

# Water Clarification and Selected Heavy Metal Biosorption by *Maerua decumbens* root tubers

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**ABSTRACT** :This research considers *Maerua decumbens* root tuber powder as natural coagulant and biosorbent. The aim was to investigate efficiency of *Maerua decumbens* root tuber powder, as primary coagulants and coagulant aid in reducing turbidity from turbid Ndarugo river water and to determine reduction of physical, chemical and bacteriological parameters in clarified Ndarugo river water. The other objective of this research was to optimize various sorption parameters during biosorption of Cd (II), Cr (II), Cu (II) and Pb (II) on *Maerua decumbens* root tuber powder. The optimum dosage was established at 1.25mg/L *Maerua decumbens* root tuber powder. Physical, chemical and bacteriological parameters of clarified water were reduced to international guidelines. In biosorption studies, optimum pH was 5.0 for Cu(II) and Cd(II), 5.2 for Cr and Pb(II) had optimum pH of 5.5. Contact time was established after 30 min for Cr(II), Cd(II) and Cu(II) and 40 min for Pb(II). FTIR analysis for *Maerua decumbens* root tuber powder revealed the presence of amino, carboxyl, hydroxyl, and carbonyl groups. These functional groups could be responsible for biosorption of Cr (II), Cr (II), Cu (II) and Pb (II). Biosorption kinetics data fitted well with the pseudo-second-order kinetic model. The biosorption isotherms for the biosorbent fitted well with Freundlich isotherm model than with Langmuir isotherm model. However, the separation factor  $R_L$  of Langmuir model was between 0 and 1 indicating favorable sorption process. The study showed that this natural coagulant had good coagulation processes and was a good biosorbent for the removal of Cr (II), Cd (II), Cu (II) and Pb (II) from aqueous solutions.

**Key words:** Biosorption, *Maerua decumbens*, Langmuir, Freundlich, coagulant, bacteriological, sorption parameters and FTIR

## 1. INTRODUCTION

Groundwater is the available source for drinking water in most rural areas of developing countries especially in Africa and it generally requires no or minimal treatment. In the event that no suitable aquifers are available, relatively clean waters from lakes or streams are preferred which is easily boiled for purification. However, only simple, practical technologies such as gravity chemical feed with solutions and flocculation, horizontal-flow sedimentation, and manually operated filters should be used for treatment of such waters (13). Natural polyelectrolytes of plant origin (seeds, stems and roots) have been used for many centuries in developing countries for clarifying and purifying turbid water (13). For home water treatment, the materials have to be used in the form of powder or paste, most of which consists of substances other than the polyelectrolytes. Even under such conditions, a few plants make effective coagulants (10); for example, seeds of the plant species of the family *Loganiaceae*- *Strychnos potatorum*, and *Moringaceae* – *Moringa oleifera* and *Moringa stenopetala*. *S. potatorum* is a small tree occurring abundantly in tropical regions. Among the different species of *Moringaceae*, *M. oleifera* is the most widely occurring tree. Reference to the use of *S. potatorum* seed to clarify turbid water is available in some countries (2), and it is still in use in the villages of Maharashtra and Tamil Nadu in India. The village women of Sudan also use *M. oleifera* seed for home water treatment (9). In laboratory and field studies, seeds

of *S. potatorum* and *M. oleifera* have shown promise as coagulant in the clarification of turbid water (14,4). Direct filtration with *S. potatorum* seed as coagulant appeared effective in clarifying a low turbidity water (1,12). The aim of this work was to investigate whether *Maerua decumbens* are suitable biosorbent for the removal of Cd, Cr, Cu and Pb from several model solutions. Concentration of heavy metals in treated model solutions were determined using atomic absorption flame emission spectrometry AAFES (AA-6200 Shimadzu).

Fourier Transform Infrared Spectrophotometer 8400CE (Shimadzu, Japan) fitted with a pellet cell, was used to identify the functional groups of *Maerua decumbens* biosorption properties. The adsorption capacities were evaluated from the equilibrium of adsorption isotherms.

