

Levels and Source Apportionment of Trace Metals in Bed Sediment of the Bharalu Tributary of Brahmaputra River

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Abstract: Bharalu Tributary originates in the foot-hills of Khasi Hills and meets the Brahmaputra River flowing northwards through Guwahati city. Trace metals viz., Pb, Ni, Cr, Cd, Cu and Zn were estimated in the bed sediment of Bharalu and their concentrations were found to be 39.54 ± 31 , 40.62 ± 19 , 32.00 ± 18 , 0.93 ± 1 , 65.45 ± 33 and $435.08 \pm 246 \mu\text{g g}^{-1}$ respectively. Levels of Cd, Pb and Zn in the bed sediments were 3, 1.5 and 5 times, respectively, higher than the concentrations in shale. Enrichment factors (EF%) of the metals showed substantial loading of the sediments with trace metals. Muller's Geoaccumulation index (I_{geo}) of the metals was calculated which showed that the river was moderate to strongly polluted. Pearson's correlation showed stronger associations for the metal pairs—Pb-Cd ($r = 0.90$), Ni-Cu ($r = 0.68$), Ni-Zn ($r = 0.61$), Cu-Zn ($r = 0.87$) and Pb-Cu ($r = 0.75$)—indicating similar sources. A multivariate statistical tool, Principal Component Analysis (PCA), was used to define the possible input sources of the trace metals and three main sources viz. industrial discharge, vehicular contribution and waste dumping were identified.

Key words: Bharalu, trace metals, sediments, I_{geo} , PCA.

Introduction

In large cities, water bodies receive loads of contaminants from untreated sewage discharge, urban runoff and atmospheric deposition. In the Indian cities, water bodies are also often seen as sites for waste dumping leading to their degradation.

Bed sediments, which conserve important environmental information, have been extensively used as environmental indicators to understand the extent of pollution (Gutiérrez et al., 2004). Sediments are also increasingly recognized as both carriers and possible sources of contamination in aquatic systems. At the bottom of water column, sediments play an important role in identifying the pollution scheme of the river systems by pollutants such as trace metals (Borovec,

1996; Duzzin et al., 1988; Huang et al., 1994; Lapaquellerie et al., 1995; Pardo et al., 1989; Wardas et al., 1996). They may also reflect both present and past discharges to the water system and can be of immense value while assessment of contaminations is addressed. The bed sediments in surface water systems act both as a sink and a source of metals. Metal accumulation in sediments provides a record of the spatial and temporal history of pollution (Martin and Meybeck 1979). Distribution of trace metals in water, sediments, plants, and fish plays a key role in detecting sources of trace metal pollution in aquatic ecosystem (Forstner and Wittman, 1981).

Trace metals have a great ecological significance due to their toxicity and accumulative behaviour. These elements, contrary to most pollutants, are not

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biodegradable and undergo a global eco-biogeochemical cycling (Nurnberg, 1984), in which natural waters are the main pathways. Sediment monitoring, therefore, can provide important information on various pollution events. Metal contaminated sediments may release metals back to the overlying water column and, thus, pose risk to aquatic life and ecosystems (Nriagu, 1989).

Besides assessment of pollution, the interest in the studies of metal constituents in sediments is also to understand their biogeochemical cycling. Rivers carry the continental weathered products to oceans and play a major role in global sea-water evolution; therefore, such studies help in understanding the geochemistry of an area (Chakrapani, 2005). Geographic variations, also, clearly define the trace elements chemistry due to the differences in climatic, weathering and anthropogenic conditions prevailing in the region (Martin and Meyback, 1979; Martin et al., 1993).

Almost all the important rivers of India have been studied in detail for trace metal pollution in water and sediments. There are several reported studies from Indian rivers—the Ganges (e.g. Mathur et al., 1987; Modak et al., 1992; Prasad et al., 1989; Singh et al., 2004; Singh 2001; Subramanian et al., 1979, 1987), the Yamuna (Chakrapani and Subramanian 1996; Gadh et al., 1993), the Narmada (Gupta and Chakrapani 2005), the Krishna (Ramesh et al., 1990) the Godavari (Balakrishna et al., 2006), the Mahanadi (Chakrapani and Subramanian, 1993), the Hindon (Jain and Sharma, 2006), the Achankovil (Prasad et al., 2006), etc.

