Distribution and pollution status of metals in some water bodies of mid-Brahmaputra valley

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ABSTRACT

A comprehensive assessment of metals in wetlands of Guwahati were done in three systems – water, sediments and aquatic plants. Enrichment factor calculated for sediment showed that lead is significantly enriched. HPI and HEI values indicate that both lotic and lentic water bodies were critically polluted. TF shows that the metals were mostly translocated to the aerial parts. The correlation calculated among metal pairs showed that they were of similar origin. analysis of variance (ANOVA) reveals that the source of metals in all the three matrices is similar. The SEM-EDX micrograph of root of *Eichhornia crassipes* revealed the accumulation of metals in the tissues and this confirms the internalization of the metals. The FT-IR spectra of *Eichhornia crassipes* may be attributed to the interaction of metal ions with the hydroxyl, amide, thiol, and amino groups present in the plant biomass.

Keywords: Metals; Enrichment factor; HPI; HEI; TF; ANOVA; SEM-EDX; FT-IR; *Eichhornia crassipes*

1. Introduction

Wetlands are marked by biologically diverse and productive transitional areas situated between land and water. This ecotone renders many important services in the ecosystem. Various laws have been enacted to protect the wetlands like the Ramsar Convention on Wetlands of International Importance, especially as Waterfowl Habitat in 1971. However, due to rapid urbanization, a significant depletion is observed in terms of total area of the wetlands. In addition, the remaining flats are also contaminated with metals and organic compounds discharged from various sources. Topography of the wetland makes it a natural sink for the metals. Waste water, city runoff and traffic related pollutants are the main carriers of toxic substances to the wetlands and other lentic water bodies. Once the metal enters into the natural waters bodies, it becomes a part of the system and is partitioned between the sediment and water column

phase. Due to strong affinity between the sediment organic carbon and metals, sediments behave as large repository of metals. The determinant for stabilization of metals in wetlands depends on adsorption, ligand exchange, sedimentation etc [1]. The pH and redox condition of the sediment greatly influence the mobility and bioavailability of metals to benthonic organisms and also acts as a sink by absorbing them from the water column [2].

Metals are of high ecological significance since they are not degraded in the environment and get accumulated and biomagnified in the food chain [3]. Various aquatic plants are known to accumulate metals from their environment [4–9]. The ability of water hyacinth to accumulate high concentration of heavy metals from their immediate environment in roots and above ground parts has been studied extensively. The research pertaining to uptake and bioaccumulation of trace elements from aqueous solution by some vegetation collected from Deepor beel has already been published elsewhere by our earlier research [10]. The root system and rhizomes of water hyacinth provide an

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expanded area to absorb metal ions and has some unique characteristics which is favorable for metal accumulation [11–13], such metal accumulating plants are generally being used to clean up water in small-scale sewage treatment technology [14].

Besides, populations living in the vicinity of these water bodies may be at a greater risk of exposure to metals as people traditionally harvest vegetables, flowers, aquatic seeds, medicinal plants, fish etc. for their day to day use.

The objective of this study is to investigate the degree of contamination caused by metals in three different compartments viz. water, sediment and plants in some of the lotic and lentic water bodies of Guwahati. Both lentic and lotic water bodies were taken into consideration while this study was carried out with a view to get lucid insight of the behavior and fate of metals in different types of water bodies.

2. Materials and methods

2.1. Physiography and climate

Guwahati city is situated on the southern bank of the river Brahmaputra. It lies at the point of intersection of 26°11′ North latitude and 91°45′ East longitude. The undulating and bowl shaped topography of Guwahati valley is surrounded by hills on all three sides with varying altitudes of 49.5 m to 55.5 m above Mean Sea Level (MSL). About 61.8% of the total area of the city is covered by hills, water bodies and low lying areas. The river Brahmaputra and its three rivulets Bharalu, Khanajan and Bondajan receives water from their catchment areas and inundated low lying areas of most part of the city. The untreated city run off, industrial effluents and municipal waste were discharged in the annual and perennial water bodies and finally the waste water outflows into the Brahmaputra.

The climate of the city is humid, sub-tropical and characterized by high rain and is influenced by the southwest monsoon. Yearly rainfall average stands at 162 cm with about 110–115 rainy days per year. Rainfall intensity is higher during monsoon period and causes water logging in some parts of the city [15]. The climate is characterized by high rainfall (more than 295.6 mm), high temperature and high humidity. Maximum temperature goes up to 32°C in July–August and minimum falls to 11°C in January.