## 2. MATERIALS AND METHODS

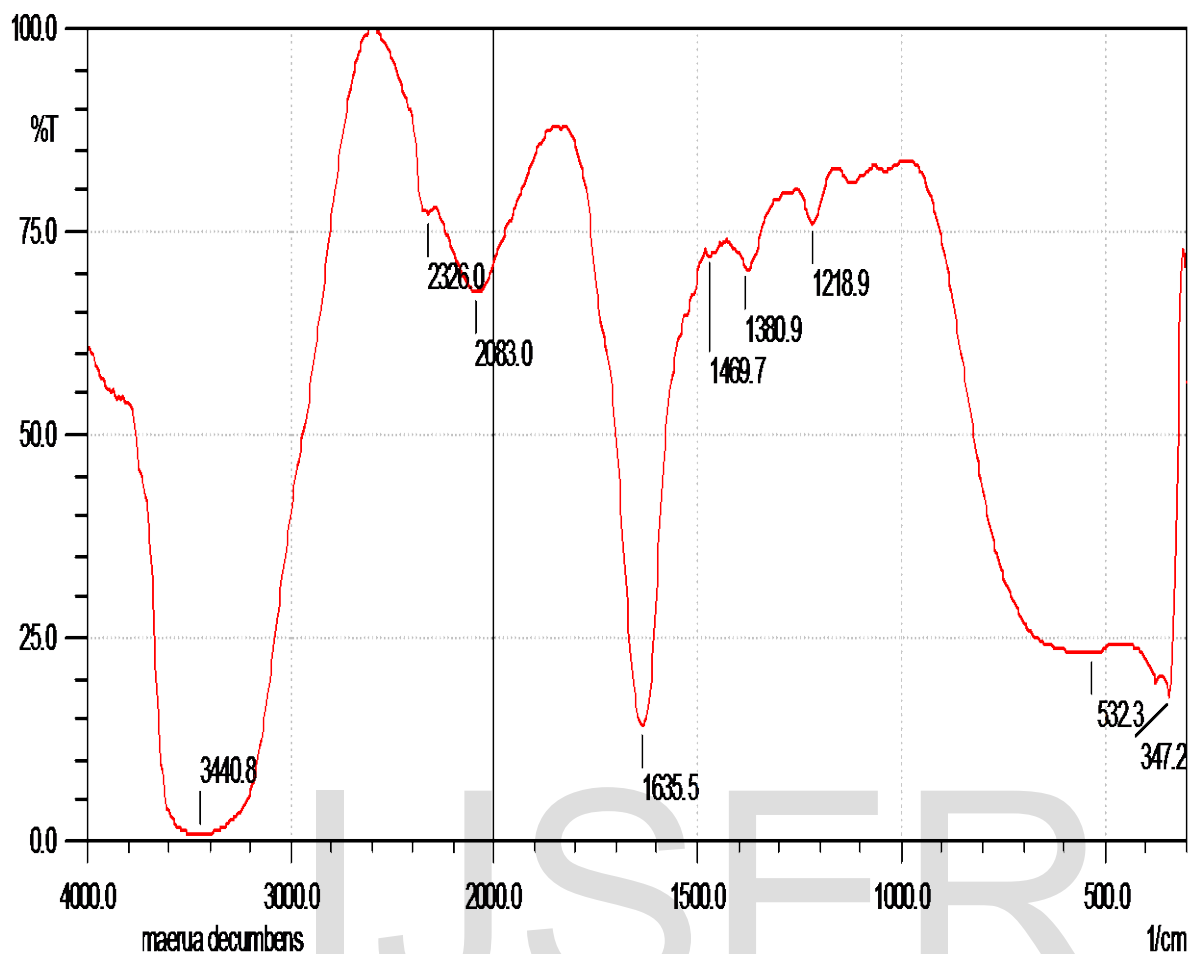
### 2.1 Equipment and Reagents

atomic absorption flame emission spectrometry AAFES (AA-6200 Shimadzu) using air acetylene Flame system and hollow cathode lamps was used for the metals(Cd,Cr,Pb and Cu) determination. pH measurements were done using a pH meter fitted with a temperature probe and a glass electrode (pH 211, HANNA Instruments) while filtration was done using a Millipore filter funnel equipped with a 0.45 µm cellulose acetate filter membrane for the fine texture powder. The *Maerua decumbens* spectra was generated from a Fourier Transform Infrared Spectrophotometer 8400CE (Shimadzu, Japan) fitted with a pellet cell(solid).

All chemicals used in this work, were of analar grade. All solutions were prepared using distilled water. Stock solutions were prepared by dissolving appropriate amounts of analytical grade salts(Cd,Cr Pb and Cu). in 250 mL distilled water, this was acidified by adding 5 mL of concentrated nitric acid and the solution was made to a liter using distilled water. Working solutions were then prepared from stock solutions.

### 2.2 Sampling

*Maerua decumbens* was collected from Marigat in Baringo County in Kenya, washed in tap water several times and rinsed with distilled water. The *Maerua decumbens* samples were sun-dried in the open for two days then oven -dried at 60 °C for eight hours. Finally it was grinded, sieved to 0.5 mm particle size and stored in a plastic bottle at room temperature until use. The first batch was used to generate spectra from the FTIR to identify the functional groups.



**FIGURE 2 0**

### 2.3 Optimization of pH

Batch biosorption experiments were conducted on model solutions of Cd, Cr, Cu and Pb. For each element, the stock solution was diluted to 200 mg/L using 0.5 molar acetate buffer (acetic acid and sodium acetate) and divided into two series of appropriate number of 50 mL batches, for which pH values were adjusted to 3.0, 3.5, 4.0, 4.5, 5.0, 5.5, 6.0 or 6.5 using concentrated sodium hydroxide and hydrochloric acid solutions. The samples from the first series were each equilibrated with 0.20 g of crushed *Maerua decumbens* for twelve hours while the second series (control) was allowed to stand for twelve hours without addition of biosorbent. The first solution was filtered through a 0.45 µm membrane and the filtrate analyzed for residual metal ion concentration by AAFES and also the control. All experiments were done in triplicate and the mean values reported. A plot of biosorption against pH was used to determine the optimum pH.

### 2.4 Optimization of contact time

The stock solutions of the selected metal ions were each diluted with 0.5M acetate buffer solution to obtain 1000 mL of 100 and 200 mg/L solutions of Cd, Cr, Cu and Pb. The pH of these solutions were adjusted to 5.0 for cadmium and chromium, and 4.5 for Copper and lead, respectively. Exactly 2.00 g of ground root tuber of *Maerua decumbens* was weighed out on a weighing balance and added to 500 mL of each solution and equilibrated for 140 minutes using magnetic stirring rod at 300 rpm. Mixture (10 mL) was withdrawn at

measured time intervals between 0 and 60 minutes. Each portion was filtered immediately through a 0.45 µm membrane and the residual metal ion concentration determined by AAFES (AA-6200 Shimadzu). All experiments were done in triplicate. The order of the reaction was determined and the experimental data was fitted to 1<sup>st</sup> and 2<sup>nd</sup> order rate equations. Linear correlation coefficient (R<sup>2</sup>) values were used to deduce the order of reaction.

### 2.5 Optimization of initial metal concentration

Initial concentrations ranging from 50 to 1000 mg/L and the respective optimum pH values was used for respective metals. 50 mL of each solution was equilibrated with 0.2 g of finely ground *Maerua decumbens* root tuber for two hours with stirring at 300 rpm. The solution was filtered through a 0.45 µm membrane and the residual metal ion concentration was determined by by AAFES (AA-6200 Shimadzu).The data was fitted to the adsorption isotherms from which the adsorption capacity was calculated. A plot of metal uptake (mg/g) against initial metal ion concentration was done for all metals and used to determine the optimum initial metal concentration for each metal.

