



KTH Land and Water
Resources Engineering

BATCH SORPTION STUDIES OF AQUEOUS CADMIUM AND LEAD FROM CONTAMINATED WATER ONTO SELECTED BIOSORBENTS

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PhD Thesis

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DEDICATION

Ekitabo kbino nkihareyo eri Abamaama abasa; O'Maama Dimitria (Dimintiriya), O'Maama Cecilia, O'Maama Esther (Sirista), O'Maama Anna, O'Maama Margaret (Mangarita), O'Maama Florence nbo'Maama Anthonia (Ntwanita) abankoleire ebikulu okuzhwa omubutobuto bwange, mwebale inho, mvebarire kimbo. O'Kibumba ababe enkabi n'emikisa mingi inho, nabatuzwile okumaiso bonabona ababe ekibumulo ekisa.

SAMMANFATTNING

Kadmium och bly är föroreningar som har skadliga hälsoeffekter på ekosystem genom endera kronisk exponering för låga halter eller akut exponering från höga halter. Även om metallerna fastläggs till jordpartiklar och organiskt material så att deras förekomst i markvatten är låg, så kan de frigöras och transporteras ned till grundvattnet om jordens mättnaskapacitet uppnås eller om pH sjunker. Halterna av spårmetaller beror av naturliga källor t ex bakgrundshalter, vittring och vulkanisk aktivitet såväl som antropogena källor. Men även om naturliga källor kan bidra till förhöjda halter så är det vanligen de antropogena källor som dominerar. Ytvatten, dagvatten, lakvatten från deponier och ytligt grundvatten provtogs i Lake Victorias avrinningsområde i Uganda. Resultaten från analyser visade att grundvattnet från utvalda skyddade källor hade förhöjda halter av spårmetaller speciellt Pb^{2+} och anjonerna NO_3^- , Cl^- och katjonerna Fe, Mn och K. Dagvattnet från de skyddade källornas avrinningsområde var framför allt förorenat med Pb^{2+} . Ytvattnet från de utvalda vattendragen hade $pH > 7$ och var förorenade med järn och mangan. Lakvattnet från en deponi innehöll höga halter av arsenik och i Lubigi channel hade höga halter av föroreningar från skrotansamlingar och bilverkstäder. Föroreningshalterna i ytvatten var relaterad till förändrade demografiska fördelningar och antropogena aktiviteter kopplade till urbanisering och industrialisering i avrinningsområdet. För att bestämma fördelning av metalljonerna i oorganisk och organisk fas i vattnet användes jämviktsmodellen Visual MINTEQ 3.1. Riskbedömningen av exponering från utvalda spårmetaller genomfördes med de Svenska bedömningsgrunderna för ytvatten och med ett Bio-met software verktyg. För utvärdering av föroreningshalterna i grundvatten användes WHO och Ugandas riktvärden.

Flera konventionella reningsmetoder för att åtgärda toxiska spårmetaller i olika typer av vatten så att tillåtna koncentrationer uppnås är tillgängliga. Tyvärr så har metoderna höga installations- och drifts kostnader samt att de producerar giftiga slam avfall som måste omhändertas och inte alltid fungerar effektivt för att fastlägga spårmetaller. Då sorptionsstudier har visat att olika biomassor är effektiva för att rena vatten från spårmetaller i låga koncentrationer har de därför blivit intressanta alternativ. De biomassor som ingick i studien var avfall från odlingar av *Albizia coriaria*, *Erythrina abyssinica*, *Coffea canephora*, *Cyperus papyrus* och *Musa spp.*

De valdes studien valdes ut eftersom stora kvantiteter finns tillgängliga och det saknas andra användningsområden. Experiment utfördes på biomassorna för att karakterisera de strukturella komponenter som bidrar till bindningen av metall joner, konkurrerande katjoner, katjonbyteskapaciteten, materialens effektivitet och verkningsgrad för fastläggning av utvalda spårmetaller. Biomaterialen behandlades med natriumhydroxid och väteperoxid och fastläggningen före och efter behandlingen jämfördes. För bestämning av de vattenlösliga oorganiska komponenterna i lakvatten användes ICP-MS och de fasta strukturella beståndsdelarna analyserades med XRD. Med jämvikts- och kinetiska skakförsök utvärderas biomassorna förmåga att fastlägga kadmium och bly i förorenat vatten. Lakvattnet från skakförsöken analyserades för DOC och spårmetaller och data från experimenten anpassades till kinetiska modeller och isotermer. Resultaten visade att behandlingen med natriumhydroxid ökade fastläggningen till biomassorna i högre grad än väteperoxidbehandlingen. En

Pseudo-second-order kinetisk modell och Langmuir isotherm hade god anpassning till data från experimenten med enskilda metaller, medan Freundlich isotherm passade bättre när fler joner konkurrerade om sorptions ytorna. Den underliggande mekanismen är troligen chemisorption som förekommer i ett monolager men som har heterogena ytegenskaper. Experiment med lösningar som innehöll både bly och kadmium visade att blyupptaget överskred upptaget av kadmium vilket indikerar att affiniteten är högre för bly än för kadmium. Även om det fortfarande finns osäkerheter så visar resultaten från studien att de undersökta biomassorna är lovande åtgärdsmaterial för att fastlägga Cd och Pb i förorenat naturligt vatten

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LIST OF ABBREVIATIONS

AAS	-	Atomic absorption spectrometry
BGS	-	British Geological Survey
BOD	-	Biological Oxygen Demand
BLM	-	Biotic Ligand Model
COD	-	Chemical Oxygen Demand
CEC	-	Cation Exchange Capacity
EA	-	Exchangeable Acidity
EC	-	Electrical conductivity
EPA	-	Environmental Protection Agency
FA	-	Fulvic acid
DOC	-	Dissolved organic carbon
DOM	-	Dissolved organic matter
DWD	-	Directorate of Water Development
ICP-MS	-	Inductively coupled plasma-mass spectrometry
ICP-OES	-	Inductively coupled plasma-optical emission spectrometry
MWE	-	Ministry of Water and Environment
NWSC	-	National Water and Sewerage Corporation
PVC	-	Polyvinylchloride
RCR	-	Risk Characterisation Ratio
rpm	-	revolutions per minute
SHM	-	Stockholm Humic Model
SOGs	-	Surface oxygen groups
TIC	-	Total inorganic carbon
TOC	-	Total organic carbon
USEPA	-	US Environmental Protection Agency
WHO	-	World Health Organisation
WQC	-	Water Quality Criteria
XRD	-	X-ray diffraction

LIST OF APPENDED PAPERS

Paper I

G. K. Bakyayita, A. C. Norrström & R. N. Kulabako, 2019. Assessment of levels, speciation and toxicity of trace metal contaminants in selected shallow groundwater sources, surface runoff, wastewater and surface water from designated streams in Lake Victoria Basin, Uganda. *Journal of Environmental and Public Health*, Vol. 2019, Article ID 6734017, 18 pages, ISSN 1687-9805, E-ISSN 1687-9813, <https://doi.org/10.1155/2019/6734017>

Paper II

G. K. Bakyayita, A. C. Norrström, M. Nalubega & R. N. Kulabako, 2014. Kinetic studies of Cd (II) and Pb (II) ions biosorption from aqueous media using untreated and chemically treated biosorbents. *Water Science and Technology* 69(11),pp 2230-2236 , doi: [10.21216/wst.2014.147](https://doi.org/10.21216/wst.2014.147)

Paper III

G.K. Bakyayita, A.C. Norrström, & R.N. Kulabako, 2018. Characterisation and application of un-treated and base-treated biosorbents from *Albizia coriaria*, *Erythrina abyssinica* and *Musa* spp.in the uptake of Cd (II) and Pb (II) ions from contaminated water (Manuscript)

Paper IV

G. K. Bakyayita, A. C. Norrström, and R. N. Kulabako, 2015. Competitive and Noncompetitive Batch Sorption Studies of Aqueous Cd (II) and Pb (II) Uptake onto *Coffea canephora* Husks, *Cyperus papyrus* Stems, and *Musa* spp. Peels, *Journal of Chemistry*, vol. 2015, Article ID 696098, 17 pages, <http://dx.doi.org/10.1155/2015/696098>

AUTHOR'S CONTRIBUTION TO PAPERS

- I. I was responsible for this paper. I was mainly responsible for the planning and design of the experiments, data analysis, modelling and writing of the manuscript.
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- III. I was the main author of this paper. I was responsible for the planning and experimental designs, data analysis and writing of the manuscript.
- IV. I was responsible for all parts of this paper. I was responsible for the planning and experimental designs, data analysis and writing of the manuscript..

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ABSTRACT

Groundwater, wastewater, surface runoff and surface water samples from Lake Victoria basin, Uganda was assessed for trace metals contamination. Untreated, base-treated and peroxide-treated biosorbents from *Albizia coriaria*, *Coffea canephora*, *Cyperus papyrus*, *Erythrina abyssinica* and *Musa spp* were investigated for removal of selected trace metals from contaminated water in batch studies. The assessed shallow groundwater and surface water was contaminated with iron and manganese. Selected speciation studies using Visual MINTEQ showed that in leachates from Municipal dumpsites 74% of the metal ions were bound to DOM, 13% were free ions and 13% were in inorganic forms moreover for urban streams 37% of the metal ions were bound to DOM, 44% were free ions and 19% were in inorganic forms. The metal levels in surface water, landfill leachate and surface runoff showed elevated levels and revealed increased risks to environmental health. Risk analysis based on the Swedish EPA showed that varied risks of negative effects in 30% – 76% of the sample sites ranging from high to increased risk in surface water whereas the results from Bio-met tool showed potential risk to toxicity effects of Cu^{2+} , Ni^{2+} , Zn^{2+} and Pb^{2+} in 15.3% - 30.8% surface water samples and 8.3% - 62.5% groundwater samples. Batch sorption studies revealed that the optimal conditions for Cd^{2+} and Pb^{2+} ions uptake were; pH 3.5 – 5.0 for contact time 3.0 – 3.5 hours and biosorbent dosage 10 – 12.5 g/L. Base-treated biosorbents showed 10 – 17 % sorption enhancement for Cd^{2+} ions and 1.6 – 2.3 % uptake reduction for Pb^{2+} ions. The biomass negative potential for binding base cations was in the order; *Musa spp.* > *A. coriaria* > *E. abyssinica* and base treatment reduced DOC leaching from biosorbents in the order; *E. abyssinica* > *A. coriaria* > *Musa spp.* Speciation studies showed that more ions were complexed to DOC in solutions at various pH levels. The maximum sorption intensities for both Cd^{2+} and Pb^{2+} ions uptake onto biomass occurred for low initial metal concentration; 5 mg/L. Freundlich model best fitted data for Pb^{2+} ions uptake whereas Temkin model fitted the sorption data for Cd^{2+} ions onto both treated and untreated biomass. For peroxide treated biomass, the maximum sorption efficiencies for both Cd^{2+} and Pb^{2+} ions were between 95.2 – 98.7% for *C.canephora*, 79.9 – 92.2% for *Musa spp.* and 42.0 – 91.3% for *C.papyrus* in non-competitive media and 90.8 – 98.0% for *C.canephora*, 56.4 – 89.3% for *Musa spp.* and 19.5 – 90.4% for *C.papyrus* in competitive media. The Langmuir model fitted non-competitive sorption data with $0.769 \leq R^2 \leq 0.999$ and the Freundlich model fitted competitive sorption data with $0.867 \leq R^2 \leq 0.989$. The pseudo second order kinetic model fitted the sorption data for Cd^{2+} and Pb^{2+} ions for untreated, peroxide treated and base treated biomass with $0.917 \leq R^2 \leq 1.000$. The sorption of trace metals was a complex potentially monolayer chemisorption with heterogeneous surface properties exhibited. In competitive sorption, sorption suppression effects observed were greater for Cd^{2+} than Pb^{2+} ions. The comparative studies on sorption performance presented agreement and no significant difference between the untreated and base treated biosorbents.

Key words: Biosorbents, cadmium, competitive uptake, Freundlich, pseudo-second-order kinetics, Langmuir, lead, remediation, risks, speciation, toxicity effects

1. INTRODUCTION

Uganda is a developing country with a huge potential for urbanization and industrial growth. Currently there is a high rate of growth of unplanned settlements, enterprises and cottage industries in upcoming urban centres. Consequently this socio-economic development has positively impacted on the country's resident peri-urban low income population. Kampala Capital City, the country's political and business hub also doubles as the national industrial heart, therefore, it takes the greatest share of industrial, infrastructural and demographic growth and the consequential environmental anthropogenic contamination.

The anthropogenic contamination of the environment is mainly due to the indiscriminate release of unsorted wastes, scraps, wastewater, sewage, domestic and agricultural wastes, as well as industrial effluent and byproducts into the wetlands, streams, channels and water catchment areas. Despite the existing environmental protection laws, there has not been a matched increase in concern, effort and measures to counteract the anthropogenic contamination and pollution of land, water resources and shallow groundwater in peri-urban areas. This implies that there is a serious threat to the proper functioning of the aquatic ecosystems, the social wellbeing, the economic and health status of the unsuspecting peri-urban residents.

The peri-urban residents mainly depend on the shallow groundwater sources for their daily water needs although due to intermittent water supply by the National Water and Sewerage Corporation (NWSC) even those with piped water connections on many occasions use water from the protected springs and wells. The quality of water from the peri-urban shallow groundwater sources has deteriorated as indicated by the studies undertaken on protected springs in Kampala Capital City. These studies have indicated widespread fecal contamination and high nitrate levels above recommended standards for drinking water (Howard, et al., 2003; KUSP, 2004; ARGOS, 2001; Kulabako, et al., 2007). Studies by Kulabako et al (2008) also showed that the quality of the shallow groundwater in peri-urban areas is a potentially major environmental problem. The main contaminants that have been studied are nutrients such as phosphorus and nitrates as well as microbial contaminants. However, there are scanty reports on trace metal contamination of shallow groundwater in the Lake Victoria basin, Uganda.

Only about 0.01% (200,000 km³) of all water on earth is usable freshwater for ecosystems and humans (Gleick, 1993). Freshwater is therefore, a meagre resource whose quality is very difficult and costly to reverse to pristine and usable condition when it deteriorates as outlined in Paper IV. Uganda is home to the source of the River Nile which is the longest river in the world that moves 6,850 km to the Mediterranean Sea from Lake Victoria. Lake Victoria is the second largest freshwater lake in the world with a

surface area of 68,800 km² and a volume of 2,760 km³ (Kayombo & Jorgensen, 2006). The Nile river basin covers almost 10% of the total area of the African continent with an area of 3100 km² (Ali, et al., 2003). There are eleven countries that make up the Nile River drainage basin, namely; Rwanda, Burundi, Democratic Republic of Congo, Uganda, South Sudan, Sudan, Kenya, Tanzania, Ethiopia, Eritrea and Egypt. Some of those riparian countries only have a small part of their area within the basin while others are located virtually entirely within the basin (Kayombo & Jorgensen, 2006). River Nile drainage basin serves the domestic, industrial and agricultural needs of over 150 million people. The Lake Victoria basin, Uganda forms a bigger part of the upper Nile basin that covers much of Uganda where it serves over 40 million people. There are socio-economic as well as industrial activities of the urbanizing region with consequential unprecedented anthropogenic contamination of the basin's surface water system which may eventually irreversibly impact the shallow aquifers as discussed in Paper I.

To conserve and protect the water environment, many countries around the world have environmental laws that require the treatment of contaminated wastes, industrial effluents and leachate before discharge into the environment. However, several types of contaminated waters such as surface runoff, landfill leachate, mine tailings, surface water in urban and industrial streams and others are hard to connect to the wastewater treatment systems due to challenges of sludge disposal, low levels of trace metals and transportation. .

2. BACKGROUND

2.1. The trace metals

Trace metals are those metals or metalloids or their compounds that are stable in the environment. They have densities above 4.5 g/cm³ (UNECE, 1979). Trace metals naturally exist in the earth's crust in minute quantities and as stable oxides, sulphides, carbonates, phosphates or hydroxides. The levels of trace metals in the environment increase due to natural or anthropogenic activities. The natural sources include; weathering and volcanic activity whereas anthropogenic sources include; agricultural activities, mining, processing and manufacturing industries, wastewater and waste dumpsite leachate among others. Industrialization, urbanization, exploitation of mineral resources and other anthropogenic activities that trigger a rise in trace metal contamination of the environment as highlighted in Paper II. The trace metal contaminants are divided into two groups; the essential metals have known biological roles while the non-essential metals have known toxicity to life processes. Trace metals such as cadmium, lead, mercury, copper and zinc are regarded as serious pollutants of aquatic ecosystems due to their environmental persistence, toxicity and ability to be incorporated into food chains (Kishe & Machiwa, 2003). The revelation of detrimental health

effects in humans occurs after acute exposure to high levels or chronic consumption of low concentrations of toxic trace metals. Plants usually adapt to toxic trace metals in polluted environments with little or no observable effects on growth and plant health presented but ordinarily animals express notable detrimental health effects after exposure to toxic trace elements as stated in Paper IV. In this study, in addition to other elements, the non-essential trace metals; cadmium and lead that are listed amongst the hazardous metals were studied. Non-essential trace metals can be detrimental to plant and animal life even in very minute concentrations in environmental matrices to which animals and plants are exposed.

2.1.1. Cadmium

Cadmium, Cd is a trace metal that mainly occurs in minute quantities in association with zinc ores. It is a byproduct of the smelting of zinc and copper. Cadmium is used as a protective layer in steel, in alloys as a stabilizer for plastics, in pigments for plastics, enamels and glazes as well as in Ni-Cd dry cells (Aylett, 1979, Wilson, 1988). The sources of soil pollution by cadmium are; motor vehicle tyres, brake pads, (Leitao, 2005), smelting of zinc, disposal of cadmium containing wastes, burning of plastics, batteries and fossil fuels as well as sewage sludge application (Hutton, 1982) to agricultural soils. The pollution of soils may impact on the soil water and eventually on the shallow groundwater that is abstracted for domestic use. Atmospheric deposition of aerosol particles is also a serious source of cadmium in industrialized countries. In studies on speciation of cadmium in soil solution, it was found that free ions of Cd^{2+} are predominated with neutral species CdSO_4 or CdCl_2 wherein the pH was greater than 6.5 (Tills and Alloway, 1975). Ingestion of food is the main route of entry of cadmium into the body for most people but only about 5%-10% gastrointestinal absorption occurs (Dobson, 1992). The other routes of exposure can be due to tobacco smoking and occupational exposure to CdO fumes and dusts.

Cadmium toxicity

Cadmium, Cd is a trace metal that mainly occurs in minute quantities in association with zinc ores. It is a byproduct of the smelting of zinc and copper. Cadmium is used as a protective layer in steel, in alloys as a stabilizer for plastics, in pigments for plastics, enamels and glazes as well as in Ni-Cd dry cells (Aylett, 1979, Wilson, 1988). The sources of soil pollution by cadmium are; motor vehicle tyres, brake pads, (Leitao, 2005), smelting of zinc, disposal of cadmium containing wastes, burning of plastics, batteries and fossil fuels as well as sewage sludge application (Hutton, 1982) to agricultural soils. The pollution of soils may impact on the soil water and eventually on the shallow groundwater that is abstracted for domestic use. Atmospheric deposition of aerosol particles is also a serious source of cadmium in industrialized countries. In studies on speciation of cadmium in soil solution, it was found that free ions of Cd^{2+} are predominated

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2.1.2. Lead

The most important sources of lead are igneous and metamorphic rocks. Lead oxides; PbO , Pb_3O_4 and PbO_2 are used in the lead-acid accumulator batteries. The lead-tin, Pb/Sn alloy is used as solder. Lead is used in motor vehicle tyres, brake pads, ball bearings, PVC pipes, glazed ceramics, lead pipes and sanitary ware. Lead oxides are used as constituents of many paints while the yellow chromate is used in road markings. Sources of soil pollution by lead are; smelting of lead, disposal of lead containing wastes such as old batteries, motor vehicle tyres, brake pads, solder droppings, bearings, PVC pipes, glazed ceramics, lead pipes and sanitary ware, fossil fuels, sewage sludge and fecal application (Leitao et al., 2000, Coker and Matthews, 1983) to agricultural soils. In the terrestrial environment, lead is strongly adsorbed to environmental components such as soil particles, organic matter and biota which reduce its availability to organisms although in acidic conditions lead solubilizes and soil solution lead may cause toxicity effects to plants and animals that depend on the water abstracted from the contaminated aquifers.

Lead plays no metabolic role and it shows toxicity effects to both plants and animals. The uptake of Pb into the body from the gastrointestinal tract is about 10% per day of the Pb content in ingested food and beverages (Kehoe, 1961). On the other hand, exposure to lead shows toxicity effects in both plants and animals.

Lead toxicity

Lead contamination is of great concern since even when the blood lead levels are below $80 \mu\text{g dL}^{-1}$ above which clinical symptoms of lead toxicity are manifested; it may cause mental impairment in children (Bearer, 2000). Lead toxicity has been linked to effects such as nervous system disorders (Needleman, 1991), anaemia, loss of appetite, high blood pressure, convulsions, damage to the brain, liver and kidney (Krishnani & Ayyapan, 2006), disorders in bone metabolism and cardiovascular disease. Both cadmium and lead interfere with the body's ability to regulate essential metals such as calcium, copper and zinc thence the need for their removal from the aqueous environment to ensure definite public health as outlined in Paper I.

2.2. Removal of trace metals from water

Leppert (1990), Namasivayam and Ranganathan (1995), Yang and Lin (1998) and Volesky (2001) have discussed several conventional techniques for remediation of trace metals in contaminated water including chemical precipitation, evaporation, electrochemical

treatment, ion exchange, electrodialysis, solvent extraction, adsorption, membrane separation flushing and fixation as well as reverse osmosis. Conventional remediation techniques have high installation and operation costs whereas they are inefficient and non-selective in removing trace metals in minute concentrations from contaminated surface water, surface runoff and landfill leachate as well as industrial and municipal effluents. Therefore, in the recent four decades, scholars have extensively and intensively studied low cost alternative methods of removal of trace metals from aqueous media using natural materials which are renewable, and environmental friendly as stated in Paper IV. Due to their abundance in nature biosorbents have been studied as low-cost sorbent materials for removal of trace metal contaminants from aqueous media. Biosorbents have shown high efficiency in sorption of trace metals even in minute concentrations and they present minimal chemical and biological contamination of the remediated solutions, they can be regenerated and have high desorption capacities (Pagnanelli, et al., 2003; Conrad, et al., 2007; Patnukao, et al., 2008; Guo, et al., 2008). Biosorbents have been studied for removal of aqueous trace metals from environmental samples in passive treatment systems for removal of trace metal contaminants in contaminated landfill leachate by Kalmykova, 2009, surface runoff by Nehrenheim, 2008, storm water by Färm, 2002 and mine tailings by Benavente, 2008.

2.3. Methods of remediation of contaminated water

There are several methods for remediation of trace metals contaminated water but the household water treatment is thought to be very cost effective and most reliable method of water treatment. Most efforts have been geared to reducing the costs and burden of the use of conventional water treatment technologies that involve chemical precipitation, use of ion exchange resins, adsorption, evaporation, electrodialysis, electroflotation, solvent extraction, membrane separation, reverse osmosis, (Leppert, 1990; Yang & Lin, 1998). The conventional remediation methods are not user friendly especially in the developing countries with a lot of upcoming industrial and urban facilities. Scientific studies aimed at fabricating low-cost adsorbents for large scale industrial applications took root almost three decades ago with a greater focus on adsorbents. Low-cost adsorbents are considered as such if they are abundant in nature, require little processing and are by-products or wastes from industry (Bailey et al., 1999). Adsorbents that have been extensively studied include clay minerals, carbon nanotubes, zeolites and biosorbents. The clay minerals were reported to be good, effective and more efficient in removal of trace metal ions from solution after modifications (Abollino et al., 2003; Unuabonah et al., 2008). The carbon nanotubes were suggested as adsorbents with superior sorption capacities and efficient adsorption and effective desorption of divalent heavy metal ions such as Cd^{2+} , Cu^{2+} , Ni^{2+} , Pb^{2+} and Zn^{2+} (Li, et al.,

2003; Rao, Lu, & Su, 2007). The zeolites were reported to have excellent ion-exchange capacities, high specific surface areas, and low cost (Chutai et al., 2009) qualifying them as alternatives in decreasing the mobility and toxicity of high valence heavy metal ions in the environment.

2.4. Biosorbents

Biosorbents are non-living biological materials of plant and animal origin that have been studied in unmodified and modified forms for remediation of contaminated aqueous solutions. Studies on biosorbents' from agricultural waste materials have shown that they are very efficient in adsorption of trace metals, have minimal chemical and biological contamination, require no nutrient additions, can be regenerated and have high desorption capacities (Conrad et al., 2007; Guo et al., 2008; Pagnanelli et al., 2003; Patnukao et al., 2008) as well as being abundant in nature all over the world. These agricultural wastes mainly contain lignocellulosic substances. The lignocellulosic substances including extractive polymers such as cellulose, pectin, protein, lignin and hemicellulose of which the polyphenols; tannin and lignin have been suggested as the active sites for trace metal ions adsorption (Maranon & Sastre, 1992). The lignocellulosic substances are very porous and have high free surface volume that allows accessibility (Rowell, 2006) of aqueous solutions to the biosorbent cell wall components contact. The mechanisms by which inorganic ions have been removed from aqueous media using adsorbents from wastes have been argued out by many filter technology scientists to include ion-exchange, chelation through bonding to oxygen, sulphur or nitrogen containing species at the surfaces such as COO^- , OH^- , NH_2^+ , SH , and others. The adsorption efficiency has been attributed to the biosorbents' large surface area and micro porosity.

