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Lead remediation of contaminated water using *Moringa Stenopetala* and *Moringa oleifera* seed powder

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ABSTRACT: The increasing influx of heavy metals into water bodies from industrial, agricultural, and domestic activities is of global concern because of their well documented negative effects on human and ecosystem health. A recent study of streams in Blantyre and Zomba, Malawi revealed lead levels of up 0.118 mg/L, exceeding the World Health Organisation acceptable level of 0.01 mg/L. Our ongoing study on low cost effective heavy metal remediation techniques in developing countries has already demonstrated that *Moringa oleifera*, the well known source of natural water clarifiers, is effective in heavy metal detoxification of water. This paper presents the first reported use of a related species, the African moringa, *Moringa stenopetala* for lead detoxification and preliminary investigation of the interaction of the metal with the polyelectrolytes of *M. oleifera* and *stenopetala*. The potential of *M. stenopetala* for lead removal was tested by means of jar tests. With an initial lead concentration of 7 ppm, *M. stenopetala* seed powder, at doses of 0.50, 1.00, 1.50, 2.00 and 2.50 g/100mL, reduced the concentration of lead by 20.00 ± 0.00, 46.19 ± 2.06, 71.19 ± 2.06 and 89.43 ± 0.60 and 96.23 ± 0.12 % respectively. *M. stenopetala* was more effective than *M. oleifera* in removing lead from water (p=0.001 at 95% confidence level). For *oleifera*, lead levels decreased exponentially during the first 5 h. of the reaction and then equilibrium was established; for *stenopetala*, a linear decrease was observed. The pH of the mixture rose from 2.30 to a maximum of 2.53 and 2.57 and then fell to an equilibrium value of 2.30 and 2.29 for *oleifera* and *stenopetala* respectively. Lead removal was also affected by pH, ionic strength, and water hardness. Our results show that *M. stenopetala* has potential in lead remediation of contaminated waters. Further studies are being carried out on remediation of other metals and the mechanism of the metal moringa interaction.

Key words: *Lead, Moringa oleifera, Moringa stenopetala, heavy metals, water pollution, remediation.*

