Removal of heavy metals from aqueous solution by water hyacinth (*Eichhornia crassipes*)

N. W. Ingole and A. G. Bhole

ABSTRACT

Water hyacinth (*Eichhornia crassipes*) is a fast growing, free-floating aquatic weed. Floating aquatic plants are capable of assimilating large quantities of trace elements and heavy metals, some of which are essential for plant growth. The uptake of these elements is often increased when plants are cultured in wastewater containing high levels of macronutrients. They have the ability to absorb heavy metals. Heavy metals and other trace contaminants enter surface and groundwater in various ways and adversely affect flora and fauna. Hence, the removal of such impurities is necessary. Batch studies were conducted and the uptake of arsenic, chromium, mercury, nickel, lead and zinc from the aqueous solution for six different concentrations ranging from 5 mg/l to 50 mg/l was studied. The daily uptake of heavy metals for all concentrations was recorded and the results analysed. Efficiency of removal was determined when each metal was present separately. Samples were analysed by using a UV visible spectrophotometer. Results indicated that at lower concentrations i.e. 5 mg/l of heavy metals, the plant growth was normal and removal efficiency was greater. At higher concentrations, greater than 10 mg/l, the plant started wilting and removal efficiency was reduced. It was observed that in aqueous solutions containing 5 mg/l of arsenic, chromium and mercury the maximum uptake was 26 mg/kg, 108 mg/kg and 327 mg/kg of dry weight of water hyacinth respectively. The heavy metal removal efficiency was also checked by evaluating the uptake rate constant (k) for water hyacinths. A generalized polynomial model was proposed for kinetics of nickel removal. Both models were verified and found to work satisfactorily.

Finally, it was concluded that by using water hyacinth, heavy metals could be effectively removed from wastewater when their concentrations were less than 10 mg/l. Lead and zinc were removed more efficiently at higher concentrations than other metals, but plants started to discolour at high concentrations of metals and this luxury uptake leads to destruction of plants in most cases.

Key words | absorption, concentration, heavy metals, kinetics, uptake rate constant

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INTRODUCTION

Heavy metals enter wastewater from a variety of sources, such as domestic, industrial and mining operations. Many of these dissolved metal ions such as copper, zinc, nickel, chromium, arsenic, mercury, lead, etc. are toxic to living organisms (Kar *et al*. 1992). The most important features that distinguish heavy metals from other toxic pollutants are their non-biodegradability. Arsenic, chromium, mercury, nickel, lead and zinc are important industrial metals, which are well known for causing pollution problems. Simultaneous exposure to arsenic and lead can occur in areas of extensive industrial development, particularly where there are lead smelters (Chigbo *et al*. 1982).

Heavy metals present in water can pose a serious threat to human health. It has been reported that cadmium produces kidney, cardiovascular and respiratory diseases. High concentrations of hexavalent chromium in water are lethal to fish, corrosive to flesh and a potential carcinogen

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to human beings (Omprakash *et al.* 1987; Singaram 1994).

Plants accumulate heavy metals (Wolverton & McDonald 1975). Aquatic plants are particularly important organisms to study since the analysis of plants can give an indication of the water environment to which they have been exposed. Metals in plant materials are also present in higher concentrations than in water, and thus detection limits are less restrictive (Chigbo *et al.* 1982). The plant chosen for this study was the water hyacinth (*Eichhornia crassipes*), which has become important in pollution treatment systems (Wolverton & McDonald 1975). Water hyacinth can be used successfully to remove cadmium (Omprakash *et al.* 1987), cadmium and nickel (Wolverton & McDonald 1975), lead and mercury, arsenic and cadmium (Chigbo *et al.* 1982) from water.

In an aquatic macrophyte-based system (AMS), removal of heavy metals occurs through:

- plant uptake, especially by roots (Hao *et al.* 1993; Ding *et al.* 1994; Low *et al.* 1994);
- chemical precipitation (Nipaney 1990);
- ion exchange with, or absorption of, settled clay and organic compounds (Abbasi & Nipaney 1993).

