soils in Tanzania. The work

covered the use of plants and sorbent materials for removal of heavy metals from polluted soils. The specific objectives of the studies were:

- i) To determine the chemistry of soil in selected mining areas in terms of concentrations of heavy metals and other field environmental conditions.
- ii) To identify and select indigenous plants for use in phytoremediation of heavy metal polluted soils in Tanzania.
- iii) To identify and document terrestrial plants naturally growing in study area.
- iv) To conduct a laboratory experiment on the capacity of sorbent materials to remove metals from the soil *in situ*.

The study is particularly relevant at this time because of the dramatic increase in industrial scale mining activities in various parts of Tanzania, as mentioned above. Several studies have shown that soils and sediments in various areas of Tanzania have been polluted by heavy metals (Kishe and Machiwa 2003; Kisamo 2003; Machiwa, 1992; Makundi 2001), but no previous study has examined appropriate remediation methods for polluted soils in Tanzania.

The study results are intended to provide the platform on which remediation efforts dealing

with the whole heavy metal soil pollution problems in the country can be based.

2. MATERIALS AND METHODS

2.1. Materials

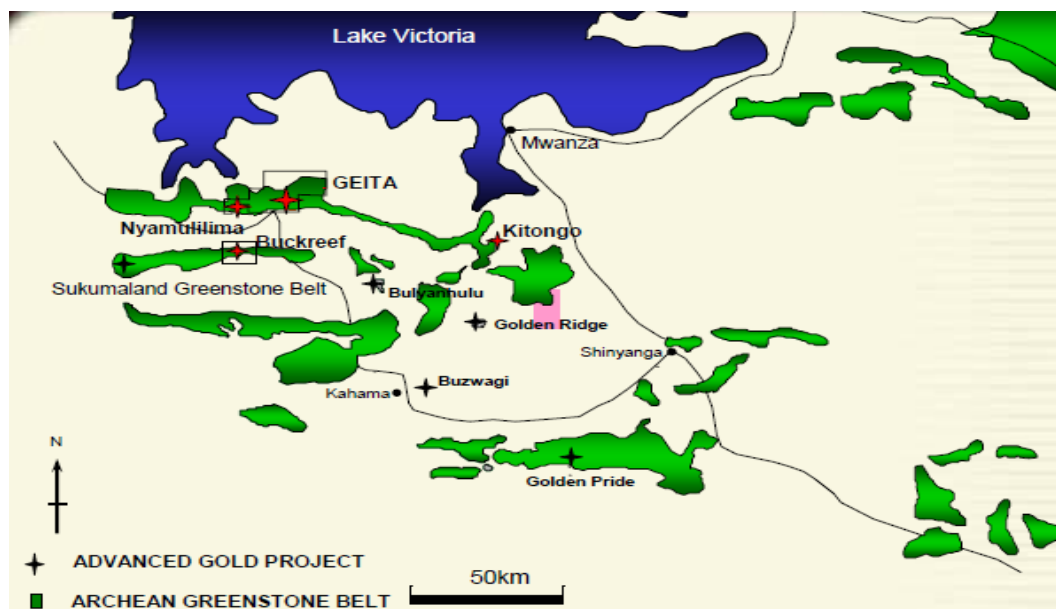
Description of the study area

The study on identification of locally growing plants with hyperaccumulating characteristics was carried out in Geita district, Mwanza region, Tanzania, between July 2010 and July 2011. Geita District is located between 2°42'0"-3°15'0"S, 32°10'0"-32°45'0"E and in administrative terms it is divided into seven divisions, 27 wards and 163 villages.

This area is endowed with gold-bearing rocks (Figure 1), as the result it has attracted a lot of domestic and foreign companies to invest in mining gold in the region.

These mining activities are carried out in a sensitive environmental setting. The area is pre-dominantly occupied by peasants and pastoralists. In addition, the mining centres are located within 20 km of Lake Victoria and form part of the Lake Victoria catchment area. According to Kisamo (2003), wastes from mining activities are one of the main anthropogenic pathways of heavy metal pollutants into the Lake Victoria environment (Figure 1). Thus the proximity of the mining activities to the Lake can potentially result in

Figure 1.
Map of Geita District in Tanzania showing the distribution of gold ore.



pollution of the lake and ultimately affect the lake ecosystem.

Geita has the highest population of all districts in the Mwanza region. The majority of people in this district are concentrated in the mining centres. Most of these centres are occupied by artisanal miners, relying on poor sanitation facilities, which could lead to outbreaks of communicable diseases. Besides clearance of forest for mineral exploitation, the high population in this area depends on the forest as the sole source of energy.

For the past few years, Tanzanian soils and sediments have been receiving huge amounts of heavy metals from industrial and mining activities (Kitula, 2006). These high concentrations of heavy metals in the soils have potential long-term environmental and health consequences for the region because of their persistence and accumulation tendency in the environment and along the food chain (Wong, 2004).

According to Kitula (2006), environmental pollution is a major problem in the mining areas of Geita District. Continuous disposal of mine wastes contributes to soil, water and air pollution, which is detrimental to human health, livestock and wildlife biodiversity, and can have serious effects on the welfare of the mining communities, especially women and children.

Mining activities generally increase the rate of metal release into the environment. Crushing, grinding, washing and all the other processes used to extract and concentrate metals make elements environmentally labile through normal biogeochemical pathways, to sinks such as sediments, soils or biomass (Davies, 1980; 1983). Mining activities produce large quantities of wastes, composed of barren rocks, ores that cannot be economically extracted and smelting rejects, which must be disposed of in the environment, usually in the surroundings of the mining area. The waste rock dump contains high metal concentrations and continues to be a source of metal

pollution long after extraction activities have ceased (Davies, 1983).

Metal leaching is greatly accelerated where acid mine drainage occurs (Bitala, 2008). Heavy metals, even in trace amounts, can be toxic to humans and wildlife.