Such studies on the tributaries of Brahmaputra River, however, are limited till date. A couple of earlier studies reported sediments characteristics of the Jhanji River (Baruah et al., 1996) and the Bharali River, Himalayan tributary of the Brahmaputra River (Hoque et al., 2010). In a study, Girija et al. (2007) reported the water quality assessment of Bharalu River and pointed out that urban runoff played a vital role in alkalinity, hardness and BOD. Their results also showed spatial characteristics of the water quality of the river. However, a detail study of the Bharalu River sediment was lacking.

The present study is the first attempt to estimate the trace metal levels in bed sediments of the Bharalu River and thereby to investigate the extent of pollution on the basis of the enrichment and geoaccumulation of the trace metals in the sediment. The apportionment of the contributing sources was done by PCA.

Methodology

Study Area

The Bharalu tributary of the Brahmaputra River originates in the foothills of the Khasi Hills of the Meghalaya state in Northeast India. It enters through the southeastern corner and flows through the heart of the Guwahati city crossing densely populated residential, industrial and commercial areas to meet the Brahmaputra River at Bharalumukh (Figure 1).

The region receives 200-400 cm of annual rainfall, and the Bharalu is the single outlet of the watershed. It also receives domestic wastewater from a large densely urbanized area and meets a major drain carrying storm water runoff from a refinery before it conflues with the Brahmaputra River at Bharalumukh.

Nearly two decades ago, the Bharalu was a fishing ground and a water source for bathing and cleaning during the festivals. Relatively recent urbanization and industrial growth, at a faster rate, have caused conspicuous environmental pollution in and around Guwahati city, which has resulted in a situation of increased pollution threat to the Bharalu. As a result, the Brahmaputra River receives the entire pollution load. The tributary carries a large portion of the city's municipal and other wastes and also serves as the natural drainage for storm water runoff (Girija et al., 2007).

Based on the prevailing local characteristics, five representative sites were selected for the collection of sediment samples. The sampling sites are shown in Figure 1. The local activities around the sampling sites, which lead to input of pollutants to the river, are described in Table 1. Except site 1, all the four sampling sites are within city area of Guwahati.

Sampling and Analysis

The sampling was carried out in the first quarter of year 2007 before the onset of monsoon. Freshly deposited sediments (10-15 cm deep) from the banks and the middle of the river were collected by a plastic shovel and were mixed to make composite samples. From each site, three composite samples were collected. Samples were air dried, ground, homogenized, and sealed in clean polypropylene bags and then stored in a refrigerator (Subramanian et al., 1987).

One gram sediment sample was first digested in a mixture solution of concentrated HClO_4 (2 mL) and HF (10 mL) to near-dryness; subsequently a second