2.4.1. Unmodified biosorbents

Unmodified biosorbents that have been studied and reported to be very efficient, effective, reusable and having minimal contamination of the treated water include among others; algal biomass (Lodeiro, et al., 2005; Aksu & Dönmez, 2006; Razaee, et al., 2006; Jha, et al., 2009; Somerville & Norrström, 2009; Dekhil, et al., 2011; Rajasimman & Murugaiyan, 2012), microbial biomass (Li, et al., 2009; Luo, et al., 2010; Gomes, et al., 2011), leaves (Babarinde, et al., 2006; King, et al., 2006; Hanafiah, et al., 2007; Sawalha, et al., 2007; Kumar & Gayathri, 2009), husks and hulls (Johnson, et al., 2002; Tarley, et al., 2004; Ahalya, et al., 2006; Bhattacharya, et al., 2006), peels (Amarasinghe & Williams, 2007; Kaewsam, et al., 2008; Feng, et al., 2011; Foo, et al., 2011), barks (Masri, et al., 1974; Horsfall Jr, et al., 2006) and sawdust (Bulut & Tez, 2003; Taty-Costodes, et al., 2003). Unmodified biosorbents from tree barks, peels, stems and husks have been studied in batch and reported in Papers II, III and IV. In several studies involving unmodified biosorbents, both good adsorptive capacities for trace

metals and regeneration of the biosorbent materials in desorption studies have been reported in literature. These characteristics give biosorbent remediation techniques an edge over conventional remediation techniques where spent adsorption materials' disposal poses environmental challenges. Gaballah et al. (1997) indicated that application of unmodified plant wastes in water treatment would alter the pH, increase the chemical oxygen demand (COD), biological oxygen demand (BOD) and dissolved organic carbon (DOC) of the resultant filtrate. Unmodified biosorbents of plant origin contain high content of tannin, lignin, polysaccharides cellulose, humic and fulvic acids, therefore, they tend to release soluble organics which impacts on the remediated water quality when used to remediate contaminated aqueous media. Soluble organic matter has both advantages and disadvantages; they form complexes with metallic ions in the liquid phase which are less toxic than the free ions but also increase the mobility of metals in the environment. In spite of the importance of dissolved organics in biosorption, only few studies have considered their effects in the liquid phase although Gaballah, et al., (1997) and Bailey, et al., (1999) suggested that association of soluble organics and metal ions leads to formation of complex species. Studies that include the release and effects of soluble organics in treated aqueous media are limited. The soluble organics are extractable with chemicals such as hot water, sodium hydroxide; diethyl ether and ethanol (Browning, 1967; Rowell, 2006). Wan Ngah and Hanafiah, (2008) and Rivera-Utrilla et al., (2011) have reported that although unmodified biosorbents in several studies showed good sorption capacities, chemically modified materials presented enhanced sorption capacities for trace metal ions in several studies.

2.4.2. Modified biosorbents

Several chemical and physical modification methods have been used to improve the sorption properties of the biosorbents to varying degrees. The polar solvents such as water and ethanol dissolve polar substances from the biosorbents such as the phenolics and tannins whereas the non-polar solvents such as diethyl ether dissolve the non-polar substances such as fats and waxes. In other studies biosorbents have been modified using base solutions; sodium hydroxide, calcium hydroxide and sodium carbonate (Min, et al., 2004; Kumar & Bandyopadhyay, 2006; Memon, et al., 2007), oxidizing agents; hydrogen peroxide, methanol, sodium chlorate and nitric acid (Ganji, et al., 2005; Horsfall Jr. & Spiff, 2005), water (Rowell, 2006) and, organic and mineral acids; ethanoic acid, phosphoric acid, nitric acid and sulphuric acid (Bulut & Tez, 2003; Taty-Costodes, et al., 2003; Wong, et al., 2003; Horsfall Jr, et al., 2006; Li, et al., 2006; Özer & Pirincci, 2006) to enhance adsorption capacities for trace metal ions as well as to reduce soluble organics transfer to the liquid phase. Sodium hydroxide and hydrogen peroxide have been used

in modifying biosorbents for remediation of trace metals in batch studies and have been reported in Papers II, III and IV.

2.5. Biosorption mechanisms

Biosorbents remove contaminants from the aqueous environment through three mechanisms; cation-exchange, absorption and adsorption. Together the three mechanisms constitute a process generally referred to as the biosorption process. The biosorption process involves a biosorbent and a solvent containing the sorbate which is also the contaminant to be removed. During the interaction between the biosorbent and the contaminated solution, the sorbate species are attracted to the biosorbent surface structures and are removed through the aforesaid mechanisms. The process continues until a steady state is established between the sorbate species in solution and those bound to the biosorbent. The distribution of sorbate species between the liquid and solid phases is governed by factors including; temperature, pH, time of interaction, concentration of the contaminant ions in solution, competing metallic ions, nature of the biosorbent surface structure, biosorbent size, biosorbent dosage and components of the biosorbent surface structures. The factors mentioned above have been studied and reported in several studies. Biosorbents have got exchangeable cations; Na^+ , Ca^{2+} , K^+ and Mg^{2+} ions that are thought to contribute to the uptake of contaminant metallic ions through ion exchange between the biosorbent and the aqueous solution. Absorption occurs when metal ions get entrapped in the biosorbent pores within the structure by oxygen, nitrogen and other electron rich species whereas adsorption pertains to the surface of the biosorbent which is governed by the presence of electron rich species and exchangeable cations. On the other hand the biosorbent surface oxygen groups as well as other electron rich species which are potential metal binding groups; tannin, lignin, polysaccharides cellulose, humic and fulvic acids, are responsible for binding the metallic ions during the remediation process. Maranon and Demirbas (1992), Chen and Wu (2004) as well as Zhao et al. (2005) have attributed the efficiency and effectiveness of biosorbents in removing trace metal ions from solution to the existence of functional groups such as carboxyl, hydroxyl and amino groups and aromatic species among the structural components that present active binding sites for trace metal ions besides ion exchange. In addition to sorption the aqueous metallic ions concentration tend to bind to the released soluble organics; dissolved organic matter which can be precipitated.

2.6. Competitive and non-competitive uptake

Most metal ions sorption studies that have been reported are non-competitive which leaves a scientific validation gap in the search for alternative remediation technologies for field scale applications. Competitive metal sorption studies give sorption intensities that nearly complement environmental conditions than the non-

competitive metal ions sorption results. The sorption intensities for metal ions in competitive sorption studies in comparison to those for metal ions in non-competitive sorption studies from selected studies indicated that the sorption intensities and sorption efficiency of divalent metal ions were affected by the presence of other metal ions. Various biosorbents showed reductions in multi-component solutions as well as natural water samples due to the presence of competing divalent ions as opposed to no effect for mono-valent ions.

In a study by Sandau, et al. (1996), the sorption efficiency for cadmium ions onto *Fucus vesiculosus* and algal biomass reduced in the presence of calcium by 3% and for magnesium by 6% but no change was observed in the presence of potassium and sodium. Aksu and Dönmez (2006) reported 18 – 30% suppression in binary component solutions as compared to single component solutions for sorption efficiency of cadmium and nickel onto *Chorella vulgaris* whereas Ahmed (2010) reported reduction in uptake of copper, cadmium and zinc by 3 – 5% onto Date palm waste in binary components solutions. In the report about competitive sorption of trace metals onto *Aspergillus niger* by Kapoor et. Al. (1999) it was indicated that the sorption reduction for lead was 25% and 52% in the presence of cadmium and copper, 52% and 84% for cadmium in the presence of lead and copper and 50% and 31% for copper in the presence of lead and cadmium correspondingly.

The effect of reduced metal ions sorption intensities increases in the order of single < binary < ternary < natural aqueous solutions. In these studies the suppression of cadmium and lead uptake onto *Coffea canephora* and *Musa spp.* were reported in Paper IV. Therefore, biosorption suppression and complementary effects on sorption of different metals onto biosorbents occurs during remediation of natural and multi-component contaminated media.

2.7. Aim

The aim of this study was to characterize and evaluate the potential of low-cost agricultural wastes; *Albizia coriaria*, *Coffea canephora*, *Cyperus papyrus*, *Erythrina abyssinica* and *Musa spp* with emphasis on *Coffea canephora* husks and *Musa spp* peels as filter media in removing Cd^{2+} and Pb^{2+} ions from trace metals contaminated water.

The specific objectives of the study were to:

- i. Evaluate the levels of trace metal contaminants, establish the speciation of trace metals and the assessment of eminent toxicity effects of trace metals on aquatic organisms in the aqueous environment; shallow groundwater, surface run-off, surface water and wastewater in selected sites in the Lake Victoria basin, Uganda (Paper I).
- ii. Investigate the ability of plant wastes; *Musa spp* peels, the barks of *Albizia coriaria*, and *Erythrina abyssinica* to uptake trace metal ions Cd^{2+} and Pb^{2+} ions from spiked aqueous media under

- batch conditions. The effects of initial pH and agitation time on trace metal ions sorption were evaluated for untreated and pretreated biosorbents (Paper II). Examine and characterise the sorption intensities through models for varying initial trace metal concentrations at constant pH and agitation time (Paper III).
- iii. Investigate the efficacy of *Coffea canephora* husks, *Cyperus papyrus* and *Musa spp* peels in removing trace metals from contaminated water through the evaluation of the kinetics and the equilibrium isotherms of the competitive and non-competitive sorption of Cd^{2+} and Pb^{2+} ions in batch. The total ionic content dissolved organic carbon (DOC), Cation Exchange Capacity (CEC) and pH of the biomass were evaluated and compared to the sorption efficiencies and capacities. Evaluation of the effects of solution pH, agitation time, initial metal ions concentration and competition on the sorption of Cd^{2+} and Pb^{2+} ions were studied and the experimental data was fitted to kinetic and equilibrium isotherm models to predict the sorption dynamics and mechanisms (Paper IV).

2.8. Scope

2.8.1. Study area and sampling sites

The study area is the Lake Victoria basin, Uganda with sampling points for surface water along streams and water channels, some of which are shown on the map (a) in Figure 1. The streams and leachate channels are associated with Lake Victoria basin. The protected springs and associated surface runoff sampling points are located in Kampala City and some of them have been shown on the map (b) in Figure 1. The choice of the sampling points in the study area was based on the resident population density and anthropogenic activities in the immediate vicinities which was predictive of environmental contamination.

The protected springs and associated surface runoff sampling points are located in Kampala Capital City. Figure 3 gives a brief state of affairs at protected springs in Kampala Capital City where contaminated surface water may eventually impact shallow ground water due to contamination from unconventional dumping of unsorted wastes, leachates and effluents in the catchment areas. The choice of the sampling points in the study area was random but based on the resident population density, demand for water from the springs and anthropogenic activities in the immediate vicinities which was predictive of contamination.

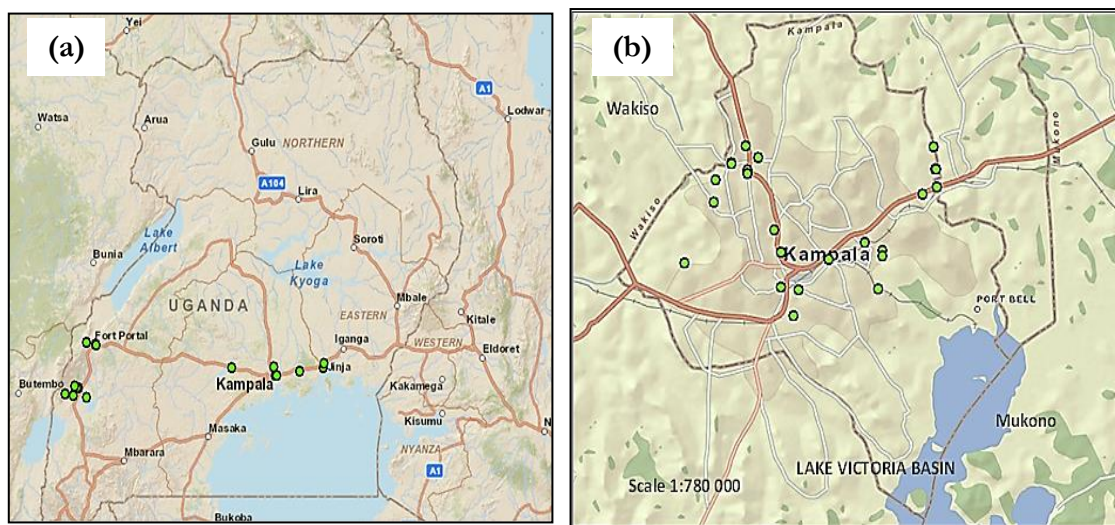


Figure 1. Map of (a) Uganda showing the sampling points for water and map of (b) Kampala city showing shallow groundwater and surface runoff sampling points.

2.8.2. Water sampling

The shallow groundwater from protected springs was drawn as it flowed from shallow aquifers. The sampling of shallow groundwater was done once a year from March, 2012 – February, 2015. Grab samples of surface runoff in the catchment area of the protected springs were picked during heavy rainstorms during September – October, 2013. The surface water and leachate samples were picked at points 1m from the peripheral at depths within 0.5m of the surface whereas for Lake Katwe it was done at approximately 100m from the shores at selected sites.

For streams, the sampling was done twice from June, 2014 – February, 2015. The plastic bottles used for collection of water samples were cleaned and rinsed twice with the water to be sampled before use. Field measurements of pH, temperature and electrical conductivity (EC) were measured using a pH ION meter. The water samples for determination of DOC were filtered into sample bottles using 0.2µm filters and cooled before being transferred to the laboratory. The water samples were sub-sampled twice; one sample was filtered and cooled while the sample for trace metals analysis was filtered and acidified with concentrated nitric acid before being transferred to the analytical laboratory at room temperature (25°C).

2.8.3. Metals, anions and DOC analyses

In this study the trace metals that were studied were lead and cadmium. The choice of these metals was based on earlier environmental studies during cycle 1 and 2 at Makerere University as well as studies by other scholars that showed trace metals prevalence in the environmental samples (Ljung, 2001; Larsson, 2002; Muwanga & Balifaijo, 2006; Nyangababo, Henry, &

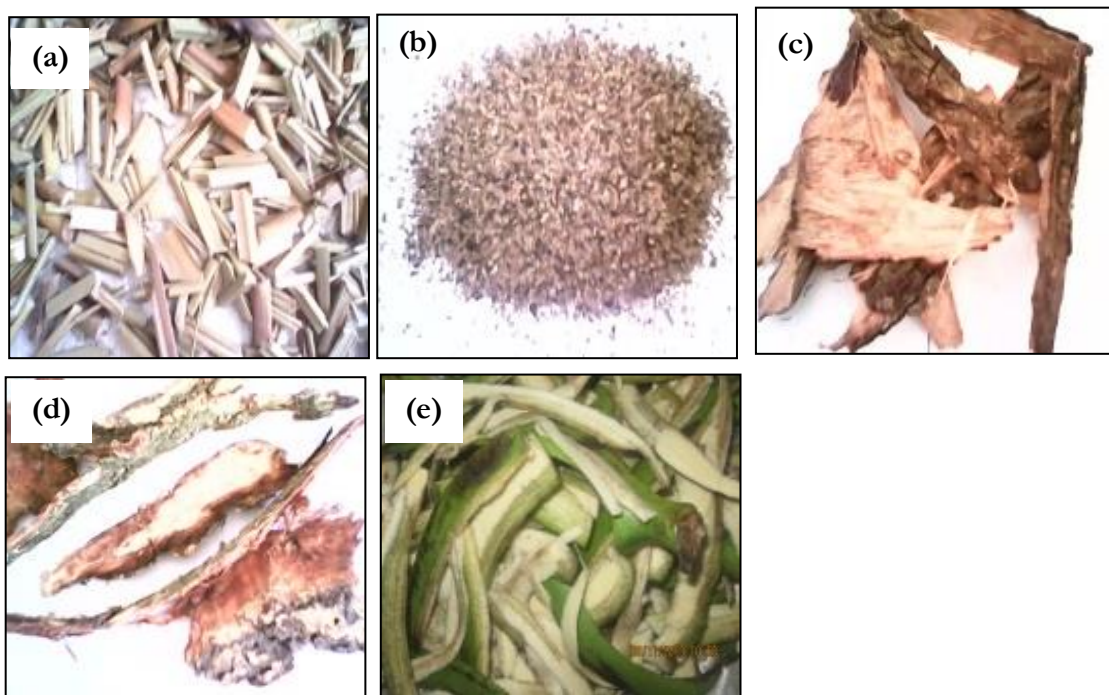


Figure 2. The plant materials used in these studies to prepare biosorbents; (a) *Cyprus papyrus*, (b) *Coffea Canephola*, (c) *Erythrina abyssinica*, (d) *Albizia coriaria* and (e) *Musa spp*

Omutunge, 2005). The acidified water supernatants were analysed for trace and major elements using an ICP-MS, ICP-EOS, and AAS whereas the samples for DOC and anions analysis were evaluated using the Shimadzu TOC-5000 without acidification. The leachate samples were digested using the wet acid digestion procedure. Pre-treatment of 50mL of leachate was done with 2 mL nitric acid and 5 drops of 30% hydrogen peroxide added in a closed bottle and placed in a constant temperature water bath at 60°C for 1 hour. The clear supernatant were filtered through 0.45µm filters into plastic sample bottles and analysed for cations whereas the samples for DOC and anions analysis were only filtered before analysis. The analyses were done over a period of three years and mean values of the measurements have been reported. The analyses were done over a period of three years and mean values of the measurements have been reported. The major anions; Cl^- , NO_3^- and SO_4^{2-} were analysed at KTH, Stockholm using a Dionex DX-120 ion chromatograph. (Paper I).

2.8.4. Metals Speciation, bioavailability and risk assessment

The sorption effectiveness and efficiency of uptake of trace metals by biosorbents depends on the speciation of Pb^{2+} and Cd^{2+} ions in a heterogeneous solution. The chemical species distribution in organic and inorganic phases would impact on the sorption process since the metal ions in free ionic state and inorganic state would be available for sorption and would also be important for toxicity. Pb^{2+} and Cd^{2+} ions speciation in selected contaminated surface water and leachate at various conditions of DOC and pH

was performed with Visual MINTEQ 3.1 which was originally coded by the U.S. Environmental Protection Agency (EPA) and has been further developed by Gustafsson, 2013. For the speciation studies of the metal ions in selected surface water, the Stockholm Humic Model (SHM) was used. The assumption used by US EPA to set Water Quality Criteria was which considered all DOC in natural waters to be active and that organic matter consists of 90% fulvic acid and 10% humic acid was applied. Toxicity studies were done by assessing the environmental risks due to trace elements through modelling using the Bio-met software tool. The Bio-met software tool version 4 is based on calculations from Biotic Ligand Models (BLM) to estimate the potential risk to the aquatic environment posed by copper, nickel, zinc, and lead after considering bioavailability as stated in Paper I.

2.8.5. The choice of biomass studied

The biomass studied (Figure 2) was chosen from that which normally has little or no specific reuse such as *Musa spp.* peels, *C. canephora* husks, *C. papyrus* cuttings and the barks of *E. abyssinica* and *A. coriaria*. The plant wastes accumulate as an urban waste burden after the useful products such as timber, food and herbal medicine have been removed. Although waste plant materials are quite varied in chemical structure, they have attracted a lot of research for application as low cost biosorbents for remediation of trace metals in contaminated aqueous media. Plant materials; peels of *Musa spp* (cooking bananas), *Cyperus papyrus* cuttings, husks of *Coffea canephora*, the barks of *Erythrina abyssinica* and *Albizia coriaria* shown in Figure 2 were sampled from a rural farming district in Uganda and used in this study to prepare biosorbents.

In 2011 Uganda was ranked as the second largest producer of bananas in the world with 11.1 million tonnes (Wikipedia contributors, 2014). Uganda is not amongst the renowned exporters because in most parts of the country, bananas are an important staple food. The bananas cultivated in Uganda are the East African highland bananas (matooke), the cooking bananas which are scientifically referred to as *Musa spp*. When the cooking banana fruit is harvested just before ripening, the banana fingers are peeled and cooked for food; Matooke.

The banana peels are used as fresh animal feeds or as compost manure in farms although they have little suitable reuse in urban centres where they form part of the urban waste burden for municipal authorities.

For the period 2012 – 2014, Uganda was ranked as the second producing country of *Coffea canephora* in the world with 294,000 tonnes of *Coffea canephora* exports (International Coffee Organization, 2014) and it is the country's biggest agricultural export. Dry *C. canephora* is partially processed before the coffee beans are exported. Husks of *C. canephora* are common by-products of coffee processing industries in the country.



Figure 3. Current state of affairs at selected protected springs (a) Kawempe (b) Bwaise (c) Katoogo and (d) Banda in Kampala Capital City, Uganda

The husks are recycled as poultry farmhouse fills, as raw materials for compost manure and as fuel for firing clay works kilns. The industrial and agricultural consumption of *C. canephora* husks by reapplication does not surpass its production, hence the waste burden. On the other hand, *C. papyrus* is a wetland plant with tall stems commonly found in most swamps in Uganda which is used in the local craft industry for making mats, carpets, sanitary towels, shutters and shelters although the stem-cuttings by-products have no reuse. *E. abyssinica* and *A. coriaria* are common tropical tree species that are grown for wood fuel and herbal medicine.

When *E. abyssinica* and *A. coriaria* trees are harvested for wood and timber, waste bark materials whose extracts by local herbalists are used as local dermatological medications have little reuse elsewhere. Whereas *C. papyrus* reeds are harvested for making crafts, *C. canephora* husks are threshed before exporting the beans, and *Musa spp* peels are removed before cooking the bananas for consumption. In all the cases, the plant wastes contribute to the waste burden of urban areas. The waste plant materials were, therefore, chosen for study as biosorbents because of their abundance, low cost and limited reapplication in both production and consumer industry in the country. In Figure 2, the raw biomass

of *C. papyrus*, *C. canephora*, *A. coriaria*, *E. abyssinica* and *Musa spp* are shown.

2.9. Structure of the thesis

This thesis is divided into two main parts, a cover essay and the appended papers. An outline of both these parts is provided below.

Chapter 1, The Introduction chapter briefly outlines the baseline information about the study.

Chapter 2, The Background to the study describes highlights the literature on Lake Victoria basin, surface and groundwater, trace metals, biosorbents and biosorption mechanisms.

The scope of the study describes and discusses the study area, samples and sampling, trace metals, speciation, bioavailability, risk assessment and choice of biosorbents.

Chapter 3, Materials and Methods chapter, presents the water quality, laboratory experiments and data analyses that were done.

Chapter 4, Results chapter, consists of the presentation of data from measurements, experimental work, risk assessment, kinetic and dynamic modelling and statistical treatment of data

Chapter 5, Discussion section, deals with the elaborate arguments of the results and findings of the study in relation to earlier studies.

Chapter 6, Conclusion chapter, gives the deductions, implications and recommendations based on the results of the study.

The second part of the thesis comprises Papers I, II, III and IV which are briefly summarised below:

Paper I; the title of the paper was, “Assessment of levels, speciation and toxicity of trace metal contaminants in selected shallow groundwater sources, surface runoff, wastewater and surface water from designated streams in Lake Victoria Basin, Uganda” argues that the shallow groundwater had elevated levels of metal ions above which remediation action is implied. The surface water contamination varied with sample site but revealed increased risks to environmental health whereas speciation studies showed that metal ions were mainly bound to organic matter.

Paper II; Kinetic studies of Cd (II) and Pb (II) ions biosorption from aqueous media using untreated and chemically treated biosorbents. The paper develops that *A. coriaria*, *E. abyssinica* and *Musa spp.* biomass in treated and untreated forms showed very good sorption intensities for aqueous cadmium and lead under different experimental conditions. The results indicated the biosorption mechanism to be mainly chemisorption.

Paper III; Characterisation and application of un-treated and base-treated biosorbents from *Albizia coriaria*, *Erythrina abyssinica* and *Musa spp.* in the uptake of Cd (II) and Pb (II) ions from contaminated water.

The biosorbents contained electron rich surface organic species capable complexing metal ions during the ions uptake. The



Figure 4. (a) Sieving of *coffea canephora* husks (b) drying of biomass in the oven (c) determination of pH in the lab and (d) XRD machine used in the study

biomass negative potential for binding base cations was high. The treatment of biosorbents reduced the levels of DOC leaching into remediated solutions. Equilibrium studies showed that the sorption processes differed with complex sorption chemistry and mechanisms for metal ions uptake.

Paper IV; Competitive and Noncompetitive Batch Sorption Studies of Aqueous Cd (II) and Pb (II) Uptake onto *Coffea canephora* Husks, *Cyperus papyrus* Stems, and *Musa spp.* the kinetics and dynamic studies revealed that competitive sorption efficiencies of all biosorbents were lower than the non-competitive sorption efficiencies which showed sorption suppression effects on the sorption of both metal ions. The sorption kinetics for both metal ions adequately followed the pseudo-second-order model which predicted chemisorption as the main reaction mechanism of the sorption process. The Langmuir isotherm fitted the non-competitive sorption data whereas the Freundlich isotherm fitted the competitive sorption data better which inferred that the sorption process was complex and involved several mechanisms.

