

Heavy metals contamination in water and sediments of an urban river in a developing country

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ABSTRACT: Water and sediment samples were collected from 20 location of the Buriganga river of Bangladesh during Summer and Winter 2009 to determine the spatial distribution, seasonal and temporal variation of different heavy metal contents. Sequential extraction procedure was employed in sediment samples for the geochemical partitioning of the metals. Total trace metal content in water and sediment samples were analyzed and compared with different standard and reference values. Concentration of total chromium, lead, cadmium, zinc, copper, nickel, cobalt and arsenic in water samples were greatly exceeded the toxicity reference values in both season. Concentration of chromium, lead, copper and nickel in sediment samples were mostly higher than that of severe effect level values, at which the sediment is considered heavily polluted. On average 72 % chromium, 92 % lead, 88 % zinc, 73 % copper, 63 % nickel and 68 % of total cobalt were associated with the first three labile sequential extraction phases, which portion is readily bioavailable and might be associated with frequent negative biological effects. Enrichment factor values demonstrated that the lead, cadmium, zinc, chromium and copper in most of the sediment samples were enriched sever to very severely. The pollution load index value for the total area was as high as 21.1 in Summer and 24.6 in Winter season; while values above one indicates progressive deterioration of the sites and estuarine quality. The extent of heavy metals pollution in the Buriganga river system implies that the condition is much frightening and may severely affect the aquatic ecology of the river.

Keywords: *Enrichment factor; Geochemical distribution; Pollution assessment; River water and sediments; Seasonal variation*

INTRODUCTION

The Buriganga river is the main river flowing beside Dhaka, the capital of Bangladesh, which is a megacity of about 12 million people. City dwellers largely depend on the Buriganga's water for drinking, fishing and carrying merchandise. The river is now threatened by pollution and possession (Nouri *et al.*, 2009). The unpleasant odor of the polluted black water of Buriganga can be sensed even from half a kilometer distance. Intensive human intervention, unplanned urbanization and population pressure have created the present unwanted situation of the river. As a result of insensible human actions on the one hand, and failure by the authority to enforce rules and regulations to

save the river on the other hand, the Buriganga is dying biologically (Alam, 2008). Nowadays, no fish and other aquatic organisms can be found in the river during the dry season.

Heavy metals are among the most common environmental pollutants, and their occurrence in waters and biota indicate the presence of natural or anthropogenic sources. Their accumulation and distribution in soil, water and environment are increasing at an alarming rate causing deposition and sedimentation in water reservoirs and affecting aquatic organisms as well (Cataldo *et al.*, 2001; Hobbelen *et al.*, 2004; Koukal *et al.*, 2004; Okafor and Opuene, 2007; Mohiuddin *et al.*, 2010). Heavy metals like chromium, lead, cadmium, arsenic, etc. exhibit extreme toxicity even

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at trace levels. Rivers are a dominant pathway for metals transport (Miller *et al.*, 2003; Harikumar *et al.*, 2009) and heavy metals become significant pollutants of many riverine systems (Dassenakis *et al.*, 1998). The behavior of metals in natural waters is a function of the substrate sediment composition, the suspended sediment composition, and the water chemistry. During their transport, the heavy metals undergo numerous changes in their speciation due to dissolution, precipitation, sorption and complexation phenomena (Dassenakis *et al.*, 1998; Akcay *et al.*, 2003; Abdel-Ghani and Elchaghby, 2007) which affect their behavior and bioavailability (Nicolau *et al.*, 2006; Nouri *et al.*, 2011). Hence, heavy metals are sensitive indicators for monitoring changes in the water environment. However, to assess the environmental impact of contaminated sediments, information on total concentrations is not sufficient and particular interest is the fraction of the total heavy metal content that may take part in further biological processes (Jain, 2004; Nwuche and Ugoji, 2008). The overall behavior of heavy metals in an aquatic environment is strongly influenced by the associations of metals with various geochemical phases in sediments (Morillo *et al.*, 2004). Geochemical distribution results have also been used as an aid in

predicting potential contaminant mobility and bioavailability (Kabala and Singh, 2001; Pueyo *et al.*, 2003).

Previous studies on the Buriganga River have focused on the river water chemistry and physicochemical properties in the river water (Ali *et al.*, 2008; Moniruzzaman *et al.*, 2009) and few studies on seasonal and spatial distribution of heavy metals (Alam, 2003; Ahmad *et al.*, 2010). However, no detailed study on geochemical fractionation of river sediments concerning heavy metals have so far been conducted. This research work was conducted to determine the spatial distribution, seasonal variation, geochemical fractionation of heavy metals content in the sediments of the Buriganga river and to assess the pollution load in water and sediments and to distinguish the sources by conducting source apportionment using multivariate statistical analysis. This study was carried out in 2009-2010 in the Laboratory of Geochemistry, Keio University, Yokohama, Japan.