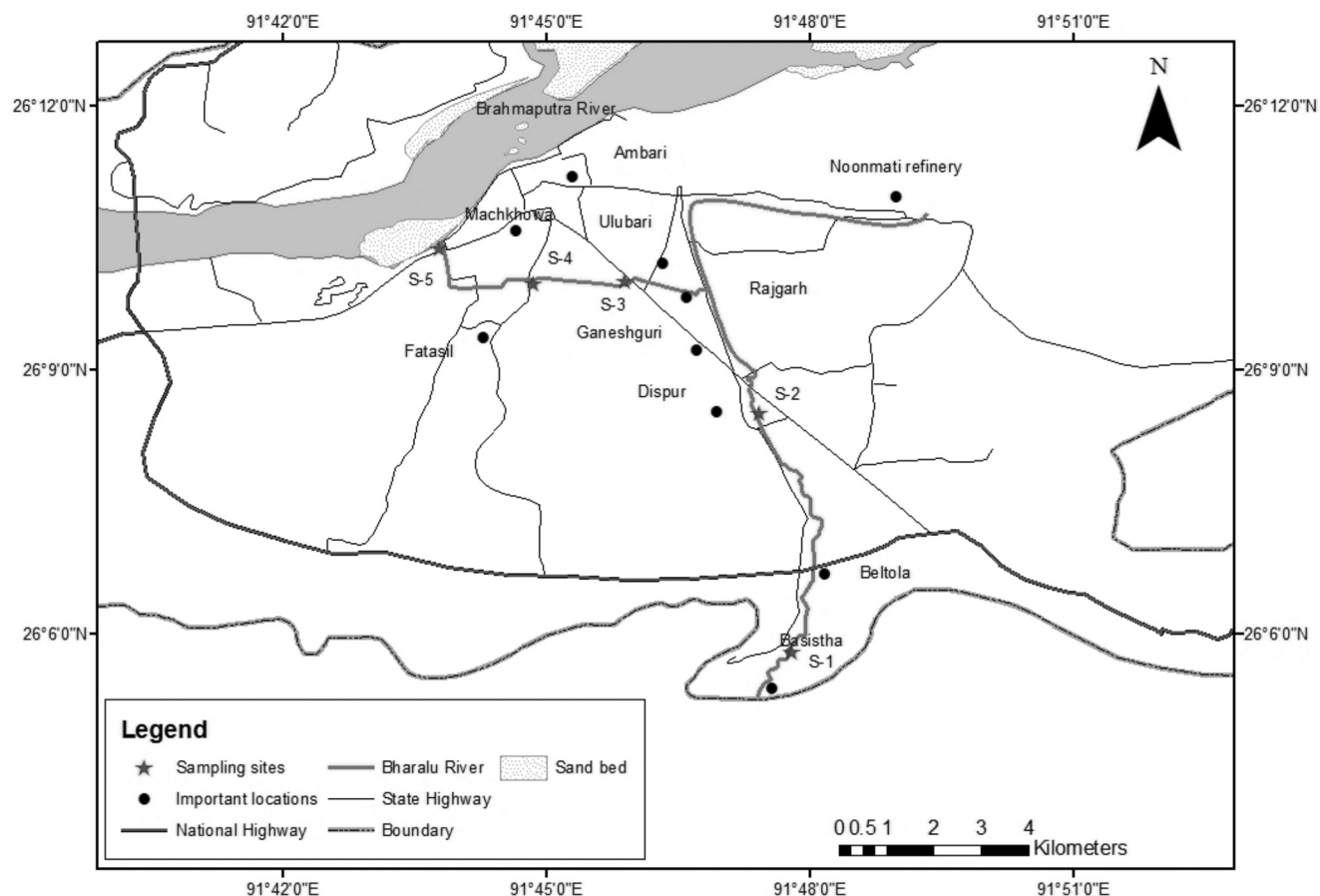


Figure 1: The study area—Bharalu River flowing through Guwahati city.

Table 1: Description of sampling sites

Sites	Coordinates	Prevailing activities
Site 1: Basistha	91°47.797' E 26°5.822' N	Close to the origin of the Bharalu in the foothill of Khasi Hills. No pollutants are expected.
Site 2: Dispur	91°47.424' E 26°8.519' N	The capital region of Assam state, a busy commercial and thickly populated area. The location has the busiest roads within Guwahati city and surrounded by residential complexes, industrial estates, office buildings, schools, automobile workshops and business houses. Dispur is about 22 km downstream of Basistha.
Site 3: Bhangagarh	91°45.904' E 26°10.011' N	A commercial area and has retail units and house the largest hospital of the Northeast India – Guwahati Medical College Hospital (GMCH) in the close vicinity.
Site 4: Sherabhatti	91°44.861' E	A site surrounded by varieties of activities. The locality has metallic fabrication units, motor vehicle repair workshops, schools, colleges, residential complexes, private hospitals, retail shops besides having urban sprawls, cow sheds, crematories and graveyard.
Site 5: Bharalumukh	91°43.794' E 26°10.39' N	The snout of Bharalu where it meets the Brahmaputra. At Bharalumukh, the river passes through congested residential colonies.

addition of HClO_4 (1 mL) and HF (10 mL) was made and again the mixture was evaporated to near dryness. Finally, HClO_4 (1 mL) alone was added and the sample

was evaporated until the appearance of white fumes. The residue was dissolved in 12 N HCl and diluted to 25 mL (Tessier et al., 1979).