3. MATERIALS AND METHODS

3.1. Laboratory preparations of reagents, biosorbents and biosorbates

All the chemicals used in this study were of analytical reagent grade and obtained from J.T. Baker H, Deventer, Holland. Contaminated aqueous media (biosorbates) were prepared using water purified by reverse osmosis and standard solutions of lead and cadmium containing 1000 mgL^{-1} of trace metal. The water used to for the reagents was prepared by the reverse osmosis system (Sartorius Arium 61315, Elga Model, Germany) and purified using the Pure lab Utra, Model: Elga Utra Analytic (UK). The pH meter (Orion model: 210A) was used to measure pH values in the aqueous phase. The initial pH of the contaminated aqueous solution was adjusted to the required value using drops of 0.1 M HCl and 0.1 M NaOH (Papers II, III and IV).

The plant materials were thoroughly washed with distilled water to remove particulate matter and dried in an oven (Figure 4, b) (E series: ED – E2 Binder, Germany) at 60°C for 72 hours to a constant weight. The dry biosorbent materials were hammer-milled and sieved to 2 mm (Figure 4, a). The powder was stored in air-tight self-sealing plastic bags. Each of the biosorbent materials was divided into two portions; one was used in batch studies unmodified while the second was chemically pre-modified.

Chemical treatment methods using hydrogen peroxide (Ganji, et al., 2005) and sodium hydroxide (Tarley, et al., 2004) were modified and employed. In the first modification, 100 g of the biosorbent was soaked in 0.25 M NaOH for 2 hours and neutralized using 0.25 M HCl to $\text{pH } 7.0 \pm 0.2$. The modified portion was generously washed with distilled water, dried in an oven at 105°C for 24 hours and stored dry in sealed plastic bags (Papers II and III).

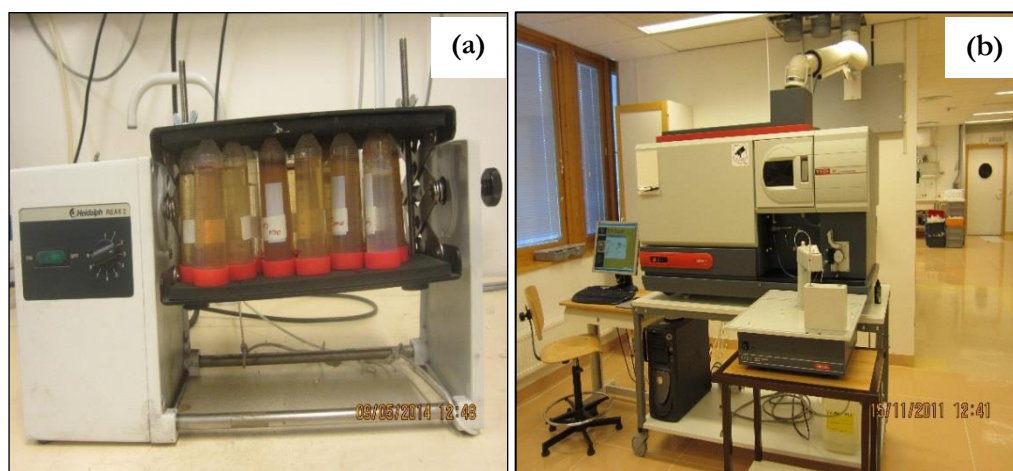


Figure 5. (a) The rotary shaker used in batch studies and (b) The ICP-EOS used to analyse for trace metals

In the second modification, 100 g of each plant material were soaked in a 250 mL of 20 % H_2O_2 solution for 24 hours. The materials were then removed from the peroxide solution and thoroughly washed with distilled deionized water and immersed in 0.25 M NaOH for 2 hours. The biosorbents were removed and excess NaOH was neutralized using 0.25 M HCl to $\text{pH } 7.0 \pm 0.2$. The modified biosorbent was generously washed with distilled water and oven dried at 105°C for 48 hours then stored in sealed plastic bags (Paper IV).

3.2. Determination of pH of the biomass

One gram of biomass was mixed with 20 mL of distilled deionized water and shaken in a closed centrifuge bottle for 25 minutes. The bottle and contents were centrifuged at 200 rpm for 15 minutes after which 15 mL of the clear supernatant were transferred to a sample bottle and the pH determined using a pH/ION meter model pHM95 with sensitivity 97.9% (Figure 4, c).

3.3. Cation Exchange Capacity

For each biomass 1.5 g and 100 mL of 0.1M BaCl_2 were placed in plastic bottles and capped. The bottles were shaken with a rotary shaker for 6 hours and clear extracts were filtered into clean bottles using coarse filters. To 15 mL of sample solutions, 35 mL of distilled water were added and the exchangeable acidity measured using an auto-titrator by successively adding 0.01M NaOH until pH 7.0 was attained. Another 20 mL of sample was filtered through $0.45\mu\text{m}$ filters for metal ions analyses. The experiments were done in triplicate and the mean values used to calculate the cation exchange capacity (CEC) (Paper IV).

3.4. X-ray diffraction (XRD) analyses

The identification of chemical composition and crystallographic structures of surface chemical species in untreated biosorbents was achieved by X-ray diffraction analysis using the X Ray Diffractometer Siemens D5000 machine (Figure 4, d).

The biomass samples were crushed in a ceramic mortar to fine powder of diameter $\leq 1\text{mm}$ and consolidated into aluminium holders. The samples were scanned at 35kV and 40mA from 10° to 80° the diffraction interval with a step size of 0.01° and scan duration of 0.5 seconds for exposure time 20 of 30 minutes for each sample. The diffraction patterns were analysed using the DIFFRAC Plus software and plotted using Microsoft Excel. (Papers III and IV).

3.5. Nitric acid digestion

Nitric acid digestion procedure was used to prepare biomass for total metal constituents' analysis. One gram of dry biomass was placed in a 250 mL digestion tube and 10 mL of concentrated HNO_3 acid was added. The mixture was heated at 100°C for 2 hours and then treated with hydrogen peroxide. After digestion the clear supernatant was diluted using distilled deionized water to 50

mL. The solution was filtered through 0.20 μm sterile non-pyrogenic hydrophilic filters into plastic sample bottles and analyzed for metal ions using an Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) (Varian Vista AX model).

3.6. Sorption experiments

Batch experiments were chosen and executed in order to evaluate the biosorbent efficiency, efficacy, optimal experimental conditions, biosorption rate and possibility of biomass regeneration and reuse. Kinetic studies were done in two separate sets of closed 50 mL bottles at $24 \pm 1^\circ\text{C}$. Agitation was done at 60 rpm for 3.5 hours using a rotary shaker shown in Figure 5, a and the samples were then centrifuged at 2000 rpm for 25 minutes. A portion of the equilibrated solution was then filtered through 0.45 μm and 0.20 μm sterile non-pyrogenic hydrophilic filters into polypropylene sample bottles to which 0.5 mL HNO_3 was added and trace metal ions analysed using Inductively Coupled Plasma-Mass Spectrometry (ICP-MS). All experiments in the first set were done in three replicas for both metal ions and all biosorbents; therefore, the results are average values of the replicas.

In the first set, studies were done in triplicate with 10mgL^{-1} of biosorbent agitated with 50 mL of aqueous solutions containing 20mgL^{-1} of either Pb^{2+} or Cd^{2+} ions. The pH range was 2.0 to 9.0 for agitation time, 0.5 – 6.5 hours (Paper II).

In the second set, studies were carried out in duplicate with 12.5gL^{-1} sorbent dose and 20 mL of contaminated solution containing 20mg L^{-1} of trace metal ions in 50 mL plastic bottles. The pH range was from 2.5 to 6.5 and a biosorbent dose of 12.5gL^{-1} (Paper III).

Isotherm studies were done in duplicate in the third set. Studies were carried out with 12.5gL^{-1} biosorbent dose and 20 mL of synthetic solution containing 20mg L^{-1} of trace metal ions in 50 mL plastic bottles at $24 \pm 2^\circ\text{C}$. The agitation was done at 60 rpm for 3.5 hours using a rotary shaker. The isotherm studies were done at pH 4.5 for metal ion concentrations in the range of 5 to 100mgL^{-1} . For studies on co-ion effects of trace metals, experiments were done with increasing mixed metal concentrations in equal proportions of 5 – 100 mg L^{-1} and . The effects of increasing the initial concentration of Cd^{2+} ions while maintaining that of Pb^{2+} ions were studied for competitive biosorption. The equilibrated solutions were centrifuged at 2000 rpm for 25 minutes and then filtered through 0.20 μm sterile non-pyrogenic hydrophilic filters into polypropylene bottles with a drop of concentrated HNO_3 as described in our earlier studies in Papers II and IV.

3.7. Determination of residual metal ions and dissolved organics

The filtrates that were analysed for trace metals were acidified using 0.5 mL HNO_3 . Residual metal concentrations in the filtrates were determined with Inductively Coupled Plasma-Mass

Spectrometry (ICP-MS) (Varian Vista AX model) and Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES) (ICAP 6300 Series) (Figure 5, b). Total Inorganic Carbon (TIC) and Dis-solved Organic Carbon (DOC) were determined using a TOC analyser (Shimadzu TOC-5000A). The experiments were done in duplicates unless otherwise stated to ensure that the experimental data showed consistence.

3.8. Data analysis

The quality of a biosorbent material depends on the amount of sorbate species removed from solution and retained by the biosorbent. The units of biosorption intensity, q_e used were milligrams of sorbate biosorbed per gram of the dry biosorbent material which is the basis for engineering process–mass balance calculations. The quantity q_e (mg/g) was determined according to equation (1),

$$q_e = (C_0 - C_e)V/m \quad (1)$$

The biosorption efficiency, of the metal ions, it was calculated according to equation (2),

$$E = (C_0 - C_e)/C_0 \times 100\% \quad (2)$$

where,

C_0 (mg L⁻¹) is the initial metal ion concentration in solution

C_e (mg L⁻¹) is the equilibrium concentration of metal ions in the filtrate

V (L) is the volume of solution agitated

m (g) is the mass of the biosorbent material

3.9. Kinetic modelling of biosorption

Experimental data of the time-dependent sorption of Cd²⁺ and Pb²⁺ ions were fitted to kinetic models to understand the sorption speed and predict the probable sorption mechanisms. The pseudo-first order and pseudo-second order kinetic models were applied to the experimental data obtained in order to predict the biosorption mechanisms for Cd²⁺ and Pb²⁺ ions. The Lagergren (pseudo-first order) model and the Ho (pseudo-second order) model are the most widely used models in sorption studies to predict the mechanisms involved in the biosorption processes (Ho & McKay, 1998). The pseudo-first order kinetic model (Lagergren, 1898), can be expressed as shown in equation (3);

$$\frac{dq_t}{dt} = k_1(q_e - q_t) \quad (3)$$

where,

q_t (mg/g) is the biosorption intensity at time t

q_e (mg/g) is the biosorption intensity at equilibrium

k_1 (min^{-1}) is the rate constant of the first-order biosorption. Integrating Eq. (3) and applying boundary conditions $q_t = 0$ at $t = 0$, and $q_t = q_t$ at $t = t$, the resultant equation is the linear form of the pseudo-first-order kinetic model expressed as shown in equation (4);

$$\ln(q_e - q_t) = \ln q_e - k_1 t \quad (4)$$

The pseudo-second-order kinetic model or Ho model (Ho & McKay, 1998) can be expressed as equation (5) shows;

$$\frac{dq_t}{dt} = k_2 (q_e - q_t)^2 \quad (5)$$

where, k_2 (g/mg/min) is the pseudo-second-order rate,

Integrating Eq. (5) and applying boundary conditions $q_t = 0$ at $t = 0$, and $q_t = q_t$ at $t = t$, the resultant equation, the pseudo-second-order kinetic model can be rearranged as equation (6);

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (6)$$

3.1. Isotherm models

The isotherm models that were used to fit data in this study were the three parameter isotherm models; Langmuir, Freundlich and Temkin models, which are commonly used in sorption studies.

The Langmuir isotherm model (Langmuir, 1916) is based on three assumptions; namely biosorption is a monolayer process, all biosorbent sites are similar and can only accommodate one sorbing constituent, and the capacity of a site to be occupied by the sorbate is independent of the nature of occupancy of sites in its vicinity. The linearized form of Langmuir isotherm is represented as in equation (7),

$$\frac{C_e}{q_e} = \frac{1}{b q_{\max}} + \frac{C_e}{q_{\max}} \quad (7)$$

where,

q_{\max} is the monolayer sorption intensity (mg/g),

b is the biosorption constant (L/mg)

A plot of $1/q_e$ versus $1/C_e$ gives a straight line with slope of $1/bq_{\max}$ and an intercept of $1/q_{\max}$ if the biosorption follows the Langmuir equation. The Langmuir isotherm is the most widely applied model.

The Freundlich isotherm (Freundlich, 1906) model is an empirical isotherm which suites biosorption onto heterogeneous surfaces. It is based on multilayer biosorption process which implies that the biosorbent surface is heterogeneous and the biosorption is due to non-uniform distribution of heat of sorption. The linearized Freundlich isotherm is described by the following equation (8),

$$\ln q_e = \ln(A_F) + b_F \ln C_e \quad (8)$$

where,

C_e is equilibrium metal concentration (mg/L),

A_F and b_F are the isotherm empirical constants

A_F the value of q when $\ln C_e = 0$ for the Freundlich isotherm. The value of $1/b_F$ has to do with site heterogeneity. A value of $1/b_F$ greater than unity would indicate heterogeneous site-binding affinity for the trace metal ions by the biosorbent.

The other isotherm model considered in this study was the Temkin model. The derivation of Temkin isotherm assumes that the fall in the heat of sorption is linear rather than logarithmic, as implied in the Freundlich equation (Aharoni & Ungarish, 1977). The linearized form of Temkin isotherm is defined by the equation (9),

$$q_e = \frac{R.T}{b_T} \ln(C_e) + \frac{R.T}{b_T} \ln(A_T) \quad (9)$$

where,

T is the temperature (K),

R is the universal gas constant, 8.314 Jmol^{-1} ,

A_T is the equilibrium constant (L mg^{-1})

b_T is related to heat of biosorption, (Jmol^{-1})

When a particular equilibrium model fits the sorption data, then the model assumptions, predictions of the biosorption mechanism and the underlying assumptions are dependable for description of the sorbate biosorption reaction, sorbent surface properties and the sorbent-sorbate interactions during the process.

3.1. Biotic ligand Model

The Bio-met software tool version 4 is based on calculations from Biotic Ligand Models (BLM) to estimate the potential risk to the aquatic environment posed by copper, nickel, zinc and lead after considering bioavailability. The basis of biotic ligand modelling is that metal accumulation at the “biotic ligand” is proportional to toxicity and that this accumulation can be predicted by solving the appropriate simultaneous equilibria among the biotic ligand(s) and the dissolved components (aqueous ligands and competing cations) in the exposure water (Bio-met, 2013). Environmental risks due to trace metals were studied using the Swedish environmental protection agency criteria and the bioavailability based approach through simulations using the biotic ligand model, Bio-met software tool that is applied over a range of different organisms to calculate the 5th percentile of the species sensitivity distribution (SSD) as discussed in Paper I.

Presented in Figure 6, is the remodified schematic diagram of the biotic ligand model (BLM) that explains the assumptions and interactions between the inorganic, organic and biotic ligands in

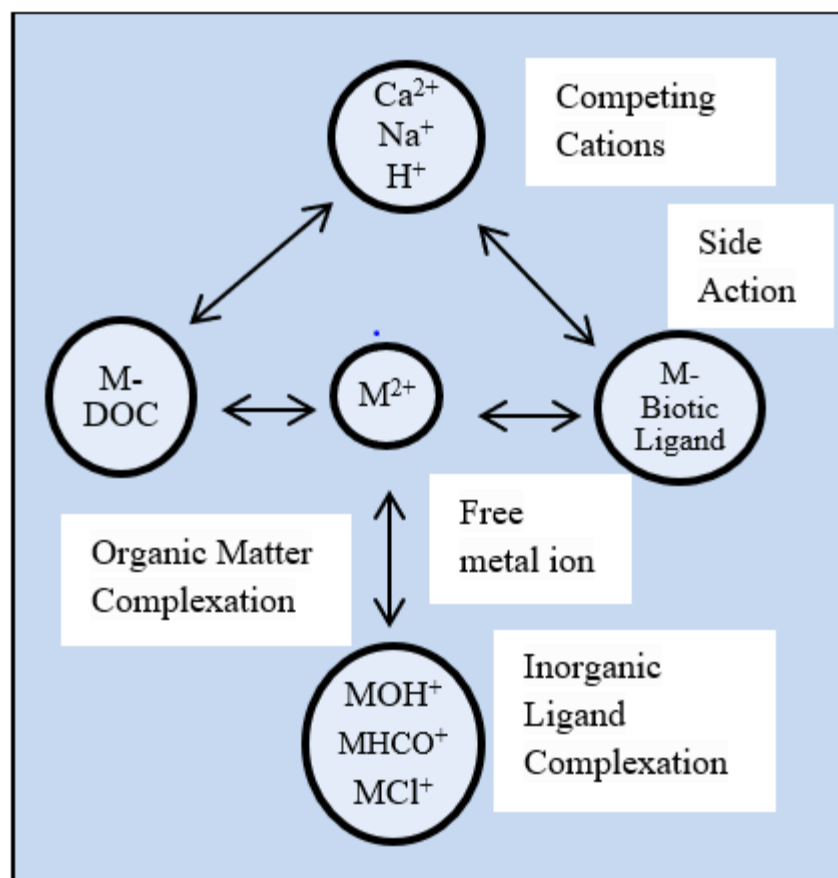


Figure 6. The remodified schematic diagram of the biotic ligand model; the M^{2+} is the free metal ion (after Di Toro et. al. 2001)

the aqueous media. Those interactions are shown in Figure 6 with the free metal ion, M^{2+} and naturally occurring competitive cations; Na^+ , H^+ , Mg^{2+} , Ca^{2+} and abiotic ligands by DOC , CO_3^{2-} , HCO_3^- , OH^- and Cl^- whereas the site toxic action is represented by the biotic ligand or test organism under study.

3.2. Comparison of biosorbent performance

3.2.1. The T-test and correlation studies

In biosorption and adsorption studies done by several scholars over the past four decades, both treated and untreated biosorbents have been reported as successful and potential alternative materials for remediation of organics such as dyes and inorganics such as trace metals. Although it is claimed by several studies that treatment enhances sorption performance for biosorbents.

Published reports on comparison of treated and untreated materials' performance are rare; therefore, studies were necessary to inform the best choice for treatment and application of either category of biosorbents to small and large scale remediation processes. The treated biosorbents have known advantages enlisted by several researchers but such treatments come with

input procedures and chemicals that would impact on the total cost of the biosorbent materials.

The two types of biosorbents were subjected to similar experimental conditions of initial trace metal concentrations, initial pH levels, agitation time and biosorbent dosage and the resultant sorption intensities were subjected to correlation studies and the T-test.

Table 1. Major elements (mg/L) in water from selected protected springs in Kampala city ($n=3$, \pm standard deviation) compared with WHO guideline values, Canadian guidelines and the Ugandan Standards (WHO, 2011; MWE, 2013; Health Canada, 2014)

Water sample	WHO Guideline values						
	Na	Mg	Al	K	Ca	Mn	Fe
	200	100	0.2	10	200	0.4	0.3
	Ugandan Standards						
	200	50	0.1	-	75	0.1	0.3
	Canadian Guideline values						
	200	-	0.1	-	-	0.05	0.3
Nakawa Division							
Nabuze	23 \pm 0.3	4.7 \pm 1.2	0.002	3.6 \pm 0.4	11.6 \pm 1.3	0.14 \pm 0.01	0.3 \pm 0.01
Katarina	14 \pm 0.6	2.2 \pm 0.7	0.002	4.4 \pm 0.5	6.7 \pm 0.8	0.02 \pm 0.01	0.2 \pm 0.01
Buwooya	10 \pm 1.5	1.8 \pm 0.2	0.191	3.6 \pm 0.3	5.7 \pm 1.5	0.02 \pm 0.01	0.7 \pm 0.03
Bukoto 1	10 \pm 1.2	1.3 \pm 0.3	0.098	2.6 \pm 0.2	4.9 \pm 1.0	0.06 \pm 0.02	0.5 \pm 0.02
Bukoto 2	23 \pm 1.2	2.0 \pm 0.4	0.360	5.7 \pm 0.5	9.2 \pm 1.3	0.24 \pm 0.01	0.7 \pm 0.02
Kawempe Division							
Nabukalu	37 \pm 1.9	4.1 \pm 0.4	0.012	8.3 \pm 0.5	13.5 \pm 1.0	0.33 \pm 0.01	0.3 \pm 0.02
Abdu	61 \pm 2.3	6.5 \pm 0.6	0.167	17.2 \pm 1.0	21.4 \pm 2.1	1.14 \pm 0.02	0.4 \pm 0.01
SP4306K	40 \pm 3.0	4.4 \pm 0.8	0.001	15.1 \pm 1.3	17.1 \pm 1.2	0.84 \pm 0.02	0.3 \pm 0.02
Kikoni	20 \pm 2.9	3.1 \pm 0.7	0.001	4.9 \pm 0.6	11.1 \pm 0.6	0.62 \pm 0.03	0.2 \pm 0.01
Rubaga Division							
SP1414R	31 \pm 2.5	4.1 \pm 0.2	0.001	6.5 \pm 0.5	12.0 \pm 1.3	0.24 \pm 0.02	0.2 \pm 0.01
Sentamu	27 \pm 2.6	3.8 \pm 0.1	0.001	7.9 \pm 0.6	13.0 \pm 1.1	0.24 \pm 0.01	0.2 \pm 0.01
Kunnya	23 \pm 2.2	1.9 \pm 0.1	0.107	8.6 \pm 0.3	8.3 \pm 0.4	0.28 \pm 0.04	0.4 \pm 0.02
SP1212R	22 \pm 1.7	3.7 \pm 0.2	0.001	8.3 \pm 0.5	11.0 \pm 0.5	0.14 \pm 0.02	0.1 \pm 0.03
Kibumbiro	23 \pm 1.8	2.7 \pm 0.3	0.299	5.7 \pm 0.4	11.2 \pm 0.3	0.78 \pm 0.02	0.7 \pm 0.02
Kabaale	19 \pm 1.3	3.2 \pm 0.4	0.169	7.6 \pm 0.2	14.3 \pm 0.3	0.09 \pm 0.01	0.5 \pm 0.03
Nababirye	8.0 \pm 0.2	2.4 \pm 0.2	0.002	4.2 \pm 0.1	7.1 \pm 0.1	0.02 \pm 0.01	1.9 \pm 0.05
SP1010R	35 \pm 1.9	4.2 \pm 0.3	0.002	9.7 \pm 0.4	12.7 \pm 0.4	0.53 \pm 0.02	0.2 \pm 0.03
Makindye Division							
Nsambya	45.8 \pm 2.3	4.4 \pm 0.2	0.002	11.1 \pm 0.6	14.7 \pm 0.3	0.58 \pm 0.01	0.2 \pm 0.02
Kapeke	66.4 \pm 3.3	4.9 \pm 0.1	0.015	12.2 \pm 0.2	17.2 \pm 0.2	1.20 \pm 0.02	0.2 \pm 0.05
Yusuf	23.2 \pm 1.1	2.8 \pm 0.1	0.001	7.4 \pm 0.5	11.7 \pm 0.1	0.09 \pm 0.01	0.4 \pm 0.06
Kasanvu	21.0 \pm 1.3	4.0 \pm 0.2	0.001	5.2 \pm 0.1	18.7 \pm 0.3	0.02 \pm 0.00	0.2 \pm 0.02
Central Division							
Nabagereka	43.7 \pm 2.3	5.3 \pm 0.2	0.150	15.1 \pm 0.4	23.5 \pm 0.1	0.74 \pm 0.01	0.5 \pm 0.02
Barracks	25.1 \pm 1.2	4.6 \pm 0.3	0.001	5.9 \pm 0.1	20.2 \pm 0.1	0.04 \pm 0.00	0.2 \pm 0.01
SP5003C	37.5 \pm 2.4	5.1 \pm 0.1	0.101	14.9 \pm 0.3	17.3 \pm 0.2	0.27 \pm 0.01	0.4 \pm 0.03

To inform decision on whether to treat or not, the sorption intensities for treated and untreated biosorbents' performances for removal of Cd^{2+} and Pb^{2+} ions were evaluated using correlation studies and subjected to the T-test. Since the application of the two types of biosorbents was intended to give the same result remediation of aqueous trace metal contaminants, the test of significance was relevant to show relationship between the biosorbents' sorption performances.