MATERIALS AND METHODS

Sampling

Water and sediments samples were collected from twenty sites of the Buriganga river in Summer and

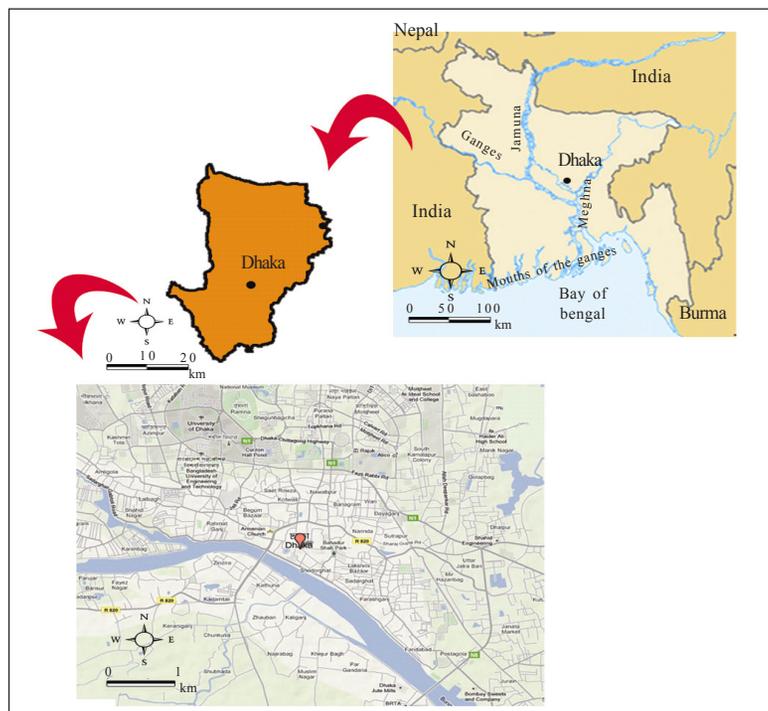


Fig. 1: Location of different sampling sites of Buriganga River, Dhaka, Bangladesh

Table 1: Name of the locations of different sampling sites of the river Buriganga, Dhaka, Bangladesh

Sample No.	Location
1	Kholamura lonch terminal
2	Signboard area
3	Borishur lonch terminal
4	Kamrangirchor tara mosjid
5	Losongonj, Zingira
6	Jahuchar, Hazaribagh
7	Kalunagar, Hazaribagh
8	Comanighat Hazaribagh
9	Hatirghat, Nowabgonj
10	Purba Rasulpur bridge
11	Sohidnagar jama mosjid
12	Sohidnagar bridge
13	Kamalbagh
14	Korimbagh
15	Raghunathpur
16	Swarighat
17	Badamtali bridge
18	Sadharghat
19	Merarbagh
20	Postogola bridge

Winter 2009 (Table 1 and Fig. 1). One sample was collected from every point at each time. The river bed sediment samples were taken at a depth of 0-15 cm. The sample mass collected in each case was about 500 g. Sub-samples of the material were oven dried at 45 °C for 48 h and ground using mortar and pestle. Then the samples were sieved with the help of a sieve (aperture 63 µm). The lower particle size fraction was homogenized by grinding in an agate mortar and stored in carefully marked glass bottles until chemical analyses were carried out. Water samples from the same points were also collected and immediately filtered with ADVANTEC® 0.2 µm size sterile syringe filter and transferred into acid-cleaned 50 mL polypropylene bottles. One ml of ultrapure nitric acid was added in each polypropylene bottle to achieve a pH of ~1 (Cenci and Martin, 2004).

Analysis of water and sediments

Organic carbon content in sediment was determined volumetrically by wet oxidation method as outlined by Walkley and Black (1934). Heavy metals like chromium (Cr), lead (Pb), cadmium (Cd), zinc (Zn), copper (Cu), nickel (Ni), cobalt (Co) and arsenic (As) in acidified water samples were analyzed using Inductively coupled plasma-atomic emission spectroscopy (ICP-AES). For the determination of total heavy metals, the extraction was carried out in Teflon containers provided with screw stoppers, using strong acid mixtures, as described by

Tessier *et al.* (1979) and heavy metals concentrations in the extract were determined by a Hewlett-Packard (HP 4500, USA) ICP-MS at National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Japan. The analytical precision was verified by using certified reference stream sediment samples (JSd-2 and JSd-3) provided by the Geological Survey of Japan. All strong acid mixtures were prepared just before the analysis of total heavy metal contents. All the reagents and chemicals used were of analytical reagent grade suitable for ultratrace analysis.

Sequential extraction experiment

A 5-step sequential extraction procedure described by Hall *et al.* (1996) was employed for the sediment samples collected at Winter season. The five steps are as follows: Step-1: AEC (Adsorbed, Exchangeable and Carbonate) Phase, Step-2: Amorphous Fe oxyhydroxide, Step-3: Crystalline Fe oxide, Step-4: Sulphides and organics, Step-5: Silicates and residual oxides. During extraction, extractant quality (especially the required pH) was maintained carefully. After each step, the solution was filtered by suction through a 0.45 µm millipore filter and the filtrate was collected in a polyester container. Then the solutions for each step were prepared accordingly for ICP-MS measurement following the manufacturer's recommendations. All the operations were carried out in 50 mL polypropylene centrifuge tubes (Nalgene, New York) and Teflon (PTFE) containers provided with screw stoppers. As a quality assurance measure, each sediment sample was subjected to triplicate analyses and the measurements are given as mean. The recovery values of the tested metals were calculated, although some recoveries deviated from acceptable values (between 90 and 110 %) at the 90 % confidence level.

Enrichment factors (EF_c)

EF_c is considered as an effective tool to evaluate the magnitude of contaminants in the environment (Franco-Uria *et al.*, 2009). The following equation was used to calculate the EFC:

$$EFC = (C_M/C_{Al})_{sample} / (C_M/C_{Al})_{Earth's\ crust}$$

Where, $(C_M/C_{Al})_{sample}$ is the ratio of concentration of heavy metal (C_M) to that of aluminum (C_{Al}) in the

sediment sample, and $(C_M/C_{Al})_{\text{Earth's crust}}$ is the same reference ratio in the Earth's crust. Samples having enrichment factor >1.5 was considered indicative of human influence and (arbitrarily) an EF of 1.5-3, 3-5, 5-10 and >10 is considered evidence of minor, moderate, severe, and very severe modification (Birch and Olmos, 2008). Average shale values taken from Turekian and Wedepohl (1961).