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INTRODUCTION

The United Nations Environment Programme's (2004) Water Policy and Strategy identifies several water focal areas including fresh water scarcity, land based pollution sources, aquatic biological diversity, resource use and management, and knowledge and technology transfer in integrated water management. The Malawi State of the Environment Report notes that water degradation is a major environmental problem that threatens the health and well being of humans and ecosystems (Malawi Government, 2002). Improper disposal of various types of waste, deforestation, and poor agricultural practices that encourage soil erosion and deposition of sediments into the water bodies were identified as the major causes of water degradation in Malawi. Studies of Malawian urban water bodies and wastewater treatment plants revealed that some water quality parameters including heavy metals exceed the World Health Organization (WHO, 2004) acceptable limits (Matope, 2002; Banda *et al.*, 2001 and Sajidu *et al.*, 2005). Of particular concern are the high levels of lead in some of the water bodies. The Sanitation Master Plan for the City of Blantyre (Matope, 2002) reported lead levels of 0.73-0.96 mg/L in Mudi and Limbe streams and Banda *et al.*, (2001) recorded 0.23 ± 0.00 mg/L lead for the Lunyangwa river basin, all above the WHO acceptable limits of less than 0.01 mg/L. Our recent quality inventory compilation of Blantyre streams (Limbe, Nasolo, and Mudi) and wastewater treatment plants (Limbe and Soche) recorded levels of lead from 0.027 to 0.118 mg/L with most values exceeding the WHO limit (Sajidu *et al.*, 2005). Lead is a pollutant of global concern with well known toxic effects (Alloway and Ayres, 1990). Major sources of lead pollution in water are lead-acid batteries, lead pipes, solders, agricultural chemicals, lead mining, and vehicle exhausts especially where, as in Malawi, leaded petrol has not been completely phased out. Lead is a powerful neurotoxin and a range of pathological conditions is associated with acute lead poisoning, the most characteristic being cerebral oedema. Clean-up technologies for the removal of lead and other heavy metals from water include chemical coagulation using aluminium and ferric salts (Fatoki and Ogunfowokan, 2002) and cationic surfactants (Evans, 2003); physical precipitation using ion exchange and adsorption (Singh *et al.*, 2001) and phytoremediation that includes rhizofiltration, phytostabilisation, phytoextraction (Lyte *et al.*, 1998 and Lambert *et al.*, 2003). However, water and wastewater treatment in Malawi does not include heavy metal removal or monitoring due to the high costs and/or the lack of technical expertise. Recently there has been increased interest in the subject of natural coagulants for treatment of water and wastewater in developing countries (Jahn, 1986; Ndabingesere *et al.*, 1995; Sutherland *et al.*, 1994; Gebremichael, 2004 and Henry *et al.*, 2004). The tropical plants of the family of *Moringaceae* are amongst some of the natural coagulants that have been studied for clarification of turbid water. *Moringa oleifera* is the most widely distributed, well-known and studied species of the family *Moringaceae* because of its previous economic importance as a source of the commercially important 'Ben oil' and more recently, as a multipurpose tree for arid lands and a source of water purifying agents for developing countries (Morton, 1991). *M. oleifera* is native to sub-Himalayan North-western India and Pakistan but the plant was distributed to other areas of tropical Asia in prehistoric times and to other parts of the world including Malawi during the British colonial era. *M. stenopetala,* often referred to as the African Moringa Tree, originates from southern Ethiopia and Kenya (Jahn, 1991). The food, fodder, water clarifying and medicinal uses of the *Moringaceae,* especially *oleifera* are well documented and the trees are recommended for live fencing, intercropping, and pollution control (Morton, 1991; Moges, 2004; Coote *et al*., 1997; Pratt *et al*., 2002; Williamson, 1975; Palgrave, 1983 and van Wyk and van Wyk, 1997). The water soluble *Moringa* seed proteins possess coagulating properties similar to those of alum and synthetic cationic polymers. Jahn (1981) first studied and confirmed the coagulating properties of *Moringa* seeds after observing women in Sudan use the seeds to clarify the turbid Nile waters. The active agents in *Moringa* extracts responsible for coagulation are dimeric cationic proteins with molecular weight of 13,000 Da and isoelectric points between 10 and 11 (Jahn, 1981 and Ndabigengesere *et al.*, 1995). The mechanism of coagulation was suggested to be adsorption and neutralisation of charges, or adsorption and bridging of destabilised particles, the two assumed to take place simultaneously. Gassenschmidt *et al.*, (1995) reported the isolation from *M. oleifera* of a flocculating protein of 60 residues with molecular mass of about 6.5 kDa, isoelectronic point above pH 10, high levels of glutamine, arginine and proline with the amino terminus blocked by pyroglutamate, and flocculant capacity comparable to a synthetic polyacrylamide cationic polymer. However, a non-protein coagulant has also been reported but not characterised (Okuda *et al.*, 2001). Our earlier studies showed that *M. oleifera* seed powder is effective in heavy metal remediation of water (Sajidu *et al.*, 2005). The African moringa, *M. stenopetala* is a quick growing tree 6-10 m tall with a trunk 60 cm in diameter at breast height, strongly branched, thick at base with a bark white to pale grey or silvery, smooth wood and soft leaves up to 55 cm long (Jahn, 1991 and Moges, 2004). The species is known by different vernacular names such as Kallanki and Haleko (Ethiopia), and Cabbage Tree (English). *M. stenopetala* is less widely distributed than *M. oleifera* but *stenopetala* is reportedly more resistant to insect pests than other members of the family and its seeds are larger and easier to process than those of *oleifera* (Kayambazinthu, 2003). Although the water clarifying properties of *M. stenopetala* have not been as extensively studied as those of *M. oleifera*, Jahn (1986) reported that 100-150 mg/L of *M. stenopetala* was as effective in water clarification as 200 mg/L of *M. oleifera* which indicates that *stenopetala* is more effective than *oleifera*. However, no work has been reported previously on the potential of *M. stenopetala* to remove heavy metals from water. The objectives of this study, therefore, were to evaluate the potential of *M. stenopetala* in removing lead from water and compare its effectiveness with that of *M. oleifera* reported earlier by our group (Sajidu *et al.*, 2005) and therefore this is the first report on the potential of *Moringa stenopetala* on heavy metal removal.