Once the soil is polluted, it requires treatment so as to safeguard the adjacent environments. Polluted soils cannot be abandoned since they are not in isolation. Thus they can turn out to be the source of pollution to adjacent land areas or groundwater. Besides, they pose a threat to human health through the food chain.

Most polluted soils in Geita District are still in use for crop production and grazing animals, forming an excellent pathway to humans (Wong, 2004). Polluted soils also form part of the catchment areas for various rivers and lakes. Through erosion, the heavy metals reach surface water bodies. Furthermore, the polluted soils pose a high risk of contaminating the groundwater as the result of soluble components being carried down through the soil.

Decontamination of polluted soils, aquifers and surface water bodies is an expensive undertaking that is not feasible for developing countries such as Tanzania (Mato, 2002). In order to prevent future problems, there is thus a need to remediate polluted soils before they have a far-reaching effect.

Soil sampling

Hand augers were used for soil sampling. The soil samples were taken at a depth of 0-20 cm. The auger was cleaned with distilled water between sampling points and between sampling fields. The samples were stored in coolboxes and transported to the laboratory in Dar es Salaam for analysis.

Positioning of sampling locations

Geographical Positioning System (GPS) was used to locate the sampling points. Accurate positioning of sampling points was important in case additional samples were needed at previously sampled sites.

A range of laboratory methods and equipment were used in the preparation, digestion and subsequent analysis of the samples.

Weighing of materials

Sample weight was measured using a Boeco electronic weighing scale model number BBI 31.

Preparation of distilled-deionised water

Distilled deionised water was prepared using a Bibby Merit Waster still W4000 (prd0984) distillation column. The distilled water was then passed through a Elgastat Micromeg deionisation column (Elga[®]).

2.2. Methods

2.2.1. Soil and plant sampling

Six sampling sites were established in Geita district. These sites were Nyakabale, where the area is affected by both small-scale mining processes and large-scale mineral processing (GGM waste rock dump), Katoma, Nyaruyeye, Nyarugusu, Nyankumbu and Mugusu, which are all artisanal gold mining areas as shown in Figure 1.

Soil and plant samples were collected in all six areas. The choice of the sampling site was based on the human activities taking place in the area. All the selected areas had some relation with mining or mineral processing activities. The sampling location was based on the areas where growth of plants was observed. The reason for doing this is because many parts of these areas have no vegetation cover this being because of the forest clearance to pave for mining activities and the effects of metal pollution. Thus selection of plants was done on the bases of the areas where there was vegetation.

2.2.2. Plant sample collection and preparation

Different plant samples and their corresponding surface soil samples (0-20 cm) were collected from the six sampling locations, around mining and mineral processing areas and adjacent farmland, between July 2010 and July 2011.

The plants sampled were annual species, the reason being that perennial crops would require a very long period of study in order to establish factors such as heavy metal degradation kinetics and their potential for phytoremediation applications. With annual crops the study was able to estimate the time required for remediation by examining the trend of extraction that could be accomplished by each growth cycle. These allowed the number of crop growth cycles that may be needed to decrease contaminant concentrations to permissible levels to be calculated.

Samples were collected for the purposes of:

1. Identifying the types of plants naturally growing in the study area
2. Analysing the heavy metal content of the sampled plants

A total of 117 plant samples were collected. For each type of plant selected, a total of 3 samples were collected. Plant (shoots and roots) were uprooted at the growing site gently to ensure that the whole plants together with their roots were collected. At the sampling site, the plant samples were cleaned to remove the soil residues attached to the roots. The samples were then wrapped in polyethylene bags, labelled and stored in coolboxes during transport to the Environmental Engineering laboratory at Ardhi University in Dar es Salaam for pre-treatment and analysis in accordance with (Princewill-Ogbonna & Ogbonna, 2011).

At the laboratory, the plant samples were separated into two parts, roots and shoot sections using a stainless steel knife. The plant samples were then thoroughly washed with flowing tap water to remove dust and other particles, and finally rinsed with distilled deionised water to remove all ions.

The cleaned plant samples were air-dried for a period of two weeks and thereafter dried in a hot air oven at 80 °C to constant weight for two days. After complete drying, the plant samples were ground to powder using mortar

and pestle. The powder was used in the extraction of heavy metals such as Cu, Zn and Pb.

At every location where a plant sample was collected, a soil sample was also collected. This was intended to provide an insight into the relationship between the concentration of a particular heavy metal in the plant and the concentration of the same metal in the soil.

After transportation of the soil samples to the laboratory, large stones and plant debris were removed. The samples were then air-dried at room temperature for 6 days to relatively low moisture content. Thereafter, they were dried to a constant weight at 105 °C using a Toshniwal hot air oven for one day. The dried samples were sieved through a 2-mm polyethylene sieve and packed in plastic bags ready for further analysis.

2.2.3. Characterisation of soil

All soil samples were analysed for their chemical and physical characteristics, such as content of sand, silt and clay, organic matter content, pH and electrical conductivity.

Determination of soil pH and electroconductivity

Soil samples were dissolved in water at 1:2.5 and 1:5 (w/v), soil:distilled deionised water suspension for pH and electroconductivity (EC) measurements, respectively. This was done by weighing 10 g air-dry soil (<2 mm) into a bottle. Then 25 or 50 mL of deionised water were added and the bottles were shaken manually at 20 minute intervals for 1 hour to allow soluble salts to dissolve and ionic exchange to reach equilibrium prior to measuring pH.

Soil EC was measured using a calibrated Hanna EC meter HI98353 DiST-3 and pH using a Hach H130 Rugged Pocket pH meter.

Determination of soil particle distribution and soil texture

Soil particle distribution was analysed using both manual and mechanical sieving. The manual analysis was carried out at Ardhi University, while the mechanical sieving was performed at KTH. In both cases the soil

aggregates were broken manually before sieving. The results were computed to find the percentage composition of textural fractions in the soil.

Determination of soil organic matter content

Organic matter content was measured by the loss of ignition (LOI) method, as described by Schulte & Hopkins (1996).