Metal uptake is always stimulated by the presence of metal-binding ligands such as thiols (Ding *et al.* 1994), or surfactants such as linear alkylbenzene sulphonate (Singh *et al.* 1994), whereas pairs of metals may either exert a mutually synergistic or antagonistic influence on the uptake (Dirilgen & Inel 1994). When concentrations of metals in the wastewaters are not high enough to cause toxicity to the plants, the uptake can be copious (Zaranyika *et al.* 1994). Organic mercury has been found to be more readily taken up than inorganic mercury (Ribeyre & Boudou 1994). Removal through plant harvest is less significant compared to the other two mechanisms (Williams 1994).

The water hyacinth can be used to partially absorb and concentrate toxic substances such as chromium, cadmium, mercury, phenol and copper in leaves and stems, and these substances are also adsorbed on the root zone (Wolverton & McDonald 1975; Chigbo *et al.* 1982; Selvapathy & Subhash Babu 1995; Panda 1996).

The water hyacinth (*Eichhornia crassipes*) has been the subject of intensive study in recent years. Due to its vegetative reproduction and extremely high growth rate, it has proved to be a persistent, and expensive to eradicate, aquatic weed problem in every part of the world it has invaded. It spreads rapidly, interfering with shipping, commercial fishing and recreation, forcing other vegetation out, even rice paddy, clogging drainage ditches and increasing water loss. Much effort and money has been devoted to the control of this prolific weed. Despite all efforts, the problem continues (Omprakash *et al.* 1987). In this study the advantages of the plant's voracious nutrient requirement, explosive growth rate and unique survival capacity have been used to absorb heavy metals such as arsenic, chromium, mercury, nickel and lead from synthetic solutions prepared in a laboratory. The mathematical model developed earlier for batch studies (Metcalf & Eddy Inc. *et al.* 1998) was verified for evaluation of the uptake rate constant (k), and a polynomial model was proposed by the authors for the kinetics of nickel removal.

MATERIAL AND METHODS

Free-floating, fully matured, equal-sized and undiscoloured water hyacinth plants were obtained from a pond near Akola (Maharastra). The plants were left in a curing tank for 2 days to regain normal growth. The synthetic solutions for different concentrations of arsenic, chromium, mercury, nickel, lead and zinc were prepared by dissolving the required amount of sodium arsenite, potassium dichromate, mercuric nitrate, nickel sulphate, lead acetate and zinc chloride (E-merk grade) in tap water. The concentrations were measured by using a UV visible spectrophotometer (model Chemito 2000). The concentrations of heavy metals selected for study were 5 mg/l to 50 mg/l for different heavy metals. The adsorption of heavy metals per unit area on the glass container was found to be the minimum but, due to practical difficulties, plastic tubs were used for batch studies.

Experimental set-up

Batch studies were conducted in plastic tubs of 15-l capacity. A known volume of clear effluent

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(BOD = 175 mg/l) of domestic waste was put in each tub as a nutrient supply for plant growth. The total volume of the sample in each tub was 10 l. A sufficient quantity of heavy metal solution (stock solution) was added to get the desired overall metal concentration. Three or four fully matured plants, with a total weight of 1 kg, were kept in each tub so that 70% to 80% of the water surface was covered (Behara *et al*. 1983), while one tub without water hyacinth plants served as a control. The tubs were exposed to natural sunlight (5 h/day) in an open place for a period of 7–10 days. Periodically, aliquots of solutions were removed and metal concentrations determined. To counter water loss due to evaporation at the ambient air temperature of 30 ± 2°C, distilled water was added to maintain the same level in each tub.

The entire plants were then taken out and dried at 110°C in a hot air oven. Both wet and dry weights were recorded. The dried plants were ground to a coarse powder in a ball mill. A 5 g aliquot of the homogenized sample was accurately weighed and burnt to ashes in a muffle furnace at 500–550°C. The ash was digested using 10 ml nitric acid. The clean digest was diluted to 100 ml and stored for metal estimation. From the volume and concentration of metals in the residual solution, the amount of unabsorbed metals was calculated. Metal concentrations were measured using a UV visible spectrophotometer (model Chemito 2000) (APHA 1992). From the difference in the amount of metals in the tubs before and after treatment, the amount of metal uptake was calculated and expressed as mg/g of dry water hyacinth plant tissues.