## 3. RESULTS AND DISCUSSIONS

### 3.1 The functional groups in *Maerua decumbens*

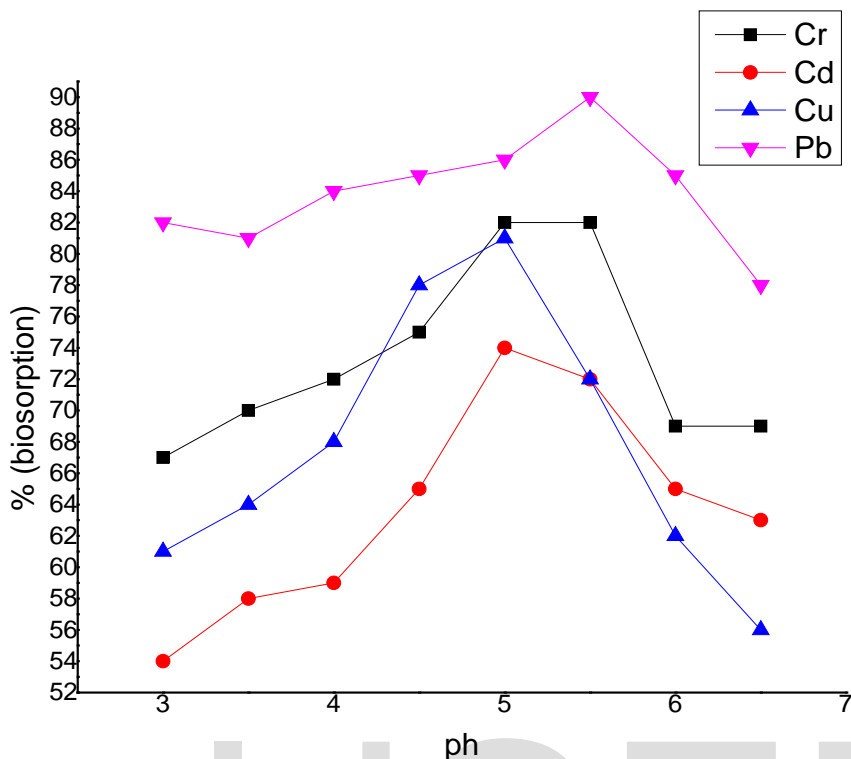
The functional groups Involved in biosorption of heavy metal by *Maerua decumbens* were investigated by Fourier Transform Infrared Spectrophotometer 8400CE (Shimadzu, Japan) analysis. The spectrum of ground root tuber of *Maerua decumbens* is shown in table 3 3 The position of absorption bands and corresponding functional groups able to interact with metal ions are summarized in Table 1.

The results of FTIR analysis confirm the presence of carboxylic, amino, hydroxyl and carbonyl groups in the root tuber powder, peaks at (3440.8,2326,2083,1635.5,1469.7,1380.9,1218.9,532.3 and 347).

PEAK	ASSIGNMENT
3440.8	Alcohol, phenol,O-H stretch,
2326	
2083.0	
1635.5	Amines, C=O(stretch),C=C(bend),C-H,
1469.7	Phenyl,C-H,Alkanes,CO <sub>3</sub>
1380.9	C-O, C-H
1218.9	Aliphatic amines, C-O(esters)
532	Alkyl halides, Amines.
347	

**Table 3 3-0;FTIR peaks and their possible loading.**

### 3.2 Effect of pH



**Figure 3.0: Effect of pH on the biosorption of Cr (II), Cd (II), Cu (II) and Pb (II) by *M. decumbens* root tuber.**

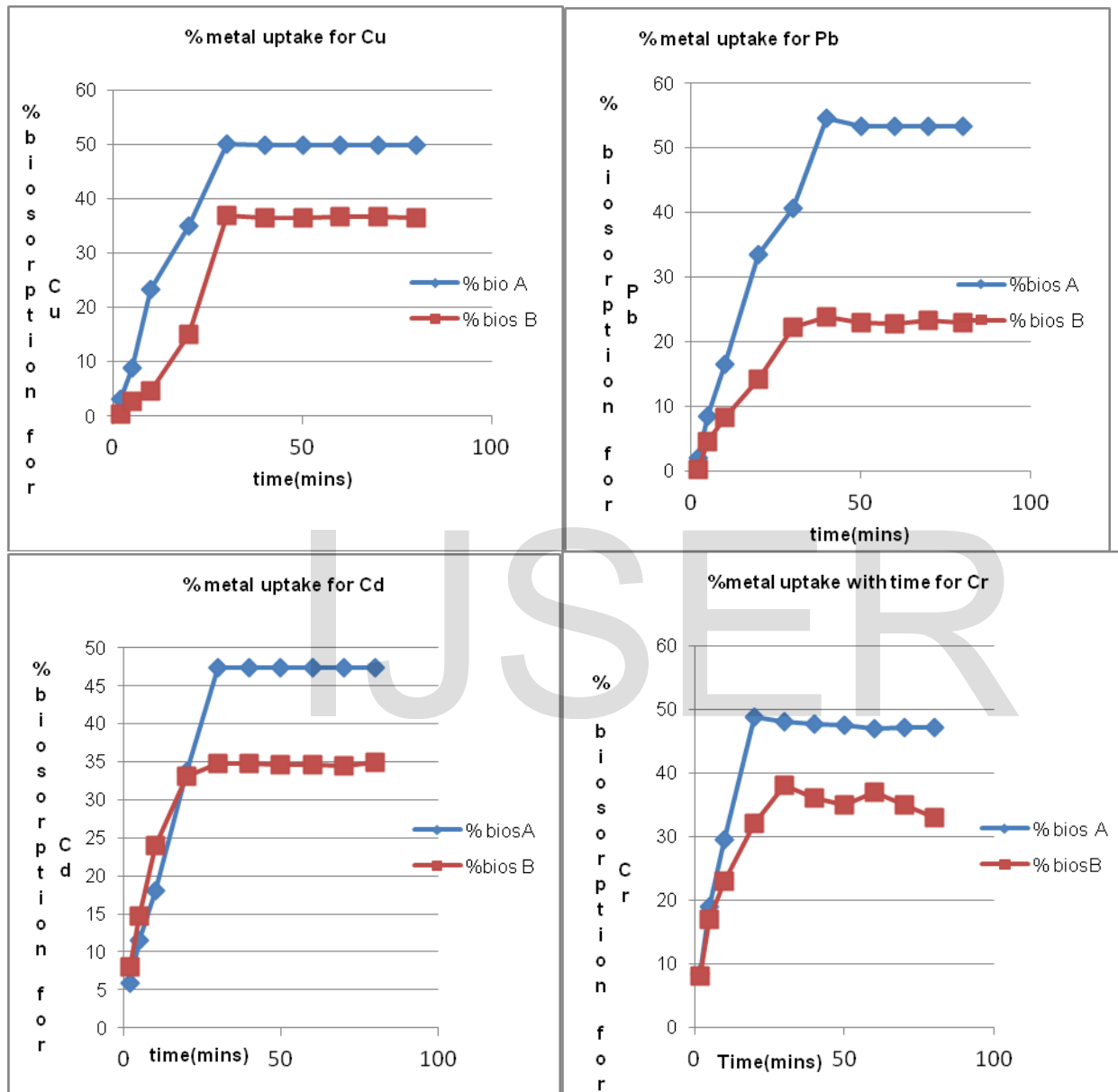
(Experimental conditions:  $C_0 = 50 \text{ mg L}^{-1}$ , dosage = 0.2 g per 50 mL; particle size  $\leq 0.5 \text{ mm}$ ; mixing rate = 300 rpm;  $T = 25 \pm 1 \text{ }^\circ\text{C}$ )

