Study of low cost biosorbent for biosorption of heavy metals

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Abstract. The efficacy of the Banana peel (*Musa sapientum*) biomass was tested for the removal of lead, copper, zinc and nickel metal ions using batch experiments in single and binary metal solution under controlled experimental conditions. It is found that metal sorption increases when the equilibrium metal concentration rises. At highest experimental solution concentration used (150 mg/L), the removal of metal ions were 92.52% for lead, 79.55% for copper, 63.23% for zinc and 68.10% for nickel while at lowest experimental solution concentration (25mg/L), the removal of metal ions were 94.80% for lead, 86.81% for copper, 84.63% for zinc and 82.36% for nickel. Biosorption equilibrium isotherms were plotted for metal uptake capacity (q) against residual metal concentrations (C_f) in solution. The q versus C_f sorption isotherm relationship was mathematically expressed by Langmuir and Freundlich models. The values of separation factor were between zero and one indicating favourable sorption for four tested metals on the biosorbent. The surface coverage values were approaching unity with increasing solution concentration indicating effectiveness of biosorbent under investigation. The non-living biomass of *Musa sapientum* present comparable biosorption capacity for lead, copper, zinc and nickel metal ions with other types of biosorbent materials found in literature and is effective to remove metal ions from single metal solutions as well as in the presence of other co-ions with the main metal of solution.
Keywords: Biomass; single metal; multi metal; sorption; isotherm; Efficacy.

1. Introduction

Present age of rapid increase in metal concentration as well as increase in awareness of the toxicological effects of metals released into environment, a number of studies for metal recovery and removal for metal solution have been done. Conventional methods for metal removal include chemical precipitation, lime coagulation, ion exchange, reverse osmosis and solvent extraction [1]. These conventional methods for the removal of heavy metals from wastewaters, however, are often cost prohibitive having inadequate efficiencies at low metal concentrations, particularly in the range of 1–100 mg/l. Some of these methods. Furthermore, generate toxic sludge, the disposal of which is a burden on the techno-economic feasibility of treatment procedures [2].

The search for new technologies involving the removal of toxic metals from wastewaters has directed attention to biosorption, based on metal binding capacities of various biological materials. Biosorption can be defined as the ability of biological materials to accumulate heavy metals from wastewater through metabolically mediated or physico-chemical pathways of uptake [3]. The major advantages of biosorption

over conventional treatment methods include low cost, high efficiency of metal removal from dilute solution, minimization of chemical and/or biological sludge, no additional nutrient requirement, regeneration of biosorbent and the possibility of metal recovery [4,5,6]. Biosorption for the removal of heavy metal ions may provide an attractive alternative to physico-chemical methods [7].

Only within the past decade has the potential of metal biosorption by biomass materials been well established. Biosorption is considered to be a fast physical or chemical process. A significant number of biosorption studies on the removal of heavy metal from aqueous solutions have been conducted worldwide. Nearly all of them have been directed towards optimizing biosorption parameters to obtain the highest removal efficiency while the rest of them are concerned with the biosorption mechanism. The biosorption rate depends on the type of the process. According to literature, biosorption can be divided into two main proceses: adsorption of the ions on cell surface and bioaccumulation within the cell [8].

Natural materials that are available in large quantities or certain waste from agricultural operations may have potential to be used as low cost adsorbents, as they represent unused resources, widely available and are environmentally friendly [9]. Different researchers have used different biomass such as Azadirachta indica bark [10], Neem biomass [11], citrus pectin [12], tamarad bark [13], potato peel waste, Bengal gram husk (husk of channa dal, Cicer arientinum) [14]. The binding ability of Cd (II) has been studied on seven different species of brown, red and green seaweeds [15] while the biosorption of cadmium metal ion from waste water using Hypnea valentiae biomass [16].

The objective of this study is to develop inexpensive and effective biosorbent that is easily available in large quantities and feasible economically for multiple metal ions in solution. Banana peel (Musa sapientum) is tested during this study for the biosorption of multiple metal ions in single metal system (SMS) and binary metal system (BMS).