2.2. Study area

The study area covers mainly the Guwahati Municipal Corporation (GMC) area with a total area of about 243.2 sq. km. The location of all the sampling stations for surface water, bed sediment and *Eichhornia crassipes* were shown in Fig. 1. Selection of sampling sites were decided keeping in mind the locations and pollution aspects of the water bodies. The sampling stations, type of water bodies and description of the site were presented in Table 1.

2.3. Collection of surface water, bed sediment and water hyacinth

Surface water samples were collected in 1 L polyethylene containers pre-cleaned with 10% reagent grade HNO_3, followed by rinsing with deionized water and drying [16]. Sediment samples were collected from the same sites from the top 5 cm layer of bottom sediments using a grab sampler in self-sealing polythene bags and transported to laboratory as early as possible. The collected samples were air dried, ground, sieved and stored for further analysis [17]. *E. crassipes* was also collected from all the sampling locations. They were first washed with tap water and finally with distilled water to remove all the particles from the plant [18]. *E. crassipes* was also collected from an unpolluted site which has been used as a control.

2.4. Analytical techniques

For determination of metals in the water sample, it was first acidified to a pH < 2.0. After acidification, the samples were stored in refrigerator at 4°C till analysis for the metals was undertaken. The water samples were mostly turbid with an appreciable load of suspended particulates, so samples were digested to obtain the concentration of both dissolved metals and those associated with the particles. In this work, nitric acid digestion technique [16] was used.

Sediment samples were acid digested for metal analysis by the method earlier used by Aksoy et al. [17]. One gram of sample was transferred into a teflon bomb and 10 ml of aqua-regia ($1HNO_3:3HCl$) was added to digest the samples. After digestion, samples were filtered and the filtrate volume was made up to 25 ml with ultrapure water.

The *E. crassipes* collected from different sites were dried at room temperature and then oven dried at 80°c for 24 h. Dried plant samples weighing 1.00 gm were digested in 10 mL concentrated HNO_3. After cooling, the suspension was filtered in a 25-mL volumetric flask and diluted to the mark [19]. The heavy metals were estimated by ICP-OES (PerkinElmer/Optima 2100DV).

2.5. Statistical analysis

The correlation matrix of the metals was built to find out the associations between the variables. Significant positive correlations can be explained in terms of common source or chemical similarity among the metal pairs [20].

One way analysis of variance (ANOVA) was carried out between different environmental matrices – water and sediment, sediment and *E. crassipes*, and water and *E. crassipes*, to examine that source of metal for all the three environmental matrices was similar or different.