RESULTS AND DISCUSSION

The removal of heavy metals from aqueous solution by water hyacinth was studied for a detention period of 7–10 days using selected heavy metals such as arsenic, chromium, mercury, nickel, lead and zinc at different concentrations. Each metal was tested separately at an initial concentration of 5 mg/l, and the decrease in metal concentration over the study period is given in Table 1. From the results it is obvious that as the detention time increases, the concentration of metal remaining in solution decreases, indicating its removal. The corresponding removal efficiencies were calculated and the values presented as cumulative percentage removals in Table 2 and in Figures 1–6. It is quite clear that the removal of heavy metals initially is very fast, and the final removal efficiency depends upon the initial concentration of the heavy metal, as shown in Table 3. The results indicate that the removal efficiencies were higher in solutions in which water hyacinth was grown as compared to those solutions in which the plants were not grown. The reason is that in the solutions in tubs where plants were not grown, the removal of metals was due to adsorption on the walls of the tub and sedimentation, while in solutions where the plants were grown, the plants absorbed the metals in addition to the other factors.

Similar experiments were also conducted for initial concentrations of 10, 15, 20, 25 and 50 mg/l and the results are summarized in Table 3.

A maximum removal efficiency of 65.4% was noticed when mercury was present at an initial concentration of 10 mg/l. Arsenic, chromium, lead, nickel and zinc were removed up to 32%, 27.80%, 40.20%, 41.98% and 33.60% respectively, at an initial concentration of 5 mg/l.

It was observed that for concentrations up to 10 mg/l of lead, the general health of the plants was normal until the end of the seventh day of the detention period. At higher concentrations, plants started wilting, leading to death. A similar observation was made in the case of mercury for concentrations up to 10 mg/l. However, in the case of arsenic and chromium, the plants started wilting after only 3 days when the concentration of metal was 10 mg/l, and the uptake of metal was less when compared with lead, mercury, nickel and zinc. In spite of the poor health of the plants, no ‘release’ of heavy metals by the plants was observed. The aquatic plants used in the batch study were also weighed, and the average size of leaves, roots, etc. was measured before and after the study for the concentration of 5 mg/l. It was observed that the average weight of plants increased by 13%.

An examination of Table 4 shows that the maximum uptake by water hyacinth of arsenic, chromium, mercury, nickel, lead and zinc was 0.0309, 0.108, 0.0327, 0.753, 0.107 and 2.270 mg/g dry plant tissue respectively. The

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Table 1 | Heavy metals remaining in solution*

Detention period (days)	Concentration of solution remaining in solution (mg/l)											
	Arsenic		Chromium		Mercury		Nickel		Lead		Zinc	
	X	Y	X	Y	X	Y	X	Y	X	Y	X	Y
1	3.61	4.50	4.03	4.50	2.95	4.55	4.26	4.43	3.28	3.91	4.00	4.65
2	3.13	4.31	3.52	4.30	2.52	4.33	3.95	4.25	2.40	3.41	3.63	4.38
3	3.00	4.22	3.14	4.21	2.14	4.10	3.28	3.93	2.08	3.15	3.02	4.14
4	2.55	4.15	2.99	4.10	1.99	3.90	2.42	3.52	1.67	2.90	2.38	3.48
5	2.50	3.85	2.50	3.83	1.80	3.90	2.10	3.16	1.16	2.83	2.12	3.25
6	2.50	3.75	2.33	3.50	1.53	3.89	1.67	3.00	0.80	2.81	1.86	3.16
7	2.45	3.75	2.00	3.39	1.20	3.90	1.07	2.83	0.80	2.60	1.53	2.95
9	—	—	1.86	3.10	—	—	0.69	2.79	—	—	1.39	2.86
10	—	—	1.80	2.80	—	—	0.60	2.65	—	—	0.94	2.62

*Initial concentration of heavy metals = 5 mg/l.
X = Solution in which water hyacinth was grown.
Y = Solution in which water hyacinth was not grown (control).

mass of metal per mass of plant increased with higher aqueous concentrations of nickel, lead and zinc. No trend was seen for chromium and mercury, and no apparent significant difference was noted for arsenic.