2. Material and Methods

2.1. Preparation of Biosorbent
Musa sapientum biomass was collected from local market. The biomass was dried in sun for fifteen days. The buds were removed and further dried in sun for another fifteen days. This biomass was washed with tap water to remove any dust or foreign particles attached to biomass and thoroughly rinsed with distilled water. The washed biomass was dried at 50°C and ground to powder with kitchen grinder. Grinded biomass was further thoroughly washed with distilled water till the color of washing water clear. The powdered biomass was dried in oven at 50°C to a constant weight. The biosorbent was again ground to powder and was sieved with three different sized meshes. Four different grade particle sizes were obtained. First particle size was less then 255 mm, second 255-355 mm, third 355-500 mm and fourth was 500-710 mm.The aforesaid particle size no. 2 was selected for further study because the amount of no. 1 size was too less and no.3 and no.4 sizes will show less efficiency because of less surface area as compared to no. 2 size.

2.2. Stock solutions of metal ions
Stock solution were prepared in deionized water from the respective salts of four metals (Pb^{2+}, Cu^{2+}, Zn^{2+}, Ni^{2+}) under investigation. The resulting stock solutions were stored in the air tight plastic bottle.

2.3. Batch Studies of SMS and BMS
For SMS, solutions of fixed volume (100 ml) with varying concentrations in conical flasks were thoroughly mixed with 0.5 g of biosorbent dose, size of 255 to 355 micron at 30°C and 100 revolutions per minute (rpm) shaking speed for 12 hours. Twelve hours of equilibrium period for sorption experiment were used to ensure equilibrium after conducting equilibrium studies of the biosorbent. The pH range was adjusted from 4-6 by using 0.1M HNO_3 and 0.1M HCl solutions. The flasks were kept on rotating shaker with constant shaking. At the end of experiment the flasks were removed and the solution was separated from the biomass by filtration through filter paper. For BMS 100 ml volume of solutions with varying concentrations (25, 50, 75,100,125 and 150 mg L^{-1}) of main metal and 25mgL^{-1} of affecting metal concentration in the same solution in conical flasks were thoroughly mixed under same conditions stated for SMS solutions. At the end of

experiment the solution was separated from the biomass by filtration through filter paper. Filtrates of SMS and BMS were diluted to 10 mg/ L^{-1} or 20 mg L^{-1} with deionized water and analyzed for metal concentration using flame atomic absorption spectrometry (Perkin Elmer A. Analyst 300). After metal concentration analysis, the final concentration was subtracted from the initial concentration in order to find the metal to be sorbed.

2.4. Calculation of Metal Uptake

The quality of biosorbent is judged by the metal uptake (biosorption capacity), q.

Amount of metal bound by the biosorbent which disappeared from the solution was calculated based on the mass balance for the biosorbent in the system.

$$q = \frac{V\,(C_i - C_f)}{S}$$

q = Metal ion uptake capacity (mgg^{-1})
C_i = Initial concentration of metal in solution, before the sorption analysis (mgL^{-1}).
C_f = Final concentration of metal in solution, after the sorption analysis (mgL^{-1}).
S = Dry weight of biosorbent (g)
V = Solution volume (L)
The difference between the initial metal ion concentration and final metal ion concentration was assumed to be bound to the biosorbent.

2.5. Freundlich and Langmuir Model

To characterize the biosorption for SMS, Langmuir and Freundlich models are used. The Langmuir model makes assumptions such as monolayer adsorption and constant adsorption energy while the Freundlich model deals with heterogeneous adsorption

Langmuir equation of adsorption isotherm is $1/q = 1/q_{max} + 1/(b.q_{max})\,(C_f)$, Where q_{max} and b are the Langmuir constants. The Freundlich equation of adsorption isotherm is $\log q = \log K + (1/n)\log C_f$, Where q is the amount adsorbed per unit mass of adsorbent and C_f is equilibrium concentration. The plot of log q vs log C_f is linear and constants K and n is evaluated from slopes and intercepts.

2.6. Separation Factor

The shape of the isotherm can be used to predict whether adsorption system is favorable or unfavorable in a batch adsorption system. Accordingly, the essential feature of Langmiur isotherm was expressed in term of dimensionless constant called the separation factor. It is defined by the following relationship.

$$Sf = 1/(1+bC_i)$$

where *SF* is the a dimensionless equilibrium parameter or separation factor, b, the constant from Langmuir equation and C_i the initial metal ion concentration. The parameter, *SF,* indicates the shape of the isotherm and nature of the sorption process. If

SF > 1 then unfavourable isotherm
SF = 1 then linear isotherm
SF = 0 then irreversible Isotherm
0 < *SF* < 1 then favourable isotherm [16]

2.7. Surface coverage (θ)

To account for adsorption behavior of the metal ions on the biomass the Langmiur type equation related to surface coverage is used. The equation is expressed as follow.

$$bC_i = \theta/(1-\theta)$$
and
$$\theta = bC_i/(1+bC_i)$$

where b is the adsorption coefficient, Ci the initial concentration and θ the surface coverage.