The LOI was calculated using the equation:

$$LOI_{550} = \left(\frac{DW_{105} - DW_{550}}{DW_{105}} \right) \cdot 100$$

Where LOI_{550} represents LOI at 550 °C,

DW_{105} represents the dry weight of the sample before combustion and DW_{550} the dry weight of the sample after heating to 550 °C (both in g).

The samples from the furnace were cooled in desiccator before measurements were done.

All measurements, except particle size distribution, were carried out in triplicate and their mean was taken to represent the given measurement.

2.2.4. Analysis of total heavy metal concentration in the soil and sorbent materials

Total heavy metal concentration was determined in soil digested using *aqua regia* (HNO₃: HCl 1:3). In this process, 1 g of the soil sample was added to a conical flask with 5 mL *aqua regia* and the mixture was digested on a hot plate at 95 °C for a period of about 1 hour. The resulting mixture was diluted using 30 mL distilled deionised water and filtered. After filtration, the resulting solution was analysed using an Atomic Absorption Spectrophotometer (AAS) PerkinElmer[®] AAnalyst 100.

2.2.5. Speciation of heavy metals in the soil

Speciation of heavy metals was determined for all the soil samples collected from the field and for the soils used in the sorption experiments. The ultimate goal of conducting speciation analysis was to accurately determine the distribution of the metal into various fractions in the soil.

Understanding the speciation helps in the design, selection and optimisation strategies that need to be applied to clean up polluted soil (D'Amore *et al.*, 2005).

The speciation of heavy metals in soils on which the sampled plants were growing was determined using the sequential extraction method proposed by Silveira *et al.* (2006). This method is adapted for tropical soils. Speciation of heavy metals in experimental soils was determined using a modified Community Bureau of Reference (BCR) three-step sequential extraction procedure for the fractionation of Pb and Zn in the soil, as described by Žemberyova *et al.* (2006).

2.2.6. Analysis of heavy metal concentrations in plants

In the laboratory, plant samples were cleaned using tap water to remove the residual soil particles from the leaves, stems and roots. This was followed by rinsing the sample with distilled deionised water. A stainless steel knife was used to cut the plant samples into different parts (shoot and roots), which were oven-dried separately at 80 °C for 48 hours and then ground using pestle and mortar.

For analysis of heavy metals in plant fractions, 1.0 g dry, powdered plant sample was placed in a test tube with 5 mL of a 5:1 mixture of *aqua regia* (HNO₃ – HCl) and perchloric acid (HClO₄). The mixture was then digested at 95 °C for approximately 1 hour until complete digestion (Lin *et al.*, 2003; Liu *et al.*, 2008).

After digestion, the samples were left to cool and then transferred to 100-mL volumetric flasks. The test tubes used for digestion were rinsed with distilled water and the rinsing water was added to the volumetric flasks to bring the volume to 100 mL. Metal analysis was carried out using an Atomic Absorption Spectrometer (AAS).

Plant ability to take up heavy metals from the soil was evaluated by calculating the ratio of element concentration in the plant to that in the soil. This is called Biological Absorption Coefficient (BAC) (Behrouz *et al.*, 2008).

Phytoextraction means that plants remove metals from the soil and concentrate them in the harvestable parts of plants (Pulford & Watson, 2003).

Phytostabilization is the use of plants to reduce the mobility and bioavailability of pollutants in the environment either by immobilisation or by prevention of migration (Pulford & Watson, 2003).

Plants with a high BAC value (BAC>1) are suitable for phytoextraction, while plants with a high bioconcentration Factor (BCF) (*i.e.* ratio of metal concentration in plant roots to metal concentration in soil) (BCF>1) and low translocation factor (TF) (*i.e.* ratio of metal concentration in plant shoots to metal concentration in roots) (TF<1) have the potential for phytostabilisation (Cheraghi *et al.*, 2011).

2.2.7. Selection of materials and experimental set up

The criteria for selection of materials were on the bases of the capacity of the materials to sorb the metal, their simplicity of removing them from the soil and their availability. Zeolite can be obtained at Olduvai Gorge in Tanzania. The AAC is not a naturally occurring materials but it can easily be manufactured since all the ingredients can easily be obtained within the country.

The sorption experiment was made in plastic boxes with dimensions 40 cm length, 21.5 cm width and 9 cm height. Each box was filled with 3.6 kg of soil. The soil used in the experiment was polluted with Zn and Pb. The set up comprised of nine boxes which include control, reference soil (original soil) treated with AAC and Zeolite one each, Zn polluted soil treated with AAC and Zeolite one each, Pb polluted soil treated with AAC and Zeolite one each, soil polluted with both Zn and Pb and treated with AAC and Zeolite one each, thus making a total of 9 boxes. The Zeolite and AAC nodules were produced in rectangular shapes with dimensions 2 cm width, 3.5 cm length and 2 cm height. The

AAC nodules were cut to that size from larger pre-fabricated blocks. Zeolite grains with a size of 1-2 mm were mixed with cement as binder and formed to the final rectangular shape. These nodules were buried at medium depth in the boxes (Zn- and Pb-free) filled with soil. The arrangements of the sorbent nodules in a box were as shown in Figure 2.

2.2.8. Artificial pollution of soil for sorption experiment

The soil used in the sorption experiment was artificially polluted by adding salts of Pb and Zn, *i.e.* lead nitrate $\text{Pb}(\text{NO}_3)_2$ and zinc chloride (ZnCl_2), to pollute the soil with Pb and Zn, respectively (Paper II).

For this, 2.8 g $\text{Pb}(\text{NO}_3)_2$ were dissolved in 3 L distilled water and thereafter mixed uniformly with 14.4 kg soil. After uniform mixing, the suspensions were transferred to plastic boxes and left uncovered at ambient temperature for two weeks. Over the course of these two weeks, the soil samples lost their water content. The reason for leaving them to dry was to allow the test metal to stabilise and speciate before the experiment started. This artificially polluted soil was subdivided into



Figure 2. Arrangement of the sorbent material in the experimental box

four equal portions, each weighing 3.6 kg, and these were placed in the plastic boxes used for experiment.