MATERIALS AND METHODS

Seeds were identified by the Forestry Research Institute of Malawi. *Moringa stenopetala* seeds were obtained from Kenya (Whizpop Products Ltd., Nairobi) in 2005. *Moringa oleifera* seeds were collected from Chikwawa district in southern Malawi in 2004 and 2005. Seeds were deshelled by hand and the deshelled seeds were ground in a coffee mill (National MX-J210PN), until a consistent powder was obtained. Defatted cakes were prepared by cold solvent extraction of the powdered seed with hexane fraction and traces of fat were removed from the residue by washing with diethyl ether until the residue was confirmed as fat free using a paper test and then dried in a vacuum oven (Gallenkamp OVL 570 010 J) at 40 ÚC and 600 mbars for 24-48 h. The following analytical grade chemicals were used: lead nitrate (BDH Chemicals Limited, Poole, UK); sodium carbonate, magnesium sulphate, sodium bicarbonate, hexane fraction (Sarchem (Pty) Krugersdorp Ltd, RSA); sodium chloride, sodium hydroxide, calcium chloride and nitric acid (Associated Chemical Enterprises (Pty) Ltd, RSA); hydrochloric acid, diethyl ether (Glassworld, RSA). pH was determined using a pH meter (Metrohm 744) checked with buffers at pH 4 and 7; constant temperature treatments were done in a constant temperature water bath (Bath: Haake Type 000-5584, Thermostirrer: Gallenkamp No. 85) and shaking was done using a Griffin shaker. Microsoft Excel was used for descriptive statistics and plots of treatment data. Analyses of variance were carried out using GenStat Discovery Edition.

Synthetic lead water

Synthetic lead water was obtained by dilution of a stock lead solution (1000 ppm) prepared by dissolving lead nitrate in de-ionised water or sodium chloride solution as described by the American Public Health Association (APHA, 1990).

Determination of lead content

Lead concentration was determined using atomic absorption spectroscopy (Shimadzu AA-680G V-5) at 283.3 nm with an air-acetylene flame as described by APHA (1990).

Lead removal

100 mL of synthetic lead water containing 7 ppm Pb was prepared using deionised water and appropriate masses of whole seed or defatted powder were added and the mixture stirred for 1 h. The mixture was filtered by gravity through Whatman No.1 filter paper and the lead concentration of the filtrate was determined.

Kinetics of lead removal

Synthetic lead water containing 7 ppm Pb was treated with 1.0 g of *Moringa oleifera* and 0.5 g of *Moringa stenopetala* whole seed cake and the residual Pb concentration of the solution quantified after different times of treatment.

Temperature dependence

7 ppm synthetic lead water was treated with 1.0 g *M. oleifera* or *stenopetala* at different temperatures. The *moringa* suspension was immersed in a constant temperature water bath with shaking for 1 h. and the residual lead concentration quantified.

pH changes during treatment

2.5 g portions of *M. stenopetala* or *oleifera* were added to 100 ml of 7 ppm or 3 ppm Pb in deionised water or 0.1 M sodium chloride and the pH recorded at intervals.

Effect of pH on lead removal capacity

Synthetic lead water of different pH's were prepared by adjusting the pH of a solution of 7 ppm Pb in deionised water using 1 M sodium hydroxide or hydrochloric acid and the resulting lead solutions treated with moringa whole seed cakes for 1h.

Effect of water hardness

7 ppm Pb solutions were treated with 1.5 g of moringa whole seed powders suspended in different concentrations of Mg^{2+}/Ca^{2+} or HCO_3^-/CO_3^{2-} mixtures. The Mg^{2+}/Ca^{2+} was prepared as described in Texas specification No. 485-54-09A. The HCO_3^-/CO_3^2 mixture was prepared by dissolving 1.3768g $NaHCO_3$, and 1.7662g Na_2CO_3 to make a 1000 ppm mixture and this was diluted to different appropriate concentrations.

Effect of sodium chloride

Solutions containing 7 ppm Pb in 0.0, 0.2, 0.4, 0.6, and 0.8 M sodium chloride were prepared by combining and diluting the stock lead solution (1000 ppm) and a 1 M stock solution of sodium chloride. The solutions were treated with 1.5 g of *M. oleifera* or *stenopetala* whole seed cake and the residual lead concentration determined.