Pollution load index (PLI)

The PLI proposed by Tomlinson *et al.* (1980) provide some understanding to the public of the area about the quantity of a component in the environment. The PLI of a single site is the n th root of n number of multiplied together Contamination factor (CF) values. The CF is the ratio obtained by dividing the concentration of each metal in the sediment by the baseline or background value.

$$PLI = (CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n)^{1/n}$$

Site indices can be treated in exactly the same way to give a zone or area index. Therefore, PLI for a zone is the n th root of n number of multiplied together PLI values. A PLI value of zero indicates perfection, a value of one indicates the presence of only baseline levels of pollutants, and values above one would indicate progressive deterioration of the site and estuarine quality (Tomlinson *et al.*, 1980).

Statistical analysis

Principal component analysis (PCA) was performed in normalized data by varimax rotation using statistical software package STATISTICA®. Cluster analysis (CA) was applied to identify different geochemical groups, which enable clustering the samples with similar metal contents. CA was formulated according to the single linkage method, and the linkage distance $\{(D_{link}/D_{max}) \times 100\}$ was employed for measuring the distance between clusters of similar metal contents.

RESULTS AND DISCUSSION

Extent of contamination of water with heavy metals

Among all the heavy metals (Cr, Pb, Cd, Zn, Cu, Ni, Co and As) analyzed, Cu content in the water was the higher and Co concentration was the lower for both seasons. The average Cr concentration was 1.43 $\mu\text{g}/\text{mL}$ in Summer and 1.96 $\mu\text{g}/\text{mL}$ in Winter (Table 2). Ahmad *et al.* (2010) found higher concentration of Cr

in same river in pre-monsoon (0.62 $\mu\text{g}/\text{mL}$) than that in monsoon and post-monsoon water samples collected before 2009 (0.54 and 0.59 $\mu\text{g}/\text{mL}$, respectively), which was lower than that of the present study. The chromium concentration was much higher than the standard level for drinking water (0.05 $\mu\text{g}/\text{mL}$) proposed through Environment conservation rules (ECR, 1997). The average concentration of Pb was higher in Summer (0.50 $\mu\text{g}/\text{mL}$) than that in Winter season (0.23 $\mu\text{g}/\text{mL}$). The content of cadmium (0.16 $\mu\text{g}/\text{mL}$ in Summer and 0.22 $\mu\text{g}/\text{mL}$ in Winter) greatly exceeded the drinking water standard value (0.005 $\mu\text{g}/\text{mL}$). The lower level of Cd during Summer than in Winter may be due to dilution effect of rise in water level in Summer. However, seasonal industrial discharges may also have direct effect on these variations, as some metals in water are higher in Summer and some others in winter. No significant seasonal variation was observed in Zn content (0.26 $\mu\text{g}/\text{mL}$ in Summer and 0.22 $\mu\text{g}/\text{mL}$ in winter) and the values were lower than the drinking water standard (5 $\mu\text{g}/\text{mL}$) level (Table 2). The Cu content also exceeded the drinking water standard level. However, Ahmad *et al.* (2010) reported much lower content of Cu (0.16 $\mu\text{g}/\text{mL}$) in water of the Buriganga river collected before 2009. The average concentration of As and Ni also exceed the drinking water standard level in both the seasons. Considering the Toxicity reference values (TRV) proposed by US EPA (1999) almost all the heavy metals greatly exceeded the limit for safe fresh water and for Cr, Pb, Cd and Cu the values exceed ~ 100 times of TRV.

Content of heavy metals in sediment samples

The concentration of Cr in sediment ranged from 105-2017 $\mu\text{g}/\text{g}$ in Summer and 105-4249 $\mu\text{g}/\text{g}$ in Winter (Table 3). The sampling point 6 located in Hazaribagh area where the tannery industries are situated, contains maximum amount of Cr in both the seasons. The Cr concentration is very high in all the sampling points located in the branch of Buriganga, but the trend was decreasing towards the downstream sites. The untreated Cr containing huge leather tanning wastes mix with the river water might results in these extreme Cr contamination. The US EPA has classified Pb as a probable human carcinogen (Adriano, 2001). Lead is a good indicator of traffic related sources or battery recycling plants, and is considered as an indicator of pollution by urban runoff water (Mukai *et al.*, 1994). The analytical results depicts that Buriganga river

Table 2: Concentration (ig/mL) of heavy metals in sediments of different sampling sites of Buriganga river