Zinc pollution of the test soil was done by dissolving 9.1 g zinc chloride (ZnCl_2) in 3 L distilled water, followed by mixing with 14.4 kg of soil. The mixture was left to dry for a period of two weeks.

In addition, a batch of soil was subjected to mixed pollution by both Zn and Pb. This was done by adding a solution containing 9.1 g ZnCl_2 and 2.8 g $\text{Pb}(\text{NO}_3)_2$ dissolved in 3 L distilled water and then mixed into 14.4 kg soil. The resulting mixture was divided into three portions of 3.6 kg each, placed in plastic boxes and left to dry for a period of two weeks.

After two weeks, six blocks of AAC and zeolite nodules were buried in each box spiked with Pb alone, Zn alone or a mixture of Pb and Zn. For each type of metal spiking, one box was left without any adsorbent as control. The experimental set-up was irrigated once in every 3 days, by adding 250 mL distilled deionised water. The experiment was conducted at a constant room temperature of 18 °C and in darkness to prevent any plant growth in the experiment, which could have affected the experiment through metal uptake by plants. Whenever there was a sign of plants germinating in the soil, they were uprooted immediately.

3. RESULTS AND DISCUSSION

The following sections present and discuss the results of the soil chemistry analyses, identified potential hyper-accumulator plants naturally growing in mining areas in Tanzania and the adsorption capacity of the sorbent materials used in the experiments.

Table 1. Physical and chemical parameters of soil

Characteristic	Range	Mean (n=60)
EC ($\mu\text{S m}^{-1}$)	110-940	548.06 \pm 172.3
pH	3.5-7.3	6.25 \pm 1.04
OM (%)	2.9-26.5	8.8 \pm 3.9
Soil content %	Sand	26.35
	Silt	41.98
	Clay	31.35

3.1. Soil properties

Chemical analyses (Paper I) showed that the sampled soils from mining areas in Geita District, Tanzania, were slightly acidic, with an average pH of approximately 6.3 ± 1.1 . Most sampling locations had pH ranging between 5.2-7.3, values that are suitable for plant growth.

Soil pH is a key parameter controlling heavy metal transfer behaviour in soils. Decreasing pH in the soil increases the competition between H^+ and dissolved metals for ligands such as CO_3^{2-} , SO_4^{2-} , Cl^- , OH^- , S^{2-} and phosphates (Jieng-feng *et al.*, 2009). This increased competition decreases the metal adsorption capacity of soil particles, leading to increased mobility of heavy metals, which ultimately boosts the bioavailability of the metals in the soil.

The average EC of the soils in the study area was $548.06 \mu\text{S m}^{-1}$ and they had a high organic matter (OM) content of $8.8 \pm 3.9\%$ (Table 1).

The source of the elevated OM was probably plant litter deposited on the surface over the years, since OM is mainly composed of humic and fulvic substances (Jieng-feng *et al.*, 2009).

The complexation reaction between heavy metals and organic complexes is usually recognised as the most important reaction pathway, due to this reaction largely determining the speciation and bioavailability of metals and then influencing the mobility of trace metals in the environment.

3.2. Metal concentrations in the soil

The results showed that the soils had wide ranges of heavy metal concentrations (Paper I). These heavy metal concentrations were compared with reference concentrations in non-polluted soils (Table 2) (*cf.* Fifield & Haines, 2000).

It can be pointed out that the sampling sites were heavily polluted, with mean soil concentrations that were 15.95, 9.25 and 21.01 times higher than the reference concentration of Pb, Zn and Cu respectively, in non-polluted soils (Fifield & Haines, 2000).

Beside the evidence that the soils are polluted, speciation analyses of these metals in the soil revealed that a small fraction was available for plant uptake (Figure 3). According to Prokop *et al.* (2003), metal speciation in the soil solution phase is one of the key factors that regulates metal uptake by plants, as well as toxicity for soil.

Table 2. Comparison of heavy metal concentration in the soil to the reference concentration in non-polluted soils

Metal	Concentration in polluted soils (mg kg^{-1} dry weight)		Factor, times ref. conc.	Mean conc. in non-polluted soils* (mg kg^{-1} dry weight)	
	Range	Mean (n=60)		Range	Mean
Pb	29.64 - 3457	318.91	15.95	1.50-80.00	20.00
Zn	37.53 - 6544.2	555.38	9.26	17.00-125.00	60.00
Cu	30.7-3625	315.25	21.01	6.00-60.00	15.00

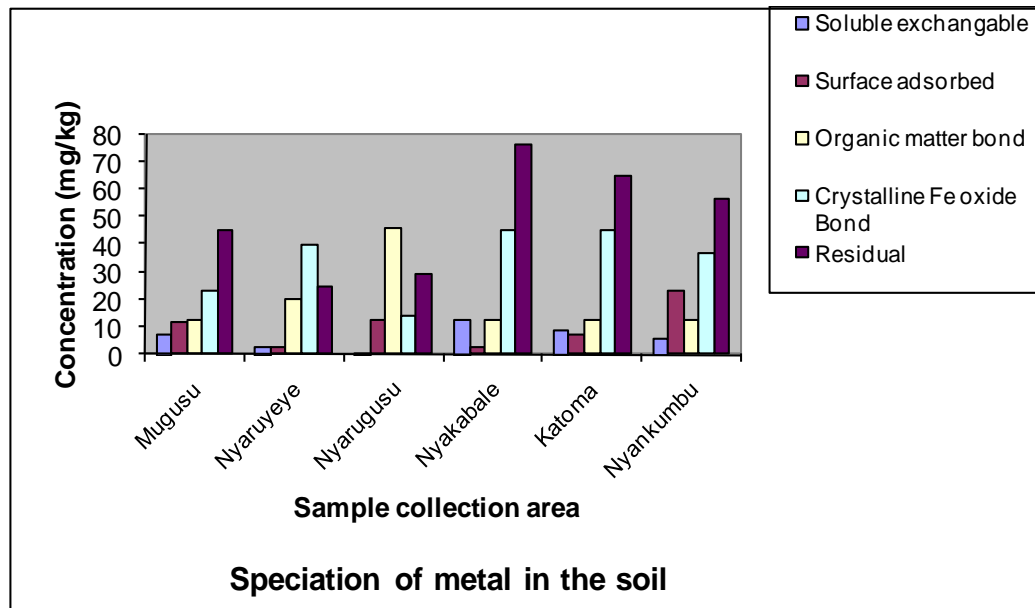


Figure 3.
Speciation of metal in soils collected from different sampling sites in Paper I.