3. Results and Discussion

Efficiency of the non conventional biosorbent of plant origin is tested for removal of metal ions Pb^{2+} Cu^{2+} $Ni2^+$ and Zn^{2+} from synthetic solutions in SMS and BMS solutions.

The rate of absorption is a function of the initial concentration of metal ions, which makes it an important factor to be considered for effective biosorption. From (Fig.1) in general, the data reveal that capacity of biosorbent increases with increase in initial concentration of metal ions. This characteristic represent that surface saturation was dependent on the initial metal ion concentrations. At low concentrations biosorbent sites take up the available metal more quickly. However, at higher concentrations, metal ions need to diffuse to the biomass surface by intraparticle diffusion and greatly hydrolyzed ions will diffuse at a slower rate [16].

It is found that as metal ions concentration is reduced, biosorption rate increased and when the metal ions concentration is high the metal removal rate decreased. Such decline in %removal rate is probably caused by the saturation of some adsorption sites. The results are in agreement to [17]

The selectivity order for metal ions towards the studied biomass is

Pb >Cu >Ni>Zn
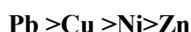
Here is anomalous behaviour of zinc and nickel as in the previous study with *Mangifera indica L.* biomass which has been done by the writer [18] where is more uptake of zinc than nickel.This differential sorption of metal ions may not be ascribed to the difference in their ionic radii as it follow in the previous study [18]. It may be due the reductant behaviour of banana biomass [19]. Zinc ions have much less reducable behaviour as compared to other three metal ions so it is lowest uptake by *Musa sapientum* biomass.

Modeling the equilibrium data is fundamental for the industrial application of biosorption since it gives information for comparison among different biomaterials under different operational conditions, designing and optimizing operating procedures [19]. To examine the relationship between uptake capacity (q) and aqueous concentrations (C_i) at equilibrium, sorption isotherm models are widely employed for fitting the data, of which the Langmuir and Freundlich equations are the most widely used [20]. The Langmuir and Freundlich adsorption constants evaluated from the isotherms with correlation coefficients are presented in (Table 4) which illustrates the relationship between absorbed and aqueous concentration at equilibrium.The Langmiur and Freundlich adsorption constants are evaluated from the isotherms with correlation coefficients. Both the models represent better absorption process due to high value of correlation coefficients (R^2).

Constant b which is related to the energy of absorption. The higher b, the higher is the affinity of the biosorbent for the metal ions. q_{max} can also be interpreted as the total number of binding sites that are available for biosorption and q as the number of binding sites that are in fact occupied by the metal ions at the concentration C_f. To get the equilibrium data, initial metal concentrations were varied while the biomass weight in each sample was kept constant. Twelve hours of equilibrium periods for sorption experiments were used to ensure equilibrium conditions. This time was chosen considering the results of kinetics of metal removal found in literature. The Freundlich model better represented the sorption process for Pb^{2+}, Cu^{2+} and Ni^{2+} in comparison to the model of Langmuir due to very close experimental values of q_{max} with the theoretical values calculated from these models. Both models well represent for Zn^{2+} due experimental and theoretical values of q_{max} are compareable. Although correlation coefficient (R^2) values show best fitting of both models but Freundlich is best. The Langmuir and Freundlich parameters are determined from a linear regression presented in (Table 4). In the view of above mentioned comparison, the values of Freundlich constant K represent the sorption. Overall, Freundlich model of sorption indicates the hetrogenecity of biomass.

TABLE I.   PERCENTAGE REMOVAL OF METAL IONS ON BIOMASS MUSA SAPIENTUM IN SINGLE METAL SYSTEM

Concentration	Pb	Cu	Zn	Ni
			Removal (%)	
25	94.80	86.81	84.63	82.36
50	96.37	83.99	71.05	78.50
75	95.25	80.78	72.96	77.96
100	93.23	81.08	73.47	69.83
125	93.37	80.30	73.95	69.27
150	92.52	79.55	63.23	68.10

TABLE II.   UPTAKE CAPACITY OF MUSA SAPIENTUM AND PERCENTAGE REMOVAL OF COPPER IN SMS AND BMS

A		
Ci	q	Removal (%)
24.65	4.28	86.81
51.87	8.71	83.99
73.90	11.94	80.78
102.05	16.55	81.08
124.14	19.93	80.30
147.00	23.39	79.55