Sites	Cr		Pb		Cd		Zn		Cu		Ni		Co		As	
	su	wi	su	wi	su	wi	su	wi	su	wi	su	wi	su	wi	su	wi
1	1.78	1.87	0.32	0.14	0.17	0.22	0.23	0.20	1.22	2.73	0.16	0.13	0.05	0.09	0.18	0.36
2	1.44	1.87	0.47	0.13	0.14	0.22	0.19	0.20	1.62	2.73	0.12	0.13	0.08	0.09	0.21	0.23
3	1.38	1.88	0.43	0.17	0.09	0.22	0.22	0.22	1.62	2.73	0.13	0.14	0.08	0.09	0.18	0.36
4	1.72	1.88	0.50	0.17	0.18	0.21	0.24	0.20	1.76	2.75	0.13	0.14	0.08	0.10	0.22	0.45
5	1.83	1.88	0.52	0.15	0.12	0.21	0.23	0.21	1.84	2.74	0.13	0.15	0.08	0.09	0.23	0.56
6	2.35	2.00	0.53	0.18	0.11	0.22	0.24	0.20	1.86	2.71	0.17	0.15	0.09	0.09	0.25	0.26
7	2.47	2.15	0.58	0.22	0.17	0.22	0.32	0.22	1.83	2.71	0.18	0.18	0.10	0.10	0.27	0.57
8	2.12	2.66	0.59	0.24	0.16	0.22	0.28	0.22	1.77	2.74	0.17	0.18	0.10	0.11	0.25	0.67
9	2.02	2.10	0.58	0.25	0.23	0.22	0.29	0.21	1.69	2.73	0.18	0.19	0.09	0.11	0.25	0.52
10	1.63	1.95	0.53	0.28	0.24	0.22	0.42	0.25	1.86	2.74	0.20	0.18	0.10	0.11	0.23	0.47
11	1.04	1.92	0.52	0.27	0.20	0.22	0.37	0.25	1.67	2.76	0.16	0.21	0.09	0.12	0.26	0.22
12	1.04	1.95	0.47	0.29	0.15	0.22	0.25	0.34	1.77	2.74	0.14	0.21	0.09	0.12	0.26	0.15
13	1.04	1.88	0.51	0.26	0.12	0.22	0.23	0.23	1.72	2.73	0.14	0.17	0.09	0.11	0.26	0.78
14	1.15	1.87	0.50	0.30	0.17	0.22	0.22	0.22	1.81	2.75	0.14	0.17	0.09	0.11	0.29	0.06
15	1.04	1.87	0.50	0.27	0.16	0.22	0.25	0.23	1.65	2.74	0.15	0.17	0.08	0.11	0.26	0.29
16	0.93	1.88	0.52	0.21	0.17	0.22	0.26	0.21	1.68	2.72	0.16	0.16	0.09	0.11	0.23	0.20
17	0.99	1.88	0.45	0.29	0.14	0.22	0.29	0.21	1.74	2.74	0.13	0.16	0.10	0.11	0.25	0.46
18	0.88	1.88	0.51	0.32	0.15	0.22	0.26	0.21	1.74	2.75	0.14	0.16	0.09	0.11	0.22	0.43
19	0.82	1.89	0.49	0.31	0.10	0.22	0.23	0.21	1.65	2.75	0.16	0.18	0.09	0.11	0.21	0.65
20	0.88	1.87	0.50	0.19	0.16	0.22	0.27	0.21	1.64	2.75	0.15	0.16	0.10	0.10	0.22	0.29
Mean	1.43	1.96	0.50	0.23	0.16	0.22	0.26	0.22	1.71	2.74	0.15	0.17	0.09	0.10	0.24	0.40
DWSB ^a	0.05		0.05		0.005		5.0		1.0		0.10		-		0.05	
TRV ^b	0.011		0.0025		0.0022		0.118		0.009		0.052		-		0.15	

Note: su- Summer and wi- Winter, respectively; ^aDrinking water standard for Bangladesh proposed through ECR (1997); ^bTRV for fresh water proposed by US EPA (1999)

sediment is severely polluted by Pb and highest Pb pollution is observed at sampling point 14 in Summer containing 1592 $\mu\text{g/g}$ and at sampling point 13 in Winter containing 1584 $\mu\text{g/g}$. Sediment contained more than 250 $\mu\text{g/g}$ Pb is considered to be exceed the Sever effect level (SEL) values; proposed by Ontario Ministry of Environment and Energy through aquatic sediment quality guidelines (Persuad *et al.*, 1993). At SEL the sediment is considered heavily polluted and likely to affect the health of sediment-dwelling organisms and a management plan may be required. More than 50 % sampling sites contained Pb higher than SEL values and the lowest Pb containing site also contained double than the Lowest effect level (LEL) values proposed by

Ontario Ministry of Environment and Energy (Persuad *et al.*, 1993). Spatial and seasonal variation of Pb content in sediments reflects their pollution may be from different point as well as non-point sources; such as leaded gasoline (Mukai *et al.*, 1994), chemical manufacturing and storage facilities and steel works in Old Dhaka. Total Cd content ranged from 3.5-7.8 $\mu\text{g/g}$ in Summer and 4.1-9.5 $\mu\text{g/g}$ in Winter. Sampling site 15 in Summer and sampling site 16 in Winter contained maximum Cd in sediments. Swarighat, the oldest and one of the main wholesale fish markets of the Dhaka city is situated in the high Cd containing area. The average Cd concentrations of all sampling sites were around 50 times higher than continental

Table 3: Concentration (ig/g) of heavy metals in sediments of different sampling sites of Buriganga river