Change in pH affects the protonation-deprotonation equilibria of the functional groups in the tuber as well as metal chemistry. As the pH rises, the hydrogen ion concentration level goes down leading to less competition for the sorption sites therefore resulting in an increase in biosorption of heavy metals. The high pH also leads to precipitation of low solubility metal hydroxides and oxides. Precipitation interferes with the biosorption process because it immobilizes the metal ions thus making them unavailable for biosorption. Precipitation process takes place due to the formation of low solubility metal hydroxides in this study;  $\text{Cr}(\text{OH})_3$ ,  $\text{Cu}(\text{OH})_2$ ,  $\text{Cd}(\text{OH})_2$ ,  $\text{Pb}(\text{OH})_2$  at higher pH (Cotton and Wilkinson 2004, Chen *et al.*, 2006, Gupta and Rastogi, 2007, Gupta *et al.*, 2005). The optimum pH for biosorption is a compromise between interference from precipitation at high pH and competition with hydrogen ions for sorption sites in the specific metals at low pH. To obtain optimum pH, a graph of percentage metal removal by biosorption against pH was plotted. Biosorption studies were conducted in determination of the adsorption parameters (pH, contact time and adsorption capacity) of the selected metals in model aqueous solutions using green algae, *spirogyra* species. The optimum pH values were found to be 5.0, 5.5, 5.8, and 5.9 for lead, cadmium, chromium and copper, respectively. (Matei *et al.*, 2011). The curves obtained for the selected metal ions are shown in Figure 3.3. From these curves the optimum pH values for the selected metals were found to be 5.2, 5.5, 5.0, and 5.0 for chromium, lead, cadmium and copper, respectively.

### 3.3 Effect of contact time

The minimum time required for quantitative uptake of the selected metal ions from solution was determined. The contact time was obtained by plotting the mean percentage metal ion uptake(% biosorption) against time as

shown in Figure 3.4. For all metals considered, metal adsorption was very rapid and went to completion in less than an hour. For chromium, cadmium and copper adsorption was the fastest with quantitative uptake being achieved in thirty minutes while lead took forty minutes. Therefore the contact times for the selected metals were found to be 30 minutes for chromium cadmium and copper while 40 minutes for lead. The short contact times demonstrate the potential of *Maerua decumbens* as a suitable biosorbent for fast removal of heavy metals from contaminated water.



**Figure3 0: Pb (II) Cu (II),Cd(II) and Cr(II) percentage biosorption onto *Maerua decumbens* root tuber powder with time.**

(Experimental conditions:  $C_0$ ; A= 50 mg L<sup>-1</sup> and B=150 mg L<sup>-1</sup>; dosage = 2.0 g per 500mL, particle size ≤ 0.5 mm, mixing rate = 300 rpm, T = 25 ± 1 °C, pH = 5.0 ± 0.2 for Cu(II) and Cd(II); Ph 5.2 ± 0.2 for Cr(II) and pH = 5.5 ± 0.2 for Pb(II) )

### 3.4 Sorption Isotherms Models

The metal uptake per gram of sorbent and the percentage removal is calculated according to equations below;

$$q_e = \frac{C_0 - C_e}{m} * V \dots\dots\dots 1$$

$$\% \text{Sorption} = \frac{C_0 - C_e}{C_0} * 100 \dots\dots\dots 2$$

Where:  $q_e$  is the equilibrium adsorption capacity ( $\text{mg g}^{-1}$ ),  $C_0$  is the initial concentration ( $\text{mg L}^{-1}$ ) of metal ions in solution,  $C_e$  is the equilibrium concentration ( $\text{mg L}^{-1}$ ) of metal ions in solution,  $V$  is the volume of aqueous solution (L) and  $m$  is the dry weight of the adsorbent (g).