Table 2. Levels of trace elements ($\mu\text{g/L}$) in water from selected protected springs in Kampala Capital City in comparison to guideline values of WHO (2011), MWE (2013) and Health Canada (2014)

Water sample	WHO Guideline values							
	B	Sb	Ni	Cu	Zn	Cd	Ba	Pb
	2400	20	70	2000	3000	3	700	10
	Ugandan Drinking Water Standards							
	-	-	-	1000	5000	10	1000	50
	Canadian Guideline values							
	5000	6	-	1000	5000	5	1000	10
Nakawa Division								
Nabuze	2.0 \pm 0.1	1.44 \pm 0.03	9.8 \pm 0.3	1.4 \pm 0.1	9.4 \pm 0.5	0.05 \pm 0.002	150 \pm 5.5	0.54 \pm 0.01
Katarina	2.3 \pm 0.4	0.45 \pm 0.01	6.8 \pm 0.4	3.2 \pm 0.2	16.9 \pm 0.8	0.04 \pm 0.001	146 \pm 4.3	0.94 \pm 0.02
Buwooya	2.1 \pm 0.9	0.38 \pm 0.01	3.9 \pm 0.6	1.3 \pm 0.1	16.0 \pm 0.4	0.23 \pm 0.011	73 \pm 3.4	4.09 \pm 0.09
Bukoto 1	2.1 \pm 0.5	0.42 \pm 0.02	3.2 \pm 0.2	0.8 \pm 0.2	13.6 \pm 0.1	0.04 \pm 0.001	140 \pm 5.6	0.70 \pm 0.01
Bukoto 2	2.5 \pm 0.1	0.39 \pm 0.02	5.4 \pm 0.3	1.8 \pm 0.3	26.4 \pm 0.4	0.05 \pm 0.002	234 \pm 7.7	1.91 \pm 0.02
Kawempe Division								
Nabukalu	2.4 \pm 0.1	0.40 \pm 0.01	7.4 \pm 0.1	1.6 \pm 0.2	20.9 \pm 0.4	0.07 \pm 0.01	332 \pm 4.5	0.87 \pm 0.03
Abdu	2.7 \pm 0.2	0.55 \pm 0.02	12 \pm 0.1	2.0 \pm 0.1	22.9 \pm 0.6	0.11 \pm 0.02	821 \pm 8.3	1.55 \pm 0.02
SP4306K	2.7 \pm 0.2	10.98 \pm 0.02	8.6 \pm 0.1	2.0 \pm 0.2	14.7 \pm 0.1	0.06 \pm 0.01	422 \pm 7.2	1.35 \pm 0.01
Kikoni	3.2 \pm 0.3	0.21 \pm 0.01	3.8 \pm 0.3	0.5 \pm 0.1	6.9 \pm 0.2	0.05 \pm 0.01	180 \pm 6.3	0.04 \pm 0.01
Rubaga Division								
SP1414R	2.0 \pm 0.4	9.19 \pm 0.01	9.1 \pm 0.2	1.7 \pm 0.5	17.1 \pm 0.7	0.05 \pm 0.01	397 \pm 5.1	0.76 \pm 0.01
Sentamu	1.7 \pm 0.5	0.41 \pm 0.02	7.4 \pm 0.1	0.6 \pm 0.3	9.3 \pm 0.0	0.05 \pm 0.01	303 \pm 7.7	0.22 \pm 0.03
Kunnya	2.4 \pm 0.1	0.90 \pm 0.01	6.1 \pm 0.7	1.8 \pm 0.4	24.5 \pm 0.7	0.07 \pm 0.02	249 \pm 6.5	1.71 \pm 0.02
SP1212R	1.7 \pm 0.0	0.86 \pm 0.02	8.5 \pm 0.4	0.7 \pm 0.3	9.8 \pm 0.0	0.06 \pm 0.02	246 \pm 8.0	0.23 \pm 0.01
Kibumbiro	3.9 \pm 0.1	1.34 \pm 0.04	15.3 \pm 0.0	3.7 \pm 0.5	48.0 \pm 0.5	0.16 \pm 0.03	454 \pm 7.9	5.06 \pm 0.05
Kabaale	5.1 \pm 0.5	0.90 \pm 0.03	4.9 \pm 0.1	1.7 \pm 0.8	16.5 \pm 0.6	0.10 \pm 0.02	286 \pm 8.3	2.11 \pm 0.02
Nababirye	1.9 \pm 0.6	1.80 \pm 0.00	4.9 \pm 0.2	5.5 \pm 0.8	7.7 \pm 0.8	0.02 \pm 0.01	108 \pm 3.1	0.43 \pm 0.03
SP1010R	2.2 \pm 0.5	0.22 \pm 0.01	8.1 \pm 0.2	0.7 \pm 0.7	12.1 \pm 0.1	0.07 \pm 0.02	442 \pm 5.8	0.34 \pm 0.01
Makindye Division								
Nsambya	2.5 \pm 0.2	1.75 \pm 0.02	15.2 \pm 0.4	1.4 \pm 0.6	11.7 \pm 0.6	0.06 \pm 0.01	506 \pm 6.3	0.72 \pm 0.02
Kapeke	2.8 \pm 0.7	4.15 \pm 0.03	25.5 \pm 0.8	3.4 \pm 0.8	38.0 \pm 0.4	0.14 \pm 0.02	1132 \pm 8.9	1.81 \pm 0.04
Yusuf	2.6 \pm 0.1	0.64 \pm 0.01	2.1 \pm 0.1	0.9 \pm 0.5	11.5 \pm 0.3	0.04 \pm 0.01	118 \pm 5.3	0.78 \pm 0.05
Kasanvu	7.2 \pm 0.8	0.44 \pm 0.01	3.5 \pm 0.0	0.9 \pm 0.2	4.1 \pm 0.2	0.02 \pm 0.00	117 \pm 6.9	0.12 \pm 0.04
Central Division								
Nabagereka	3.5 \pm 0.2	0.38 \pm 0.02	8.94 \pm 0.4	1.5 \pm 0.9	15.9 \pm 1.3	0.08 \pm 0.01	508 \pm 9.0	1.72 \pm 0.02
Barracks	3.0 \pm 0.8	5.74 \pm 0.08	8.22 \pm 0.3	1.1 \pm 0.8	7.6 \pm 0.7	0.05 \pm 0.01	213 \pm 5.1	0.51 \pm 0.01
SP5003C	2.3 \pm 0.2	0.52 \pm 0.05	4.52 \pm 0.1	1.4 \pm 0.4	25.1 \pm 1.5	0.19 \pm 0.02	424 \pm 6.4	2.82 \pm 0.01

4. RESULTS

4.1. Trace metal contamination of the Lake Victoria basin,

Although these studies were focussed on the trace metals contamination, the levels of the major elements in the shallow groundwater samples from selected protected springs in Kampala Capital City were also studied. The results are presented in Table 1. The results show the levels of major elements in shallow groundwater, surface runoff, landfill leachate, wastewater and surface water that were analysed. Table 1 presents the guidance of WHO guideline values, Ugandan Standards and the Canadian guideline values (Health Canada, 2014; WHO, 2011; MWE, 2013). According to the WHO guidelines, the results showed that 46% of the sampled protected springs were contaminated with Fe which exceeds 0.3 mg/L and was in the range of 0.34 – 1.86 mg/L.

This exceedance of the guideline value could be attributed to weathering and since the surface water in the associated wetlands appears with a rusty flimsy layer in slow and stagnant waters in the catchment areas. The shallow groundwater samples from peri-urban protected springs were studied and the levels of trace metals were determined.

The results presented in Table 2 showed that the concentrations of most trace metal elements studied were high except Boron whose levels are below contamination levels in all springs. Antimony levels are low and only elevated in one spring (>10 mg/L) that was on the peripheral of a busy urban market, car washing bay and motorcycle service workshop which are suspected to be the sources of contamination.

Nickel levels in 5% of the springs were above the WHO guideline value, 20mg/L and the affected springs were characterised by nearness to motorways and residential areas where wearing of car parts, surface runoff and domestic wastewater are assumed to impact the metal levels.

Eight percent of the protected springs exceeded the WHO guideline value for barium whereas 25% had elevated levels. On the other hand, cadmium, copper and zinc levels were all below the WHO guideline values in all the protected springs studied. Lead levels in all springs were well below the WHO guideline value; 10 mg/L. The mobility of trace metals increases under acidic conditions with high dissolved organic matter content. The other parameters of shallow groundwater and associated surface runoff from peri-urban Kampala were studied and compared to Guidelines for Canadian Drinking Water Quality and Ugandan Standards. The results are presented in Table 3.

The temperature of the water samples varied slightly around the guideline value of 25°C moreover the pH values were all below the guideline value range; 6.5 – 8.5.

Table 3 Other parameters of shallow groundwater and associated surface runoff from peri-urban Kampala compared to Canadian Drinking Water Quality and Ugandan Guidelines; MWE (2013) and Health Canada (2014)

Water sample	Shallow groundwater values						Surface runoff values		
	pH	DOC (mg/L)	EC (μ S/cm)	NO ₃ ⁻ (mg/L)	Cl ⁻ (mg/L)	F ⁻ (mg/L)	EC (μ S/cm)	Cd ²⁺ (μ g/L)	Pb ²⁺ (μ g/L)
Canadian values	6.5-8.5	-	-	45	-	0.5	-	5	10
Ugandan values	6.0-8.5	-	-	10	250	1.0	-	10	50
Nakawa Division									
Nabuze	5.46	1.0	665	34.1	11.9	0.1	750	1.18	178
Katarina	5.36	1.0	372	19.9	6.9	0.1	450	1.30	81
Buwooya	5.45	1.0	211	10.4	3.8	0.2	350	2.62	560
Bukoto 1	5.07	1.0	226	27.0	13.3	0.3	300	2.52	915
Bukoto 2	4.80	2.0	470	64.3	24.2	0.2	520	3.10	905
Kiddumu	5.72	1.0	862	44.8	16.8	0.1	1208	0.32	108
Kawempe Division									
Nabukalu	4.93	2.0	873	78.7	30.3	0.1	980	2.39	129
Abdu	4.74	1.0	2650	78.5	46.5	0.1	3050	2.64	1294
SP4306K	4.98	1.0	786	38.6	16.6	0.1	824	1.35	95.8
Kikoni	5.35	1.0	497	42.5	18.2	0.1	532	8.00	519
Bukuku	5.86	2.0	643	65.4	31.2	0.1	835	1.19	92.1
Rubaga Division									
SP1414R	4.93	1.0	774	63.6	27.0	0.1	812	1.82	121
Sentamu	4.98	1.0	722	74.6	32.3	0.1	820	0.42	69
Kunnya	5.30	1.0	574	107.3	41.7	0.1	642	0.31	11
SP1212R	5.03	1.0	631	1.0	10.2	1.0	722	0.13	28
Kibumbiro	5.14	2.0	586	68.4	40.1	0.1	650	0.96	185
Kabaale	5.32	1.0	607	70.2	28.4	0.2	600	0.90	45
Nababirye	5.68	1.0	272	42.5	18.3	0.1	472	0.35	31
Nsereko	4.89	1.0	786	24.7	8.8	0.1	786	24.7	8.8
Lugala	5.18	1.0	272	57.9	21.6	0.1	1090	0.30	30.8
SP1010R	5.18	1.0	916	64.5	28.4	0.1	1200	0.03	78
Makindye Division									
Nsambya	5.37	1.0	1098	101.2	42.1	0.2	1250	0.68	140
Kapeke	5.18	1.0	1472	102.5	64.4	0.2	1670	0.29	106
Yusuf	5.45	1.0	648	101.4	42.7	0.2	1210	0.43	215
Kasanvu	5.68	1.0	764	38.7	11.8	0.5	825	0.18	36
Kibuuka	5.89	1.0	873	45.6	18.6	0.2	972	1.20	201.4
Kisugu	5.68	1.0	764	125.3	58.5	0.2	1250	0.68	140.0
SP7009M	5.24	2.0	607	45.6	18.6	0.2	648	0.82	141.0
Central Division									
Nabagereka	5.54	2.0	1208	58.3	35.6	0.3	779	0.03	34
Barracks	5.36	1.0	779	74.7	23.2	0.1	864	0.22	121
SP5003C	5.55	2.0	1051	53.7	35.5	0.3	1251	0.29	57
Kisekka	4.97	2.0	772	5.1	49.6	5.0	820	18.3	15.4
Kinyoro	5.20	1.0	421	18.0	5.9	0.1	774	1.05	294.9
Bativa	4.95	1.0	832	38.8	14.8	0.1	870	0.68	55.7
SP5508C	5.24	1.0	680	44.4	29.4	0.2	680	44.4	29.4
SP5303C	5.25	2.0	648	53.7	36.5	0.3	648	53.7	36.5
SP4902C	4.92	1.0	786	60.5	23.2	0.1	1670	0.29	105.8

The shallow groundwater was therefore, generally acidic with pH < 7.0 whereas acidic water increases the weathering of rocks and the release of metals from soil surfaces into soil water. Acidic water is aggressive to surfaces and has a high capacity to corrode ferrous materials which explains why surface water in the catchment areas exhibits a flimsy rusty layer. On the other hand, Olade (1987) stated that groundwater tapped from weathered regolith is acidic which may promote oxidation and corrosion of steel casings and screens. The DOC levels were all below the contamination level, 2.0 mg/l. Mobility of trace metals is known to be dependent on the dissolved organic matter content and pH, although trace metals redistribution in groundwater may be affected by other factors such as redox conditions. Electrical conductivity values were high but only exceed the guideline value of 1000 μ S/cm in a few scattered cases which indicated that inorganic species occurred mainly in ionic form. Only 3% of nitrate levels were below the WHO guideline value and the rest were highly polluted with 11 – 125 mg/L. Of the sampled shallow groundwater, 52.8% had nitrate levels above the guideline value; 50mg/L and 21.6% of them had elevated levels. (Paper I)

On the other hand, the surface water, wastewater and landfill leachate from selected streams and waterways in the Lake Victoria basin, Uganda was sampled and studied, and the results for the levels of pH, EC and major elements are presented in Table 4.

Table 3. Major elements (mg/L), pH and EC (μ S/cm) for surface water from selected streams and waterways in the Lake Victoria basin, Uganda compared to WHO guidelines (2011) and Ugandan Standards (Uganda Government, MWE, 2013)

Sampling site	WHO water guidelines								
	pH	EC	Na	Mg	Al	K	Ca	Mn	Fe
	6.5-8.5	1000	200	50	0.1	10	200	0.1	0.3
	Ugandan Standards								
	6.0 – 8.0	–	–	100	0.5	–	100	1.0	10
Lubigi channel	7.49	1530	51.5	5.1	0.066	25.03	26.3	0.80	2.61
Mulago channel	7.02	1420	50.7	4.6	0.062	22.60	25.5	0.83	2.63
Nakivubo channel	7.20	1723	56.4	5.4	0.154	28.62	24.3	0.53	2.84
Lugogo stream	7.16	930	31.8	5.7	0.001	17.32	26.3	0.83	3.49
Bugoloobi stream	7.09	700	19.9	5.1	0.002	6.28	18.0	0.13	0.33
Kiteezi Landfill	8.44	5100	5245	82.8	0.140	0.03	26.2	0.29	5.35
Lugazi Steel Mills	7.68	133	40.7	4.6	0.739	27.06	27.5	0.62	14.98
Jinja Landfill	7.89	3120	400	184.8	0.002	0.07	135.4	2.87	6.49
Jinja Steel Mills	7.46	1772	36.1	22.5	0.001	4.66	62.6	0.68	1.14
Jinja Walukuba	7.90	300	11.9	1.7	0.001	3.49	5.8	0.01	0.20
River Mpanga	6.80	768	27.8	15.2	0.001	6.84	82.7	1.57	1.27
Kibenge hot springs	7.60	924	600.2	4.4	0.040	19.99	199	0.03	0.97
River Nyamwamba	7.03	127	6.4	3.8	0.002	3.62	10.0	0.13	1.21
Kilembe Mines	7.45	1569	509.8	25.1	0.924	518.10	78.6	0.45	0.93
Kitagata hot springs	7.80	1182	205.6	0.2	0.001	10.50	30.5	0.02	0.33

The pH values for all the sampled sources were within the limits of the WHO guideline values, 6.5 – 8.5.

The electrical conductivity values for 53.3% of the surface water, surface runoff and landfill leachate samples were higher than 1000 $\mu\text{S}/\text{cm}$ whereas samples from hot springs and municipal landfills had 3 – 6 times higher than the WHO guideline value. One of the most serious of the inorganic contaminants reported was fluoride with concentrations above the guideline value, 1.5mg/l for samples of groundwater in interaction with crater lakes in Uganda (Mungoma, 1990; BGS, 2001), although the fluoride levels in shallow groundwater in Kampala were lower than the guideline values which indicated a difference in sources of fluorine which of geochemical origin.

These values may be due to mobilisation of conducting ions during the decay processes of landfills, thermal mobilisation of ions as hot water rises up through mineralised soils in hot springs moreover the salt lake has a rich matrix of conducting ions. The mine tailings from an old copper mine had higher EC values attributed to copper contamination of water. The temperatures of surface water sources were within the guideline values.

In Table 5, the results of studies to determine the levels of trace metal elements in the surface water samples from various sources in the Lake Victoria basin, Uganda are presented.

Table 4. Comparison of Trace metal elements ($\mu\text{g}/\text{L}$) in surface water from selected sources with WHO guidelines (2011) and Ugandan Standards (Uganda Government, MWE, 2013)

WHO water guidelines	B 2400	Sb 20	Ni 70	Cu 2000	Zn 4000	As 10	Se 40	Cd 3	Ba 700	Pb 10
Ugandan Standards	5000	–	1000	1000	5000	200	1000	100	10000	100
Lubigi channel	485	30.47	178.9	133.2	584.7	25.15	27.71	1.11	6227	98
Mulago channel	14	23.10	5.8	4.9	25.9	0.70	0.88	0.05	215.1	2.9
Nakivubo channel	32	0.72	10.9	10.9	66.2	1.0	1.19	0.26	181.9	11
Lugogo stream	18	0.99	6.9	3.3	15.7	0.88	1.02	0.02	218.7	0.9
Bugoloobi stream	199	92.80	126.8	38.4	506.8	6.11	16.46	2.31	2724	27
Kiteezi Landfill	1338	15.74	271.2	119.6	248.3	23.32	71.37	1.14	214.5	99
Lugazi Steel Mills	126	0.14	19.6	16.4	210.3	2.08	0.69	0.19	73.3	29
Jinja Landfill	953	5.65	104.7	74.1	241.1	11.98	25.62	0.44	186.3	30
Jinja Steel Mills	29	0.26	6.2	1.0	4.9	0.46	2.55	0.03	104.8	0.3
Jinja Walukuba	10	0.51	0.9	0.5	6.1	0.21	0.21	0.01	30.2	1.0
River Mpanga	11	1.24	5.5	1.8	15.2	0.38	0.47	0.48	341.7	5.1
Kibenge springs	296	1.34	9.1	7.6	12.8	2.96	23.18	0.14	25.8	2.0
River Nyamwamba	7.5	2.17	18.0	172.7	24.2	0.27	0.29	0.19	28.6	2.3
Kilembe Mines	6.8	1.67	90.0	1604	53.4	2.03	0.60	0.45	31.0	2.6
Kitagata springs	375	1.08	2.4	3.6	21.5	0.82	2.60	0.47	36.8	4.9

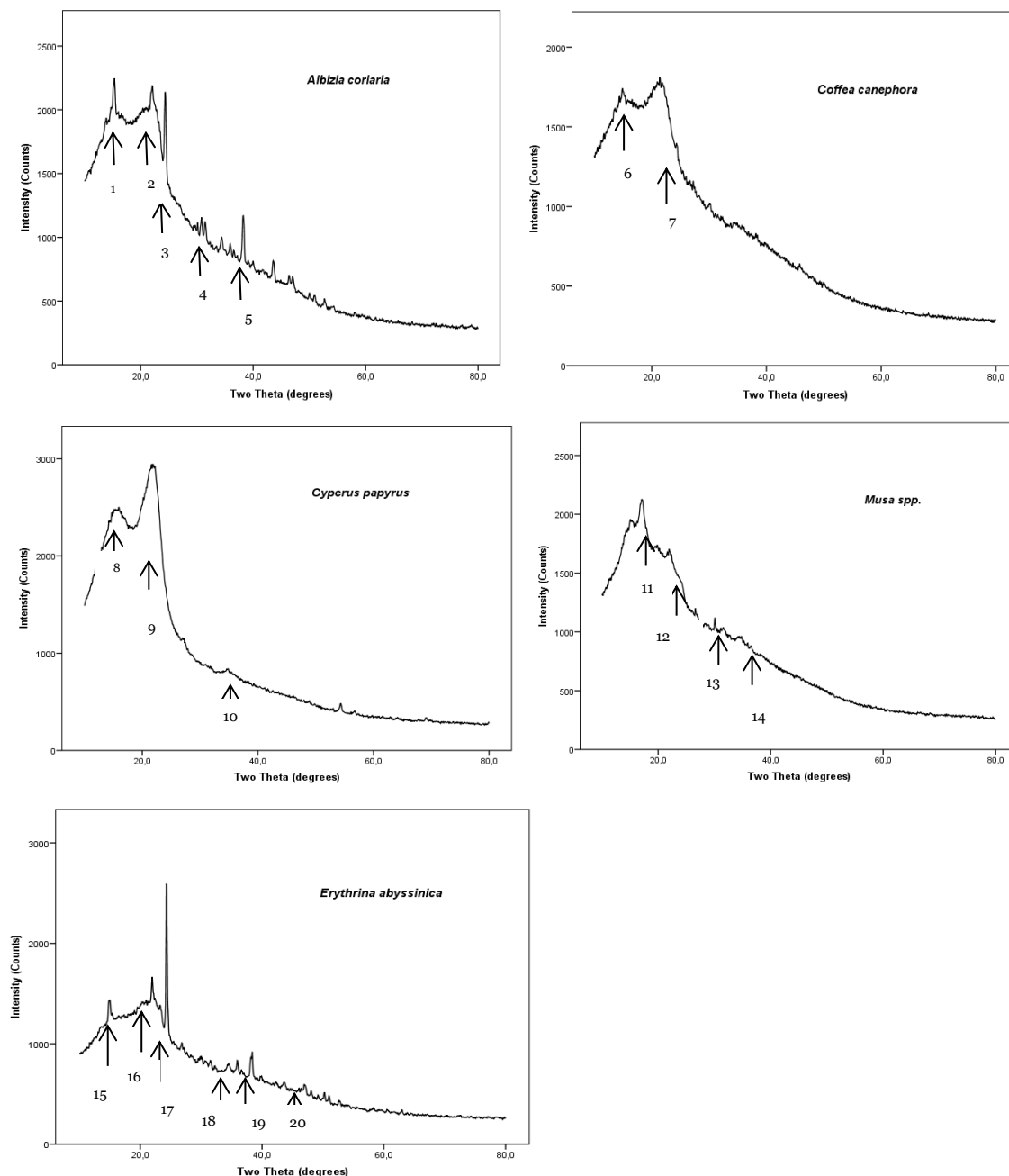


Figure 7. XRD characteristic patterns for untreated *Albizia coriaria*, *Erythrina abyssinica*, *Coffea canephora*, *Cyperus papyrus* and *Musa spp.* biosorbent powders with numbered notable organic species

The percentage of levels of trace elements exceeding the guideline values in surface water were; 25% for boron, 25% for antimony, 31% for nickel, 6% for copper, 13% for zinc, 25% for arsenic, 38% for selenium, 13% for barium and 31% for lead although Taylor and Howard (1994) reported minor exceedances above the WHO guideline values for barium, nickel, lead, uranium and cadmium in their study of groundwater in Uganda.

Table 6. Selected trace metal constituents in samples of the biosorbent materials

	Main trace metal constituents (µg/g)						
Biosorbents	Mn	Fe	Ni	Cu	Zn	Cd	Pb
<i>Albizia coriaria</i>	19.2	18.4	6.96	2.85	15.8	0.01	0.56
<i>Erythrina abyssinica</i>	43.0	79.3	4.11	5.12	19.9	0.02	0.97
<i>Coffea canephora</i>	11.5	4.20	0.92	18.6	5.31	0.02	0.46
<i>Cyperus papyrus</i>	34.6	1.98	0.27	1.26	11.0	0.04	0.18
<i>Musa spp.</i>	11.7	1.53	0.91	2.93	17.8	0.01	1.23

Table 7. Exchangeable acidity (EA) and cation exchange capacity (CEC) (cmol/kg) of selected biomass (Netz and Salmonsson, 2014).