Sites	Cr		Pb		Cd		Zn		Cu		Ni		Co		As	
	su	wi	su	wi	su	wi	su	wi	su	wi	su	wi	su	wi	su	wi
1	243	187	82	110	4.1	4.2	279	166	92	55	81	91	29	27	16	13
2	161	130	98	166	4.3	4.3	218	317	88	63	79	88	29	26	13	12
3	193	105	56	154	4.1	4.3	279	479	53	60	56	73	32	21	11	11
4	184	150	79	162	4.2	4.3	362	318	115	126	75	81	26	23	13	12
5	169	140	1186	129	4.0	4.3	326	255	187	145	64	89	23	36	15	10
6	2017	4249	567	147	4.2	4.4	476	466	345	109	160	92	33	27	10	9
7	1456	2910	171	115	4.1	4.8	357	580	242	119	68	94	27	34	11	12
8	1447	1224	354	208	3.5	4.7	439	327	137	174	67	91	22	22	9	15
9	1245	1014	539	544	4.6	8.4	368	1322	297	313	71	165	22	32	11	16
10	610	968	283	470	4.2	7.9	714	1257	355	346	158	154	19	32	9	16
11	466	689	408	240	6.5	5.6	1835	999	405	267	171	244	35	32	22	15
12	433	390	460	429	5.8	4.6	2163	1873	250	363	137	142	27	39	21	20
13	410	287	740	1584	5.1	7.3	1182	1950	346	364	156	206	29	36	21	25
14	197	481	1592	1468	6.3	5.5	1310	3002	398	257	98	142	32	47	16	34
15	195	237	541	475	7.8	7.9	1749	1639	743	302	165	217	37	45	21	18
16	151	294	501	858	4.1	9.5	663	917	158	318	186	198	37	49	15	19
17	154	149	368	938	4.4	8.3	445	1199	144	459	176	186	47	46	13	11
18	105	190	753	976	4.1	8.5	288	1432	191	354	153	202	48	58	9	17
19	190	151	554	202	4.2	4.6	677	386	193	135	84	91	31	36	9	14
20	188	243	165	182	4.0	4.1	129	279	30	162	58	101	24	39	10	16

Note: su- Summer and wi- Winter, respectively

upper crust (0.09 ig/g) values. Higher Cd concentration might be related to industrial activity, atmospheric emission and deposition of organic and fine grain sediments, leachates from defused Ni-Cd batteries and Cd plated items. Zinc concentration in Buriganga river sediment ranged from 129-2163 ig/g in Summer and 166-3002 ig/g in Winter season respectively and extremely Zn polluted areas were between sampling points 9 to 18. The LEL and SEL values for Zn are 120 and 820 ig/g respectively. Whereas, the average content of Zn in the sediments was 713 and 958 ig/g for Summer and Winter sediment samples respectively. Moreover one fourth sampling sites of Summer and half of the Winter seasonal samples analyzed exceeded the SEL values for Zn. [Sorme et al. \(2002\)](#) identified domestic construction and car related source and untreated waste water as the main sources of Zn. Buriganga

river sediment samples contained excessive Cu and sampling sites 10 to 18 contained maximum amount of Cu. TRV for Cu is 16 ig/g, whereas, average Cu content in the sediment was 238 and 225 ig/g for Summer and Winter respectively. High level of Cu indicates its higher input in these sites, which might be originated from urban and industrial wastes. Nickel constitutes about 47 µg/g of the Earth's upper crust ([Rudnick et al., 2003](#)) having the toxicity reference value of 16 ig/g ([US EPA, 1999](#)). However, Ni in sediments ranged from 56-186 ig/g in Summer and 73-244 ig/g in Winter. The Co content was 31 and 35 ig/g for Summer and Winter respectively, which is also higher than the geochemical background values. Sampling sites 10-18 contains higher amount of Ni and Co compared to other sites. Content of As is almost similar to shale values but higher than continental upper crust values. Almost

in all cases the average concentration of heavy metals in Winter samples was higher than those in Summer samples. The sampling sites 10 to 18 contained the maximum amount of toxic metals analyzed except Cr, which was dominating in sampling sites 6 to 10.

Geochemical fractionation of different heavy metals

AEC (adsorbed / exchangeable / carbonate) fraction

A large proportion of exchangeable and carbonate-bound Zn (54 % of total) was found in sediments (Table 4). Recovery of Co and Ni were also relatively high for AEC fraction (22 and 24 % of the total, respectively). Gleyzes *et al.* (2002) cautioned that heavy metals extracted from soils and sediments with 1 M sodium acetate adjusted to pH 5 may have also been specifically sorbed to low energy sites on the surfaces of clay minerals, organic matter, and oxide minerals, as well as coprecipitated with carbonate minerals. Therefore, it is acknowledged that heavy metals associated with this fraction may also be weakly sorbed to other noncarbonate phases. However, the recovery of Cr, Pb and Cu in AEC fraction was comparatively low (8, 13 and 9 % of the total, respectively).

Amorphous Fe oxyhydroxide fraction

Amorphous Fe oxyhydroxide phase is well recognized for its scavenging properties of heavy metals in the surface environment (Hall *et al.*, 1996). Oxyhydroxide minerals, along with organic matter, have long been recognized as the predominant

heavy metal sorbents in aquatic systems. In comparison with carbonate minerals, amorphous oxyhydroxide minerals have relatively large surface area and surface site density (Benjamin and Leckie, 1981; Forstner and Wittmann, 1983; Bilinski *et al.*, 1991). Relatively high affinity of Pb (62 % of total), Cu (48 % of total), Cr (45 % of total), Co (31% of total), Zn (30 % of total) and Ni (30 % of total) for amorphous Fe oxyhydroxide minerals was observed in Buriganga river sediments (Table 4). Metals extracted in this fraction might be associated with amorphous Fe oxyhydroxide minerals, such as goethite and may transfer to AEC fraction easily.