2.6. Scanning electron microscope (SEM) and energy dispersive X-ray (EDX) studies

Roots, shoots and leaves of *E. crassipes* were subjected to primary fixation using 3% glutaraldehyde in 0.05 M phosphate buffer for 90 min, followed by secondary fixation in 2% osmium tetroxide in 0.01 M Sodium cacodylate buffer for 30 min [21]. The samples were dehydrated in an acetone series. SEM images were recorded for the samples using SEM model JEOL JSM-6390 LV, attached with EDX unit, with an accelerating voltage of 20 kv.
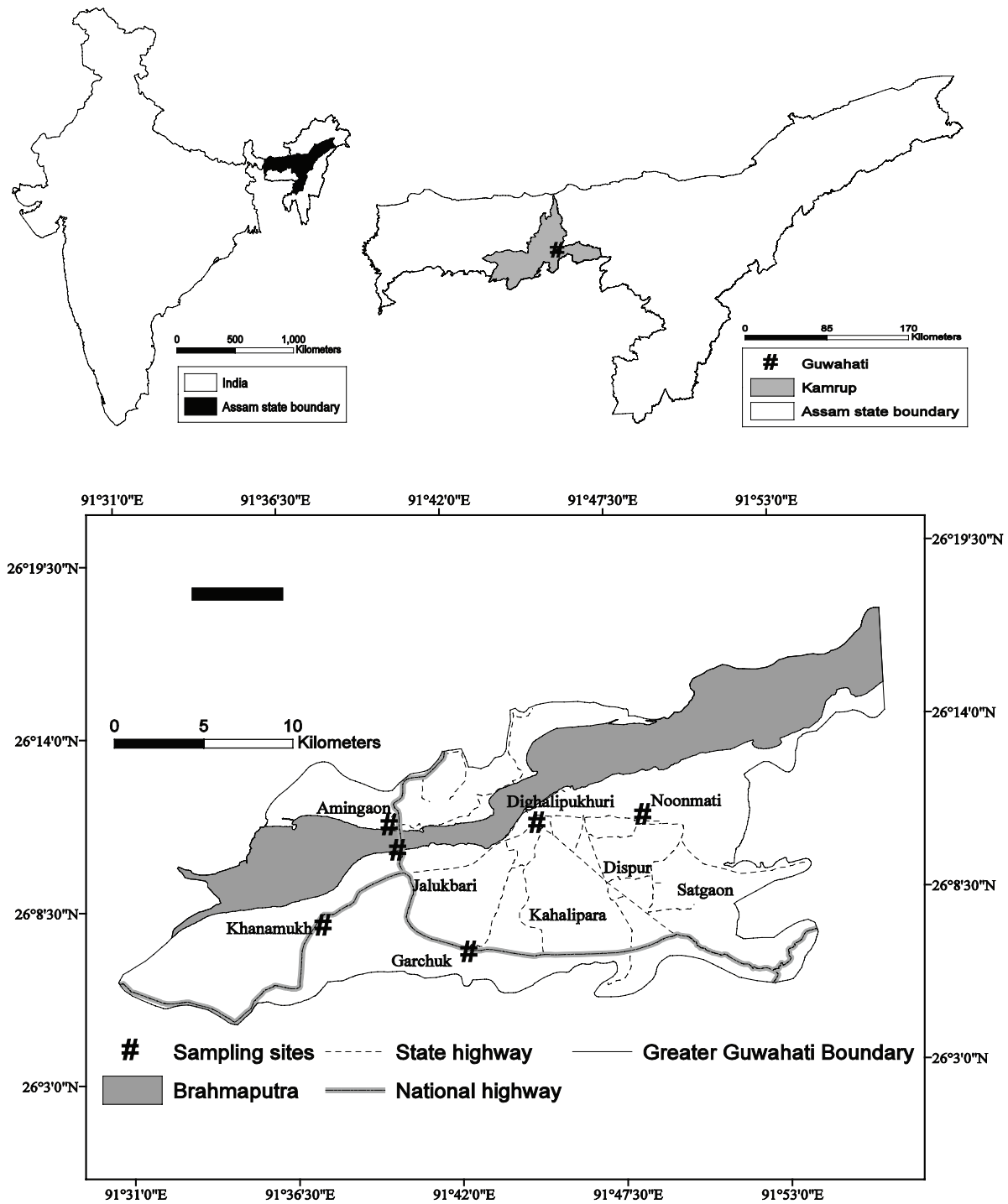
Fig. 1. Map showing sampling stations.

2.7. Fourier transform infrared spectrometer (FTIR)

E. crassipes samples were oven dried at 60°C and ground into fine powder through agate mortar. Two milligrams of the sample were mixed with 200 mg KBr (FT-IR grade) and pressed into a pellet. The pellet was immediately put into the sample holder and FT-IR spectra were recorded in the range 4,000–450 cm^{-1} for all samples [22].

3. Results and discussion

3.1. Metal concentration in water, bed sediment and E. crassipes

The average concentration of all the four metals viz. Mn, Ni, Pb and Zn in the three different compartments were presented in Table 2.

Table 1
Sampling points of the study area along with its type and site description

Station	Name	Type	Description of the site
S1	Noonmati	Lotic	In this site, the untreated effluent of an oil refinery is discharged.
S2	Ambari	Lentic	This site is situated in the mid of the city. The commercial as well as domestic run off get discharged.
S3	Gorchuck	Lentic	This is a perennial wetland (Deepor beel). It is one of the largest open filled municipal dumping site of the Guwhati city.
S4	Khanamukh	Lentic	This is also a perennial wetland (Deepor beel). At this site the untreated municipal run-off is discharge.
S5	Near Saraighat bridge	Lotic	This is an intermediate zone between a wetland and river, where the domestic, industrial and commercial effluent were discharged. This site is also near a busy highway. So the water body is also polluted by the road runoff.
S6	Amingaon	Lentic	This wetland is off annual type. The main source of pollution in this site is the road run-off.