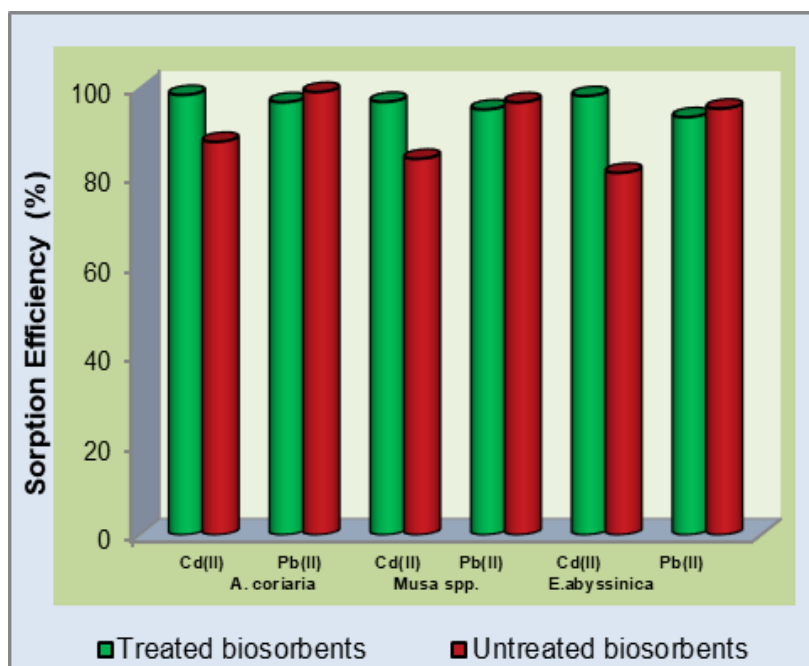
Biomass		pH	EA	CEC
<i>Musa spp</i>	Treated	5.49	0.19	53
	Untreated	5.22	1.64	142
<i>C.canephora</i>	Treated	5.41	0.22	95
	Untreated	4.91	1.66	91
<i>C.papyrus</i>	Treated	5.95	0.42	36
	Untreated	5.39	0.95	85

4.2. Sorption efficiencies of biosorbents for Cd²⁺ and Pb²⁺ ions

The results shown in Figure 8, were an average of three replicas with standard deviation between 0.07 – 0.22% for Cd²⁺ ions and 0.06 – 1.03% for Pb²⁺ ions biosorption. All the base-treated biosorbents showed higher sorption efficiencies for Cd²⁺ ions than the untreated biosorbents which were in the order; *E. abyssinica* bark (17.1%) > *Musa spp.* peels (12.7%) > *A. coriaria* bark (10.5%). In several previous studies, it has been reported that chemical treatment of biosorbents improves trace metals biosorption (Wan Ngah & Hanafiah, 2008) although in these studies, a slight reductive effect on Pb (II) ions biosorption in the order; *A. coriaria* bark (2.3%) > *E. abyssinica* bark (1.9%) > *Musa spp.* peels (1.6%) was realised (Paper II). The highest Cd²⁺ ions biosorption competences observed were; 98.3%, 98% and 96.7% for treated *A. coriaria*, *E. abyssinica* and *Musa spp* respectively. Moreover the highest Pb²⁺ ions sorption competences realised were; 98.9%, 96.6% and 95.2% for untreated *A. coriaria*, *Musa spp.* and *E. abyssinica* respectively.

Table 8. Released dissolved organic matter (DOC) (mg/L) in remediated samples at equilibrium for various pH levels for untreated and base treated biosorbents

pH	<i>A. coriaria</i>		<i>Musa spp.</i>		<i>E. abyssinica</i>	
	Treated	Untreated	Treated	Untreated	Treated	Untreated
Solutions from which Cd ²⁺ ions were removed (mgL ⁻¹)						
2	54.8	697	81.4	438	53.5	580
4	98.6	857	111	470	94.9	732
5	99.0	858	105	470	83.2	714
7	98.7	894	111	257	94.5	709
9	99.2	879	90.2	250	87.0	741
Solutions from which Pb ²⁺ ions were removed (mgL ⁻¹)						
2	54	599	92	450	58	648
4	105	856	119	455	112	747
5	105	842	126	459	97	712
7	109	879	126	461	103	753
9	107	900	129	477	103	776

**Figure 8. Sorption efficiencies for Cd²⁺ and Pb²⁺ ions treated and untreated biomass (Paper II).**

4.3. Organic carbon released and biosorption

Dissolved organics released into treated water were studied for base-treated biosorbents and the results presented in Table 8.

The results showed that NaOH treatment of biosorbents considerably reduced extractable organics in the range of 2 – 11 times in both Cd^{2+} and Pb^{2+} ions remediated solutions. It has been suggested in literature that improvement in reduction of extractable organics could further be done by employing thorough washing of biosorbents after chemical treatment. Studies were done to determine the effects of DOC (mg/L) removal on the biosorption efficiency as a function of the pH (Figure 9).

4.4. Effect of agitation time on sorption

For untreated and base-treated biosorbents, the effect of agitation time on the sorption of trace metal ions was studied and the results presented in Figure 10. The sorption intensities of both treated and untreated biosorbents increased gradually for Cd^{2+} to peak values at equilibrium time (3.5h) before declining for agitation time $>3.5\text{h}$. The sorption intensities of Cd^{2+} were similar for base-treated and untreated materials although the treated biosorbents presented higher values.

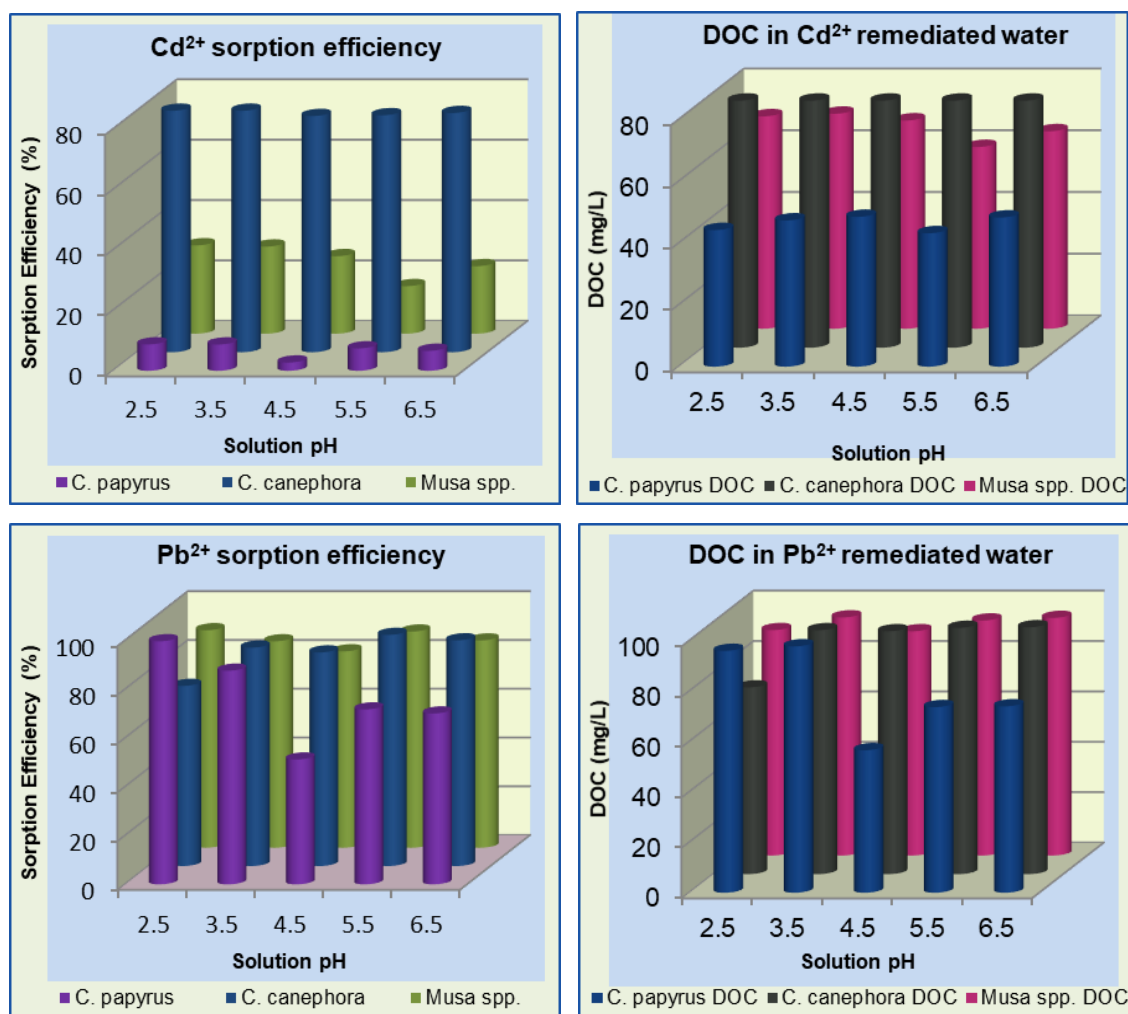


Figure 9. Comparison of sorption efficiencies and DOC released as a function of the solution pH for the various biosorbents studied (Paper IV).

On the other hand, the sorption intensities for Pb^{2+} ions (Figure 10) onto both treated and untreated biosorbents showed a marked difference with that of Cd^{2+} ions since the increase in sorption intensities with agitation time was near polynomial with peak sorption intensities at 3.5 hours.

4.5. Effect of pH on biosorption

The biosorption of trace metal ions from aqueous media by biosorbents is pH-dependent and determination of the optimal pH in biosorption studies is important. The effect of the initial pH of the aqueous solution on the biosorption intensities of untreated and base treated biosorbents were studied then compared for Cd^{2+} and Pb^{2+} ions for initial trace metal ion concentration of 20 mg L^{-1} and agitation time 3.5 hours. The results for Cd^{2+} and Pb^{2+} ions biosorption onto biosorbents for pH range 2.0 – 9.0 are shown in Figure 11.

4.6. Sorption performance of peroxide treated biosorbents

The sorption performance of peroxide treated biosorbents were studied and the results are presented in Figure 12; a and b. These show that the variations of sorption capacities with increasing initial Pb^{2+} ions concentration were comparable and increased gradually to near level from $50 - 100 \text{ mg L}^{-1}$ in both single – and binary – component solutions.

4.7. Effect of initial metal concentration on trace metal biosorption intensities

The sorption intensities for Cd^{2+} and Pb^{2+} ions uptake onto untreated and base treated biosorbents for increasing initial metal concentrations are presented in Figure 13. The Cd^{2+} ions uptake onto base treated biomass with increase in initial concentration C_0 , $5 - 100 \text{ mg/L}$ for Cd^{2+} ions was in the decreasing order for all biomass was $97.3\% - 8.449\%$ for *A. coriaria*, $94.6\% - 5.219\%$ for *Musa spp.* and $93.48\% - 1.259\%$ for *E. abyssinica*.

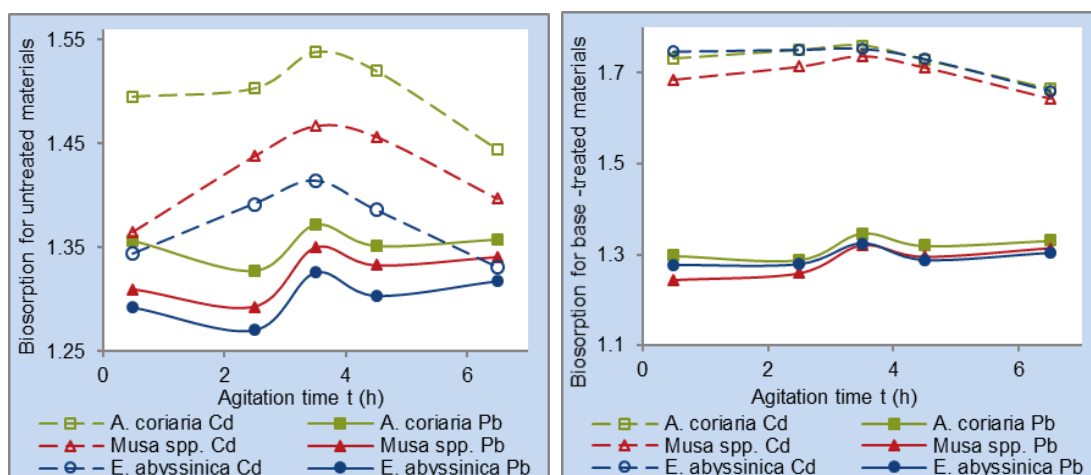


Figure 10. Sorption intensities of Cd^{2+} and Pb^{2+} ions (mg/g) onto untreated and base treated biomass (Paper II)

On the other hand, Cd^{2+} ions uptake onto untreated biosorbents was in the order of 44.9% – 7.5% for *A. coriaria*, 81.76% – 4.58% for *Musa spp.* and 44.02% – 4.766% for *E. abyssinica*. All the biosorbents exhibited decreasing sorption intensity for Cd^{2+} ions with increasing initial concentration C_0 . The biosorbent sorption performance for Cd^{2+} ions for both untreated and base treated biomass was in the order of *Musa spp.* > *A. coriaria* > *E. abyssinica*. (Paper III).

4.8. Kinetic studies

The parameters of kinetic models presented in Table 9 indicated that the Pseudo-first-order model did not fit all the experimental data for Cd^{2+} ions although part of the data for Pb^{2+} ions sorption gave good regression coefficient (R^2) values; 0.8183 – 0.9084. The Lagergren model could not explain the Cd^{2+} ions sorption mechanisms but could describe Pb^{2+} ions sorption for short durations. The Ho model kinetic parameters for untreated and base-treated biosorbents which gave very satisfactory fits for both Cd^{2+} and Pb^{2+} biosorption data for all biosorbents with regression coefficient (R^2) values; 0.9978 – 0.9999 for Cd^{2+} ions and 0.9995 – 0.9998 for Pb^{2+} ions. The Ho model parameters for all the biosorbents were a very good representation of the sorption process. In that case the q_e values were; 1.334 – 1.728 mg/g for Cd^{2+} ions and 1.306 – 1.360 mg/g for Pb^{2+} ions. Since the Ho model fits all the Cd^{2+} and Pb^{2+} ions experimental data, the biosorption mechanism predicted was chemisorption as reported in previous studies (Kalmykova, 2009; Dekhil, et al., 2011; Sari & Tuzen, 2009; Bulut & Tez, 2003). The principle on which the pseudo-second order model was established was that when it fits experimental data it deduces chemisorption as the operative reaction mechanism. Ho & McKay, (1998) asserted that the pseudo second order model predicts the kinetic behaviour of biosorption with chemical sorption being the rate controlling step.

4.9. Equilibrium studies

Biosorption data for untreated, base and peroxide treated biosorbents was fitted to the Freundlich, Langmuir and Temkin isotherm models and the resultant model parameters determined are presented in Tables 10 and 11. The correlation coefficients (R^2) for the models were used to determine the model that best fitted the experimental data. On the other hand, fitting of biosorption data for Cd^{2+} and Pb^{2+} ions by peroxide treated biosorbents from single – and binary – component solutions to the Freundlich, Langmuir and Temkin sorption isotherms was done to determine the isotherm model parameters presented in Table 10. The correlation coefficients (R^2) for the models were used to determine the best fitting model. The overall fitting of the isotherm models for Cd^{2+} ions biosorption data followed the order; Langmuir ($0.951 \leq R^2 \leq 1.000$), Temkin ($0.492 \leq R^2 \leq 0.951$) and Freundlich ($0.363 \leq R^2 \leq 0.882$). The isotherm model fitting for Pb^{2+} ions

biosorption data followed the order; Langmuir ($0.990 \leq R^2 \leq 1.000$), Temkin ($0.831 \leq R^2 \leq 0.993$) and Freundlich ($0.769 \leq R^2 \leq 0.954$).

In Table 11, Freundlich and Temkin isotherm models' parameters for Cd^{2+} and Pb^{2+} ions uptake onto base treated and untreated biosorbents are presented. The results showed that the Freundlich isotherm fits to the sorption data Pb^{2+} ions with linear regression coefficient $0.9997 \leq R^2 \leq 1$ for uptake of Pb^{2+} ions onto base treated biomass and $0.9924 \leq R^2 \leq 0.9997$ for uptake of Pb^{2+} ions onto untreated materials. The correlation coefficient values were high and indicated that the model fits the sorption data. (Paper IV) The model parameters were $0.939 \leq b_F \leq 0.992$ and $9.997 \leq A_F \leq 11.37 \text{ mg/g}$ for uptake of Pb^{2+} ions onto treated biomass whereas for the uptake onto untreated biomass the values were $1.01 \leq b_F \leq 1.11$ and $1.012 \leq A_F \leq 10.756 \text{ mg/g}$.

4.10. Comparison of base-treated and untreated biosorbents

The choice of whether or not to treat biosorbents before application to remediation studies needed a basis for which the statistical comparison and Student t-test would correctly inform decision. Figure 14 shows correlation studies for agreement between base-treated and untreated biomass by comparison of their sorption intensities (mg/g).

Comparison of untreated and base-treated biosorbents' performances revealed regression coefficients, r for comparison Cd^{2+} ions biosorption for *musa spp* as 0.997, for *Albizia coriaria* as 0.923 and for *Erythrina abyssinica* as 0.947 whereas the values of r for comparison Pb^{2+} ions biosorption for *musa spp* as 0.984, for *Albizia coriaria* as 0.971 and for *Erythrina abyssinica* as 0.973.

Table 12 shows the T-test results for both equal and unequal variances for matched pair comparisons of sorption intensities for Cd^{2+} and Pb^{2+} ions onto untreated and base-treated biosorbents. The degrees of freedom were 8, $t_{0.025}$ was 2.306.

The null hypothesis: $\mu_1 - \mu_2 = 0$ where $\mu_1 - \mu_2 = \delta$ and $\delta = 0$

The alternative hypothesis: $\mu_1 - \mu_2 > 0$ or $\delta \neq 0$

Level of significance: $\alpha = 0.05$

Criterion: Reject the null hypothesis if $t > 2.306$ or $t < -2.306$ where $t_{\alpha/2} = 2.306$ for 8 degrees of freedom.

Decision: Since all the t values were less than 2.306, the null hypothesis cannot be rejected at level $\alpha = 0.05$.

5. DISCUSSION

5.1. Shallow groundwater and surface runoff

The levels of major elements in shallow groundwater, surface runoff, landfill leachate, wastewater and surface water samples were in agreement with the report by BGS (2001) where it was stated that iron was one of the main inorganic groundwater quality problems in Uganda.

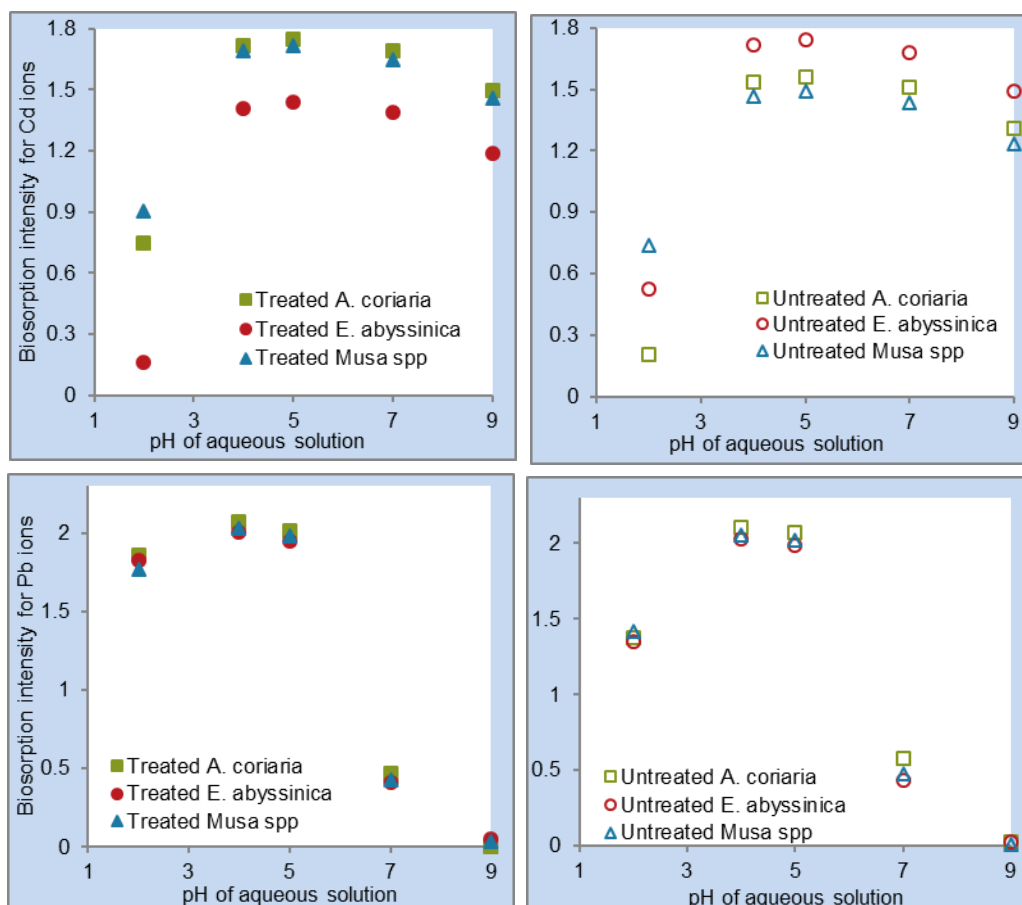


Figure 11. Variation of sorption intensities with solution pH for untreated and base treated biomass (Paper II)

Iron is mainly transported in groundwater in reduced form as Fe^{2+} ions. Therefore, the main source of Fe contamination appears to be background levels in the soils where acidic and redox conditions enhance release of Fe and Mn into soil water. There were only 4% of the springs that had Cr levels above the 0.05 mg/L level. The protected springs with elevated Cr levels are located in Nakawa division and their contamination may be attributed to the existence of paint manufacturing industries in the spring catchment zone. However, 13% of the samples had high levels of Al (0.299 – 0.360 mg/L) exceeding the 0.2 mg/L level. The contamination of the environment with aluminium may be due to anthropogenic activities besides wear and tear of motor parts since the contaminated sampling points were located in a built environment with paved roads but near a car parking lot. On the other hand, the soils in Kampala were reported to be acidic by Moulodi and Thorsell (2013) whereas under acidic conditions the natural sources of metals may contribute to high levels of aluminium since it is mobilised and solubilized from bedrock and soil into soil water at $\text{pH} < 5.5$. The springs with K contamination were 8% with levels above 10mg/L with values between 15 – 17mg/L whereas 29% had Mn levels above the 0.4 mg/L mark

with values ranging between 0.58 – 1.20 mg/L. The BGS (2001) report indicates that high concentrations of manganese are a common problem in Ugandan groundwater. The high iron and manganese occurrences have been attributed to the shallow clayey regolith that restrict aeration of underlying aquifers leading to anaerobic conditions to which the high iron and manganese are related (BGS, 2001). These results for iron and manganese concur with other studies in Uganda by GIBB (1998) where iron levels were reported as 0.3 – 4.9 mg/L moreover Taylor and Howard (1994) reported iron concentrations up to 45mg/L and the Mn levels up to 2mg/L. 95.5% of the water samples had Ca levels below 20mg/L and with the low pH it may be a health risk. According to the Ugandan standards, the levels of contamination of the groundwater samples were; 29% for Al, 4.2% for Cr, 66.7% for Mn and 58.3% for Fe whereas the the contamination levels were; 29% for Al, 4.2% for Cr, 75% for Mn and 45.8% for Fe according to Canadian guidelines.

According to the Ugandan urban drinking water standards (MWE, 2013), most of the levels of trace elements were below the guidelines except for 4.2% Ba contamination whereas the Canadian guidelines showed that 8.3% of the samples were contaminated with antimony. The sampled shallow groundwater from Kampala had elevated levels of trace metals and contamination in a few cases however, several springs have concentrations above which is the concentration when remediation action should be implied. This gives strong relevance for studies on sorption to locally available waste materials.

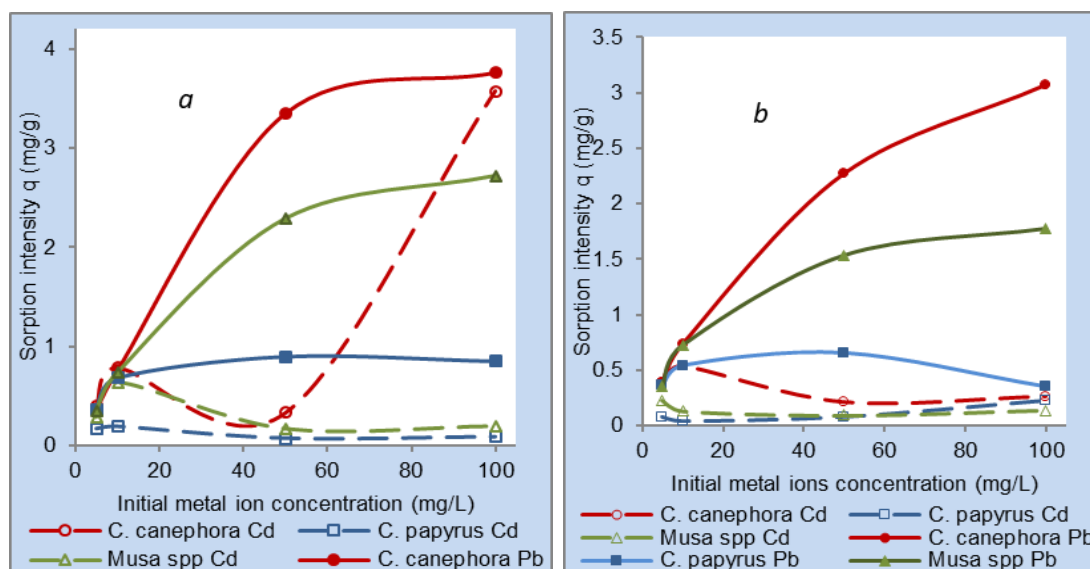


Figure 12 Sorption intensities q (mg/g) of peroxide treated biosorbents for Cd^{2+} and Pb^{2+} ions as a function of the initial metal ions concentration in (a) single-component and (b) binary-component solutions (Paper IV).