Crystalline Fe-oxide fraction

In contrast to amorphous Fe oxyhydroxide minerals, there was relatively low affinity for crystalline Fe oxyhydroxide minerals (Table 4). Only 4, 9, 15, 16, 18 % and 20 % of total Zn, Ni, Pb, Co, Cu and Cr; respectively were associated with this fraction. These trends probably reflect the much greater surface area of amorphous minerals in comparison with crystalline material (Kampf *et al.*, 2000). Heavy metals associated with oxide (both amorphous and crystalline) minerals are likely to be released in reducing condition. According to Patrick and Jugsujinda (1992), reductive dissolution of the oxide minerals occurs at Eh less than approximately +250 mV for Mn oxides and +100 mV for Fe oxides. Relatively small changes in Eh toward reducing conditions would cause reduction of Fe and Mn oxide species (Burton *et al.*, 2005). This will cause dissolution of Fe and Mn oxide minerals, thereby allowing release of associated heavy metals

Table 4: Average heavy metal content (µg/g) in different sequential extraction phases (values in parenthesis represents % contribution of total)

Metals	AEC	Am. Fe Oxide	Cry. Fe Oxide	Sulphides & organics	Silicates & residuals
Cr	49.4 (8)	276.4 (45)	121.8 (20)	101.4 (16)	69.1 (11)
Pb	62.9 (13)	310.9 (62)	89.0 (18)	25.0 (5)	14.7 (3)
Zn	486.1 (54)	269.2 (30)	35.5 (4)	15.0 (2)	89.6 (10)
Cu	18.6 (9)	94.5 (48)	31.1 (16)	25.1 (13)	28.8 (15)
Ni	28.7 (24)	35.8 (30)	11.3 (9)	13.7 (11)	30.4 (25)
Co	6.8 (22)	9.3 (31)	4.6 (15)	5.6 (18)	4.0 (13)

to bioavailable phase.

Sulphides and organics fraction

Heavy metals bound to this fraction are assumed to reflect strong association of organic materials present in sediments. On an average only 2 % Zn, 5 % Pb, 11 % Ni, 13 % Cu, 16 % Cr and 18 % of total Co were associated with the operationally defined sulphides and organics fraction (Table 4). However, the organic carbon in Buriganga river sediment samples was comparatively higher (mean 7.68 wt.%). These results inferred that the source of heavy metals in sediments were not mainly sulphides and/or organics bound (which has strong binding affinity for heavy metals), but they might be adsorbed in organic matter. It is also evident from the present study that the source of organic matter in sediments of the river Buriganga does not originate from its parent material but may be due to different anthropogenic activities.

Silicates and residual fraction

Heavy metals in the silicates and residual fraction are notably fixed within the crystalline lattice, and are usually considered to be fragments of the primary mineral phase. All other fractions can be of secondary mineral phases as they involve materials formed through physical and chemical processes of weathering of primary minerals. However, this fraction

is not available to biological or diagenetic processes except over very long time scales (Tessier *et al.*, 1979). Silicates and residual fraction retained 3 % Pb, 10 % Zn, 11 % Cr, 13 % Co, 15 % Cu and in 25 % of total Ni sediments. Amount of metals detected in this fraction was lower than respective continental upper crust values.

Comparative study with standards and some Bangladeshi river sediment values

The average concentration of Cr, Pb, Cd, Zn, Cu and Ni in sediments of the river Buriganga greatly exceeded the geochemical background i.e. average worldwide shale standard and continental upper crust value, but the average concentration of Co and As are very close to the geochemical standard values (Table 5). The mean concentrations of total heavy metals in sediments of the river Buriganga were several times higher than those of the sediments of the river Turag, Padma and Jamuna in Bangladesh. All the analyzed heavy metals were higher than TRV or LEL values and Cr, Pb, Zn, Cu and Ni content even exceeded the SEL values.

PCA

PCA was employed to evaluate the extent of metal contamination and infer the hypothetical location

Table 5: Comparison of heavy metal concentration ($\mu\text{g/g}$) in sediment of the Buriganga river with different reference values and those in some rivers of Bangladesh

Heavy metals	Present study (average)		Reference values					Other Bangladeshi rivers		
	Summer	Winter	ASV ^a	CUC ^b	TRV ^c	LEL ^d	SEL ^d	Turag ^e	Padma ^f	Jamuna ^f
Cr	511	709	90	92	26	26	110	97	97	110
Pb	475	478	20	17	31	31	250	24	17	19
Cd	4.7	5.9	0.30	0.09	0.60	0.60	10	-	-	-
Zn	713	958	95	67	110	120	820	111	76	83
Cu	238	225	45	28	16	16	110	49	25	28
Ni	113	137	68	47	16	16	75	42	28	33
Co	31	35	19	17.3	-	-	-	-	-	-
As	14	16	13	4.8	6	6	33	-	-	-

Note: ^a ASV-Average shale value proposed by Turekian and Wedepohl (1961); ^b CRC- Continental upper crust values proposed by Rudnick and Gao (2003) ^c TRV- Toxicity reference value proposed by US EPA (1999); ^d LEL- Lowest effect level; SEL- Severe effect level Ontario Ministry of Environment and Energy through aquatic sediment quality guidelines (Persuad *et al.*, 1993); ^e Zakir *et al.* (2006) and ^f Datta and Subramanian (1998), respectively