B						
	(Cu Zn) Binary solution		(Cu Pb) Binary solution		(Cu Ni) Binary solution	
Ci	q	Removal (%)	q	Removal (%)	q	Removal (%)
24.65	4.50	87.37	4.28	86.81	3.08	65.11
51.87	8.47	83.61	8.71	83.99	6.79	68.93
73.90	12.15	81.05	11.94	80.78	11.06	73.24
102.05	16.47	81.01	16.55	81.08	14.84	72.40
124.14	20.02	80.37	19.93	80.30	17.50	70.02

TABLE III. UPTAKE CAPACITY OF MUSA SAPIENTUM AND PERCENTAGE REMOVAL OF ZINC IN SMS AND BMS.

A		
Ci	q	Removal (%)
24.01	4.06	84.63
49.40	7.02	71.05
76.32	11.138	72.96
101.21	14.87	73.47
124.84	18.46	73.95
148.90	18.83	63.23

B						
	(Zn Cu) Binary solution		(Zn Ni) Binary solution		(Zn Pb) Binary solution	
Ci	q	Removal (%)	q	Removal (%)	q	Removal (%)
24.30	4.12	84.81	4.51	85.93	2.79	57.88
48.75	6.89	70.66	7.39	72.10	5.47	55.05
76.00	11.07	72.85	11.13	72.96	8..30	54.64
99.08	14.44	72.90	14.87	73.47	10.39	51.46
124.09	18.31	73.80	18.40	73.89	13.41	52.77
148.45	18.74	63.11	18.95	63.38	15.59	51.98

TABLE IV. COMPARISON OF LANGMUIR AND FREUNDLICH ISOTHERM FOR LEAD, COPPER, ZINC AND NICKEL.

Metal	Langmuir isotherm parameters			Experimental value	Freundlich isotherm parameters			
	q_{max}	b	R^2	q_{max}	q_{max}	K	1/n	R^2
Pb	71.42	0.063	0.905	27.46	28.43	5.062	0.716	0.939
Cu	37.73	0.038	0.992	23.39	22.90	1.718	0.761	0.996
Zn	20.36	0.065	0.920	18.83	20.95	1.676	0.631	0.930
Ni	28.82	0.037	0.995	20.32	20.31	1.624	0.654	0.984

According to the above data (Table 3) the affinity order of *Musa sapientum* biosorbent is

Pb > Cu > Ni > Zn

Langmuir isotherm, which represents that monolayer of metal ions (sorbate), is formed on biosorbent.

Adsorption-partition constants are determined for metals using the following log form of the Freundlich isotherm

$$\log q = \log K + (1/n) \log C_f$$

Where q is the metal ion sorbed (mgg^{-1}), C_f the equilibrium concentration of metal ion solution in mgL^{-1}, K and n are Freundlich constants. The constants K and 1/n were determined by linear regression from the plot of log q against $logC_f$. K is a measure of the degree or strength of adsorption. Small value of K indicate the minimal absorption and large value indicates the more absorption while 1/n is used as an indication of whether absorption remains constant (at 1/n = 1) or decreases with increasing metal ions concentrations (with $1/n \neq 1$). The q_{max} value is the maximum value of q, is important to identify the biosorbent highest metal uptake capacity and as such useful in scale-up considerations [20].The magnitude of the experimental q_{max} for *Musa sapientum* biomass is found to 27.46, 23.39, 18.38 and 20.32 mgg^{-1} for all the four i.e., lead copper zinc and nickel metal ions are comparable with theoretically calculated q_{max} values from Langmuir and Freundlich isotherm models.
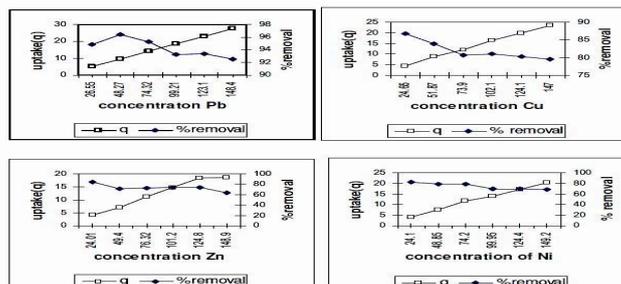