RESULTS

Lead ion removal

Lead treatment was done using different doses of moringa whole seed powder. The results indicate that *M. stenopetala* has the capacity to remove heavy metals from water (Table 1) and the effectiveness of removal increased with increasing dosage of whole seed powder with p <0.001 at 95% CL. There was no significant difference for lead removal at 1 h and 24 h. (p = 0.348). Similar results were obtained using defatted cakes (Table 2). Metal ion removal increased with increasing dosage of *M. stenopetala* defatted seed cake (p <0.001) and there was no significant difference for Pb removal at 1 h. and 24 h. (p=0.55). The effectiveness of Pb removal of the defatted cakes (Table 2) and the whole seed powder (Table 1) was also compared and the defatted cakes were more effective than the whole seed cakes (p <0.001). Comparison of the removal effectiveness of *Moringa oleifera* and *Moringa stenopetala* (Table 3) using ANOVA showed that there was a significant difference in the effectiveness of removal with *stenopetala* being more effective than *oleifera* (p = 0.001).

Kinetics of lead removal

Time dependence studies of lead removal by *stenopetala* gave a straight line plot for lead concentration versus time suggesting that removal is zero order with respect to metal ion (Fig. 1). For *oleifera*, a plot of natural log of lead concentration versus time gave a straight line indicating that removal is either first order or pseudo first order (Fig. 2).

Effects of other ions on metal removal

(a) Sodium chloride

Fig. 3 shows the effect of increasing NaCl ionic strength on lead removal using *Moringa*. Metal ion removal decreased with an increase in the concentration of NaCl from 0.0 to 0.8 M. The percentage decreased from 72.00 to 16.44 % for *oleifera* and from 65.76 to 34.84 % for *stenopetala*.

Table 1: Metal ion removal by *M. stenopetala* whole seed cake at initial metal ion concentration of 7 ppm at around 25 ÚC

Dosage (g/100 mL)	Remaining lead conc./ppm after 1 h. (% removal)	Remaining lead conc./ppm after 24 h. (% removal)
0.5	5.60± 0.00 (20.00± 0.00)	5.03± 0.25 (28.14± 3.57)
1.0	3.77± 0.14 (46.19± 2.06)	3.28± 0.00 (53.14± 0.00)
1.5	2.02± 0.14 (71.19± 2.06)	2.03± 0.00 (71.00±0.00)
2.0	0.74± 0.04 (89.43 ± 0.60)	0.99 ± 0.004 (85.85 ± 0.51)
2.5	0.26 ± 0.01 (96.23 ± 0.12)	0.71 ± 0.03 (89.83 ± 0.46)

Table 2: Metal ion removal by hexane defatted *M. stenopetala* at initial metal ion concentration of 7 ppm around 25 ÚC

Dosage (g/100 mL)	Remaining lead conc./ppm after 1 h. (% removal)	Remaining lead conc./ppm after 24 h. (% removal)
0.5	4.43±0.27 (36.76± 3.79)	4.35± 0.00 (38.00 0.00)
1.0	1.64 0.13 (76.71± 1.98)	1.87± 0.13 (73.33± 1.88)
1.5	Not detected	Not detected
2.5	Not detected	Not detected

Table 3: Comparison of 1 h. treatment of lead water using whole seed powders *M. oleifera* and *M. stenopetala*

Dosage (g/100 mL)	Remaining lead conc./ppm MOWSK (% removal)	Remaining lead conc./ppm MSWSK (% removal)
1.0	3.66± 0.00 (47.71± 0.00)	3.36± 0.17 (52.05± 2.40)
1.5	2.21± 0.00 (68.43± 0.00)	1.91± 0.064 (72.52± 0.91)

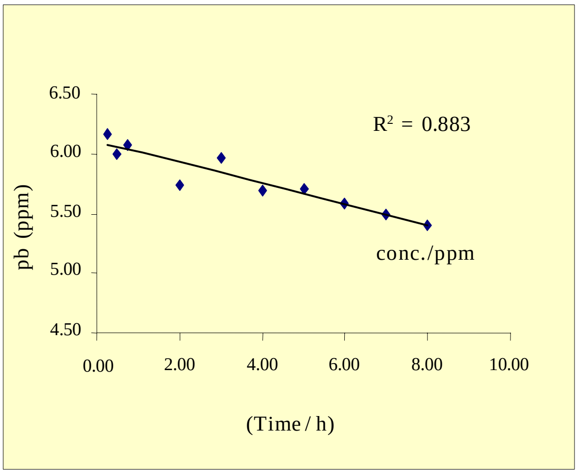

Fig. 1: Relationship between concentration and time for *M. stenopetala* whole seed cake