3.3. Identification of plants with phytoremediation potential

Of the 39 species analysed in Paper I, *Sporobolus* sp. was identified as being an hyperaccumulator of Cu, while *Launea cornuta* (Oliv & Hiern) O. Jeffrey, *Tagetes minuta* (L.) and *Blitiella glabra* (Bory) Tryon demonstrated a high potential for phytoextraction of Cu, and *Dioscorea* spp. (yam) and *Stylochaeton natalensis* Schott showed high potential for phytostabilisation of Cu (Paper I).

No hyperaccumulators of Pb and Zn were identified in the area, but *Tephrosia candida* and *Tagetes minuta* (L.) showed potential for phytoextraction of Pb, while *Sphaeranthus africanus* (L.) and *Pteridium aquilinum* (L.) Kuhl showed potential for phytostabilisation of Pb.

Conyza bonariensis (L.) Cronquist, *Launea cornuta* (Oliv & Hiern) O. Jeffrey, *Tagetes minuta* (L.), *Blitiella glabra* (Bory) Tryon, *Pteridium aquilinum* (L.) Kuhl and *Polygonum setogulum* A. Rich were shown to be potential plants for phytoextraction of Zn and *Stylochaeton natalensis* Schott for phytostabilisation of Zn (Paper I).

3.4. Plant species naturally growing in the study area

A total of 117 plants naturally growing around mining activities were collected from the study area (Paper 1). These were identified as belonging to 39 different species, as shown in Table 3. Amongst these plant species few species showed potential for phytoremediation (refer section 3.3 above).

3.5. Results of sorption experiments

The effects of the sorbent materials on Pb and Zn concentrations in the artificially polluted soils are reported in Paper II. The chemical composition of the zeolite and ACC used as sorbent materials in Paper II are summarised in Tables 4 and 5.

The results showed that the major components of zeolite are SiO₂ (73.73%), Al₂O₃ (12.10%), Fe₂O₃ (3.81%), K₂O (3.22%), Na₂O (2.82%) and CaO (2.43%).

According to Bogdanov *et al.* (2009), zeolite can hold water up to 60% of its weight. This characteristic can be used protect the soil environment, since the water held by the zeolite can be evaporated, leaving all the

Table 3. Plant species identified in the study area

Sampling site	Site type	Species
Nyankumbu	Mineral processing area and paddy farm land	<i>Imperata Cylindrica</i> , <i>Ageratum compoides</i> L., <i>Tagetes Minuta</i> L., <i>Trichodesma zeylanicum</i> (L) R.Br., <i>Celosia trigyna</i> L., <i>Glinus oppositifolium</i> (L) A.DC., <i>Ageratum conyzoides</i> L., <i>Launea cornuta</i> (Oliv & Hiern) C. Jeffrey., <i>Sphaeranthus africanus</i> L., <i>Oryza sativa</i> L.
Nyakabale	Wetland	<i>Lantana camara</i> L., <i>Stachytarpheta jamaicensis</i> Vahl, <i>Blotiella glabra</i> (Bory) Tryon, <i>Sporobolus</i> sp, <i>Cyperus Papyrus</i> , <i>Stylochaeton natalensis</i> schott, <i>Hyparrhenia rufa</i> , <i>Conyza bonariensis</i> (L) Cronquist, <i>Launea cornuta</i> (Oliv & Hiern) C.Je, <i>Nymphaea caerulea</i> sarvigny
Katoma	Mineral processing area and Wetlands	<i>Blotiella glabra</i> (Bory)Tryon, <i>Stylochaeton natalensis</i> schott, <i>Helianthus annuus</i> , <i>Pteridium aquilinum</i> (L) Kulm,
Nyaruyeye	Paddy farm and animal grazing land	<i>Vernonia pyrhopappa</i> , <i>Polygonum Senegalense</i> Meisn, <i>Oryza sativa</i> L., <i>Ludwigia stolonifera</i> (Guill & Perr) Ra, <i>Homoea aquatica</i> Forssk, <i>Guizotia scabra</i> (Vis) Chiov, <i>Polygonum senegalense</i> Meisn, <i>Rgemone mexicana</i> L., <i>Evolvucus didinoides</i> L., <i>Sphaeranthus pelycephatus</i> Oliv, <i>yam</i>
Nyarugusu	Mineral processing and paddy farm	<i>Polygonum Setogulum</i> A. Rich, <i>Carduus Nyassanus</i> (S. Moore) R. E. Fr, <i>Solanum incanum</i> L., <i>Chenopodium Cumbrosioides</i> L., <i>Oxygonum sinuatum</i> (Meisn.) Dammer, <i>A canthus</i> SP, <i>Polygonum salicifocium</i> Willd, <i>Lantana canara</i> L
Mugusu	Wetlands	<i>Cyperus Papyrus</i> , <i>yams</i> , <i>Stachytarpheta angustifolia</i> vahl, <i>Trichodesma zeylanicum</i> (L) R. Br., <i>Rgemone mexicana</i> L, <i>Euphorbia heterophylla</i> L., <i>Euphorbia heterophylla</i> L.

pollutants contained in the matrix of the zeolite.

The major components in the AAC nodules were found to be SiO₂ (51.4%), CaO (26.3%), Al₂O₃ (1.95%) and Fe₂O₃ (1.1%). According to Renman & Renman (2012), ACC is very porous, with low density ranging between 275-400 kg m⁻³.