### 3.4.1 The Langmuir Isotherm Model

The Langmuir isotherm model is valid for monolayer adsorption onto a surface containing a finite number of identical sorption sites, and it is represented by equation 3 (Langmuir, 1916):

$$q_e = \frac{q_m K_L C_e}{1 + K_L C_e} \dots\dots\dots 3$$

Where:  $q_e$  is the amount of metal adsorbed per specific amount of adsorbent ( $\text{mg g}^{-1}$ ),  $C_e$  is the equilibrium concentration of the solution ( $\text{mg L}^{-1}$ ) and  $q_m$  is the maximum amount of metal ions required to form a monolayer ( $\text{mg g}^{-1}$ ). The Langmuir equation can be rearranged to a linear form for the convenience of plotting and determination of the Langmuir constant ( $K_L$ ) as shown in equation 4. The values of  $q_m$  and  $K_L$  can be determined from the linear plot of  $C_e/q_e$  versus  $C_e$ :

$$\frac{C_e}{q_e} = \frac{1}{K_L q_m} + \frac{1}{q_m} C_e \dots\dots\dots 4$$

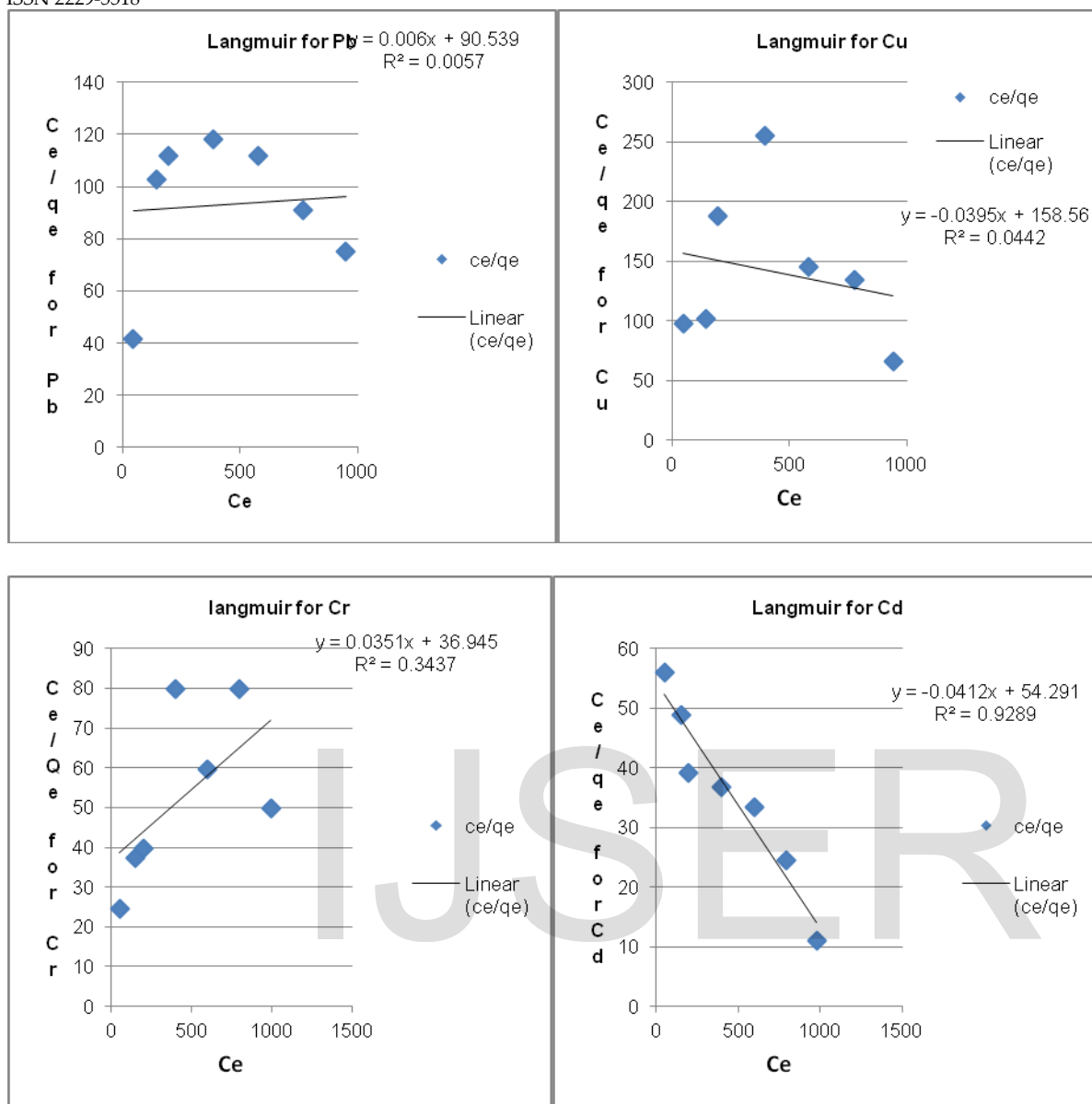
The essential characteristics of the Langmuir isotherm parameters can be used to predict the affinity between the sorbate and the sorbent using the separation factor or dimensionless equilibrium parameter ' $R_L$ ', expressed as in equation 5 (19):

$$R_L = \frac{1}{1 + K_L C_0} \dots\dots\dots 5$$

Where:  $K_L$  is the Langmuir constant and  $C_0$  is the initial concentration of the metal ion. The value for the separation factor  $R_L$  provides important information about the nature of adsorption; when the value of  $R_L$  is between 0 and 1 adsorption is favourable, when  $R_L > 1$  adsorption is unfavourable and when  $R_L = 1$  adsorption is linear while adsorption process is irreversible if  $R_L = 0$ . The  $R_L$  values were all below one (0.377, 0.783, 0.487 and 0.64) for all the metals indicating favourable adsorption.