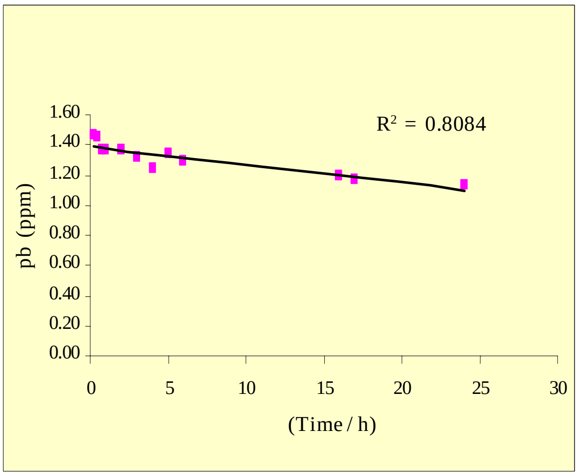

Fig. 2: Relationship between concentration and time for *M. oleifera* whole seed cake

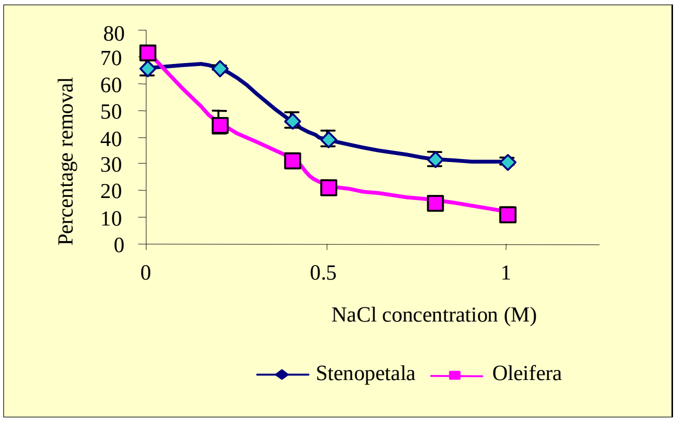

Fig. 3: Relationship between NaCl concentration and lead ion removal using *Moringa* whole seed cakes at initial lead ion concentration of 7 ppm

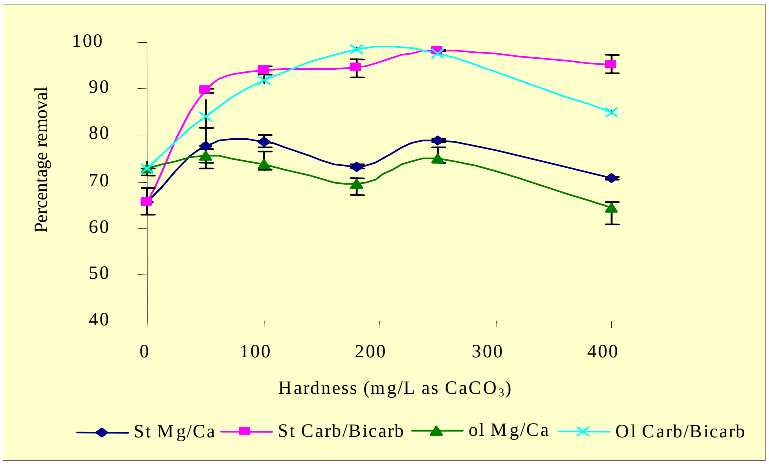

Fig. 4: Effects of water hardness on lead ion removal using *oleifera* and *stenopetala* whole seed cake at 7ppm lead concentration and 25 °C

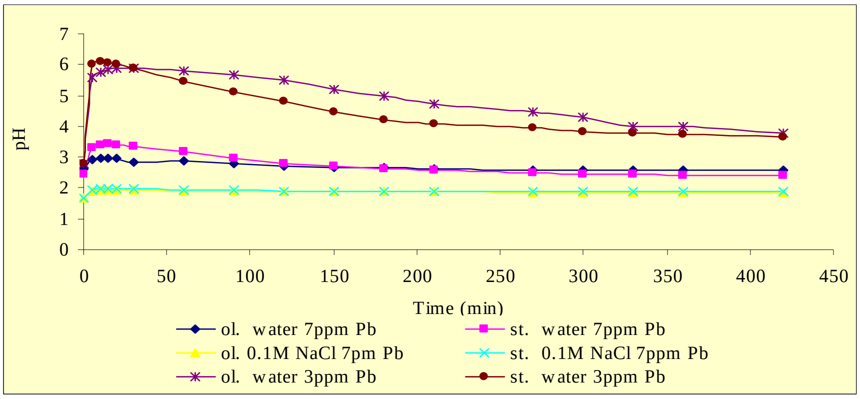

Fig. 5: Change in pH with treatment time for lead ion removal using 2.5 g *M. stenopetala* and *oleifera* in de-ionised water and 0.1M NaCl solution initial lead concentrations of 7 and 3 ppm (23.5 ÚC)