The kinetic data obtained from the laboratory studies have been mathematically analysed by determination of uptake rate constants (k) of water hyacinth in removal of different heavy metals, and a polynomial model has been obtained.

Verification of model

The time-variant material-balance equation for a batch reactor has been established (Metcalf & Eddy 1998). If first order kinetics are assumed, the resulting expression is:

$$C_o/C_t = e^{-kt} \qquad (1)$$

where C_o = initial concentration of parameter (mg/l),
C_t = concentration of parameter after time t (mg/l),
k = uptake rate constant (day^{-1}),
t = time period (days).

A linear relationship between $\log(C_t)$ and time t was observed giving an indication that the process was exhibiting first order behaviour as shown in Figure 7.

Equation (1) can be reduced to a straight-line equation as follows:

$$\log(C_t) - \log(C_o) = -kt \log e$$

$$\log(C_t) = -k \log e^{*}t + \log(C_o)$$

$$Y = mX + C \qquad \text{(Equation for straight line)}$$

The uptake constant (k) for water hyacinth in removal of heavy metals such as arsenic, chromium, lead, mercury,

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Table 2 | Cumulative percent removal of heavy metals*

Detention period (days)	Cumulative percent removal																	
	Arsenic			Chromium			Mercury			Nickel			Lead			Zinc		
	X	Y	Z	X	Y	Z	X	Y	Z	X	Y	Z	X	Y	Z	X	Y	Z
1	27.80	10.00	17.80	19.40	10.00	9.40	41.00	9.00	32.00	14.90	11.36	3.54	34.50	21.80	12.70	20.00	7.00	13.00
2	37.40	13.80	23.60	29.60	14.00	15.60	49.60	13.50	36.10	21.00	15.00	6.00	52.00	31.80	20.20	27.50	12.50	15.00
3	40.00	15.60	24.40	37.24	15.80	21.44	57.24	18.00	39.24	34.50	21.46	13.04	58.40	37.00	21.40	39.60	17.30	22.30
4	49.00	17.00	32.00	40.20	18.00	22.20	60.20	22.00	38.20	51.70	29.54	22.16	66.70	42.00	24.70	52.40	30.50	21.90
5	50.00	23.00	27.00	50.00	23.44	26.56	64.00	22.00	42.00	58.00	36.80	21.20	76.80	43.40	33.40	57.52	35.00	22.52
6	50.00	25.00	25.00	53.44	30.00	23.44	69.50	22.20	47.30	66.70	40.00	26.70	84.00	43.80	40.20	62.80	36.80	26.00
7	51.00	25.00	26.00	60.00	32.20	27.80	76.00	22.00	54.00	78.66	43.38	35.28	84.00	48.00	36.00	69.36	41.00	28.36
9	—	—	—	62.90	38.00	24.90	—	—	—	86.18	44.20	41.98	—	—	—	72.30	42.80	29.50
10	—	—	—	64.00	44.00	20.00	—	—	—	88.00	47.00	41.00	—	—	—	81.20	47.60	33.60

*Initial concentration of heavy metals=5 mg/l.
X=Solution in which water hyacinth was grown.
Y=Solution in which water hyacinth was not grown (control).
Z=Due to growth of water hyacinth i.e. (X–Y).

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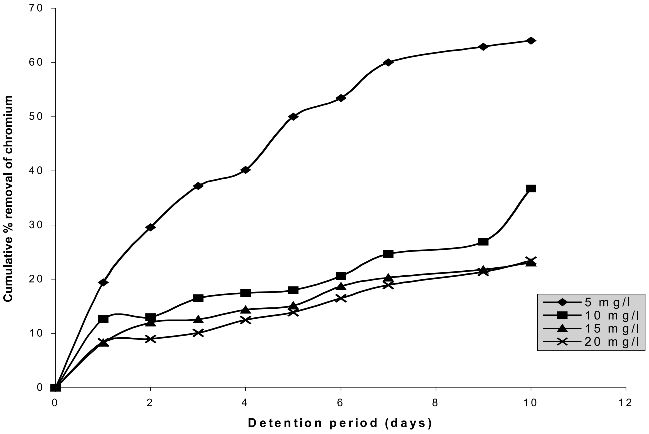

Figure 1 | Removal of chromium from synthetic wastewater.