The trace metals were estimated by Atomic Absorption Spectrophotometer (AAS)—VARIAN AA240—involving direct aspiration of the aqueous solution into an air-acetylene flame. Minimum detection limit of AAS for the trace metals were: Pb – 0.01, Zn – 0.05, Cu – 0.001, Cd – 0.001, Cr – 0.001 and Ni – 0.001 mg L⁻¹. In the different experiments, blanks were run and corrections applied if necessary. All the determinations were in triplicate and average values were reported.

Organic matter content of sediment samples was determined by potassium dichromate oxidation method (Jackson, 1973).

Enrichment Factor (EF%)

Enrichment factor is often regarded as a tool to understand changes in sediment chemistry, which is the ratio of the concentration of a given pollutant in sediments at a given location within the watershed to the corresponding concentration at location that represents background or natural concentrations. EF% has been calculated using Equation (1) (Zonta et al., 1994; Loska and Wiechula, 2003).

$$EF\% = \left[\frac{(C - C_{\min})}{(C_{\max} - C_{\min})} \times 100 \right] \quad (1)$$

where C is the mean metal concentration in the sediment ($\mu\text{g g}^{-1}$) and C_{\min} and C_{\max} are minimum and maximum concentration in $\mu\text{g g}^{-1}$ determined during the study period.

Geoaccumulation Index (*Igeo*)

Geoaccumulation index was developed by Muller (1979), which has been widely used in European trace metal studies (Forstner et al., 1990). The index has been used to evaluate the degree of pollution by trace metals in sediments by several workers in the past (e.g. Audry et al., 2004; Bermejo Santos et al., 2003; Munendra et al., 2002). *Igeo* can be calculated by using Equation (2):

$$I_{geo} = \frac{\log_2 Cn}{1.5Bn} \quad (2)$$

where Cn is the concentration of the examined metals in the sediment, Bn is the geochemical background value of a given metal in shale (Turekian and Wedepohl, 1961), and 1.5 is the background matrix correction factor. The factor 1.5 is used for possible variation in the background due to lithogenic effects. The following

classification is given for the index of geoaccumulation by Forstner et al. (1990) and inference can be drawn accordingly:

- <0 Practically unpolluted
- 0-1 Unpolluted to moderately polluted
- 1-2 Moderately polluted
- 2-3 Moderately to strongly polluted
- 3-4 Strongly polluted
- 4-5 Strong to very strongly polluted
- >5 Very strongly polluted

Source Apportionment

The correlation analysis was performed to find associations between the variables. Variables showing positive significant correlations are assumed to exhibit associations that can be explained in terms of a common source or by chemical similarity (Knudson et al., 1977). Then, a multivariate statistical model, PCA, was carried out on the correlation matrix to transform the original data set into a smaller set of linear combinations that account for most of the variance of the original data set. Each component with significant eigenvalue was considered to exhibit a characteristic source.

The first step in this analysis is to transform the elemental data into a dimensionless standardized form

$$Z_{ik} = \frac{(C_{ik} - \bar{C}_i)}{\sigma_i} \quad (3)$$

where $i = 1, 2, \dots, n$, the total number of elements in the analysis; $k = 1, 2, \dots, m$, the total number of observations; Z_{ik} (Z score) is the normalized value of the i th element for the k th observation; C_{ik} is the concentration of the i th element in the k th sample; \bar{C}_i is the mean concentration for the i th element over all observations; and σ_i is the standard deviation of the distribution of concentrations of the i th element.

The PCA assumes that the total concentration of each element is made up of the sum of elemental contributions from each of j pollution source components. Hence,

$$Z_{ik} = \sum_{j=1}^P W_{ij} P_{jk} \quad (4)$$

where P_{jk} is the j th component value for the k th observation and $j = 1 \dots P$, the number of polluting sources influencing the data and W_j is the coefficient matrix of the components. In the present study, we have used the computer software SPSS which is accurate and user friendly.

Results and Discussion

The mean and range values of trace metals in Bharalu sediments and their EF% are presented in Table 2. The mean metal concentration in the Bharalu sediments were compared with metals in shale, other rivers, lakes and estuary sediments, which is presented in Table 3.