The results in Paper II showed that AAC had a higher removal capacity for both Zn and Pb than zeolite when the soil was spiked with one metal (Figure 4). This difference can be explained by differences in the radius of the ions, which is 0.084 nm for Pb²⁺ and 0.074 nm for Zn²⁺. The smaller ionic radius allows the Zn ions to penetrate deeper into the exchangeable sites of porous materials, while the larger size of the Pb²⁺ ion impedes their diffusion to exchangeable sites in the channels of porous structures, and thus decreases the ion exchange capacity.

The results also showed that both AAC and zeolite have the capacity for adsorbing heavy metals in the soil.

The removal capacity of zeolite and AAC in a mixed metal scenario (Pb and Zn, Figure 5) showed little difference from their removal capacity for Zn but there was a significant difference for Pb absorption (Figure 4).

Summing up the amount of metal sorbed by each adsorbent; for Zn it was 151.08±28.35 kg m⁻³ in AAC and 130.16±13.3 kg m⁻³ in zeolite. This was a significant difference (p<0.001) according to t-test. For Pb 69.2kg m⁻³ was adsorbed by Zeolite while AAC adsorbed 65.5 kg m⁻³. The sorption of Pb was low, because of the low concentration of the soil and low available component.

Table 4. Chemical composition of the zeolite nodules used in the experiment

Zeolite	
Chemical from XRF	composition (%)
SiO ₂	73.73
Al ₂ O ₃	12.1
CaO	2.43
Fe ₂ O ₃	3.81
K ₂ O	3.22
MgO	0.95
MnO	0.06
Na ₂ O	2.82
P ₂ O ₅	0.13

3.6. Speciation of metals in sorption experiment soil

Speciation analysis of both Pb and Zn in the soil used for the experiment showed that the majority of these metals were present in the form of firmly bound metal in the soil (Figures 6 and 7) (Paper II). Thus most of the metals in the soil were attached to the soil particles and were not readily released on irrigation of the experimental set-up with water. The plant-available heavy component is the sum of the acid, reducible and oxidisable digested fractions of the metal. From Figures 6 and 7 it can be seen that only a small fraction of the metal was available. Compared with the total metal concentration analysed, the plant-available component accounted for 13-39% for Zn and 31-39% for Pb. This might be the reason for the low adsorption of the metals on the sorbent materials.

3.7. Combined phytoremediation and sorption

The two methods (phytoremediation and sorption of metals on zeolite and AAC) can be used together to complement each other for enhanced soil remediation.

Table 5. Chemical composition of the AAC nodules used in the experiment

AAC		
Chemical from XRF	composition (%)	g kg ⁻¹
SiO ₂	51.4	
Al ₂ O ₃	1.95	
CaO	26.3	
Fe ₂ O ₃	1.1	
Si		232
Ca		194
Al		10.0
Fe		8.2

Some indigenous plant species were identified as having good potential for metal uptake from the polluted soils. However the low bioavailable fraction of metals in the soil may be a hindrance to phytoremediation. The application of a chelating agent may be one possible solution for the problem of low bioavailability (Turan & Esringü, 2007). However, it should be remembered that any increase in the bioavailability of heavy metals may affect plant growth and in worse case an increase in metal ions can kill the plants (Alkorta *et al.*, 2004; Wu *et al.*, 2004). This can have very serious negative implications on the application of phytoremediation technology. Beside the effects of increased metal ions on the plant, the increase in soluble metals in the soil is likely to pose high environmental risks to groundwater sources, the reason being that there is no way to increase the absorption capacity of the plant. Thus it is possible to increase the soluble component of the metal in the soil, but the capacity to equally increase the metal uptake by plants is limited. It has been argued that the uptake can be enhanced by use of genetically modified plants (Wong, 2004). This may be a good idea, but is still a subject of discussion. For one reason, we cannot be sure of the ecological implications of these genetically engineered plants.

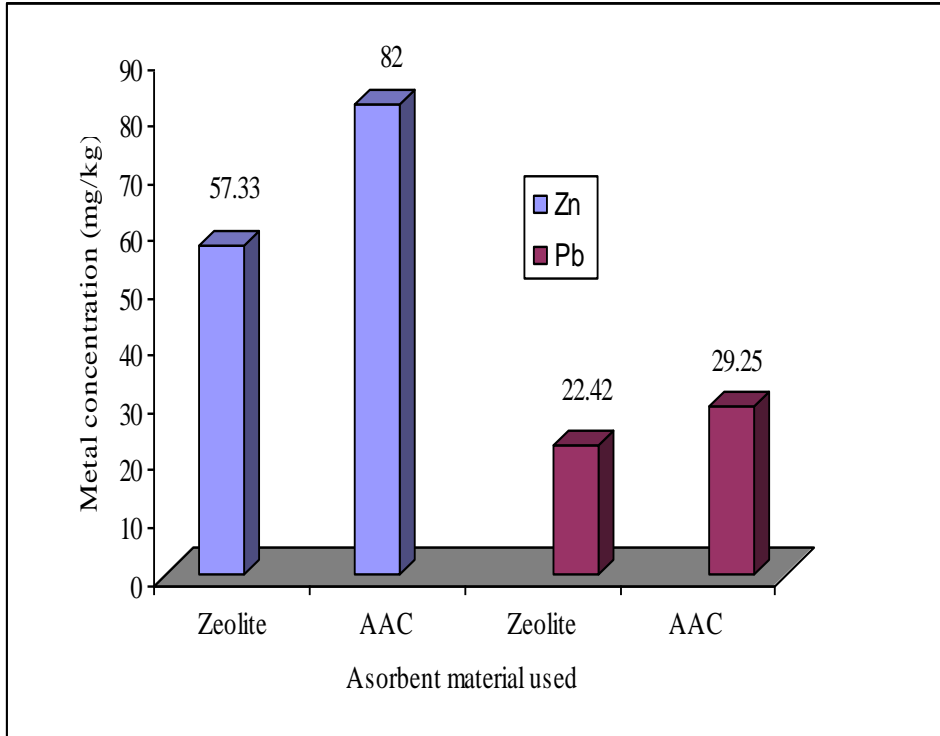


Figure 4.
Sorption of Zn and Pb on zeolite and AAC when the soil was spiked with one of these metals.