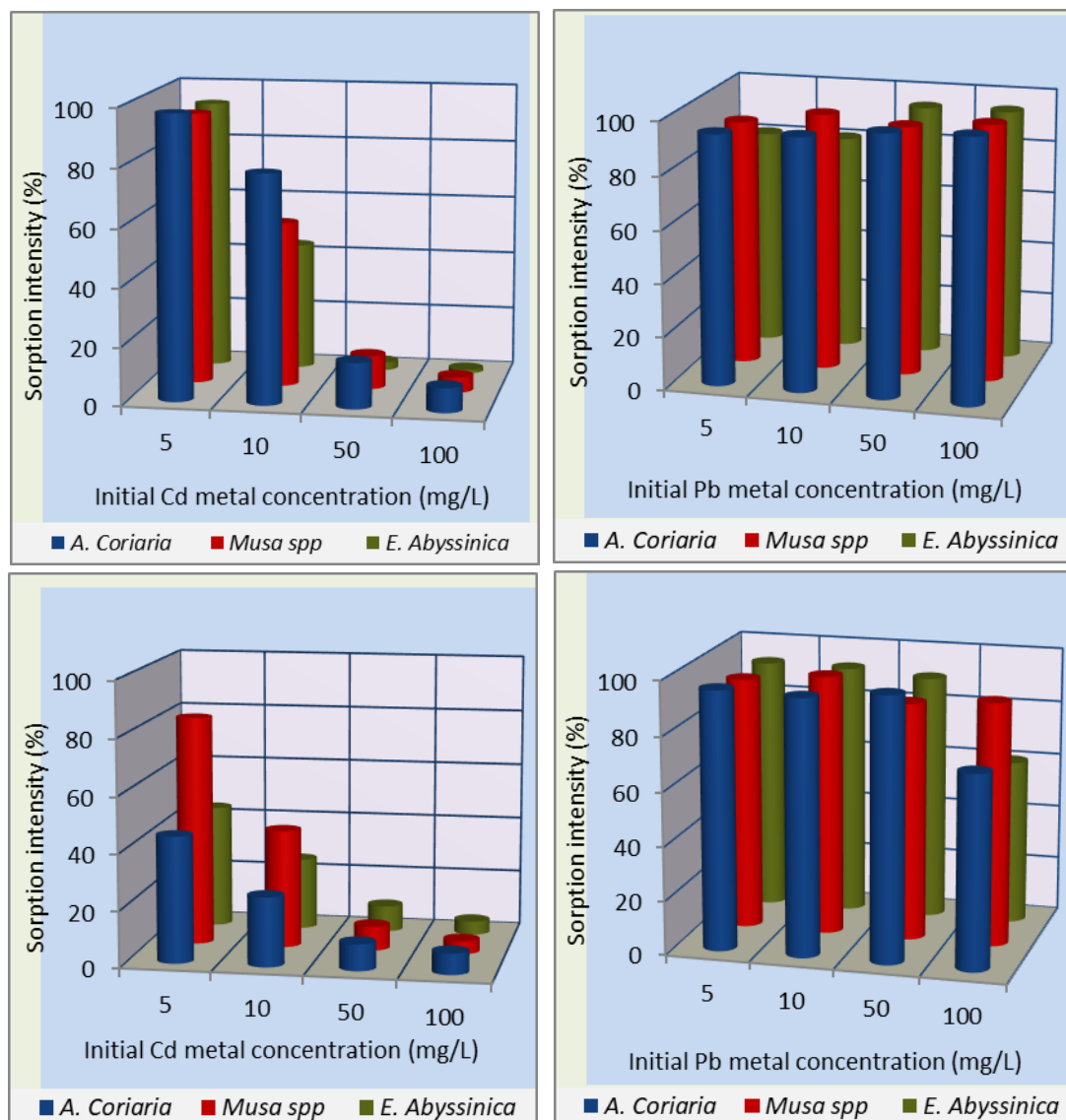


Figure 13 Sorption performance of the biomass in removing Cd²⁺ and Pb²⁺ ions was done with C₀, 5 – 100 mg/L at pH 4.5 and biosorbent dosage 10 g/L for agitation time 3.0 hours (Paper III)

Results from studies of other parameters agreed with studies by Taylor & Howard, (1994) where nitrate levels of 26 mg/L were reported in groundwater and in another study by Kulabako *et al.* (2007) the mean nitrate values reported in shallow groundwater were 67 mg/L. These high levels could be attributed to anthropogenic sources such as on site sanitation, domestic sewage disposal habits, waste dumpsites and others in the spring catchment areas. The chloride levels were high in 57% of the springs moreover 38% of the water from protected springs were contaminated and have high salinity probably due to weathering since the groundwater is acidic. The results in these studies were similar to the high salinity reported by Kulabako *et al.* (2007)

which was 59 mg/L whereas Moulodi and Thorsell (2013) reported 4.6 – 54.6 mg/L of chloride levels in shallow groundwater as outlined in *Paper I*.

Of all the surface runoff samples 25% were contaminated with cadmium compared to the WHO guideline value of 3µg/L while 97.2% of the samples had Pb levels above the WHO health-based value of 10µg/L. According to the Ugandan standards (MWE, 2007; MWE, 2013) and the Canadian standards (Health Canada, 2014), the pH levels for all the shallow groundwater samples were below the recommended range of 6.5 – 8.5 and 54% of them were contaminated with nitrates which was indicative of nutrient loading of the shallow aquifers whereas for Canadian standards, 2.9% were contaminated with cadmium and fluorine.

The sources of lead contamination are mainly anthropogenic activities since there are scrap metal yards, motor service centres and metal workshops in addition to wearing of parts of vehicles that may contribute to the elevated levels.

Table 9 Parameters of kinetic models fitting to Cd^{2+} and Pb^{2+} ions sorption data for base treated and peroxide treated biomass

		Biosorption Kinetic Models					
Biosorbents		Pseudo-first-order			Pseudo-second-order		
		k_1	q_e	R^2	k_2	q_e	R^2
<i>A. coriaria</i>							
Base treated	Cd^{2+}	0.245	0.013	0.4223	0.454	1.728	0.9999
	Pb^{2+}	0.206	0.068	0.8183	0.393	1.336	0.9996
Untreated	Cd^{2+}	0.084	0.030	0.1016	0.118	1.450	0.9978
	Pb^{2+}	0.053	0.025	0.0671	0.853	1.360	0.9998
<i>E. abyssinica</i>							
Base treated	Cd^{2+}	0.038	0.038	0.0217	0.093	1.659	0.9985
	Pb^{2+}	0.135	0.058	0.8384	0.752	1.306	0.9996
Untreated	Cd^{2+}	0.530	0.002	0.6839	0.137	1.334	0.9982
	Pb^{2+}	0.258	0.059	0.6664	0.322	1.323	0.9996
<i>Musa spp.</i>							
Base treated	Cd^{2+}	0.093	0.030	0.1306	0.112	1.647	0.9984
	Pb^{2+}	0.426	0.128	0.9084	0.207	1.323	0.9995
Untreated	Cd^{2+}	0.110	0.056	0.0751	0.210	1.408	0.9984
	Pb^{2+}	0.251	0.049	0.9802	0.307	1.348	0.9996
Peroxide treated							
<i>C. canephora</i>	Cd^{2+}	0.003	0.20	0.134	0.25	1.18	0.999
	Pb^{2+}	0.005	0.06	0.973	0.67	1.58	1.000
<i>Cyperus papyrus</i>	Cd^{2+}	0.013	0.18	0.946	4.22	0.12	0.892
	Pb^{2+}	0.016	1.34	0.713	0.02	1.73	0.894
<i>Musa spp.</i>	Cd^{2+}	0.010	0.05	0.465	0.31	0.26	0.984
	Pb^{2+}	0.010	0.19	0.877	0.17	1.56	1.000

Continued discharge of untreated surface runoff, wastewater and leachate will negatively impact the shallow groundwater when the soil water is laden during the rainy season, the surface water interacts with the groundwater in shallow aquifers without reacting with the soil chemicals thence causing groundwater contamination. (Paper I).

5.2. Surface water

The exceedances above the guideline values for major elements in surface water were; 31% for sodium, 13% for magnesium, 19% for aluminium, 50% for potassium, 69% for manganese and 94% for iron. Surface water from different sources is therefore, contaminated with major elements. The levels of metals in the sampled surface water were higher those in a study by Walakira and Okot-Okumu (2011); 17 – 39mg/L for calcium, 0.6 – 53mg/L for sodium, 0.05 – 0.26mg/L for lead, 0.02 – 0.56mg/L for copper and below detection level for cadmium. The main source of these elements appears to be weathering of mineralised soils and rocks as well as mobilisation during sorption – desorption processes for trace elements onto organic matter and soil particles as featured in Paper I.

When the Canadian Environmental Quality permissible limits for cadmium; 100 µg/L and lead; 50 µg/L were compared with surface water with 0.3 – 97.7 µg/l for Pb and 0.004 – 2.307 µg/L for Cd, 8.8 – 129.4 µg/L for Pb and 0.03 – 44.4 µg/L for Cd in surface runoff as well as 29.7 – 99.3 µg/L for Pb and 0.436 – 1.143 µg/L for Cd in landfill leachate it indicates that the levels of Pb in environmental samples were elevated. The Cd levels were below the permissible limits although a comparison with Ugandan standards showed that the surface runoff was contaminated with lead. The results in this study were higher than those for Lake Victoria water reported by Tole and Shitsama (2001) which were 0.12 -0.45 µg/L for Pb and 0.01 µg/L for Cd. The levels of lead contamination were 6% for surface water, 50% for landfill leachate and 71.4% for surface runoff samples.

The results showed that surface water from urban streams and landfill leachate are particularly contaminated with trace metals; boron, antimony, arsenic, nickel, selenium, barium and lead. The mine tailings are contaminated with copper and nickel whereas lead was very high in wastewater from the landfills, steel mills and an urban stream, Lubigi where the anthropogenic activities are the suspected sources of contamination. In earlier studies, Walakira and Okot-Okumu (2011) reported lower concentration ranges of lead (0.039 – 0.256mg/L), calcium (17.5 – 38.8mg/L), sodium (0.6 – 53mg/L) and copper (0.05 – 0.53mg/L) (PaperI).

Table 10 Sorption isotherm parameters for uptake of Cd^{2+} and Pb^{2+} ions onto peroxide-treated biosorbents

Single component solutions										
Biosorbents		Langmuir			Freundlich			Temkin		
		b	q_{max}	R^2	b_F	a_F	R^2	b_T	A_T	R^2
<i>C. canephora</i>	Cd^{2+}	0.078	4.285	1.000	0.228	0.455	0.363	6.3E3	6.0E1	0.492
	Pb^{2+}	2.880	1.381	0.997	0.459	0.803	0.850	3.4E3	5.232	0.934
<i>C. papyrus</i>	Cd^{2+}	0.623	0.131	0.983	0.266	0.254	0.780	7.7E4	9.9E2	0.780
	Pb^{2+}	0.019	45.46	0.999	0.142	0.503	0.769	2.9E4	4.9E2	0.831
<i>Musa spp.</i>	Cd^{2+}	0.566	0.334	0.993	0.216	0.463	0.600	2.0E4	3.0E2	0.951
	Pb^{2+}	3.661	0.780	0.998	0.350	0.705	0.803	5.1E3	4.663	0.992
Binary component solutions ($C_0 \text{ Cd}^{2+} = C_0 \text{ Pb}^{2+}$)										
<i>C. canephora</i>	Cd^{2+}	1.133	0.224	0.983	0.383	1.215	0.882	1.36E4	2.6E2	0.925
	Pb^{2+}	4.806	0.673	0.990	0.408	0.614	0.954	4.62E3	4.222	0.993
<i>C. papyrus</i>	Cd^{2+}	5.575	0.016	0.951	0.691	0.008	0.886	3.54E4	6.967	0.728
	Pb^{2+}	0.550	1.203	1.000	0.129	0.422	0.916	3.85E4	8.2E2	0.953
<i>Musa spp.</i>	Cd^{2+}	4.038	0.033	0.939	0.154	0.218	0.540	1.000	6.9E3	0.589
	Pb^{2+}	2.666	0.679	0.997	0.283	0.554	0.905	9.3E3	1.1E1	0.982

Table 11. Freundlich and Temkin isotherm models' parameters for Cd^{2+} and Pb^{2+} ions uptake onto base treated and untreated biosorbents

Parameters of the Sorption Isotherm Models							
Biosorbents	Freundlich (Pb^{2+})				Temkin (Cd^{2+})		
	b_F	$1/b_F$	a_F	R^2	b_T	A_T	R^2
<i>A. coriaria</i>							
Treated	0.9828	1.020	9.997	1	347	1.332	0.6473
Untreated	1.0717	0.933	1.012	0.9924	448	1.154	0.9014
<i>E. abyssinica</i>							
Treated	0.9390	1.065	11.379	0.9997	328	2.034	0.9446
Untreated	1.1190	0.894	10.756	0.9871	240	1.051	0.9643
<i>Musa spp.</i>							
Treated	0.9916	1.008	10.125	0.9998	146	1.466	0.6209
Untreated	1.0226	0.978	7.558	0.9996	104	2.718	0.8514

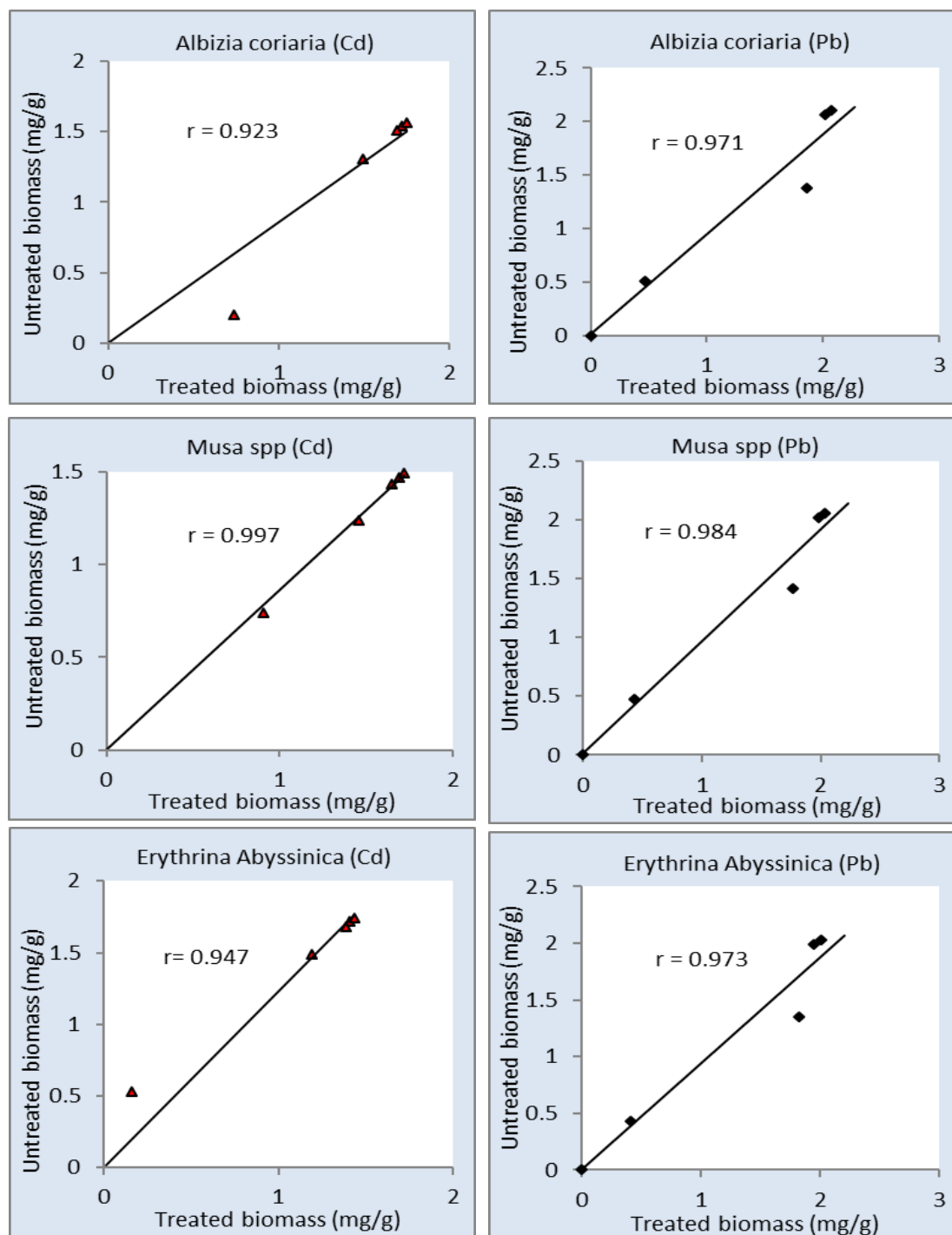


Figure 14 Correlation studies for agreement between base-treated and untreated biomass by comparison of their sorption intensities (mg/g)

5.1. Speciation studies

Table 13 shows results of metal speciation studies for selected streams, waterways and landfill leachates. The results show that in the landfill leachates, 74% of the metal ions are bound to (DOM) dissolved organic matter except for aluminium; 70% exists as inorganic aluminium whereas most metal contaminants show toxic

effects in their inorganic forms. The dominant ionic species in the landfill leachate are fulvic acid bound metals and metal hydroxides for nickel, copper, zinc, cadmium, lead and iron, whereas manganese is a free ion and aluminium is in form of aluminium tetra hydroxyl radical.

For the urban streams, 25% of the metal ions are bound to DOM, 38% metal species are free metal ions and 19% are in form of inorganic species. The dominant ionic species in urban streams are manganese, nickel, zinc and cadmium ions, iron (III) hydroxide, aluminium tetra hydroxyl radical and fulvic acid bound copper and lead. The tendency of inorganic lead to form highly insoluble salts and complexes with anions as well as its tight binding to soils reduces its bioavailability as has been outlined in Paper I. The speciation of metal ions shows to be bound to DOM or fulvic acid however, if the pH, DOC and calcium content are lowered, the metal binding to biotic ligand will increase.

5.2. Bioavailability of trace metals and Risk assessment

The effects of selected trace metals on life support systems have been studied basing on the criteria from the Swedish Environmental Protection Agency (SEPA, 2000) and are shown in Table 15. Although this method of classification has become absolute and has been replaced by the Water directive, nevertheless it was used in these studies due to a limited number of samples used.

In Table 14, a comparison of the trace metal elements in surface water with the Swedish environmental guidelines for lakes and watercourses, shows that; in some cases all metals show a high risk of negative effects on life support systems even for short time exposure where the levels are capable of causing toxicity effects (values in red) whereas in other cases, there is increased risk of negative effects on biological systems due elevated levels of contaminants (values in orange). In the selected waterways, streams and landfill leachate there were suspected toxicity effects due to lead, copper and zinc with Bugoloobi and Lubigi waterways being most risky.

Moreover the levels of the metals in the aquatic environment that show no effects to moderate effects have been accorded colours from yellow to blue. Those values in blue such as those for arsenic are suspected to cause environmental disturbance but values close to guideline values normally show no effects in organisms moreover those in yellow are concentrations to which local sources or long distance atmospheric contributions have contributed although it may represent natural deviations and no significant effects to life. The green coloured values are concentrations in water that have no anthropogenic contribution and are below guideline values.

Table 12 T-test results for matched pair comparisons of sorption intensities for Cd²⁺ and Pb²⁺ ions onto untreated and base-treated biomass

$\alpha = 0.05$	Std err	t-stat	df	p-value	t-crit	lower	upper	sig	effect r
<i>Albizia coriaria</i> (Cd) T TEST: Equal Variances									
One Tail	0.321	0.789	8	0.226	1.860			no	0.269
Two Tail	0.321	0.789	8	0.453	2.306	-0.487	0.995	no	0.269
T TEST: Unequal Variances									
One Tail	0.321	0.789	7.326	0.227	1.895			no	0.280
Two Tail	0.321	0.789	7.326	0.455	2.365	-0.506	1.013	no	0.280
<i>Musa spp</i> (Cd) T TEST: Equal Variances									
One Tail	0.207	1.018	8	0.169	1.860			no	0.339
Two Tail	0.207	1.018	8	0.339	2.306	-0.267	0.688	no	0.339
T TEST: Unequal Variances									
One Tail	0.207	1.018	7.953	0.169	1.860			no	0.339
Two Tail	0.207	1.018	7.953	0.339	2.306	-0.267	0.688	no	0.339
<i>Erythrina abyssinica</i> (Cd) T TEST: Equal Variances									
One Tail	0.335	0.938	8	0.188	1.860			no	0.315
Two Tail	0.335	0.938	8	0.376	2.306	-1.087	0.458	no	0.315
T TEST: Unequal Variances									
One Tail	0.335	0.938	7.978	0.188	1.860			no	0.315
Two Tail	0.335	0.938	7.978	0.376	2.306	-1.087	0.458	no	0.315
<i>Albizia coriaria</i> (Pb) T TEST: Equal Variances									
One Tail	0.605	0.120	8	0.454	1.860			no	0.042
Two Tail	0.605	0.120	8	0.908	2.306	-1.322	1.467	no	0.042
T TEST: Unequal Variances									
One Tail	0.605	0.120	7.986	0.454	1.860			no	0.042
Two Tail	0.605	0.120	7.986	0.908	2.306	-1.322	1.467	no	0.042
<i>Musa spp</i> (Pb) T TEST: Equal Variances									
One Tail	0.595	0.086	8	0.467	1.860			no	0.030
Two Tail	0.595	0.086	8	0.934	2.306	-1.321	1.424	no	0.030
T TEST: Unequal Variances									
One Tail	0.595	0.086	7.989	0.467	1.860			no	0.030
Two Tail	0.595	0.086	7.989	0.934	2.306	-1.321	1.424	no	0.030
<i>Erythrina abyssinica</i> (Pb) T TEST: Equal Variances									
One Tail	0.592	0.132	8	0.449	1.860			no	0.047
Two Tail	0.592	0.132	8	0.898	2.306	-1.288	1.444	no	0.047
T TEST: Unequal Variances									
One Tail	0.592	0.132	7.984	0.449	1.860			no	0.047
Two Tail	0.592	0.132	7.984	0.898	2.306	-1.288	1.444	no	0.047

Tables 15 and 16 give the description of the criteria that were used in the assessment, classification of risks and toxicity effects due to levels of metal ions in selected water channels and wastewater streams (Paper I).

The results of risk assessment using the levels of elements in the surface water using the Bio-met software tool that is applied over a range of different organisms to calculate the 5th percentile of the species sensitivity distribution (SSD) are presented in Table 17. The HC5 aims at protecting at least 95% of the species and reflects the bioavailability conditions of a specific site according to the water chemistry.

Table 13 Percentage composition of metal ions as determined by speciation studies of surface water samples from selected streams in the Lake Victoria basin, Uganda

Water source	Metal ions	Ni ²⁺	Cu ²⁺	Zn ²⁺	Cd ²⁺	Pb ²⁺	Al ³⁺	Mn ²⁺	Fe ³⁺
Jinja Landfill	Bound-DOM	92.5	100	96.3	87.7	99.9	46.9	24.5	100
	Free ion	7.0	–	2.9	5.2	–	–	65.5	–
	Inorganic	0.5	–	0.8	7.1	0.01	54.1	10.0	–
	Dominant ion	FANi ⁺	FA ₂ CuOH	FA ₂ Zn	FA ₂ Cd	FA ₂ Pb	Al(OH) ₄ ⁻	Mn ²⁺	FA ₂ FeOH
Kiteezi Landfill	Bound-DOM	96.5	100	99.4	97.5	100	13.5	37.1	100
	Free ion	3.4	–	0.4	0.9	–	–	61.9	–
	Inorganic	0.1	–	0.2	1.8	–	86.5	1.0	–
	Dominant ion	FANi ⁺	FA ₂ CuOH	FA ₂ Zn	FA ₂ Cd	FA ₂ Pb	Al(OH) ₄ ⁻	Mn ²⁺	FA ₂ FeOH
Lubigi channel	Bound-DOM	58.5	98.3	35.1	31.8	97.7	4.7	8.2	97.5
	Free ion	40.0	0.7	59.8	58.4	1.6	–	86.3	–
	Inorganic	1.5	1.0	6.1	10.8	1.3	95.3	5.5	2.4
	Dominant ion	Ni ²⁺	FA ₂ Cu	Zn ²⁺	Cd ²⁺	FA ₂ Pb	Al(OH) ₄ ⁻	Mn ²⁺	FA ₂ FeOH
Nakivubo channel	Bound-DOM	9.2	47.1	3.2	2.8	42.6	0.5	0.9	45.7
	Free ion	86.9	27.6	87.9	76.6	38.7	–	90.3	–
	Inorganic	3.9	25.3	8.9	20.6	18.7	99.5	8.8	54.3
	Dominant ion	Ni ²⁺	FA ₂ Cu	Zn ²⁺	Cd ²⁺	FA ₂ Pb	Al(OH) ₄ ⁻	Mn ²⁺	Fe(OH) ₃

In Table 18 and 19, the local HC5 values were in the ranges of 54.56 – 474.12 µg/L for Cu²⁺, 29.95 – 151.90 µg/L for Ni²⁺, 62.55 – 623.74 µg/L for Zn²⁺ and 30.24 – 92.51 µg/L for Pb²⁺ for selected surface water samples. The bioavailable metal concentration is the concentration of a metal that is bioavailable at the site or water body and were in the ranges of 0.02 – 29.40 µg/L for Cu²⁺, 0.18 – 12.45 µg/L for Ni²⁺, 0.96 – 74.19 µg/L for Zn²⁺ and 0.03 – 1.50 µg/L for Pb²⁺.