The adsorption capacities,  $q_{\text{max}}$  was calculated from the linearized Langmuir isotherm and found to be 24.39, 28.57, 25.64 and 166.66 mg/g for cadmium, chromium, Copper and lead, respectively. These results show that the adsorption capacities of *Maerua decumbens* for all selected metals compare well with those of other biosorbents in literature. e.g for green algae (12)  $q_{\text{max}}$  was found to be 22.52, 38.19, 35.59 and 94.34 mg/g for cadmium, chromium, copper and lead, respectively.





**Figure3.1: Langmuir isotherms on Pb(II) Cu(II), Cd(II) and Cr(II) onto *Maerua decumbens* root tuber powder.**

(Experimental conditions: Dosage = 0.2 g per 50 mL; particle size ≤ 0.5 mm; mixing rate = 300 rpm; T = 25 ± 1 °C; contact time = 30 min. for Cu(II),Cr(II) and Cd(II) and 40 min. for Pb(II); pH = 5.0 ± 0.2 for Cu(II)and Cd(II);pH=5.2±0.2 for Cr and pH = 5.5 ± 0.2 for Pb(II) )

### 3.4.2 The Freundlich Isotherm Model

The Freundlich equation is purely empirical based on sorption on a heterogeneous surface; it is described by the equation 6:

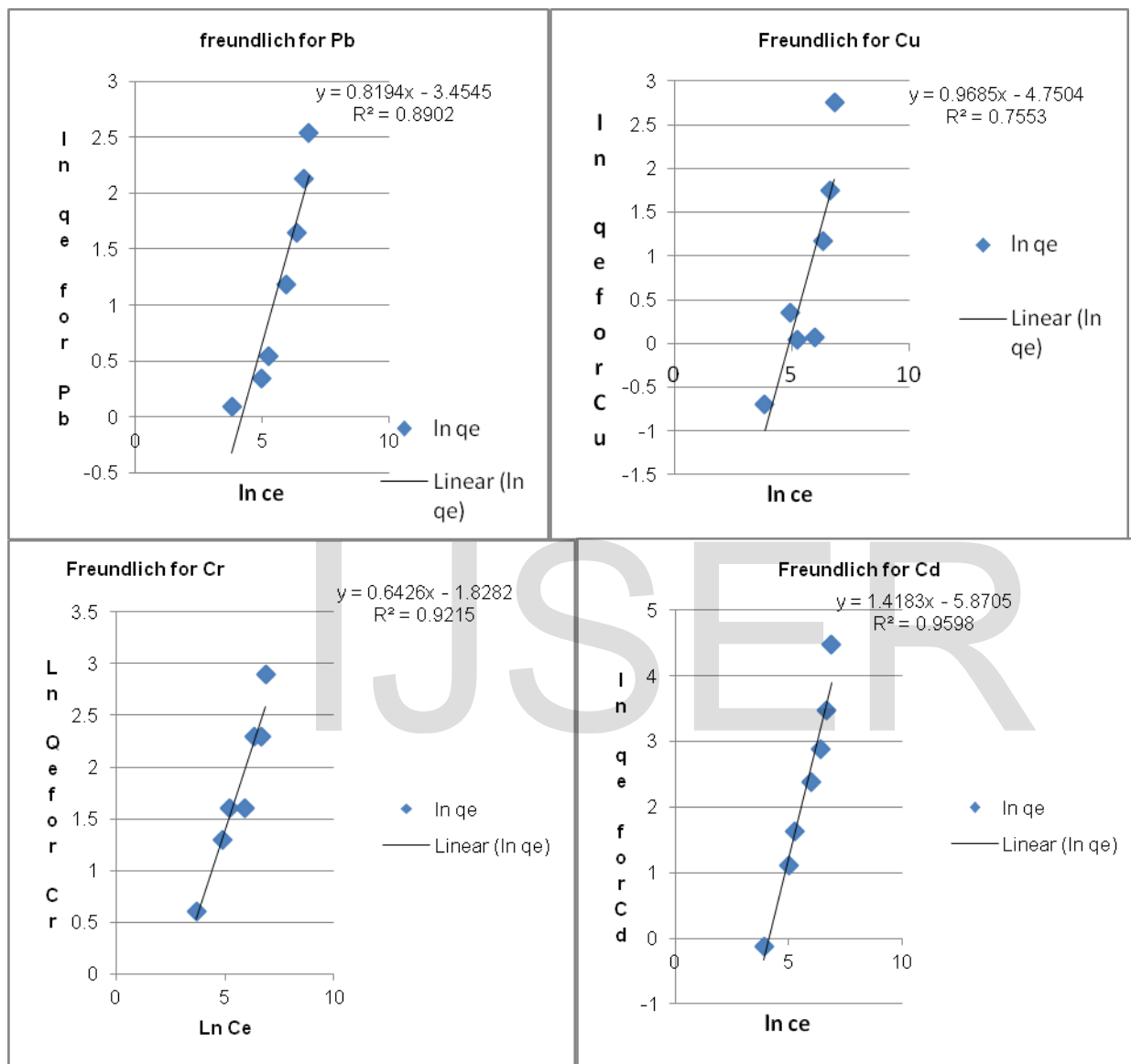
$$q_e = K_F C_e^{\frac{1}{n}} \dots \dots \dots 6$$

In its linear form, this equation assumes form of equation 7 (19):

$$\log q_e = \log K_F + \frac{1}{n} \log C_e \dots \dots \dots 7$$



Where:  $q_e$  is the metal uptake ( $\text{mg g}^{-1}$ ) at equilibrium,  $K_F$  is the measure of the sorption capacity,  $1/n$  is the sorption intensity, and  $C_e$  is the final ion concentration in solution, or equilibrium concentration ( $\text{mg L}^{-1}$ ). The Freundlich isotherm constants  $K_F$  and  $1/n$  are evaluated from the intercept and the slope respectively, of the linear plot of  $\log q_e$  versus  $\log C_e$ .