Table 2
Shows the concentration of metals in water, sediment and *E. crassipes*

Sampling station	Environmental matrices (ppm)	Pb	Mn	Ni	Zn
1	Water	0.133	0.247	0.009	0.148
	Sediment	6.3	24.9	2.4	14.4
	E. crassipes	36.85	356.55	23.9	148.5
2	Water	0.04	0.181	0.010	0.576
	Sediment	6.9	41.9	3.4	9.6
	E. crassipes	27.60	925.5	18.95	79.55
3	Water	0.056	0.191	0.055	0.194
	Sediment	4.4	31.7	3.9	5.9
	E. crassipes	25	2,938	26	80
4	Water	0.055	0.125	0.037	0.077
	Sediment	3.9	31.1	5.4	6.3
	E. crassipes	10	685.5	15	59.5
5	Water	0.057	0.044	0.04	0.092
	Sediment	5	9	2.8	2.3
	E. crassipes	29	604	20	34.9
6	Water	0.056	0.053	0.028	0.068
	Sediment	5.2	28.8	5	10.4
	E. crassipes	39.6	61.5	11.2	42.8
Transfer factor (*E. crassipes*)		5.248	36.725	5.722	10.143

The concentration of Pb in all the water samples was higher than the limit of USEPA for fresh water aquatic life (0.034 µg/ml). Similarly, Mn concentration in Stations 1, 2, 3, and 4 were higher than the limit of USEPA for fresh water aquatic life (0.1–1 µg/ml) but lower in Stations 5 and 6. Average concentrations of zinc in Stations 2 and 3 were found to be higher than the limit of USEPA for fresh water aquatic life (0.18 µg/ml) [23].

The average values of lead, zinc, manganese and nickel for bed sediment in all the six stations were found to be lower than the limit of USEPA fresh water sediment quality criteria for typical contaminated sediment, which is 36 µg/g (TEC) [24,25] for lead, 460 µg/g (TEC) for manganese, 120 µg/g (TEC) for zinc and 22.7 µg/g for nickel (TEC). Though the concentration of metals was found to be low in water, a relatively high concentration of metals was found in bed sediment. The trend of metals in bed sediment is in the order Mn>Zn>Pb>Ni. Similar results have also been reported by Bazrafshan et al. [26] for Chah Nimeh water reservoir of Iran.

In the present study, average concentration of lead, manganese, nickel, and zinc in the *E. crassipes* collected from the sampling stations were found to be significantly higher than the concentration of these metals in the reference sample. The accumulation of metal ion in *E. crassipes* of all six sampling stations was in the following order –Mn>Zn>Pb>Ni.

The concentration of manganese in the *E. crassipes* was highest among the metals. The higher average concentration of manganese as compared with concentration of manganese in reference sample could be due to its bio-absorption from polluted water.

It is interesting to note that although the concentration of metals in water was low, but the plant accumulates higher concentration of metal. This reveals that the uptake of heavy metals by *E. crassipes* not only depends on the concentration of metals in surface water but also on other factors like pH, temperature, presence of chelating agent and other ions in water. Comparatively a very high uptake value of lead was found in sampling Station S6 because the site was polluted by road runoff from busy traffic area of Amingaon.

3.2. Correlation and ANOVA of metal

A significant positive correlation was found among the metal pairs Mn-Pb ($r = 0.46$), Ni-Pb ($r = 0.86$), Ni-Mn ($r = 0.75$), Zn-Pb ($r = 0.82$), Zn-Mn ($r = 0.55$) and Zn-Ni ($r = 0.89$). The strong correlation among metal pairs suggested that the source of pollution is similar. Industrial, domestic and vehicular effluents are the main anthropogenic sources of metals within the study area. Strong correlation between zinc and lead reveals that these metals come from vehicular traffic [27]. ANOVA reveals that the source of metals in all the three matrices is similar and is significant at 0.01 level.

3.3. Assessment of pollution level

3.3.1. Enrichment factor (EF)

EF of heavy metals were calculated for each sediment samples of six stations. The minimum, maximum and mean of the EF values are presented in Fig. 2. The EF was calculated to see the influence and degree of anthropogenic activities. EF of trace metals were calculated for each samples relative to the background value of the elements in the crust, choosing zinc as the reference element as it is one of the geochemical markers. The EF of an element is calculated as follows:

$$EF = [C_x \text{ (sample) } / \, R \text{ (sample)}] / \, | \, C_x \text{ (reference) } / \, R \text{ (reference)}] \quad (1)$$

where, C_x is the metal concentration; R is the reference element concentration. Here, the reference values considered were the crustal values. EF value less than 2 are not considered significant because such enrichment may arise from differences in the composition of local soil material and reference soil used in EF calculations [28]. Five contamination categories are recognized on the basis of the EF. EF < 2 minimal enrichment; EF = 2–5 moderate enrichment; EF = 5–20 significant enrichment; EF = 20–40 very high enrichment and EF > 40 extremely high enrichment [29].