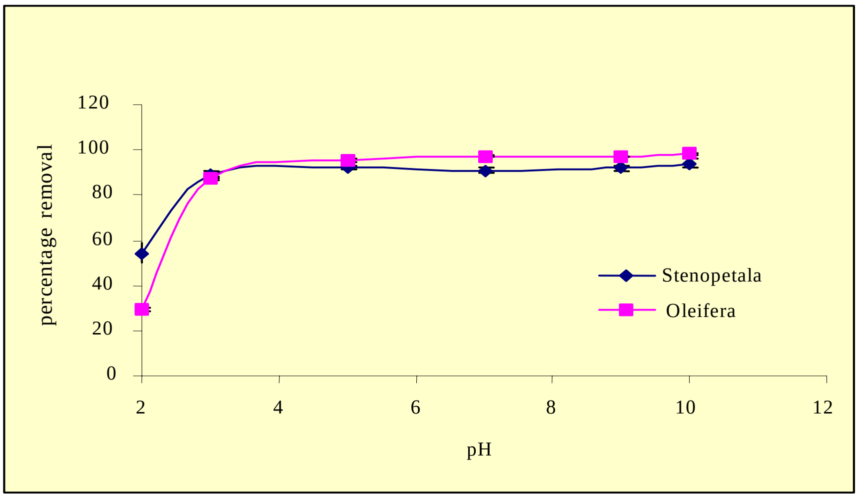

Fig. 6: Relationship between pH and percentage lead removal using Moringa whole seed cakes at initial lead ion concentration of 7 ppm and 25 °C

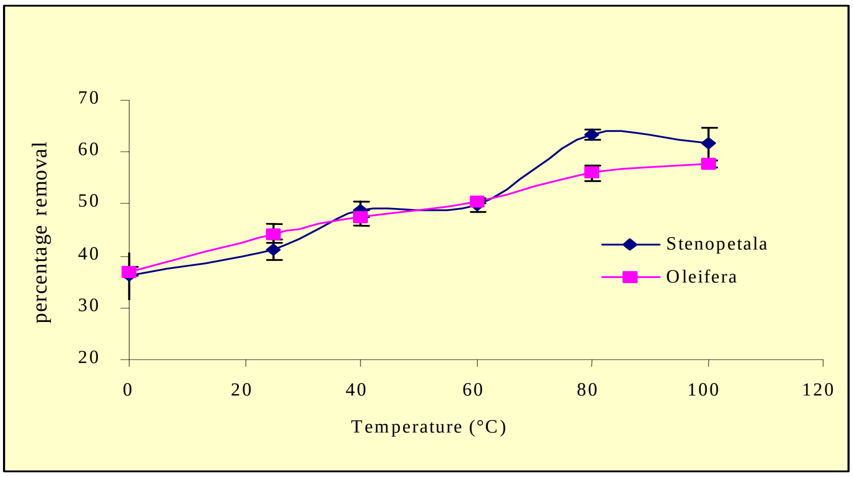

Fig. 7: The effects of temperature on lead removal using moringa seed powders at 7 ppm initial lead concentration

(b) Water hardness ions (Mg^{2+}/Ca^{2+} and HCO_3^-/CO_3^{2-})

Removal of lead ions in different concentration of magnesium/calcium or carbonate/bicarbonate mixtures was carried out to investigate the effects of increasing water hardness on the metal ions removal (Fig. 4). Percentage removal increased with the increase in carbonates/bicarbonates concentration and there was no general trend for magnesium/calcium.

pH changes

(a) pH changes in during treatment

Lead treatment was carried out in deionised water or 0.1 M sodium chloride solution while recording pH at different time intervals to investigate the effects of treatment time on pH of the mixture (Fig. 5). The removal was done for different initial lead concentration of 3 ppm for deionised water and 7 ppm for both deionised and salt water. The variation in pH was more pronounced for 3 ppm Pb^{2+} solution in deionised water and least pronounced in 0.1 M NaCl solution.