Figure 1. Comparison of uptake capacity (mgg^{-1}) and percentage removal as a function of metal ions concentration by *Musa sapientum* biosorbent
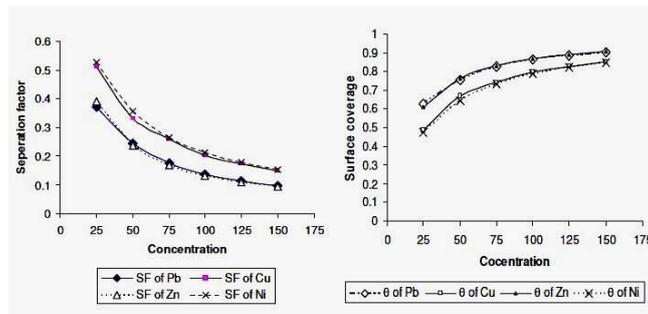
Figure 2.   A plot of SF and surface coverage (θ) against concentration of lead, copper, zinc and nickel for *Musa sapientum* biomass
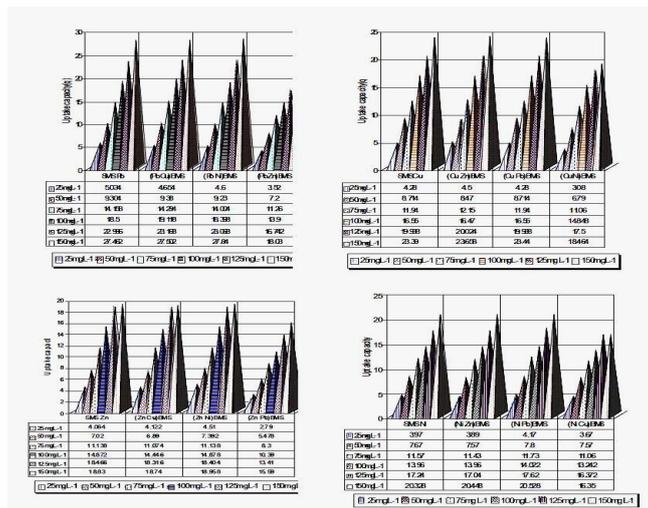

Figure 3.   Comparison of uptake capacity (q) of Pb2+, Ni2+, Cu2+, Zn2+ in SMS and BMS by Musa sapientum

The maximum absorption capacity is observed of lead on *Musa sapientum* suggesting that it is a potential biosorbent for removal of lead as well as the other three tested metals. The shape of the Langmuir isotherm can be used to predict whether a biosorption system is favorable or unfavorable in a batch adsorption process. Accordingly, the essential features of the Langmuir isotherm was expressed in terms of a dimensionless constant called the equilibrium parameter, *SF*, which is defined by the following relationship

$$SF = 1 / (1 + bC_i)$$

Where *SF* is the dimensionless equilibrium parameter or separation factor, b, the constant from Langmuir equation and C_i the initial metal ion concentration of 100 mgL^{-1}. The parameter, *SF,* indicates the shape of the isotherm and nature of the sorption process. *SF* value between 0 and 1 represents favorable isotherm. The *SF* values of Pb2$^+$ Cu^{2+} Zn^{2+} Ni^{2+} for *Musa sapientum* biomass is calculated from above equation and plotted against initial metal ion concentration.

The data in (Table 1) shows that, the sorption of metals on *Musa sapientum* biomass increase as the initial metal ion concentration increase from 25 to 150 mgL^{-1}, indicating that biosorption is even favorable for the higher initial metal ion concentrations (Fig. 1). The biosorption process is favorable for metal removal at all concentrations investigated. According to this classification, removal ability tends to be in the order:

Pb > Cu > Ni > Zn

Above given order illustrates that initially equilibrium for metals uptake is more favorable for *Musa sapientum* its sorption capacity and selectivity is the same as presented in (Table 4). The trend presented by *SF* in Fig.2A is also providing information that the *Musa sapientum* biomass is more effective and excellent adsorbent for metal at lower metal concentrations.

SF values for *Musa sapientum* are between 0 and 1 which represents favorable isotherms for all the four metal ions. The *SF* values of Pb^{2+}, Cu^{2+}, Zn^{2+} and Ni^{2+} for *Musa sapientum* biomass is calculated from above equation and plotted against initial metal ion concentration. The data in (Table 4) shows that, the sorption of metals on *Musa sapientum* biomass increase as the initial metal ion concentration increase from 25 to 150 mgL^{-1}, indicating that adsorption is even

favorable for the higher initial metal ion concentrations (Fig. 2a). The sorption process is favorable for metal removal at all concentrations investigated.