In recent decades, a rapid increase in motor vehicles in Guwahati has aggravated heavy metal pollution in the aquatic system as traffic-related metals in road dust can enter the sediments through urban runoff. In the present study, lead shows moderate enrichment in S1, S2, S3, S4, S6, whereas in S5, there is significant lead enrichment (Fig. 2). The sources of lead in the sediments of water bodies could be the leaded gasoline which was used heavily as anti-knocking substance before it was being phased out. As it is non-biodegradable, sediment is a good repository of such metals. Manganese and Nickel showed no enrichment. The source of these two metals thus may be geogenic.

3.3.2. Heavy metal pollution index (HPI)

HPI is a method of rating that shows the composite influence of individual heavy metal on the overall quality of water.

Fig. 2. Boxplot showing EF of metals.

The rating is a value between zero and one, reflecting the relative importance of individual quality considerations and defined as inversely proportional to the recommended standard (Si) for each parameter [30,31].

$$HPI = \sum_{i=1}^{n} W_i Q_i / \sum_{i=1}^{n} W_i \quad (2)$$

where Q_i is the sub-index of the ith parameter, W_i is the unit weightage of the ith parameter and n is the number of parameters considered. The sub-index (Q_i) is calculated by:

$$Q_i = \sum_{i=1}^{n} (M_i - I_i) / (S_i - I_i) \times 100 \quad (3)$$

where M_i, I_i and S_i are the monitored value of metal, ideal and standard values of the ith parameter, respectively.

The critical pollution index value, above which the overall pollution level should be considered unacceptable, is 100 [31,32]. The results of HPI are given in Table 3. HPI indicated S1 as a critically polluted area, whereas the other stations were found to be below the critical pollution index value. The reason for the high HPI value at S1 (Noonmati) may be due to the fact that at this sampling point, the untreated effluent of the oil refinery gets discharged.

3.3.3. Metal evaluation index (HEI)

The HEI method gives an overall quality of the water with respect to heavy metals [33] and is computed as follows:

$$HEI = \sum_{i=1}^{n} H_c / H_{mac} \quad (4)$$

where H_c is the monitored value of the ith parameter and H_{mac} the maximum admissible concentration of the ith parameter.

By following the approach as proposed by Edet and Offiong [33], HEI criteria for the samples are as follows: low (HEI < 10), medium (HEI = 10–20) and high (HEI > 20). The present level of HEI shows that the water quality of all the sampling stations falls within high zone of pollution (Table 3). The reason for high HEI in all the stations is due to the continuous discharge of untreated waste from various sources. If this continues, the sampling stations will become a "potential" source of metals. This is of concern as during flood, the metals from these sites may contaminate other water bodies nearby. Moreover, high HEI in Deepor Beel, which is the only Ramsar site of Assam, poses a risk to its existing flora and fauna.
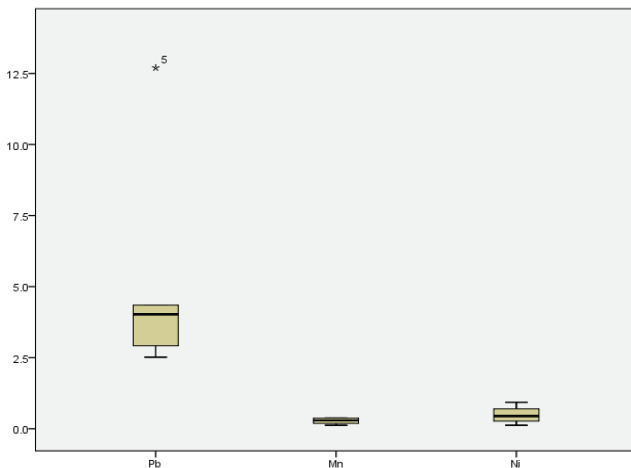
Table 3
Represents HPI and HEI values in the sampling stations

Sampling stations	HPI	HEI
S1	142.78	94.09
S2	58.83	30.90
S3	85.33	43.94
S4	71.04	41.03
S5	62.94	40.90
S6	60.86	39.81

3.3.4. Transfer factor (TF)

Translocation factor or mobilization ratio [34,35] was calculated to determine relative translocation of metals from soil to other parts (root and shoot) of the plant species.

$$\text{TF} = \text{Concentration of metals in plant tissues / Concentration of metal in corresponding soil} \qquad (5)$$

Plants with translocation factors greater than one (TF > 1) have the potential to be used in phytoextraction [36]. Metals that are accumulated by plants and largely stored in the roots of plants are indicated by TF values <1. TF > 1 indicate translocation to the aerial parts of the plant [37]. The TF values calculated for *E. crassipes* are presented in Table 2. The TF follows the order Mn>Zn>Ni>Pb. The uptake of Mn and Zn is higher as they are essential elements for plant growth [38], whereas the uptake of Pb and Ni is low as they are non-essential elements. It was observed that all the TF values were greater than one which indicates that the metals were mostly translocated to the aerial parts.