The risk characterisation ratio (RCR) values that identified potential environmental risks for the selected surface water samples were 15.4% for Cu²⁺, 30.8% for Ni²⁺, 30.8% for Zn²⁺ and 15.4% for Pb²⁺. These results require more detailed risk assessments to inform policy on appropriate action.

The water source with the highest potential risk was Lubigi Channel. Johnson et al. (2017) studied twelve metals in UK surface waters and found that the relative risk to organisms was highest for copper, aluminium and zinc whereas in this report the highest risk of exposure to metals in surface waters was due to zinc and nickel. (Paper I).

Table 14 Comparison of concentrations of selected trace metal elements ($\mu\text{g/L}$) in surface water from selected sources in the Lake Victoria basin, Uganda with Swedish environmental guidelines

Sampling site	Ni	Cu	Zn	Cd	Pb	As
Lubigi channel	178.9	133.2	584.7	1.110	97.7	25.15
Mulago channel	5.8	4.9	25.9	0.047	2.9	0.70
Nakivubo channel	10.9	10.9	66.2	0.255	10.9	1.0
Lugogo stream	6.9	3.3	15.7	0.024	0.9	0.88
Bugoloobi stream	126.8	38.4	506.8	2.307	27.4	6.11
Kiteezi Landfill	271.2	119.6	248.3	1.143	99.3	23.32
Lugazi Steel Mills	19.6	16.4	210.3	0.193	29.0	2.08
Jinja Landfill	104.7	74.1	241.1	0.436	29.7	11.98
Jinja Steel Mills	6.2	1.0	4.9	0.030	0.3	0.46
Jinja Walukuba	0.9	0.5	6.1	0.004	1.0	0.21
River Mpanga	5.5	1.8	15.2	0.477	5.1	0.38
Kibenge hot springs	9.1	7.6	12.8	0.136	2.0	2.96
River Nyamwamba	18.0	172.7	24.2	0.188	2.3	0.27
Kilembe Mines	90.0	160.4	53.4	0.445	2.6	2.03
Kitagata hot springs	2.4	3.6	21.5	0.471	4.9	0.82

Table 15 Naturvårdsverket criteria from Swedish EPA (SEPA, 2000)

Metal	Class 1	Class 2	Class 3	Class 4	Class 5
$\mu\text{g/l}$	Very low concentration	Low concentration	Moderate concentration	High concentration	Very high concentration
Cd	< 0.01	0.01 – 0.1	0.1 – 0.3	0.3 – 1.5	> 1.5
Cu	< 0.5	0.5 – 3	3 – 9	9 – 45	> 45
Ni	< 0.7	0.7 – 15	15 – 45	45 – 225	> 225
Pb	< 0.2	0.2 – 1	1 – 3	3 – 15	> 15
Zn	< 5	5 – 20	20 – 60	60 – 300	> 300
As	< 0.5	0.5 – 3	3 – 8	8 – 40	> 40

Tables 18 and 19 present results of modelling values of levels of elements in the selected shallow ground water sources using the Bio-met software tool that is applied over a range of different organisms to calculate the 5th percentile of the species sensitivity distribution (SSD). The local HC5 values in Tables 11 and 12 were in the ranges of 4.15 – 8.55 $\mu\text{g/L}$ for Cu^{2+} , 8.20 – 9.67 $\mu\text{g/L}$ for

Ni^{2+} , 10.90 – 14.81 $\mu\text{g/L}$ for Zn^{2+} and 2.70 – 5.38 $\mu\text{g/L}$ for Pb^{2+} for selected shallow ground water samples.

The bioavailable metal concentration values were in the ranges of 0.13 – 1.13 $\mu\text{g/L}$ for Cu^{2+} , 1.03 – 12.20 $\mu\text{g/L}$ for Ni^{2+} , 2.94 – 34.88 $\mu\text{g/L}$ for Zn^{2+} and 0.10 – 1.13 $\mu\text{g/L}$ for Pb^{2+} . The risk characterisation ratio (RCR) values that identified potential environmental risks for the selected surface water samples were 4.1% for Cu^{2+} , 45.8% for Ni^{2+} , 62.5% for Zn^{2+} and 8.3% for Pb^{2+} . To concretise these findings, more detailed risk assessments need be undertaken to inform policy on appropriate action since the water sources are used for domestic purposes. The shallow groundwater source with the highest potential risk was Kibumbiro in Lubaga Division whereas those that had RCR values below 1 required no action. (Paper I).

5.3. Characterisation of biosorbents

The biomass surfaces are dominated at the peaks by; 2-Benzyl naphthalene, p'-diaminodiphenylmethane, n-Diallylmelemine and $\text{C}_{18}\text{H}_{14}$, for *Erythrina abyssinica*, while 2,2-bipyridine, cyclotetradecaheptane, 2,2'-bis(2-indenyl) biphenyl and N-ethyltryptamine were dominant in *Coffea canephora*. The prominent species were $\text{C}_{17}\text{H}_{14}$, $\text{C}_{16}\text{H}_{18}\text{N}_2$, $\text{C}_{20}\text{H}_{14}$ and $\text{C}_{20}\text{H}_{18}$ in *Albizia coriaria*, 1,2-diphenyl cyclohexene in *Cyperus papyrus* and 9-methylfluorene, 4-ethylpyrene, sodium cyanurate, 3-beta hydroxy-cholestan-6-one in *Musa spp.*

Several of the surface organic species had aromatic ringed structures rich in pi-electrons as well as surface oxygen and nitrogen containing groups, ligands that are capable of bond formation during the uptake of Cd^{2+} and Pb^{2+} ions by complexation.

Table 16 Criteria used in classification of risks from metal contaminants (SEPA, 2000)

	Class 1. No or only very slight risk of biological effects. The concentrations represent an estimate of concentrations in pristine waters, displaying no signs of anthropogenic impact.
	Class 2. Slight risk of biological effects. The majority of waters in this class have elevated metal concentrations due to emissions from point sources and/or long-distance dispersal. However, the class may include concentrations that are natural in some areas with different geology, for example. The rise in concentration is such that it is not generally possible to detect any effects.
	Class 3. Effects may occur. The risk is greatest in bodies of soft, oligotrophic water, low in humus and in waters with low pH. The term "effects" here means impact on the reproduction or survival of the young of species or groups of species, which is often manifested as a reduction in the number of individuals. This may have repercussions on the communities of organisms in the water and on the structure of the entire ecosystem.
	Class 4. Increased risk of negative biological effects on organisms
	Class 5. Metal concentrations in this class affect the survival of aquatic organisms even where exposure is only short-term. High risk of negative biological effects on life support systems and aquatic organisms even for short-time exposure.

Table 17. Results for risk assessment with respect to zinc, lead, copper and nickel for selected surface water samples

Sample Name	Local HC5 (dissolved) [µg/L]	BioF	Bioavailable Zinc Conc (µg/L)	RCR	Local HC5 (dissolved) [µg/L]	BioF	Bioavailable Lead Conc (µg/L)	RCR
River Mpanga	75.47	0.14	2.05	0.19	65.88	0.02	0.09	0.08
Nakivubo Channel	149.16	0.07	4.76	0.44	75.30	0.02	0.17	0.14
Lubigi Channel	183.54	0.06	34.68	3.18	78.08	0.02	1.50	1.25
Kibenge Hot springs	105.39	0.10	1.22	0.11	68.61	0.02	0.03	0.03
River Nyamwamba	66.13	0.16	3.82	0.35	65.57	0.02	0.04	0.04
Kitagata Hot springs	233.22	0.05	0.96	0.09	86.05	0.01	0.07	0.06
Kilembe Mines tailings	84.19	0.13	6.73	0.62	30.24	0.04	0.10	0.09
Mulago Channel	62.55	0.17	4.34	0.40	40.48	0.03	0.09	0.07
Bugoloobi Channel	74.31	0.15	74.19	6.81	66.34	0.02	0.50	0.41
Lugogo Channel	75.77	0.14	2.11	0.19	40.80	0.03	0.03	0.02
Lugazi Channel	150.28	0.07	15.16	1.39	67.58	0.02	0.51	0.43
Jinja Landfill Leachate	111.33	0.10	23.50	2.16	77.56	0.02	0.46	0.38
Kiteezi Landfill Leachate	623.74	0.02	4.32	0.40	92.51	0.01	1.29	1.07
Sample Name	Local HC5 (dissolved) [µg/L]	BioF	Bioavailable Copper Conc [µg/L]	RCR	Local HC5 (dissolved) [µg/L]	BioF	Bioavailable Nickel Conc (µg/L)	RCR
River Mpanga	119.21	0.01	0.02	0.02	53.20	0.08	0.41	0.10
Nakivubo Channel	185.25	0.01	0.06	0.06	59.21	0.07	0.74	0.18
Lubigi Channel	191.68	0.01	0.69	0.69	57.50	0.07	12.45	3.11
Kibenge Hot springs	227.55	0.00	0.03	0.03	56.31	0.07	0.65	0.16
River Nyamwamba	131.48	0.01	1.32	1.32	42.11	0.09	1.71	0.43
Kitagata Hot springs	159.16	0.01	0.02	0.02	53.90	0.07	0.18	0.04
Kilembe Mines tailings	54.56	0.02	29.40	29.40	29.95	0.13	12.02	3.00
Mulago Channel	75.42	0.01	0.06	0.06	30.80	0.13	0.75	0.19
Bugoloobi Channel	144.60	0.01	0.27	0.27	43.10	0.09	11.79	2.95
Lugogo Channel	82.05	0.01	0.04	0.04	31.76	0.13	0.87	0.22
Lugazi Channel	117.74	0.01	0.14	0.14	39.35	0.10	1.99	0.50
Jinja Landfill Leachate	474.12	0.00	0.16	0.16	151.90	0.03	2.77	0.69
Kiteezi Landfill Leachate	173.20	0.01	0.69	0.69	100.41	0.04	10.80	2.70

The chemical structures of selected notable organic species of the surface structures that were dominant at the peaks in the XRD patterns in Figure 7 are shown in Table 20 and Figure 15. The chemical structures were drawn using ChemBioDraw Ultra 14.0 software.

Table 18. Results for risk assessment with respect to copper and nickel for selected shallow ground water samples

Sample Name	Local HC5 (dissolved) [µg/L]	BioF	Bioavailable Copper Conc [µg/L]	RCR	Local HC5 (dissolved) [µg/L]	BioF	Bioavailable Nickel Conc (µg/L)	RCR
Nabuze	4.95	0.20	0.28	0.28	8.20	0.49	4.78	1.20
Katarina	4.95	0.20	0.64	0.64	8.20	0.49	3.34	0.83
Buwooya	4.95	0.20	0.26	0.26	8.20	0.49	1.93	0.48
Bukoto 1	4.95	0.20	0.17	0.17	8.20	0.49	1.57	0.39
Bukoto 2	7.10	0.14	0.26	0.26	9.67	0.41	2.25	0.56
Nabukalu	7.10	0.14	0.22	0.22	9.67	0.41	3.05	0.76
Abdu	4.15	0.24	0.48	0.48	8.20	0.49	5.91	1.48
SP4306K	4.95	0.20	0.40	0.40	8.20	0.49	4.20	1.05
Kikoni	4.95	0.20	0.09	0.09	8.20	0.49	1.87	0.47
SP1414R	4.95	0.20	0.33	0.33	8.20	0.49	4.45	1.11
Sentamu	4.95	0.20	0.13	0.13	8.20	0.49	3.62	0.90
Kunya	4.95	0.20	0.37	0.37	8.20	0.49	3.01	0.75
SP1212R	4.95	0.20	0.15	0.15	8.20	0.49	4.17	1.04
Kibumbiro	7.10	0.14	0.53	0.53	9.67	0.41	6.33	1.58
Kabaale	4.95	0.20	0.36	0.36	8.20	0.49	2.40	0.60
Nababirye	4.95	0.20	1.13	1.13	8.20	0.49	2.40	0.60
SP1010R	4.95	0.20	0.16	0.16	8.20	0.49	3.96	0.99
Nsambya	4.95	0.20	0.29	0.29	8.20	0.49	7.42	1.85
Kapeke	4.95	0.20	0.70	0.70	8.20	0.49	12.20	3.05
Yusuf	4.95	0.20	0.19	0.19	8.20	0.49	1.03	0.26
Kasanvu	4.95	0.20	0.19	0.19	8.20	0.49	1.71	0.43
Nabagereka	8.55	0.12	0.17	0.17	9.67	0.41	3.70	0.92
Barracks	4.15	0.24	0.26	0.26	8.20	0.49	4.01	1.00
SP5003C	7.10	0.14	0.20	0.20	9.67	0.41	1.87	0.47

Leaching of the organic species as dissolved organic matter occurs at certain pH levels and may complex metal ions to form insoluble compounds which renders the metal ions unavailable for sorption. The functional groups; pi bonds, oxy, hydroxy, carbonyl and carboxylic groups of the organic species in the biosorbent structures are varied and therefore, they form an array of heterogeneous sorption sites for metal ions binding in phases. Biosorbents contain organics as well as major metallic ions such as Ca^{2+} and Mg^{2+} that could leach into the solution during remediation causing sorption reductive or enhancement effects for Cd^{2+} and Pb^{2+} ions remove. Al-Ahmary (2009) studied retention profiles and reported reduction in sorption capacities of lead and cadmium in the presence of calcium and magnesium ions.

Table 19. Results for risk assessment with respect to zinc and lead for selected shallow ground water samples

Sample Name	Local HC5 (dissolved) [µg/L]	BioF	Bioavailable Zinc Conc (µg/L)	RCR	Local HC5 (dissolved) [µg/L]	BioF	Bioavailable Lead Conc (µg/L)	RCR
Nabuze	11.56	0.94	7.93	0.73	2.70	0.44	0.24	0.20
Katarina	10.90	1.00	15.90	1.46	2.70	0.44	0.42	0.35
Buwooya	10.90	1.00	15.00	1.38	2.70	0.44	1.82	1.51
Bukoto 1	10.90	1.00	12.60	1.16	2.70	0.44	0.31	0.26
Bukoto 2	14.81	0.74	18.70	1.72	5.38	0.22	0.43	0.36
Nabukalu	14.81	0.74	14.65	1.34	5.38	0.22	0.19	0.16
Abdu	11.56	0.94	20.74	1.90	2.70	0.44	0.69	0.57
SP4306K	11.56	0.94	12.91	1.18	2.70	0.44	0.60	0.50
Kikoni	11.56	0.94	5.56	0.51	2.70	0.44	0.02	0.01
SP1414R	11.56	0.94	15.18	1.39	2.70	0.44	0.34	0.28
Sentamu	11.56	0.94	7.82	0.72	2.70	0.44	0.10	0.08
Kunya	10.90	1.00	23.60	2.17	2.70	0.44	0.76	0.63
SP1212R	11.56	0.94	8.29	0.76	2.70	0.44	0.10	0.09
Kibumbiro	14.81	0.74	34.67	3.18	5.38	0.22	1.13	0.94
Kabaale	11.56	0.94	14.70	1.35	2.70	0.44	0.94	0.78
Nababirye	10.90	1.00	6.80	0.62	2.70	0.44	0.19	0.16
SP1010R	11.56	0.94	10.46	0.96	2.70	0.44	0.15	0.13
Nsambya	11.56	0.94	10.18	0.93	2.70	0.44	0.32	0.27
Kapeke	11.56	0.94	34.88	3.20	2.70	0.44	0.80	0.67
Yusuf	11.56	0.94	9.90	0.91	2.70	0.44	0.35	0.29
Kasanvu	11.56	0.94	2.94	0.27	2.70	0.44	0.05	0.04
Nabagereka	19.34	0.56	8.40	0.77	5.38	0.22	0.38	0.32
Barracks	11.56	0.94	6.23	0.57	2.70	0.44	0.23	0.19
SP5003C	14.81	0.74	17.74	1.63	5.38	0.22	0.63	0.52

However, a study suggested that Ca and Mn constituent ions of the biomass did not leach to water during the sorption of aqueous metal ions in cation exchange may contribute to the observation that the biomass that contained higher levels of metallic ions showed the highest sorption efficiencies (Paper IV) which indicated occurrence of minimal interference effects of major metallic elements.

Cation Exchange Capacities of selected biosorbents were studied and the observed values for the different materials were above 10 cmol/kg. This indicated that the biosorbent materials had a high capability to hold cations. Untreated materials had a higher potential to hold cations than treated materials. The biomass order from the highest to the lowest negative potential for binding base cations was *Musa spp.* > *C.canephora* > *C.papyrus* for untreated

biomass and *C.canephora* > *musa spp.* > *C.papyrus* for treated materials.

Treating biosorbents was intended to reduce DOC released and improve biosorption of cations. The improvement in metal biosorption for Cd^{2+} was comparable to 2 – 8% reported for *alga spirogyra* (Bishnoi, et al., 2007) and 10 – 20% improvement reported for waste materials (Chen, et al., 2009; Memon, et al., 2007) and the reduction was similar to that of 5 – 13% for Pb bioremoval by treated cork residues (Mota, et al., 2006).

5.4. Biosorption studies of aqueous trace metals

The variation in the biosorption of Cd^{2+} and Pb^{2+} ions observed here may be due to different adsorption mechanisms. Cd^{2+} is well known to exist to a large part in the exchangeable fraction from which explains the biosorption increase after treatment whereas Pb^{2+} is known to bind more strongly to sorbent surfaces and it is therefore expected to be biosorbed through a different adsorption mechanisms. The sorption efficiencies in Figure 13 corresponded with the observation that the metal uptake was expected to closely relate with CEC results in Table 7.

The sorption efficiencies at different pH levels and DOC released into remediated water (Figure 14) were observed to vary in the same order for *C. canephora* husks, *Musa spp.* peels and *C. papyrus* stems. The peak Cd^{2+} ions sorption efficiency values were 83.9% for *C. canephora*, 29.3% for *Musa spp.* peels and 8.7% for *C. papyrus* at pH 2.5. While the peak Pb^{2+} ions sorption efficiencies were 94.3% for *C. canephora* at pH 5.5, 89.2% for *Musa spp.* peels and 99.7% for *C. papyrus* at pH 2.5. Although the DOC released into remediated water samples was nearly constant with increase in pH, the peak DOC values in Cd^{2+} remediated solutions were; 91.3mg/L for *C. canephora* and 69.6mg/L for *Musa spp.* peels at pH 3.5 and 48.5mg/L for *C. papyrus* at pH 4.5. Whereas the peak DOC values in Pb^{2+} remediated water were; 98.0mg/L for *C. canephora* at pH 6.5, 94.5mg/L for *Musa spp.* peels at pH 3.5 and 97.8mg/L for *C. papyrus* at pH 5.5. For all biosorbents, sorption performance for Cd^{2+} ions was lower than that for Pb^{2+} ions and nearly linear over the pH range 2.5 – 6.5.

The sorption of Cd^{2+} and Pb^{2+} ions in the pH range studied showed that they were dominant and preferred over H_3O^+ ions. The optimal pH for the removal of Cd^{2+} and Pb^{2+} ions from aqueous solutions for the biomass studied was between 4.5. The sorption performance for Cd^{2+} ions was in the order; *C. canephora* > *Musa spp* > *C. papyrus*. The sorption performance for Pb^{2+} ions was in the order; *C. canephora* > *C. papyrus* > *Musa spp* for non-competitive sorption. The conditions in batch studies differ from those in column studies, but for comparison purposes, the results showed that the effects of DOC on the sorption efficiencies of the materials were in agreement with the reports by Nehrenheim (2008) and Kalmykova (2009) which indicated that DOC had only

minor effects on metal sorption and transport in column studies. Therefore the DOC released into remediated solutions could have effects on the residual metal ions and not on sorbed metal ions.

The amounts of DOC (mg/L) released by the biosorbents into treated water were not dependent on the initial pH and had no observable effects on the biosorption capacities of the treated biosorbents for Cd^{2+} and Pb^{2+} ions. A comparison of DOC released by untreated and base treated materials showed that it did not vary significantly with increase in pH as well as the increase and decrease in biosorption capacities for Cd^{2+} and Pb^{2+} ions.

Table 20. Notable organic species identified by numbers in the XRD patterns

Peak	2Theta	Peak height(counts)	d (Å)	Compound
1	15.332	2316	5.7743	$\text{C}_{17}\text{H}_{14}$
2	22.123	2256	4.0146	$\text{C}_{16}\text{H}_{18}\text{N}_2$
3	24.403	2207	3.6448	$\text{C}_{19}\text{H}_{16}$
4	30.814	1108.8	2.8995	$\text{C}_{20}\text{H}_{18}$
5	38.223	1186	2.3528	$\text{C}_{20}\text{H}_{14}$
6	14.904	11784	5.9389	$\text{C}_{10}\text{H}_8\text{N}_2$
7	21.458	1859	4.1375	$\text{C}_{12}\text{H}_{16}\text{N}_2$
8	15.332	2527	5.7743	$\text{C}_{16}\text{H}_{17}\text{N}_5$
9	22.028	2999	4.0220	$\text{C}_{15}\text{H}_{13}\text{N}$
10	34.803	846	2.5755	$\text{C}_{30}\text{H}_{22}$
11	17.186	2169	5.1552	$\text{C}_{14}\text{H}_{12}$
12	21.889	1794	4.0570	$\text{C}_3\text{H}_2\text{N}_3\text{NaO}_3$
13	30.107	1150	2.9657	$\text{C}_{20}\text{H}_{18}$
14	34.810	1051	2.5750	$\text{C}_{19}\text{H}_{14}$
15	14.953	1322	5.1916	C_7H_{14}
16	21.984	1703	4.0397	$\text{C}_{18}\text{H}_{14}$
17	24.312	2922	3.6579	$\text{C}_{17}\text{H}_{14}$
18	34.482	806	2.5991	$\text{C}_{13}\text{H}_{14}\text{N}_2$
19	38.231	894	2.3437	$\text{C}_{54}\text{H}_{110}$
20	47.067	643	1.9291	$\text{C}_{18}\text{H}_{22}$

The results showed that removal of soluble organics from the biosorbents did not have any effects on the biosorption efficiency for the trace metal ions in this study.

A comparison of DOC released by untreated and base treated materials showed that it did not vary significantly with increase in pH as well as the increase and decrease in biosorption capacities for Cd^{2+} and Pb^{2+} ions. The results showed that removal of soluble organics from the biosorbents did not have any effects on the biosorption efficiency for the trace metal ions in this study.

To determine the relationship between sorption intensities and DOC released by the materials during sorption, correlation studies were done. The coefficient of determination, r for Cd^{2+} ions uptake was 0.992 for *Musa spp.* peels and 0.84 for *C. canephora* moreover the coefficient of determination, r for Pb^{2+} ions was 0.959 for *C. canephora* and 0.962 for *C. papyrus*. These coefficients of determination were strong whereas weak coefficients of determination, r were observed for Cd^{2+} ions sorption onto *C. papyrus*; 0.553 as well as for Pb^{2+} ions sorption onto *Musa spp.* peels; 0.707. The strong coefficients of determination, r values showed that there was an association between the amount of remediated metal ions and DOC released from biomass whereas the weak correlations indicated weak relationships. This observation is consistent with the observation by Gaballah, et al. (1997) and Bailey, et al. (1999) who suggested that association of soluble organics and metal ions leads to formation of complex species. Complexation with DOC in solution may remove aqueous trace metals from solution other than sorption onto biomass.

Sorption intensities were time dependent and studies revealed that the peak values for Cd^{2+} biosorption using untreated biomass were 1.538 mg/g for *A. coriaria*, 1.414 mg/g for *E. abyssinica* and 1.467 mg/g for *Musa spp.* in 3.5 hours. Moreover the peak sorption for Cd^{2+} ions using base treated materials were 1.760 mg/g for *A. coriaria*, 1.753 mg/g for *E. abyssinica* and 1.737 mg/g for *Musa spp.* in 3.5 hours. The treated materials showed improved performance in removing the Cd^{2+} ions. For Pb^{2+} ions sorption, the plots were near even throughout the study time. Nevertheless, the peak intensity values for Pb^{2+} ions using untreated biomass were 1.372 mg/g for *A. coriaria*, 1.326 mg/g for *E. abyssinica* and 1.350 mg/g for *Musa spp.* while for the base treated materials, the peak intensity values occurred at 1.347 mg/g for *A. coriaria*, 1.325 mg/g for *E. abyssinica* and 1.320 mg/g for *Musa spp.* in 3.5 hours. Results from these studies further indicated that biosorption intensities for Cd^{2+} and Pb^{2+} ions were in the order; *A. coriaria* > *Musa spp.* > *E. abyssinica* for untreated materials and *A. coriaria* > *E. abyssinica* > *Musa spp.* for base-treated biosorbents. The optimum agitation time was 3.5 hours for Cd^{2+} and Pb^{2+} ions sorption onto untreated and base treated materials for 10 mg/L biosorbent dosage (Paper I). The peak values for Cd^{2+} ions biosorption were 1.33 mg/g for *C. canephora* and 0.411 mg/g for *Musa spp.* peels in 1 hour, then 0.164 mg/g for *C. papyrus* in 2 hours.