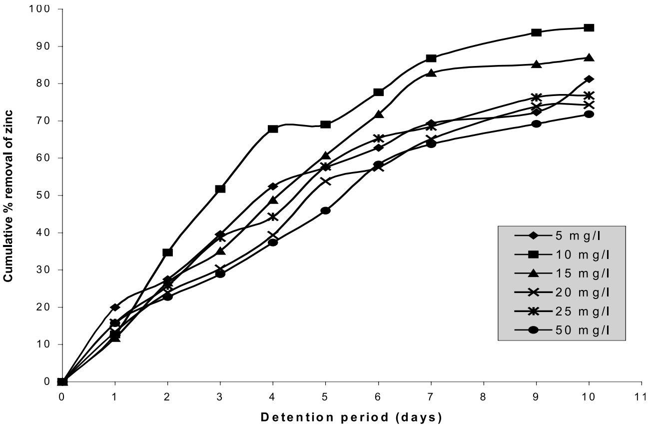

Figure 3 | Removal of zinc from synthetic wastewater.

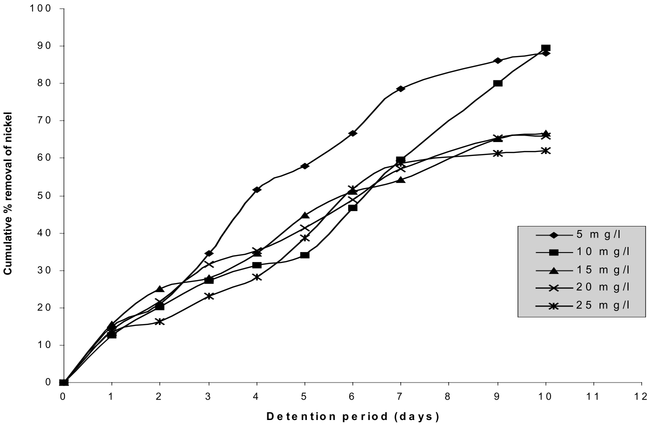

Figure 2 | Removal of nickel from synthetic wastewater.

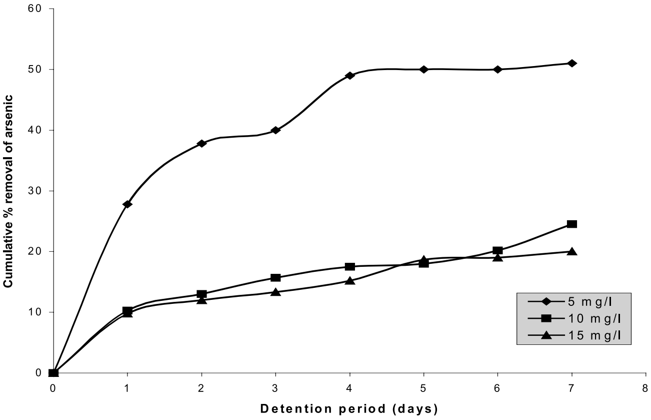

Figure 4 | Removal of arsenic from synthetic wastewater.

nickel and zinc is determined by using the results of the batch study in which the depletion of heavy metals was observed over 10 days.

From Table 5 it is observed that the uptake rate constant for water hyacinth in the removal of lead is higher than those for nickel, mercury, zinc, arsenic and chromium.

Polynomial model

An attempt was also made to establish a mathematical relationship between time and the concentration of nickel remaining in the water using a polynomial equation:

$$C = A_0 + A_1 T + A_2 T^2 + A_3 T^3 + \ldots + A_n T^n \quad (2)$$

In which C = concentration of heavy metal at time T in mg/l; T = time in days; n = order of polynomial; and A_0, A_1, A_2, $A_3 \ldots + A_n$ = constants.