**Figure3.2: Freundlich isotherms on Pb(II), Cu(II), Cd(II) and Cr(II) onto *Maerua decumbens* root tuber powder.**

(Experimental conditions: Dosage = 0.2 g per 50 mL; particle size  $\leq 0.5$  mm; mixing rate = 300 rpm;  $T = 25 \pm 1$  °C; contact time = 30 min. for Cd(II), Cr(II) and Cu(II) and 40 min. for Pb(II); pH =  $5.0 \pm 0.2$  for Cu(II) and Cd(II); pH =  $5.2 \pm 0.2$  for Cr(II) and pH =  $5.5 \pm 0.2$  for Pb(II) )

### 3.5 Order of reaction

The first and second-order kinetics are described by the rate equations (8) and (9), respectively, which upon integration yield the following solutions (10) and (11). Linear plots of  $t$  versus  $\ln (q_e - qt)$  for first order and  $t/qt$

versus t for second order were used to test the data. The Linear correlation coefficients (R<sup>2</sup> values) were used to deduce the order of reaction of the specific metals. For all studied metals (the kinetic parameters are summarized in Table 2), the results fit better in to second order rate equation than first order rate equation.

$$\frac{dq_t}{dt} = K_1(q_e - q_t) \dots\dots\dots 8$$

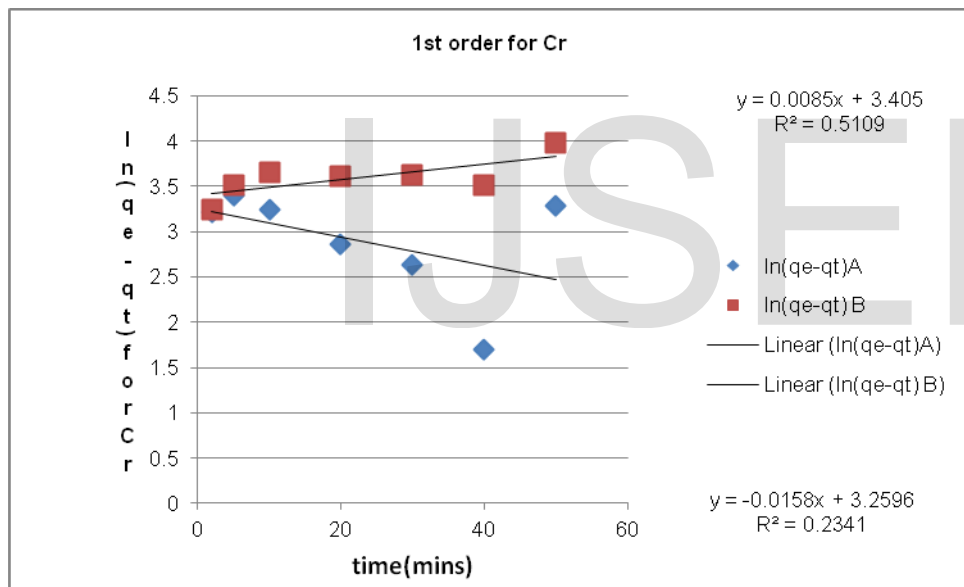
$$\frac{dq_t}{dt} = K_2(q_e - q_t)^2 \dots\dots\dots 9$$