Table 21. Co-ionic effects on the sorption process in binary-component solutions at 297K (Paper IV)

Sorption capacity ratio q_{mix}/q_s for peroxide treated biosorbents						
	<i>C. canephora</i>		<i>C. papyrus</i>		<i>Musa spp.</i>	
pH	Cd ²⁺	Pb ²⁺	Cd ²⁺	Pb ²⁺	Cd ²⁺	Pb ²⁺
2.5	0.006	0.995	0.154	0.308	0.141	0.661
3.5	0.071	0.894	0.882	0.410	0.255	0.792
4.5	0.081	0.929	0.225	0.576	0.141	0.764
5.5	0.052	0.847	0.800	0.529	0.051	0.690
6.5	0.080	0.850	0.870	0.462	0.190	0.755

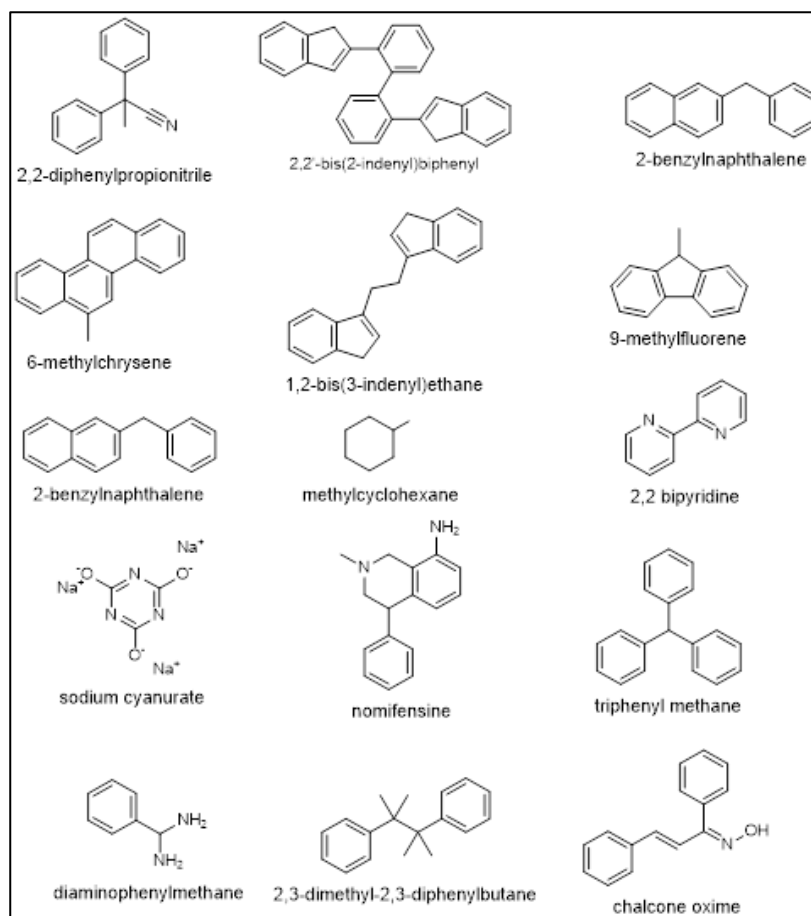


Figure 15. Some of the chemical structures of selected notable organic species in the biosorbent materials studied.

For Pb^{2+} ions sorption, the plots for *Musa spp.* and *C. canephora* were near even throughout the study time.

The plots for *C. papyrus* showed steady increase in sorption intensity from 1 to 3 hours. The peak Pb^{2+} ions sorption values were 1.574 mg/g for *C. canephora*, 1.546 mg/g for *C. papyrus* and 1.528 mg/g for *Musa spp.* in 3 hours.

Under the optimal experimental conditions, sorption studies showed that the highest sorption capacities for Cd^{2+} and Pb^{2+} ions onto biosorbents were in the order of *C. canephora* > *Musa spp.* > *C. papyrus*. The optimum agitation time was determined as 3.0 hours for Cd^{2+} and Pb^{2+} ions biosorption onto peroxide treated materials at $24 \pm 1^\circ\text{C}$ for 12.5 mg/L biosorbent dosage. These results were consistent with those in literature where optimum agitation time for Cd^{2+} and Pb^{2+} ions biosorption was reported as 2.0 – 4.0 hours (Özer & Pirincci, 2006) (Paper IV).

5.4.1. Comparative biosorption studies

On the other hand, comparative studies showed that base treated materials had higher trace metals biosorption performance than the peroxide treated materials. Both base treated and untreated biosorbents showed sharp increases in biosorption intensities for Cd^{2+} ions from pH 2.0 to pH 4.3 followed by a gradual decrease in biosorption occurred with increase in initial pH of aqueous media. The base treated biosorbents had better biosorption intensities for Cd^{2+} ions than the untreated materials at all pH levels which was in the order; *A. coriaria* > *E. abyssinica* > *Musa spp.* The Pb^{2+} ions biosorption slightly increased for treated biosorbents between pH 2.0 and 4.0, otherwise there was a decline in biosorption intensities of all biosorbents for Pb^{2+} ions to minima at pH 9 (Paper II). In aqueous solution lead hydrolyses at about pH 7.5 although even at low pH levels it is strongly bound by oxygen containing functional groups of humic substances, oxides and surface oxygen groups of the biosorbent materials. Results from different sorption studies are difficult to compare due to variations in experimental conditions adopted by different scholars, nevertheless the results of the present studies concur with results from earlier studies elsewhere where it was reported that the optimal pH for biosorption for trace metals by other biosorbents as 4.0 – 5.0 (Kaewsam et. al., 2008; Dekhil et. al., 2011).

The biosorption intensities were indicative of ionic competition for biosorption sites on biosorbents with the predominant ions; H_3O^+ ions at pH levels lower than 3.5, the trace metal cations at the optimal pH (4.2 – 4.7) and the OH^- ions at pH levels than 5.0. The data suggested the optimal pH range for biosorption of trace metal ions; Cd^{2+} and Pb^{2+} to be 4.2 – 4.7 over which the metal cations were predominantly free metal ions and had the highest interaction and binding with the biosorbents structural molecules.

At low pH levels the H_3O^+ ions shield Cd^{2+} from the negatively charged biosorption sites although due to pH dependent

dissociation of organic matter, there aren't many negatively charged sites. Whereas at pH levels above 5.0 for Pb^{2+} ions and pH above 8.0 for Cd^{2+} ions, several hydroxyl complex species are predominant in solution which greatly negatively impacts on their biosorption onto biosorbents. In other studies by Patnukao, et al., 2008 and Sari & Tuzen, 2009 clarified that at pH levels higher than 5.0, Cd^{2+} and Pb^{2+} ions complex with OH^- to form soluble hydroxylated species of the metal ions.

5.4.2. Competitive sorption studies

Comparative and competitive sorption studies done showed that Cd^{2+} ions biosorption intensity in single-component aqueous media was in the ranges of 0.045 – 1.343 mg/g and 0.008 – 0.195 mg/g in binary-component solutions. The Cd^{2+} ions biosorption suppression was most pronounced in *C. canephora* and least in *C. papyrus* during competitive biosorption. For Pb^{2+} ions, the ranges of biosorption intensities were between 0.819 – 1.595 mg/g in single-component solutions and 0.472 – 1.303 mg/g in multi-component solutions. The results implied that there was a much weaker competition for biosorption sites for Cd^{2+} ions in multi-component solutions than that for Pb^{2+} ions under similar conditions in binary component solutions.

Lead is strongly bound to humic substances and other variable-charge species such as hydroxyl, carboxyl and phenolic groups through complexation.

The Cd^{2+} ions biosorption was spasmodic compared to that of Pb^{2+} ions biosorption which was near linear. Cadmium sorption is mainly by surface complexation or ion exchange although its retention is not as strong as that of lead. Therefore, for all biosorbents, there was preferential Pb^{2+} ions sorption to that of Cd^{2+} ions. The Pb^{2+} ions adsorption was higher (0.472 – 1.303 mg/g) than that for Cd^{2+} ions (0.008 – 0.195 mg/g) in multi-component solutions. From the results of biosorption of metal ions from multi-component solutions, the performance of the biosorbents was in the order of *C. canephora* > *Musa spp.* > *C. papyrus* (Paper IV). For both metal ions, the biosorption was relatively low compared to that in sole metal solutions (Figure 22). The results from multi-component solutions were consistent with the observations by Khani (2013) in which stated that the uptake of both cadmium and lead in the presence of one another decreased.

The co-ion effects on biosorption of metal ions in binary solutions due to ionic interactions of Cd^{2+} and Pb^{2+} ions were predicted using the ratio of the sorption intensity for one metal ion in the mixture, q_{mix} to the sorption intensity of the same metal ion when it is alone in solution, q_s , such that;

$q_{\text{mix}}/q_s > 1$; Adsorption is promoted by the presence of other metal ions,

$q_{mix}/q_s = 1$; No observable net interaction effects,

$q_{mix}/q_s < 1$; Adsorption is suppressed by the presence of other metal ions (Tan, et al., 1985).

The results of co-ion effects analysis for all biosorbents studied are presented, the ratios were in the range of 0.048 – 0.800 for Cd^{2+} ions biosorption and 0.361 – 0.929 for Pb^{2+} ions biosorption which implied that biosorption for either metal ions was suppressed by the presence of other metal ions in the multi-component solutions. Cd^{2+} ions biosorption was more suppressed than Pb^{2+} ions biosorption in multi-component solutions (Paper IV). These results were consistent with findings by Khani (2013) in which ratios of 0.85 for Cd^{2+} ions and 0.47 for Pb^{2+} ions sorption in binary solutions were reported. The nature of metal ions sorption in single component solutions differs from that in binary component solutions due to the sorbate-sorbate and sorbate-sorbent interactions.

The trend of sorption intensities for Cd^{2+} ions were uncharacteristic and analogous for both single – and binary – component solutions except for *C. canephora* that was curvilinear in single – component solutions. The sorption intensities for both metal ions were in the order: *C. canephora* > *Musa spp* > *C. papyrus*. (Paper IV). From the comparative plots of sorption intensities with initial metal ions concentration in Figure 22, it was evident that increasing the initial metal ions concentration under the experimental conditions positively influenced sorption of Pb^{2+} ions although no significant effect on the Cd^{2+} ions sorption was observed between single – and binary – component solutions. The results for binary-component solutions showed that the biosorbents had stronger physical affinity for Pb^{2+} ions than Cd^{2+} ions most likely due to the difference in their ionic radii; Pb^{2+} (1.19Å) and Cd^{2+} (0.97Å) in addition to the hydrated ionic radii; Pb^{2+} (4.61Å) and Cd^{2+} (4.01Å). All the peroxide treated biosorbents showed lower sorption intensities for Cd^{2+} ions in single- and binary-component solutions than the base treated materials although the performance was in the order: *C. canephora* ≥ *Musa spp* ≥ *C. papyrus*. Pb^{2+} ions were preferentially removed from solution compared to Cd^{2+} ions although all metal ions sorption from binary-component solutions was suppressed when compared to sorption in single-component solutions which were similar to findings by Khani (Khani, 2013) (Paper IV). Generally, increasing the initial metal concentration results in an increase in the biosorption intensity since it gives the sorbate ions momentum to overcome mass transfer resistance in the system.

The Pb^{2+} ions uptake onto base-treated biomass with increase in initial concentration C_0 , 5 – 100 mg/l for Pb^{2+} ions was in the increasing order for all biomass was 94.16% – 98.67% for *A. coriaria*, 92.08% – 96.38% for *Musa spp*. and 81.4% – 94.69% for *E. abyssinica* was observed. For Pb^{2+} ions uptake onto untreated biosorbents it was observed that the sorption intensity was in the

decreasing order of 95.64% – 71.50% for *A. coriaria*, 92.86% – 71.50% for *Musa spp.* and 92.82% – 60.61% for *E. abyssinica*. The biosorbents exhibited higher sorption intensity for Pb^{2+} ions than for Cd^{2+} ions. The biosorbent sorption performance for Pb^{2+} ions for both untreated and base treated biomass was in the order of *A. coriaria* > *Musa spp.* > *E. abyssinica*. It is evident that the base treatment changes the chemical nature of the biomass structural and surface molecules which consequently alters the metal ions uptake. No saturation of binding sites for uptake of Pb^{2+} ions was observed in both untreated and base treated biomass whereas the biomass exhibited less affinity for the Cd^{2+} ions which could be attributed to different sorption mechanisms of metal ions uptake. The results in this study are comparable to those reported by Kapoor, et al. (1999) where untreated and base treated *Aspergillus niger* presented; 2.25 mg/g for untreated and 7.24 mg/g for treated biomass for Pb^{2+} ions and 1.31 mg/g for untreated and 3.43 mg/g for treated biomass for Cd^{2+} ions. For both Cd^{2+} and Pb^{2+} ions, the sorption performance for the biosorbents was in the order: *A. coriaria* > *Musa spp.* > *E. abyssinica*. The sorption intensities for the biosorbents corresponded to the observation that the metal uptake was expected to be closely related to cation exchange capacity results. (Paper IV).

5.4.3. Sorption kinetic studies

Kinetic studies of Cd^{2+} and Pb^{2+} ions biosorption onto biosorbents showed that the second-order kinetic model (Ho model) gave a better description of the biosorption processes. These findings were consistent with sorption studies reviewed by Wan Ngah & Hanafiah, (2008) and the generally held opinion that the biosorption processes follow pseudo second order kinetics.

The trace metal biosorption data for all peroxide treated biosorbents were not well represented by the Lagergren model except for *C. papyrus* and *C. canephora* where the regression coefficient, R^2 was >0.93 for agitation time 0 – 1 hour. Otherwise, the regression coefficient (R^2) values of the Pseudo-second-order model indicated that it fitted best the experimental biosorption data for both Cd^{2+} and Pb^{2+} ions for all the biosorbents and were close to unity ($0.89 \leq R^2 \leq 1.00$). It was observed that the experimental data for Cd^{2+} ions sorption for all biosorbents did not fit the Pseudo-first-order (Lagergren) model although that for Pb^{2+} ions sorption onto treated biosorbents gave good regression coefficient (R^2) values in the range between 0.818 and 0.908 for both base treated and untreated materials between 0.713 and 0.973 for peroxide treated materials.

The Lagergren model could not be used to explain the underlying Cd^{2+} and Pb^{2+} ions biosorption mechanisms. The pseudo-second-order model regression coefficients were in the range $0.9978 \leq R^2 \leq 0.9999$ for base treated and untreated biosorbents and $0.892 \leq R^2 \leq 1.000$ for peroxide treated materials. Therefore, the Cd^{2+} and Pb^{2+}

ions biosorption follows the Ho model implying that the reaction mechanism was chemisorption.

5.4.4. Sorption equilibrium studies

Equilibrium studies revealed that Temkin and Freundlich models fitted Pb^{2+} ions biosorption data for both treated and untreated biosorbents better than they did for Cd^{2+} ions biosorption data. The values of the Langmuir and Freundlich parameters showed that the models explained the sorption by untreated biosorbents better than they did for base treated biosorbents. (Paper II, III & VI). For the Langmuir isotherm, the b values for Pb^{2+} ions sorption were superior to those of Cd^{2+} ions which signified that the biosorbents had higher affinity for Pb^{2+} ions than Cd^{2+} ions. Freundlich isotherm fitted Pb^{2+} ions biosorption data for treated biosorbents. The values of Temkin isotherm parameters A_T and b_T (Table 10) were high and in the same range as those reported by Otun, et al., (2006) for the adsorption of Pb^{2+} , Ni^{2+} and Cd^{2+} ions onto powdered egg shells. The values of A_T and b_T implied that the biosorption intensities for Cd^{2+} and Pb^{2+} ions were high for all biosorbents studied and that the sorption processes were complex in nature (Paper II, III & IV). The regression coefficient, R^2 values for Temkin model fitting of Pb^{2+} ions biosorption suggested that it could describe the sorption processes onto untreated and treated biosorbents.

The three models tested fit Pb^{2+} ions biosorption data better than the Cd^{2+} ions biosorption data for both single- and binary-component solutions. For the Langmuir sorption isotherm, the b values for the sorption data of the peroxide treated biosorbents implied high affinity for Cd^{2+} and Pb^{2+} ions. For the Freundlich sorption isotherm, the high b_F values for sorption of Cd^{2+} and Pb^{2+} ions onto peroxide-treated biosorbents indicated that these sorbents have heterogeneous site binding affinities while the values of A_T and b_T in the Temkin sorption isotherm were high and implied higher sorption capacities for Cd^{2+} and Pb^{2+} ions especially in binary-component solutions. To a good extent Temkin isotherm fits sorption data for Cd^{2+} and Pb^{2+} ions in binary-component solutions (Paper IV).

The smaller values of b_F imply stronger interaction between biosorbent and Pb^{2+} ions onto treated biomass whereas the b_F values for untreated biomass indicated linear adsorption leading to identical adsorption energies for all sites. The A_F values correspond to sorption intensity and therefore the higher they are the higher the sorption intensity (Igwe & Abia, 2007). The high A_F values implied that the maximum sorption intensity for Freundlich isotherm was high for both types of biomass. The Freundlich model explains the uptake of Pb^{2+} ions and predicts a multi-layer sorption process onto both untreated and base treated biomass. The model fitted the sorption data in the order; *A. coriaria* \geq *E. abyssinica* $>$ *Musa spp.* The results show that the Temkin isotherm fits to the Cd^{2+} ions sorption data with linear regression coefficient

$0.8514 \leq R^2 \leq 0.9446$ for uptake of Cd^{2+} ions onto untreated biomass and $0.6209 \leq R^2 \leq 0.9643$ for uptake onto base treated materials. The parameter values in this study are comparable to those reported by Kapoor et al., (1999) and Otun et al. (2006). The Langmuir model generally presented the best fits for the Cd^{2+} and Pb^{2+} ions sorption data for all biosorbents. The fitting of experimental data to Freundlich and Langmuir isotherm models suggested that the biosorption process was a monolayer capacitive sorption process in which heterogeneous surface conditions co-existed.

5.4.5. Correlation and student's T-test

The correlation studies for agreement between sorption intensities for base-treated and untreated biomass presented r values that were very high and close to unity which implied that the two sets of biosorbents exhibited a higher degree of agreement although the sorption intensities for Pb^{2+} ions were more correlated and agreeable than those of Cd^{2+} ions.

Student's T-test was done and the results for both equal and unequal variances for matched pair comparisons of sorption intensities for Cd^{2+} and Pb^{2+} ions onto untreated and base-treated biosorbents were obtained. The results of computation of a paired T-test showed that $0.00003 \leq P \leq 0.0038$. P was the probability of obtaining values for the test statistic that were more to the extreme than the actual values observed. Therefore, there is very strong evidence that the sorption intensities of the base-treated and untreated biosorbents were not significantly different for biosorption of Cd^{2+} and Pb^{2+} ions for both unequal and equal variances. Since there was considerable agreement between the two types of biosorbents, then the choice for treatment of biosorbents given the advantage of reduced release of soluble organics from biosorbents into the remediated solutions should be taken on that basis.

6. CONCLUSIONS

The shallow groundwater from protected springs in Kampala capital city showed elevated levels of major and trace metals whereas the surface water, wastewater, landfill leachates and surface runoff were contaminated with trace metals and nutrients as well as nitrates. The metal levels in surface water, landfill leachate and surface runoff indicated anthropogenic contamination and revealed increased risks of negative biological effects on organisms according to the Swedish environmental guidelines. Toxicity studies using the Bio-met software tool showed that selected surface water sources and shallow groundwater sources had potential environmental risks and require further detailed risk assessments to inform policy on appropriate action. The cation exchange capacities of the biomass agreed with the trace metals'

uptake but could not be dependable to draw conclusions on the contribution of ion exchange to the overall metal sorption. The constituent Ca, K, Mg and Na ions in the materials contributed to the cation exchange capacities of the materials and the sorption efficiencies although the toxic trace metals were in minute concentrations and showed no contamination of the remediated water. The biosorbent surface structures components were electron rich species and oxygen groups that were most likely responsible for binding trace metal ions.

Chemical treatment reduced dissolved organics released into remediated water and the dissolved organics were found to have no observable effect on Cd^{2+} and Pb^{2+} ions sorption in batch studies. Base treated biosorbents showed enhanced Cd^{2+} ions removal but exhibited reductive effects on Pb^{2+} ions sorption efficiencies although comparative studies exhibited agreement between the materials and no significant differences in performance of either the treated or the untreated materials. Therefore, the materials should be treated due to the fact that it has advantages and but not that it enhances sorption performance. The maximum sorption intensities for treated biosorbents were achieved at pH range; 3.5 – 5.0 for Cd^{2+} and Pb^{2+} ions in 3.5 hours of contact time.

The batch sorption data was adequately described by the Ho model with regression coefficient (R^2) values; 0.9978 – 0.9999 for Cd^{2+} and Pb^{2+} ions which indicated that the sorption process mainly occurred with chemisorption reaction mechanism although other mechanisms have some contribution to the overall sorption process. The base treated biosorbents were more appropriate for Cd^{2+} ions removal whereas the untreated materials were more suitable for Pb^{2+} ions sorption.

The Cd^{2+} and Pb^{2+} ions sorption from binary component solutions was suppressed although the synergistic effects were greater for Cd^{2+} than Pb^{2+} ions. Pb^{2+} ions were preferentially removed in binary-component solutions even when Cd^{2+} ions concentration was more than that the Pb^{2+} ions.

Langmuir equilibrium model offered the best fits and described best the Cd^{2+} and Pb^{2+} ions sorption from aqueous media for treated and untreated biosorbents. The Langmuir isotherm fitted the non-competitive metal sorption data whereas the Freundlich isotherm fitted the competitive metal sorption data better which implied that the sorption process was complex and involved several metal distribution mechanisms onto the biomass surfaces.

Sorption of trace metals is feasible and the biosorbents; *A. coriaria*, *C. canephora*, *C. papyrus*, *E. abyssinica* and *Musa spp* were found to be potential biosorbents for removal of Cd^{2+} and Pb^{2+} ions in minute concentrations from contaminated aqueous media.

6.1. Implications

The environmental implication of this study is that waste biomass was found to be usable as low cost alternative biosorbents for removing trace metal ions from contaminated aqueous media.

On the other hand the waste biomass disposal burden will be minimised with reapplication in water treatment. The socio-economic implication of these studies is that the vulnerability of children and women due to exposure to trace metal contaminants in contaminated water can be reduced through the sorption onto biosorbents.

6.2. Recommendations

Laboratory scale experiments do not adequately represent on-site environmental conditions and variables which play an important role in trace metals uptake onto biosorbents. There is need to examine the sorption performance of biosorbents from *Coffea canephora* husks, *Cyperus papyrus* and *Musa spp* peels in continuous flow studies for removal of Cd^{2+} and Pb^{2+} ions from real contaminated surface water from industrial centres, urban streams and channels as well as leachate from municipal landfills.

There is need for future research considering pilot and field filter beds for various applications to remediation of real contaminated surface and shallow groundwater. Intervention through monitoring and remediation of wastewater is inevitable to cease the increasing trend and inflow of surface runoff into peri-urban aquifers and avoid high concentrations of metals in the soil water.

Future studies need to be done to build a time series data base on total water quality, regular monitoring of groundwater quality, enforcing regulation and preventive remediation and to predict trace metal contaminants mobility and distribution in the environment need be done to control trace metals and protect the meagre resource.

To study reapplication and sustainability biosorption process, the sorption – desorption studies need be done for the biosorbents in this study.

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8. APPENDIX

- I. **G. K. Bakyayita**, A. C. Norrström & R. N. Kulabako, 2019. Assessment of levels, speciation and toxicity of trace metal contaminants in selected shallow groundwater sources, surface runoff, wastewater and surface water from designated streams in Lake Victoria Basin, Uganda. *Journal of Environmental and Public Health*, Vol. 2019, Article ID 6734017, 18 pages, ISSN 1687-9805, E-ISSN 1687-9813, <https://doi.org/10.1155/2019/6734017>
- II. **G. K. Bakyayita**, A. C. Norrström, M. Nalubega & R. N. Kulabako, 2014. Kinetic studies of Cd (II) and Pb (II) ions biosorption from aqueous media using untreated and chemically treated biosorbents. *Water Science and Technology* 69(11),pp 2230-2236 , doi: 10.21.2166/wst.2014.147
- III. **G.K. Bakyayita**, A.C. Norrström, & R.N. Kulabako, 2015. Characterisation and use of untreated and base-treated biosorbents from *Albizia coriaria*, *Erythrina abyssinica* and *Musa spp.* in the uptake of Cd (II) and Pb (II) ions from contaminated water (Manuscript)
- IV. **G. K. Bakyayita**, A. C. Norrström, and R. N. Kulabako, 2015. Competitive and Noncompetitive Batch Sorption Studies of Aqueous Cd (II) and Pb (II) Uptake onto *Coffea canephora* Husks, *Cyperus papyrus* Stems, and *Musa spp.* Peels. *Journal of Chemistry*, Vol. 2015, Article ID 696098, 17 pages, 2015. <http://doi:10.1155/2015/696098>