The polynomial was solved, using a computer, by taking the value of n as 2 to 6 for each concentration, and choosing the best-fit equation of least order for different initial concentrations based on the coefficient of correlation.

Table 6 gives the order of the best-fit equation which should be selected using the parameter weighted coefficient of correlation from the computer program for each concentration of nickel. When the coefficient of

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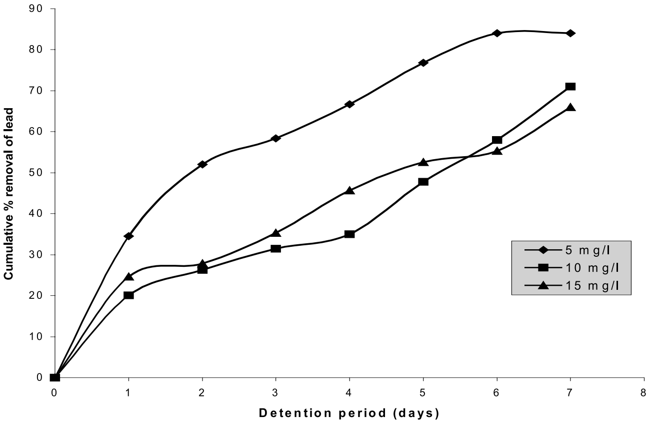

Figure 5 | Removal of lead from synthetic wastewater.

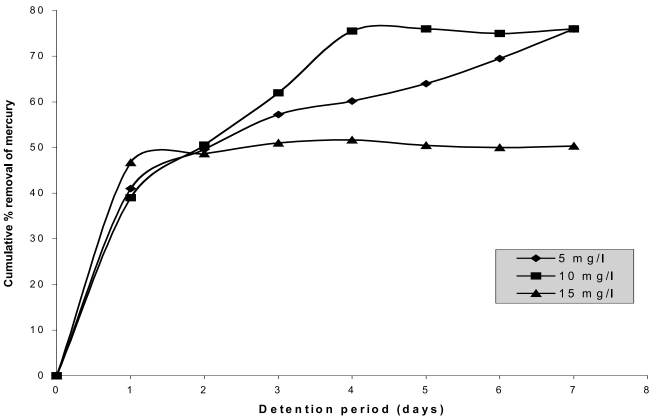

Figure 6 | Removal of mercury from synthetic wastewater.

correlation approaches 1, the corresponding equation gives results similar to actual results. Hence based on the coefficient of correlation, the degree of equation should be decided which gives significant results.

To verify the mathematical analysis, the values of concentration of nickel (in mg/l) remaining in solution at time T (days), and the concentration of nickel obtained from the mathematical formulae as f (T) are plotted. Figure 8 gives one such plot for an initial concentration of 10 mg/l and shows that the experimental values and computed values are more or less the same which indicates the 'best-fit' model.

Table 3 | Removal efficiencies of heavy metals

Metal concentration (mg/l)	Percent removal					
	Arsenic	Chromium	Mercury	Nickel	Lead	Zinc
5	26.00	20.00	54.00	41.00	36.00	33.60
10	8.50	14.45	42.00	32.00	16.00	45.00
15	3.33	3.73	2.33	28.00	34.33	38.00
20	—	4.00	—	27.40	—	34.26
25	—	—	—	27.70	—	40.80
50	—	—	—	—	—	41.80

Disposal of water hyacinth containing heavy metals

Preliminary work on the biogasification of heavy metal-spiked water hyacinth has indicated that the biomass can be anaerobically digested (Ishanand *et al.* 1982; Bhole & Ingole 2000; Ingole & Bhole 2000). However, the best method for disposal of the digested biomass remains an issue. Metal speciation studies must be conducted on both undigested and digested biomass before either is put onto land as fertilizer.

CONCLUSIONS

1. The general health of the water hyacinth plants was good up to a concentration of 5 mg/l for all metals, and was adversely affected when the metal concentration increased above 15 mg/l.
2. The removal efficiency (as mass of metal removed per mass of plant dry weight) of heavy metals was found to decrease with increase in concentration of metals.
3. The results also reveal that plants have a higher uptake value for mercury and the survival period of plants was longer in solutions containing mercury. Hence, water hyacinth is the better option for the bioaccumulation of trace mercury from the lower to the higher range.