of sources of heavy metals (Shin and Lam, 2001; Franco-Uria *et al.*, 2009; Kikuchi *et al.*, 2009; Zare Garizi *et al.*, 2011). Initially data were normalized using Fe to compensate for both granulometric and mineralogical variability of metal concentration in sediments (Daskalakis and O'Connor, 1995; Schiff and Weisberg, 1999; Seshan *et al.*, 2010); and then PCA with varimax rotation was applied to the data matrix. The PCA leads to a reduction of the initial dimension of the dataset to three components which explain 94.54 % and 89.16 % of the data for Summer and Winter samples, respectively (Table 6). Therefore, these three factors play a significant role in explaining metal contamination in the study area. The first factor (PC 1), which has the highest loadings for all parameters except Pb and Cr in Summer and Zn, Pb and Cr in Winter and accounts for 69.26 % of variance for Summer samples and 55.23 % of variance; and emerge as the most important component or factor. Factor 1 could be better explained as anthropogenic source, because most of the metals of this component are severely accumulated in the sediments. Probable sources might be the industrial discharges, municipal waste water, household garbage and urban runoff. A second

anthropogenic component (PC2) which include only Pb in Summer and Pb and Zn in Winter, explaining 13.76 % and 21.83 % in Summer and Winter respectively. Higher Pb content in the air of Dhaka city may influence this enrichment through storm drainage. Another factor which helps concluding the origin of Pb is the enrichment factor, which is discussed in next section. Organic carbon (OC) showed different pattern of correlation in different seasons. Seasonal variation in industrial discharges, river dredging as well as variable contribution by fish waste, aquatic plants etc may influence the variable tendency of association of metals in OC. The last component extracted (PC3) includes Cr in Summer, accounts for 11.51 % of variance and Cr and OC with 12.10 % of variance in winter. For both the seasons Cr evolved as single dominant metal exist as separate factor, which reflects the impact of untreated tannery waste from Hazaribagh leather processing area. CA is the complimentary of PCA. The CA was applied to the data set for identifying associations (common origin) between metals. Four main clusters can be distinguished in the dendrogram obtained from the CA performed on the analyzed parameters (Fig. 2). Chromium, Pb, Zn and Cu

Table 6: Rotated component matrix of Principal component (PC) / factors for the sediments of Buriganga river Summer and Winter sediments

Parameters	Summer			Winter		
	PC 1	PC 2	PC 3	PC 1	PC 2	PC 3
OC	-0.96	0.17	0.00	-0.30	0.46	-0.71
Cr	-0.21	0.52	-0.79	-0.09	0.07	0.95
Pb	-0.20	-0.71	-0.63	0.09	0.91	-0.08
Cd	-0.98	0.14	0.00	0.97	0.14	0.13
Zn	-0.82	-0.42	0.08	0.40	0.79	-0.06
Cu	-0.87	-0.38	0.05	0.82	0.51	0.06
Ni	-0.99	0.02	0.07	0.81	0.45	-0.07
Co	-0.95	0.26	0.05	0.98	-0.09	0.01
As	-0.98	0.15	0.06	0.95	0.18	0.03
Eigenvalue	6.23	1.24	1.04	4.97	1.96	1.09
% Total variance explained (TVE)	69.26	13.76	11.51	55.23	21.83	12.10
Cumulative % TVE	69.26	83.03	94.54	55.23	77.06	89.16

Extraction method: PCA; Factor loadings: Varimax normalized (Gray colored loadings are > 0.70)

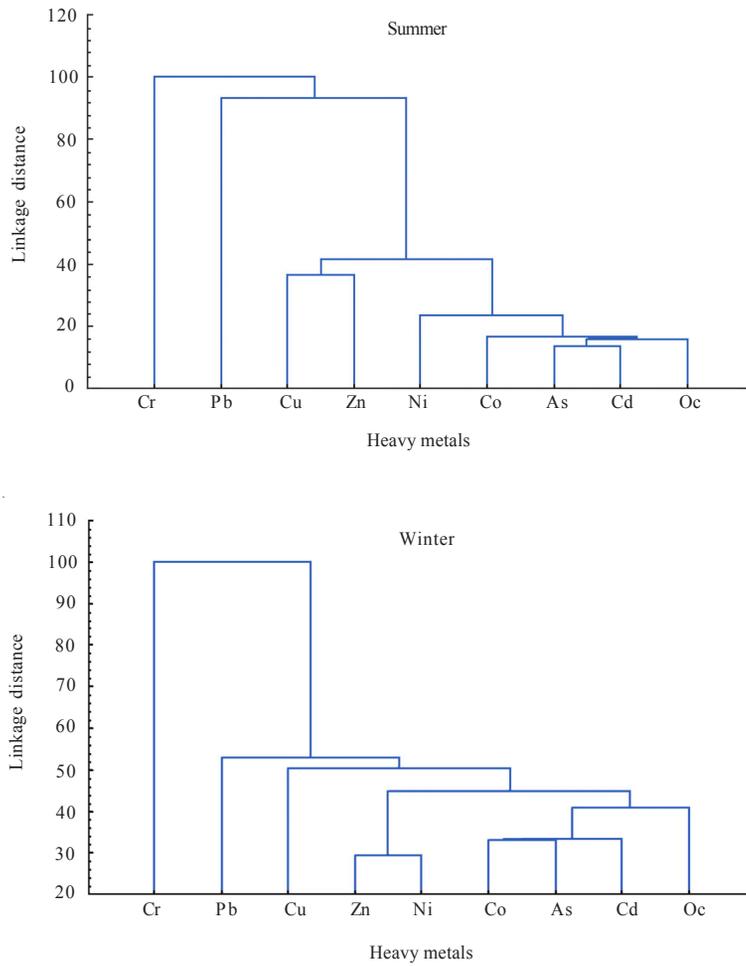


Fig. 2: Dendrogram of CA amongst the parameters of Buriganga river Summer and Winter sediment samples

showed very poor relation with organic carbon in both seasons, which indicates that these contaminants might be accumulated through anthropogenic influence. Chromium and Pb represent two independent clusters for both the seasons, which might be linked to their point source of pollution, for instance tannery industry of Hazaribagh area for Cr and automobile assembling industries of Zingira as well as leaded urban runoff for Pb.