3.4. SEM and EDX

The SEM-EDX micrograph of root of *E. crassipes* collected from sampling Station 6 revealed the accumulation of zinc, lead and nickel in the tissues which confirms the uptake of metals by the plant roots (Fig. 3(a) and 3(b)). Although very low quantity of nickel was present both in water and sediments, nickel was also up taken by the plants which is evident from both SEM-EDX and analysis of ICP results. This is because metal uptake capacity of an aquatic macrophyte are affected by some biological and non-biological factors viz. plant species and different organs, season, temperature, pH, metal concentration, and exposure time [39]. In the EDX spectra (Fig. 3(b)), Mn was not shown which may be due to absence of Mn in that portion.

3.5. Fourier transform infrared spectroscope (FTIR)

The FTIR technique was used to identify the different functional groups of *E. crassipes* involved in binding the metal ions. It is seen that there is a significant shift of the absorption peaks of functional biomass groups of plants. The characteristic N-H stretching frequency of amine/amide at 3,419.03 cm^{-1} in leaf (Fig. 4(a)), 3,429.24 cm^{-1} in root (Fig. 4(b)) and 3,428.29 cm^{-1} in shoot (Fig. 4(c)) of a *E. crassipes* plant taken from an unpolluted site (control) have been shifted to 3,448.00 cm^{-1} in leaf (Fig. 4(d)); 3,435.89 cm^{-1} in root (Fig. 4(e)); and 3,430.85 cm^{-1} in shoot (Fig. 4(f)), respectively, of a *E. crassipes* plant taken from a polluted site (contaminated). This may be due to complexation of metal ions with *E. crassipes*. The medium to strong peaks of υ(O-H) of carboxylic acid at 2,919.08 cm^{-1} in the leaf; 2,921.62 cm^{-1} in the root; and 2,922.12 cm^{-1} in the shoot of the control plant were shifted to 2,926.66 cm^{-1} in leaf; 2,924.33 cm^{-1} in root; and 2,923.78 cm^{-1} in the shoot of the plant (contaminated) after coordination of metal ions with carboxylic acid group. Very strong absorption bands of plant samples (control) are observed at 1,651.51 cm^{-1} in leaf; 1,638.05 cm^{-1} in root; and 1,624.81 cm^{-1} in shoot which may be due to the presence of bonded C=O stretching frequencies of amide, have been shifted to 1,634.21 cm^{-1} in leaf; 1,639.32 cm^{-1} in root; and 1,627.21 cm^{-1} in shoot of contaminated *E. crassipes* plant. A weak absorption band appeared at 1,372.96 cm^{-1} in leaf; 1,382.91 cm^{-1} in root; and 1,369.44 cm^{-1} in shoot of the plant (control) due to C-H assymetrical/symmetrical stretching of methyl group have also been shifted to 1,318.50 cm^{-1} in leaf; 1,319.50 cm^{-1} in root; and 1,318.79 cm^{-1} in shoot of the contaminated plant sample due to complexation with metal ions. The C-N stretching frequencies appeared at 1,061.77 cm^{-1} in leaf; 1,033.44 cm^{-1} in root; and 1,028.17 cm^{-1} in shoot of control plant which shifted to 1,036.81 cm^{-1} in leaf; 1,035.52 cm^{-1} in root; and 1,067.63 cm^{-1} in shoot due to contamination due to metal ions present in the *E. crassipes* (contaminated). A change in peak position in the spectrum of the metal loaded biomass
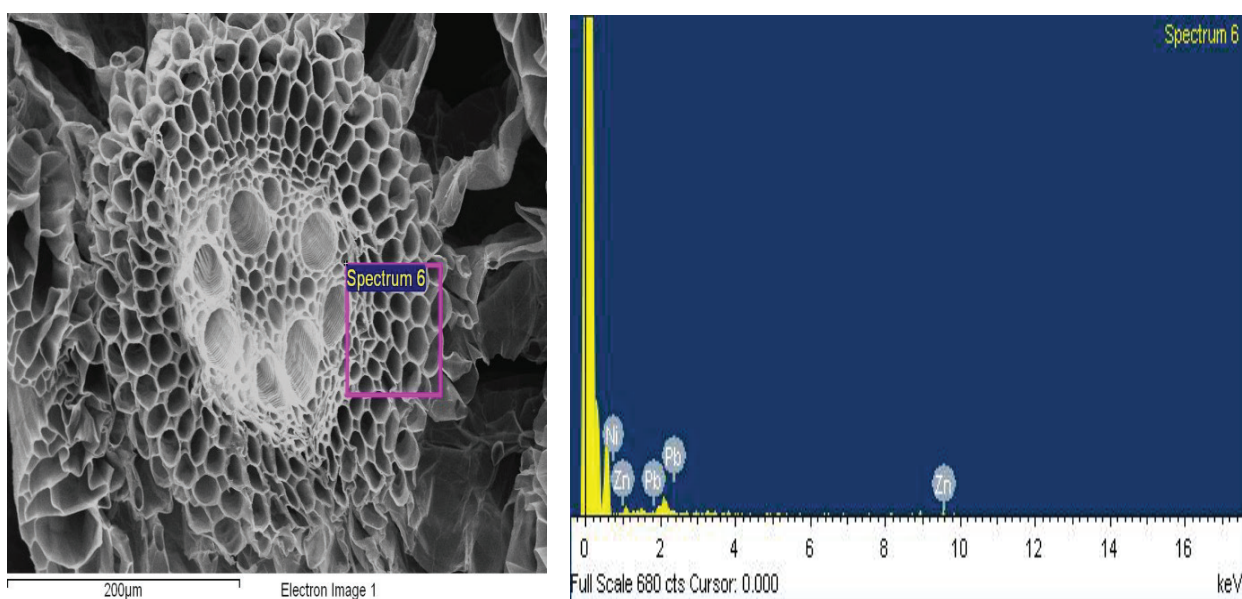