Furthermore, plants can suffer from other calamities, such as drought and outbreaks of disease and pests. Sorbent materials do not suffer from such environmental problems. In addition, the size of the adsorbent can be adjusted according to need, whereas plants roots cannot be adjusted at different levels of

metal concentration. With sorbents shaped as nodules, both size and shape adjustments are possible. The other possibility is that with these materials they can be arranged in different patterns in the soil, depending on the flow patterns of water in the soil. Thus it is proposed that clean-up of heavy metal

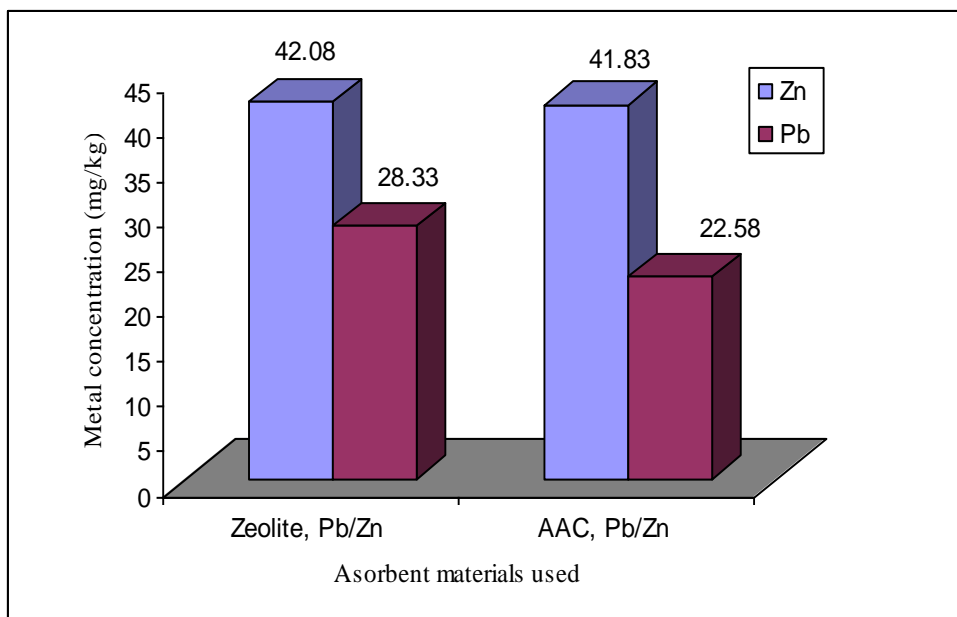


Figure 5.
Adsorption of Zn and Pb on zeolite and AAC when soil was spiked with both of these metals.

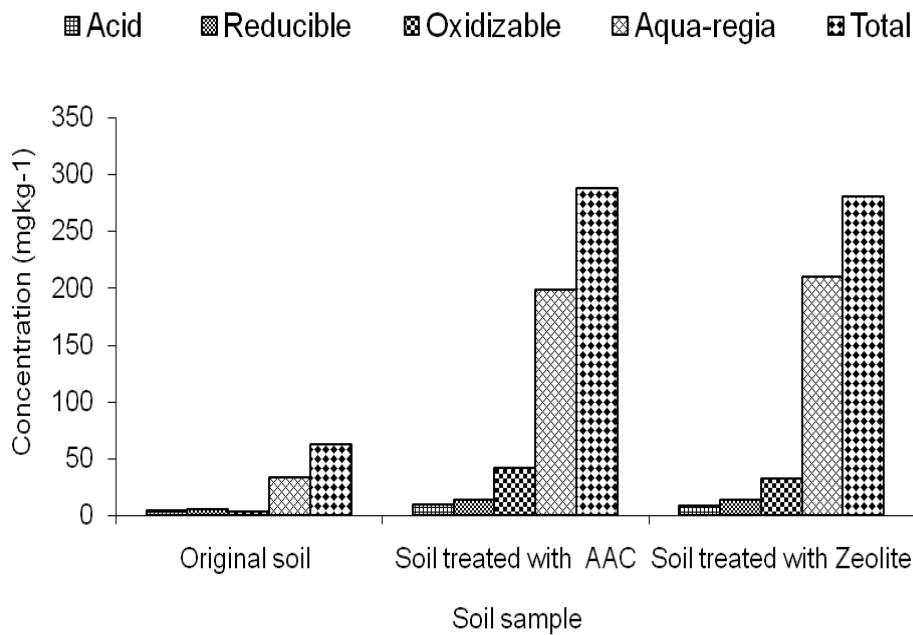


Figure 6.
Speciation of Zn in the original soil and in zeolite-treated soil samples (Box 1) and AAC-treated samples (Box 2) spiked with Zn.

polluted soils be done by application of both phytoremediation plants and heavy metal sorbent. With the application of both methods, the use of the chelating agents can be avoided, thus minimising the risky of polluting the groundwater. In soils with very high concentrations of metals, sorbents can be used to pre-clean the soil. Once the levels in soils are lowered by using sorbents, and then hyperaccumulator plants can be used to further lower the concentration to acceptable levels. The plant at this level can use their rhizosphere acidification (Brett *et al.*, 2006) to dissolve the remaining metals, thereby avoiding addition of chemicals to the soil. Plants can be affected by pests and diseases, so when they represent the sole method this can completely impede remediation, but in the combined method, the adsorbent materials can provide for metal removal (Alkorta, 2004). The effects of climate change will highly influence the applicability of phytoremediation, especially when it is used as a stand-alone technology, but when combined with other methods the impacts of the pollution can be greatly minimised.

Evaluation of the cost in terms of time wasted when using phytoremediation as the sole method (up to 18-60 months) can justify the ideal of applying a combined method to remove heavy metals from soils.