To account for absorption behavior of the metal ions on the biomass the Langmiur type equation related to surface coverage is used. The equation is expressed as follow.

$$bC_i = \theta \diagup (1-\theta)$$
and
$$\theta = bC_i \diagup (1+bC_i)$$

Where b is the absorption coefficient, Ci the initial concentration and θ the surface coverage.

The fraction of biomass surface covered by metal ion was studied by plotting the surface coverage values (θ) against metal ions concentration. The data is presented in Fig.2B. The figure shows that, increase in initial metal ion concentration for Musa *sapientum* biomass increases the surface coverage on the biomass until the surface is nearly fully covered with a monomolecular layer. Further examination of Fig. 2B reveals that the surface coverage ceases to vary significantly with concentration of metal ions at higher levels. Surface coverage value for metal ions on absorbents of *Musa sapientum* is in following order:

Pb > Cu > Ni > Zn

The fraction of biomass surface covered by metal ion was studied by plotting the surface coverage values (θ) against metal ions concentration. The data is presented in (Fig. 2b).

The figure shows that, increase in initial metal ion concentration for *Musa sapientum* biomass increases the surface coverage on the biomass until the surface is nearly fully covered with a monomolecular layer. Further examination of (Fig. 2b) reveals that the surface coverage ceases to vary significantly with concentration of metal ions at higher levels. Comparing the intercationic effect in BMS, biosorption of Pb^{2+} by *Musa sapientum* biomass in Table 1 and Fig. 3A it is observed the inhibition of Pb^{2+} sorption in the presence of $Cu2^+$, Ni^{2+} and Zn^{2+} co-ions. The sorption of Pb^{2+} enhances in the presence of Cu^{2+} and Ni^{2+} from 27.4 6 to 27.50 and 27.84 mgg^{-1} respectively compared to its sorption from SMS. It is not much significant change. In case of Zn^{2+} co-ion the uptake capacity of biososrbent reduces from 18.83 to 18.74 mgg^{-1} in the presence of $Cu2^+$. This reduction in sorption or uptake capacity (q) is observed at highest concentration of lead in solution. The % removal of lead is observed in all binary metal system of lead as compared to % removal in SMS.

Interestingly Ni^{2+} sorption increases in the presence of co-ions Cu^{2+}, Pb^{2+} and Zn^{2+} as compared to its sorption in SMS. Uptake capacity of *Musa sapientum* biosorbent for Ni^{2+} increases from 20.32 in SMS to 20.44 and 20.52mgg^{-1} in the presence of Zn^{2+} and Pb^{2+} co-ions and a slight increase in the presence of Cu^{2+} that is 18.07 mgg^{-1}.

The effect of Ni^{2+}, Pb^{2+} and Zn^{2+} on the uptake of copper by *Musa sapientum* biosorbent is observed as:

Cu^{2+} sorption is inhibited by the presence of co-ion compared to its sorption from SMS and the amount of metal ions sorbed is less as compared to sorbed from Cu^{2+} solution of SMS.

The data in (Table 2) reveal that uptake capacity of Cu^{2+} is 23.39mgg^{-1} in SMS but it reduces to 23.65, 23.44 and 18.46 mgg^{-1} in the presence of Zn^{2+}, Pb^{2+} and Ni^{2+} respectively. The reduction is significant in (Cu, Ni) binary solution.

Zn^{2+} uptake is not effected significantly in the presence of Cu^{2+} an Ni^{2+} co-ions in BMS and Pb^{2+} show significant effect on Zn^{2+} sorption. Zn^{2+} sorption reduces from 18.83 to 15.59mgg^{-1} in the presence of $Pb2^+$ as compared to sorption by *Musa sapientum* biomass in SMS. This is shown in (Table 3 and Fig. 3D).

4. Conclusion

The following conclusions can be withdrawn from present study:

1: The harvesting of the *Musa sapientum* biomass is a relatively simple procedure, and can be obtained without excessive cost.

2: The adsorption range of metal ions including Pb^{2+}, $Cu2^+$, Ni^{2+} and $zinc^{2+}$ ions from dilute acidic solutions of pH 4 – 6.

3: The non-living biomass of *Musa sapientum* present comparable biosorption capacity for Pb^{2+}, Cu^{2+}, Zn^{2+} and Ni^{2+} ions with other types of biosorbent materials found in literature.

4: Efficiency of the biomass indicates that it is effective to remove metal ions from binary metal solutions as well as in single metal solution.

Although both the biomass well performed in single and binary metal solutions under randomly controlled experimental conditions but it requires further research to investigate the optimum conditions to get the best performance.

5. Acknowledgement

6. References

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