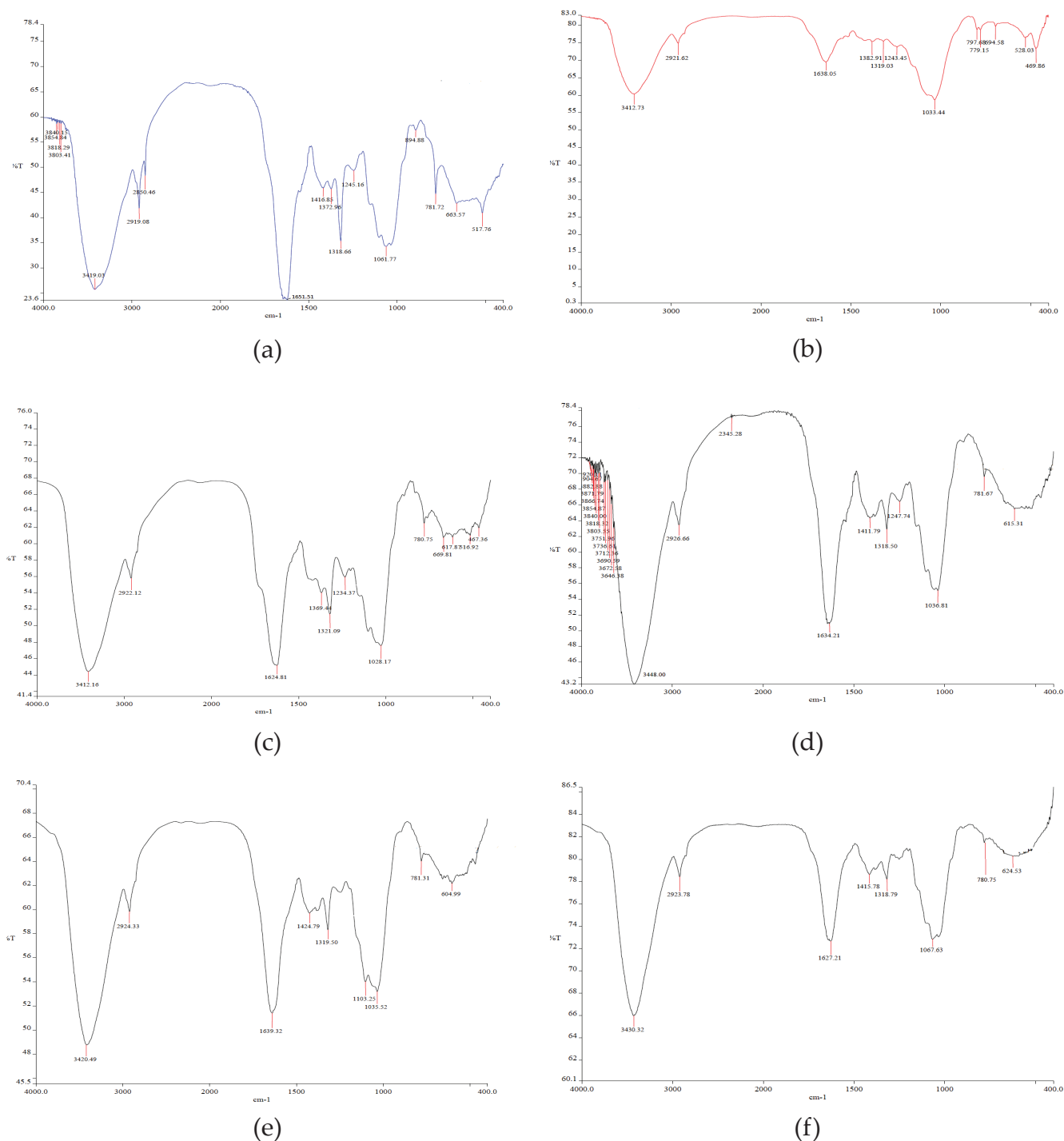Fig. 3. (a) SEM image of root of the Fig. 3(b); (b) EDX-micrograph of the root of water *E.crassipes*.