Trace Metals Concentration

Metals, in the present study, showed many fold higher concentrations at the sites within Guwahati city when compared with site 1. The average Pb concentration increases about 80 times at Dispur (Site 2), just 22 km downstream of site 1. Similar increase of concentrations of metals – Ni (3 times), Cr (2 times), Cd (20 times), Cu (12 times) and Zn (16 times) was observed at Site

2 when compared with Site 1. Metal concentrations at the four sampling stations within Guwahati city also showed higher variability. The relative abundance of the metals was Site 1: Zn > Ni > Cr > Cu > Pb > Cd; Site 2: Zn > Cu > Pb > Ni > Cr > Cd; Site 3: Zn > Cu > Cr > Ni > Pb > Cd; Site 4: Zn > Cu > Ni > Pb > Cr > Cd and Site 5: Zn > Cu > Ni > Pb > Cr > Cd. Zinc was found to be the most and Cd was found to the least abundant among the metals that were studied.

The concentration trends (Figure 2) clearly exhibit that there is enough input of toxic pollutants into Bharalu, which could be from various sources including urban run off as suggested by Girija et al. (2007). Singh et al. (1997) in a study of Gomati River sediments concluded that urbanization process is associated with higher concentrations of trace metals such as Cd, Cu,

Table 2: Concentration ($\mu\text{g g}^{-1}$) ranges and EF% of heavy metals in bed sediments of Bharalu River

Sites		Pb	Ni	Cr	Cd	Cu	Zn	pH	Org. matter (%)
Site 1	Mean ^a	1.11	13.40	12.55	0.01	7.92	36.34	6.5	0.46
	S.D.	0.12	1.30	2.10	0.01	0.93	9.28		
Site 2	Mean ^a	82.35	37.35	25.32	2.55	94.71	600.10	5.0	0.59
	S.D.	13.73	1.97	1.95	1.21	6.25	56.80		
Site 3	Mean ^a	32.44	42.64	45.65	0.32	68.91	382.20	6.5	0.60
	S.D.	8.78	3.99	4.63	0.16	5.96	44.27		
Site 4	Mean ^a	56.61	59.23	54.23	1.55	69.95	422.30	6.4	0.61
	S.D.	19.75	16.26	20.33	1.39	27.19	39.84		
Site 5	Mean ^a	25.21	50.23	22.25	0.22	86.12	734.50	5.6	0.58
	S.D.	12.06	19.02	5.70	0.16	9.61	62.43		
Bharalu	Mean ^b	39.544	40.62	32.00	0.93	65.45	435.08		
	Max	95.55	77.65	77.0	3.95	101.88	801.35		
	Min	0.98	12.3	11.8	0.01	7.0	28.35		
	S.D.	30.67	18.60	18.02	1.23	33.47	247.57		
	EF%	40.78	43.34	31.30	23.40	61.60	52.62		

^an = 3 (Composite samples – mix of sediments at 1/4, 1/2, 3/4 widths across the river)

^bn = 9 (each sample is a composite mix)

Table 3: Comparison of heavy metals of Bharalu River sediments with studies elsewhere

	Pb	Ni	Cr	Cd	Cu	Zn
Average Shale ^a	20	68	90	0.3	45	95
Guadaria River sediments ^b	20	37	38	3	25	51
Pearl River Estuary ^c	37.83	41.42	85.23		47.85	95.66
Gomti River sediments ^d	51.04	21.57	12.63	2.53	10.71	42.83
Rybnik reservoir sediments ^e	118.55	71.08	129.84	25.81	451.7	1583.90
Bharali River ^f	8			4	39	39
Ganga ^g		20	52		21	46
Present study	39.54	40.62	32.00	0.93	65.45	435.08

^aTurekian and Wedepohl (1961); ^bGonzalez et al. (2000); ^cWang et al. (2008); ^dSing et al. (2005); ^eLoska and Wiechula (2003);

^fHoque et al. (2010) and ^gSubramanian et al. (1987).

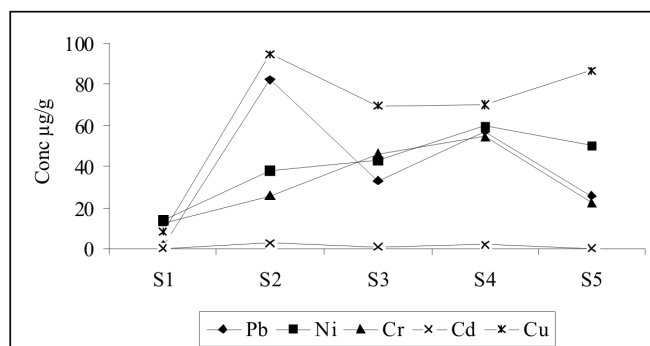


Figure 2: Concentration ($\mu\text{g g}^{-1}$) trends of the heavy metals from Basistha towards downstream of Bharalu River.