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Table 4 | Removal of heavy metals by water hyacinth

Sr. no.	Metal	Initial metal concentration (mg/l)	Metal concentration in dry plant tissues (mg/g)
1	Arsenic	5	0.0266
		10	0.0267
		15	0.0309
2	Chromium	5	0.1080
		10	0.1070
		15	0.0610
		20	0.0870
3	Mercury	5	0.0327
		10	0.0250
		15	0.0315
4	Nickel	5	0.2230
		10	0.3550
		15	0.4570
		20	0.5960
		25	0.7530
5	Lead	5	0.0456
		10	0.0520
		15	0.1070
6	Zinc	5	0.1830
		10	0.4890
		15	0.6190
		20	0.7460
		25	1.1090
		50	2.2700

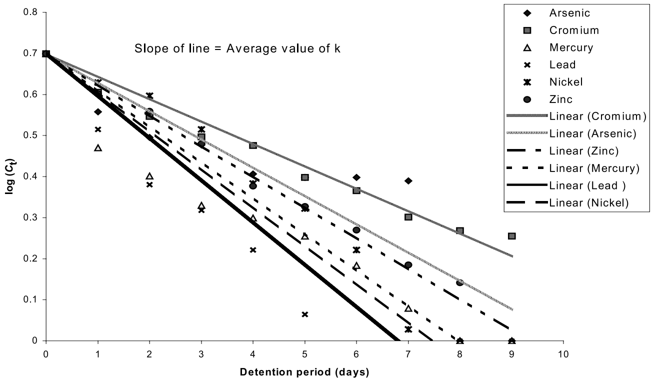

Figure 7 | Plot of log (C_t) vs time for different heavy metals.

Table 5 | Uptake rate constants for water hyacinth in removing heavy metals

Sr. no.	Metal	Uptake rate constant (k) (day^{-1})	Coefficient of correlation
1	Arsenic	0.0693	0.825
2	Chromium	0.0548	0.968
3	Mercury	0.0879	0.885
4	Nickel	0.0937	0.950
5	Lead	0.1027	0.789
6	Zinc	0.0749	0.990

4. The uptake rate constant (k) for water hyacinth in the removal of zinc is found to be higher compared with lead, mercury, arsenic, nickel and chromium.
5. The kinetics of absorption which have been developed can be used to achieve different concentrations of heavy metals in polluted waters using the hyacinth system.
6. Since water hyacinth plants grow and photosynthesize rapidly, and are adapted to an aquatic mode of existence, they could prove useful in assessing and rectifying heavy metal pollution, particularly in water containing zinc and mercury.

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Table 6 | Order of equations for polynomial model for nickel

Order of equation	Coefficient of correlation	Initial concentration of nickel (mg/l)
4	0.995	5
4	0.995	10
4	0.993	15
5	0.998	20
5	0.998	25

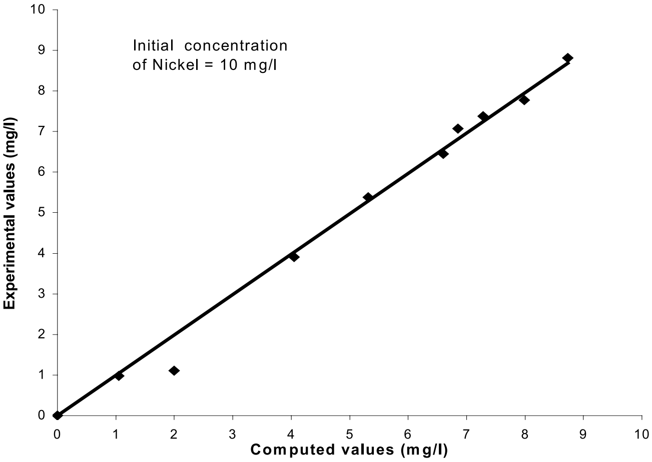

Figure 8 | Verification of kinetic model for nickel removal.

7. The system proposed is of simple technology and holds promise of economic viability in rural areas where large areas of land will be available at minimum cost.

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