Fig. 4. (a) FTIR spectra of control leaf of *E. crassipes*; (b)FTIR spectra of control root of *E. crassipes*; (c) FTIR spectra of Control shoot of *E. crassipes*; (d) FTIR spectra of polluted leaf of *E. crassipes*; (e) FTIR spectra of Polluted root of *E. crassipes*; (f) FTIR spectra of Polluted shoot of *E. crassipes*.

indicates the involvement of amide group in the bioabsorption process. The band observed at 781.72 cm⁻¹ in leaf, 797.68 cm⁻¹ in root and 780.75 cm⁻¹ in shoot of the control plant sample could be assigned to frequency (C=O) which shifted to 781.67 cm⁻¹ in leaf, 781.31 cm⁻¹ in root and 788.00 cm⁻¹ in shoot of the contaminated plant could be due to coordination of metal ions present in the *E. crassipes*. Another shift of absorption peak of biomass of *E. crassipes* (control) was observed from 663.57 cm⁻¹ in leaf, 664.58 cm⁻¹ in root and 669.81 cm⁻¹ in shoot to 615.31 cm⁻¹ in leaf, 604.99 cm⁻¹ in root, 624.53 cm⁻¹ in shoot of the contaminated *E. crassipes* corresponding to the thiol or sulfhydral group.

4. Conclusion

The average concentration of lead in water in all the stations, manganese in Stations 1, 2, 3 and 4 and zinc in Stations 2 and 3 were found to be higher than the limit of USEPA for fresh water aquatic life. Higher concentration of these metals may lead to toxicity to the aquatic dwellers. Higher concentration of Pb in water in all the stations could be due to the untreated effluent released into both lotic and lentic water bodies. The average values of lead, zinc, manganese and nickel in sediments of all the six stations were found to be lower than the limit of USEPA fresh water sediment quality criteria for typical contaminated sediment. Moderate lead enrichment was found in S1, S2, S3, S4, S6 whereas in S5, a significant lead enrichment may be due to prior use of non-degradable leaded gasoline. Manganese and nickel shows deficient to minimal enrichment in all the stations. It was found that the metal concentration in lotic water bodies were less as compared with lentic water bodies. It may be due to the fact that with due courses of time the metals were infiltrated to the deeper zones of the sediment. This is of concern as the wetlands are recharge point for the ground water and in time series the wetlands may become "potential source" of metals to the ground water. The SEM-EDX micrograph of root of *E. crassipes* revealed the accumulation of manganese, zinc and lead in the tissues which confirms the uptake of metals by the plant roots. The shifts in the absorption peaks generally observed indicates the existence of metal binding process taking place in the plant biomass. FT-IR indicates that carboxylic, alcohol or phenolic, amide, amine and thiol groups are responsible for the binding of the metal ions. *E. crassipes* is a suitable plant for phytoextraction of metals and can also be used for reclamation of wetlands.

Acknowledgments

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