(b) Effect of initial pH

Treatment was carried out at different initial pH (pH 2–10) to investigate the effects of initial pH on lead ions removal (Fig. 7). The percentage removal increased with an increase in initial pH. The maximum removals for treatment with powders, 93.99 % and 98.39 %, for *oleifera* and *stenopetala* respectively, were observed at pH 10. However significant change in percentage removal was observed between pH 2 and 3 (29.21 – 87.42 and 54.31 – 89.07 for *oleifera* and *stenopetala* respectively).

Effect of temperature on lead removal

To investigate the effects of temperature on lead removal using *M. oleifera* and *stenopetala* whole seed powders lead treatment was done at different temperatures, 0, 25, 40, 60, 80, and 100 °C (Fig. 7). The percentage removal of lead ions increased with temperature from 36.69 to 57.76 and 36.07 to 61.57 for *oleifera* and *stenopetala* respectively.

DISCUSSION AND CONCLUSION

Our previous studies have shown that *M. oleifera* is effective in removal of heavy metals including lead from water (Sajidu *et al.*, 2005). This present study indicates that *M. stenopetala* seed powders (whole seed and defatted seed) are more effective than *oleifera* in lead removal (Table 3). These results agree with Jahn's (1986) observation that *stenopetala* is a better water clarifier than *oleifera* and suggests that the same agents, the polyelectrolytes, are responsible for water clarification and lead removal. Our observed increase in removal with increasing dosage of seed powders (Tables 1 and 2) is probably due to increasing concentration of the polyelectrolytes. Similarly the superiority of the defatted cake over whole seed cake may be due to the removal of lipids from the whole seed cake, thus increasing the relative concentration of the polyelectrolytes, the likely agents of metal ion removal. Increase in sodium chloride ionic strength caused a decrease in lead removal (Fig. 3) suggesting that the lead removal reaction involves electrostatic interaction (Krishnan and Anirudhan, 2003). At high ionic strength the sodium ions can compete for binding sites with Pb^{2+} ions on the polyelectrolytes and hence reduce lead removal. Furthermore Pb^{2+} ions form stable complexes with chloride ions, which inhibit interaction between Pb^{2+} and the polyelectrolyte sites. The effect of water hardness on lead removal varied depending on the ions involved (Fig. 4). Generally increase in concentration of carbonates/bicarbonates enhances lead removal; this effect can be explained by the formation of sparingly soluble lead carbonates that precipitate out of the solution (McGinnes, 2002). However there was no noticeable trend observed for the effect of the concentration of Mg/Ca on lead removal and further studies are being carried out. For the variation in pH during the course of treatment (Fig. 5), there is a rapid increase in pH during the first few minutes as the seed powder dissolves to give free moringa polyelectrolyte in solution followed by a drop in pH as the dissolved polyelectrolyte interacts with the Pb^{2+} ions. The relative maximum pH values for the different treatments reflect the amount of free moringa polyelectrolyte in solution. Therefore at low concentration (3 ppm) of Pb^{2+} the highest concentration of free polyelectrolytes is expected due to less interaction between the sites leading to the highest maximum pH. At 7 ppm more interaction is expected due to a higher amount of Pb^{2+} hence the observed lower maximum pH. The lowest maximum pH was obtained for the treatment in 0.1 M sodium chloride solution and may be explained by the presence of a greater amount of metal ions which have a salting in effect on proteins and inhibit the extraction and dissolution of the free polyelectrolytes. The observed increase in lead ion removal with increase in initial pH, may be attributed to the formation of insoluble lead hydroxides at higher pH. The lower efficiency in lead removal at low pH probably may arise from the presence of a large amount of H^+ ions in the mixture, which compete with Pb^{2+} ions for the binding sites (Krishnan and Anirudhan, 2003 and Raji *et al*, 1997). The increase in lead ion removal with temperature is possibly due to the breakdown of the protein chain, at higher temperatures to expose more sites for binding with metal ions. In conclusion the study has shown that *Moringa stenopetala* seed powder can be used as an effective heavy metal purifier in water and that *Moringa stenopetala* is more effective than *oleifera* in heavy metal removal.

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