Cr, Pb and Zn. In the present study it was found that the levels of the metals were higher in the sampling location within the city area.

Concentrations of Cd, Pb and Zn in the Bharalu sediments were found to be 3, 1.5 and 5 times, respectively higher than that of shale; however, concentrations of Ni and Cr were on the lower side compared to levels in shale. Trace metals in the sediments of other rivers and lakes are generally reported to be higher than the corresponding background concentration for bottom sediments (Salomonas and Forstner, 1984). Metals concentrations showed many fold lower concentration levels compared with Rybnik reservoir sediments. Pb, Ni and Cu levels of the present study were, however, comparable with sediments of Pearl River estuary sediments.

Hoque et al. (2010) reported the levels of heavy metals in Bharali sediments, a Himalayan tributary of the Brahmaputra River, in Sonitpur district of Assam. Pb, Cu and Zn concentrations in the present study were found to be 5, 2 and 50 folds respectively higher than the former.

A comparison was also made with metal levels of Ganga River reported by Subramanian et al. (1987) and it was found that Zn level of the present study was about 10 folds higher than the level in Ganga River.

Enrichment Factors (EF%) and Geoaccumulation Index (I_{geo})

The enrichment factors calculated for the metals show that there is a substantial loading of the sediments with trace metals. This also elucidates that the bed sediments are possibly behaving as sink for the trace metals that reach the Bharalu system from urban activities. It was also evident from the concentration trends of the metals, shown in Figure 2, that there is no continuous rise of the concentrations from the movement of metals carried

by the water current. Should it happened, the site at the snout of the river would show the highest level of concentrations.

An EF% <1 should be dealt with caution because heavy metals may be released from the sediments contributing to their bioavailability, which was found in the study for any metal in the present study.

Table 4 presents the geoaccumulation indices (I_{geo}) for the quantification of trace metal accumulation in the Bharalu bed sediments. The geoaccumulation index consists of seven grades ranging from unpolluted to very highly polluted. Class 6 indicates a 64-fold enrichment above the background value. I_{geo} valued suggest that Bharalu is unpolluted to moderately polluted with respect to Ni and Cr at all the sampling locations. Bharalu is moderately polluted with respect to Pb, Cu and Zn at Sherabhatti. At Dispur, Bharalu is strongly polluted with respect to Cd. Jain et al. (2008), in a study of Narmada River sediments in central India, found that I_{geo} of Cd to be high while rest of other metals were under unpolluted to moderately polluted class.

Table 4: Calculated I_{geo} values at different sites

Sites	Pb	Ni	Cr	Cd	Cu	Zn
Site 1	0	0	0	0	0	0
Site 2	0.2	0.1	0	3	0.1	0.1
Site 3	0.2	0.1	0	0	0.1	0.1
Site 4	1.3	1	1	0	1.1	1.1
Site 5	0.2	0.1	0	0	0.1	0.1

Source Apportionment

The correlation matrix of the data was built to find associations between variables. Table 5 shows the matrix with significant correlation coefficients highlighted. All the trace metals have positive coefficients and several pairs are significantly correlated. Stronger associations

Table 5: Pearson's correlation matrix ($n = 15^{\#}$) of the heavy metals

	Pb	Ni	Cr	Cd	Cu	Zn
Pb	1.00					
Ni	0.45	1.00				
Cr	0.33	0.45	1.00			
Cd	0.90 ^a	0.38	0.12	1.00		
Cu	0.76 ^a	0.68 ^a	0.23	0.57 ^b	1.00	
Zn	0.51	0.61 ^b	0.16	0.29	0.87 ^a	1.00

[#]Composite samples

^aCorrelation is significant at the 0.01 level (2-tailed).