3.8. Application of existing laws and regulation to protect the Environments.

Despite the fact that Tanzania has established a number of laws and policies meant for environmental protection, these efforts have been hampered by the absence of Rule of law. Notwithstanding several penalties stipulated in the law, they have not been effective to inflict pain to polluter because they are not implemented. To facilitate the environmental protection the laws should be implemented and enforced. As part of enforcement monitoring of various development activities should be done regularly and whenever these activities are realized to have contribution to the environmental pollution the existing laws and regulations should be implemented. Principles such as 'polluter pays principle' can also be implemented through internalizing

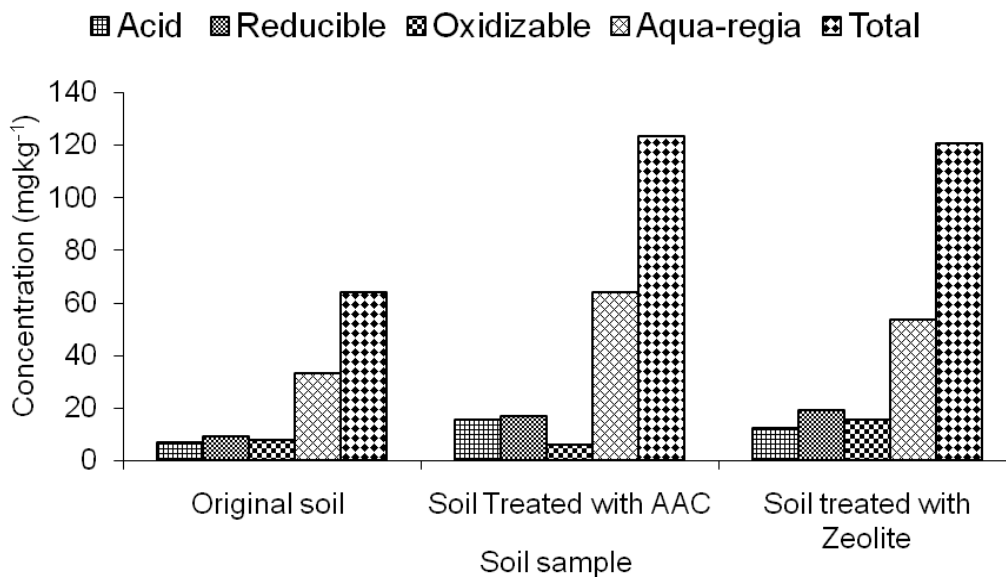


Figure 7. Speciation of Pb in the original soil and in ACC-treated soil samples (Box 7) and zeolite-treated samples (Box 2) spiked with Pb.

environmental costs by making those who benefit from the environmentally damaging activity bear the costs of the damage. The economic incentives such as introduction of pollution tax can encourage adaptation of waste management systems to decrease the pollutant discharges.

Lack of knowledge on the impacts of environmental pollution and their related consequences contributes a lot to poor public support on the environmental protection. Most of small scale miners and the general public lack the knowledge of the consequences of pollution, and hence they provide no support to the efforts of the environmental protection. Thus awareness campaign should be conducted to all small scale miners on the impacts of pollution to the environments and public health.

4. CONCLUSIONS

Some of the important conclusions drawn from this work were:

- *Sporobolus* sp. is a hyperaccumulator of Cu, as the Cu concentration in plant shoots exceeded the criterion of 1000 mg kg⁻¹. Other species, *Launea cornuta* (Oliv & Hiern) O. Jeffrey, *Tagetes minuta* (L.) and

Blotiella glabra (Bory) Tryon showed high potential for phytoextraction of Cu, while *Dioscorea* spp. (yam) and *Stylochaeton natalensis* Schott showed high potential for Cu phytostabilisation.

- No hyperaccumulators of Pb or Zn were identified in the area, but *Tephrosia candida* and *Tagetes minuta* (L.) were identified as potential plants for phytoextraction of Pb, while *Conyza bonariensis* (L.) Cronquist, *Launea cornuta* (Oliv & Hiern) O. Jeffrey, *Tagetes minuta* (L.), *Blotiella glabra* (Bory) Tryon, *Pteridium aquilinum* (L.) Kulm and *Polygonum setogulum* A. Rich were identified as potential plants for phytoextraction of Zn. *Sphaeranthus africanus* (L.) and *Pteridium aquilinum* (L.) Kulm showed potential for phytostabilisation of Pb and *Stylochaeton natalensis* Schott for phytostabilisation of Zn.
- Both zeolite and AAC nodules have the capacity to remove Zn and Pb if they are buried in soils polluted with heavy metals. However, AAC has a higher removal capacity for Zn compared with zeolite.
- The removal capacity of both zeolite and AAC is lowered when samples contain a mixture of metals. This can be attributed to

competition between the metal ions for adsorption sites and the lower concentrations of each ion in the soil.

- Speciation analysis revealed that the plant-available component of the heavy metals was low in experimental soils and in soils collected in the plant sampling areas. This could impede phytoremediation.
- A combined method using hyperaccumulator plants together with heavy metal adsorbent nodules in polluted soil could overcome some of the problems of the phytoremediation method.

5. FUTURE RESEARCH

To develop a functioning low cost remediation method for heavy metal polluted soils, some aspects need to be explored more. Such aspects include:

- Studies on the phytotoxicity of heavy metals in selected plants. This is important since the tolerance limit of the identified hyperaccumulator species needs to be established.
- Environmental factors such as pH effects on both the growth of plants and availability of the metals for uptake and adsorption. The optimum pH needs to be explored in order to gain a better understanding of the effects of pH for growth of plants and reasonable availability of metals for uptake.
- This study identified plants with potential for hyperaccumulation of heavy metals, but further studies need to be undertaken to establish the heavy metal uptake kinetics in the selected plants
- This study established that AAC and zeolite nodules have potential for use as heavy metal adsorbents in polluted soils. However, further investigations are required to evaluate the sorption limits of the materials on application of chelating agents, and the combined effects when these adsorbents are used alongside hyperaccumulating (phyto-remediating) plant species.

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