$$K_1 t = \ln q_e - (\ln q_e - q_t) \dots\dots\dots 10$$

$$\frac{t}{q_t} = \frac{t}{q_e} + \frac{1}{K_2 q_e^2} \dots\dots\dots 11$$

### Adsorption Kinetics

#### 3.5 1 Lagergren pseudo first order kinetics models

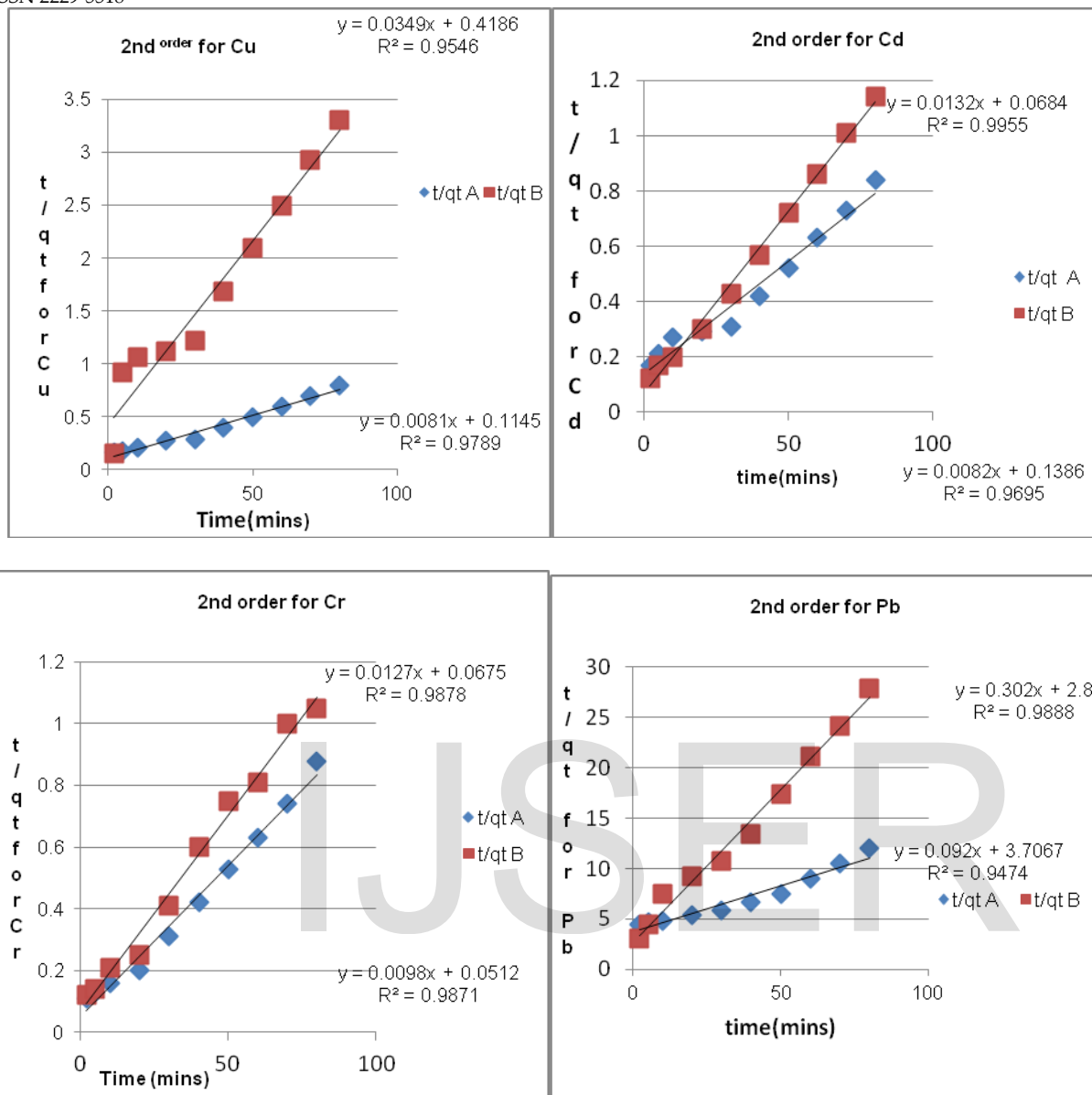


**Figure 3.3: Lagergren pseudo-first-order kinetics biosorption for Cr(II) onto *Maerua decumbens* root tuber powder.**

(Experimental conditions: C<sub>0</sub> B= 150 mg L<sup>-1</sup> and A= 50 mg L<sup>-1</sup>; dosage = 2.0 g per 500 mL, particle size ≤ 0.5 mm, mixing rate = 300 rpm, T = 25 ± 1 °C, pH = 6.5 ± 0.2 for Cu(II) and pH = 5.5 ± 0.2 for Pb(II) )

The data did not fit in the Lagergren pseudo first order kinetics model and the correlation coefficient (R) was not obtained for Cd(II), CU(II) and Pb(II) metals. However for Cr(II) the R (correlation coefficient ) was 0.510 and 0.234 for 150 mg L<sup>-1</sup> and 50 mgL<sup>-1</sup> respectively for *Maerua decumbens* in figure 3.16 indicating that the biosorption of *Maerua decumbens* onto Cd(II),Cr(II),Cu(II) and Pb(II) did not follow the Lagergren pseudo-first-order kinetics model.

#### 3.5 2 Lagergren pseudo-second-order kinetics model



**Figure 3.4: Lagergren pseudo-second-order kinetics biosorption for Cd(II), Cu(II) and Pb(II) onto *Maerua decumbens* root tuber powder.**

(Experimental conditions:  $C_0$ ; B= 150 mg L<sup>-1</sup> and A= 50 mg L<sup>-1</sup>; dosage = 2.0 g per 500 mL, particle size ≤ 0.5 mm, mixing rate = 300 rpm, T = 25 ± 1 °C, pH = 6.5 ± 0.2 for Cu(II) and pH = 5.5 ± 0.2 for Pb(II) ).

From the graphs drawn for the second-order kinetic model, the correlation coefficient (R) fitted well with the linear plot of  $t/q_i$  versus  $t$ ; giving R value between 0.947 – 0.995, for 150 mg L<sup>-1</sup> and 50 mg L<sup>-1</sup> for *Maerua decumbens* in figure 3.15 indicating that the biosorption of *Maerua decumbens* onto Cd(II), Cr(II), Cu(II) and Pb(II) followed the Lagergren pseudo-second-order kinetics model.

**The FTIR spectra of the *Maerua decumbens*.**

The band at 3440.8 cm<sup>-1</sup> represents the presence of –OH and –NH groups. The band observed at 2326 cm<sup>-1</sup> is associated to asymmetric stretches of –CH group. However, a small peak located around 2083 cm<sup>-1</sup>, which is

assigned for symmetric stretching vibrations of –CH group. The dual bands at 1635 and 1469  $\text{cm}^{-1}$  indicates the presence of –COO, –CO and –NH groups for the untreated and treated *maerua decumbens* root tuberpowder. Another band appearing around 1380 and 1218  $\text{cm}^{-1}$  can be assigned to the C–O stretching and sulphonic group respectively. The last but not least peak was at 532 and 347  $\text{cm}^{-1}$  Spectrum explains that some peaks were shifted or disappeared and that new peaks were also detected. These changes observed in the spectrum indicated the possible involvement of those functional groups on the surface of the *Maerua decumbens* adsorption process.

## DISCUSSIONS AND CONCLUSIONS

The study showed that natural coagulant *Maerua decumbens* root tuber powder compared well to alum coagulant in respect to removal of turbidity and other pollutants found in turbid Ndarugo river water. The residual turbidities of clarified water were within stipulated guidelines of NEMA and WHO; It as well evident that the natural coagulants *Maerua decumbens* was a good coagulant aid. The natural coagulants were effective against total and feecal coliforms in untreated turbid Ndarugo river water.

In the study of biosorption, *Maerua decumbens* root tuber powder had high sorption capacity from initial concentration of 1000mg  $\text{L}^{-1}$ . This study revealed that *Maerua decumbens* root tuber powder could be an important low-cost biosorbent for Cr(II), Cd(II), Cu(II) and Pb(II) removal. The sorption isotherms of *Maerua decumbens* root tuber powder correlate well with the Freundlich model than langmuir model; The FTIR analysis results indicated that hydroxyl and carboxylic functional groups were involved in Cr(II), Cd(II), Cu(II) and Pb(II) removal by *Maerua decumbens* root tuber powder through the ion exchange mechanism.

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