^bCorrelation is significant at the 0.05 level (2-tailed).

were observed for the pairs: Pb-Cd ($r = 0.90, p < 0.05$), Ni-Cu ($r = 0.68, p < 0.05$), Ni-Zn ($r = 0.61, p < 0.01$), Cu-Zn ($r = 0.87, p < 0.05$), Pb-Cu ($r = 0.75, p < 0.05$) and Cd-Cu ($r = 0.51, p < 0.01$). These associations can be explained in terms of common source or chemical similarity (Knudson et al., 1977). Thus, positive significant correlations could indicate a common source for the pairs.

Further, PCA was carried out to ascertain the possible contributing factors towards the metal concentrations and thereby determine which metals have a common origin. By extracting the eigenvalues and eigenvectors from the correlation matrix, the number of significant factors and the percent of variance explained by each of them were calculated.

Three eigenvalues were found to be >1 and cumulatively explained $\sim 85\%$ of variance. The first three eigenvalues were, therefore, selected for further analysis and other small but nonzero eigenvalues were discarded for the purpose of establishing a probable number of contributing source factors. The initial eigenvalues extracted were 'cleaned up' by means of Varimax rotation. The comparison of initial and rotated eigenvalues and their corresponding contribution towards explanation of %variance shows that both the eigenvalues and their corresponding contribution towards the explanation of cumulative %variance have not changed substantially.

The rotated component matrix (Table 6) showed that all the metals concentrations are explained by three

components. The Principal Component 1 amounts for $\sim 52\%$ of variance and includes Ni, Cr, Cu, Zn and organic matter with high loadings. This factor could be attributed to the discharges from industries and workshops. Thuy et al. (2007) considered Cd, Cr, Cu and Zn as "urban" metals and concluded that the concentrations of these metals appear to be associated with the uncontrolled and untreated industrial runoff to the discharge canals.

The Principal Component 2 explains $\sim 21\%$ of variance and showed high loading for Pb and Cd. Studies have shown that a large amount of Pb is supplied by the precipitation of aerosols in aquatic environments (Salomons et al., 1988). Atmospheric input from anthropogenic sources add Pb to the sediments (Li et al., 2000). Therefore, this component could be attributed to air-borne deposition of particulates to the water body and urban run-off carrying the deposited atmospheric dusts. As Pb in the atmospheric dust is mainly contributed by vehicular emission in large urban areas, we would like to attribute this component to atmospheric deposition of vehicular origin.

The Component 3 explains $\sim 13\%$ of variance and showed high loading for Ni and Cr. Both Ni and Cr are primary constituents of stainless steel. Wastes containing stainless steel alloy scrap could be a contributing source of Ni and Cr and these metals are ubiquitous in municipal solid waste (MSW). This component could be attributed to waste dumping both municipal and industrial.

Table 6: Principal component analysis - the rotated component matrix

	<i>Component</i>		
	1	2	3
Pb	0.26	0.92	0.25
Ni	0.83	0.27	0.14
Cr	0.68	0.28	-0.51
Cd	0.07	0.94	0.18
Cu	0.55	0.50	0.59
Zn	0.56	0.19	0.75
Org matter	0.75	-0.03	0.12
pH	0.07	-0.40	-0.88
Eigen values	4.14	1.64	1.03
% Variance	51.76	20.52	12.91
Cumulative %	51.76	72.27	85.19
Probable source	Industrial and workshops	Atmospheric deposition of Vehicular origin	Waste dumping

Extraction method: Principal Component Analysis. Rotation Method: Varimax with Kaiser normalization. Rotation converged in seven iterations.

Conclusions

Higher levels of trace metals were found in the bed sediments of Bharalu at the sampling stations within Guwahati city compared to the levels at the origin of the tributary. The enrichment factors show that there is a substantial loading of the sediments with trace metals. The bed sediments are possibly behaving as sink for the trace metals that come into the Bharalu system from urban activities. Igeo estimates suggest that trace metal pollution of Bharalu is moderately polluted to strongly polluted depending on the metal concerned and the location of sampling. Pearson's correlation matrix show stronger associations between Pb-Cd, Ni-Cu, Ni-Zn, Cu-Zn and Pb-Cu and the pairs could be from similar sources. PCA of the trace metal extracted three factors representing three possible sources of the trace metals in the Bharalu River sediments, viz., (i) discharge from industries and workshops, (ii) vehicular contribution and (iii) waste dumping of municipal and industrial origin.

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