Assessments of anthropogenic pollution in sediments

E_fc

E_fc were calculated to determine if the levels of

metals in sediments of Buriganga river were of anthropogenic origins. Lead and Cd were found to be enriched very severely (>10) in both Summer and Winter season. Chromium is severe to very severely enrich (>5~10) in the branch of Buriganga consisting sampling sites 6-10. Enrichment of Zn is also severe to very severe (>5~10) in the heavily water traffic areas of the Buriganga river consisting sampling sites 10-18. Copper also moderate to severely enriched in Buriganga river sediment. Heavy metals like Ni and Co having lower E_fc depict their lithogenic origin.

PLI

The PLI calculated from sediment data provides

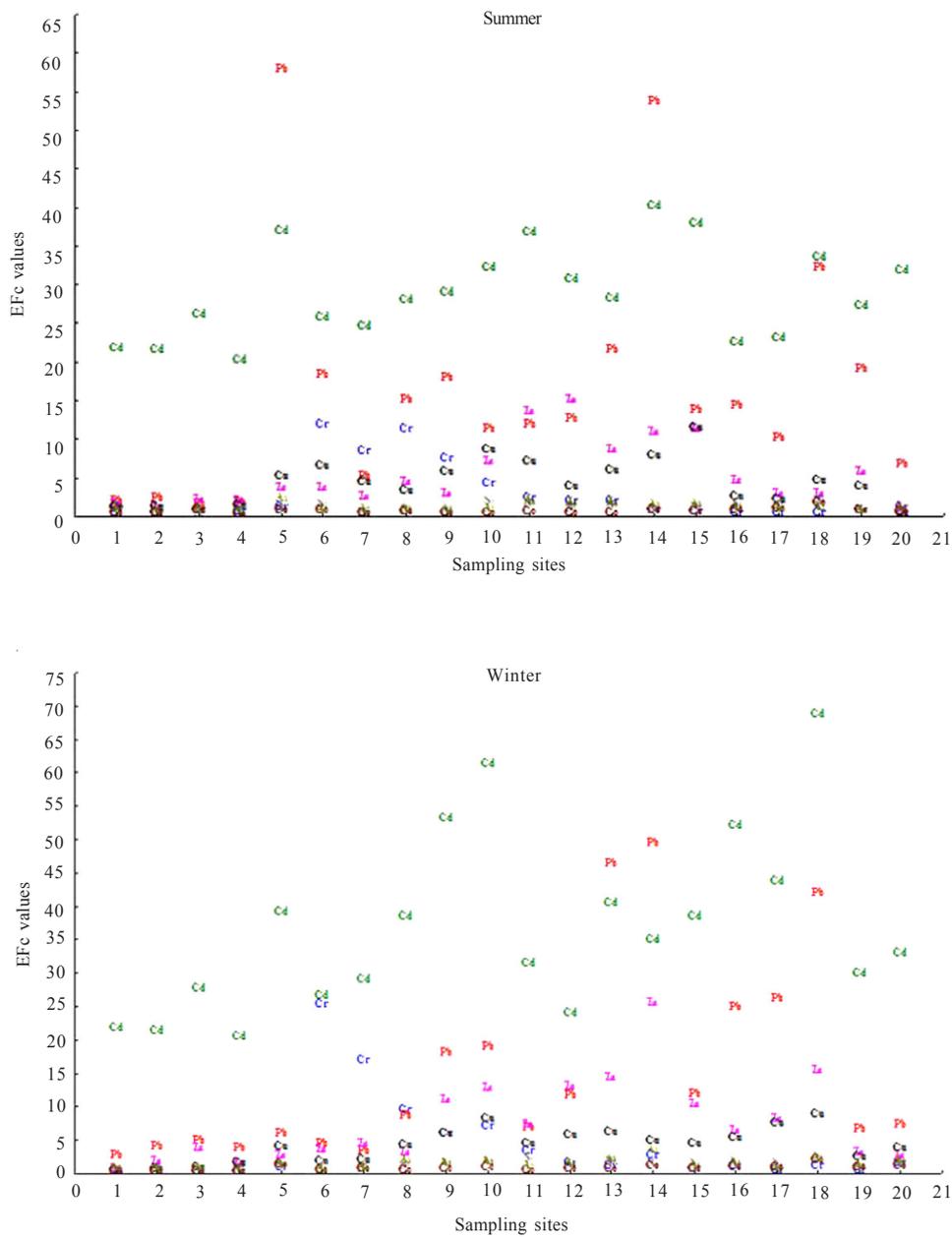


Fig. 3: Variation in EFC values of different sampling sites of Buriganga river Summer and Winter sediment samples

valuable information and advice for the policy and decision makers on the pollution level of the area. The highest PLI values were observed in sampling point 9 to 18 i.e. heavy water traffic area of the Buriganga river (Fig. 4). The main stream with heavy water pressure showed relatively lower PLI values. The PLI values for Summer and Winter ranged from

4.9-24.2 and 5.2-27.4, respectively. Most of the sites showed higher PLI values in Winter than in Summer. The area load for Winter is also higher than that in Summer having 21.1 and 24.6 in Summer and Winter respectively and Cd, Pb, Cr, Zn and As were the major 5 pollutants contributing towards the high PLI for this area.

Heavy metals contamination in river

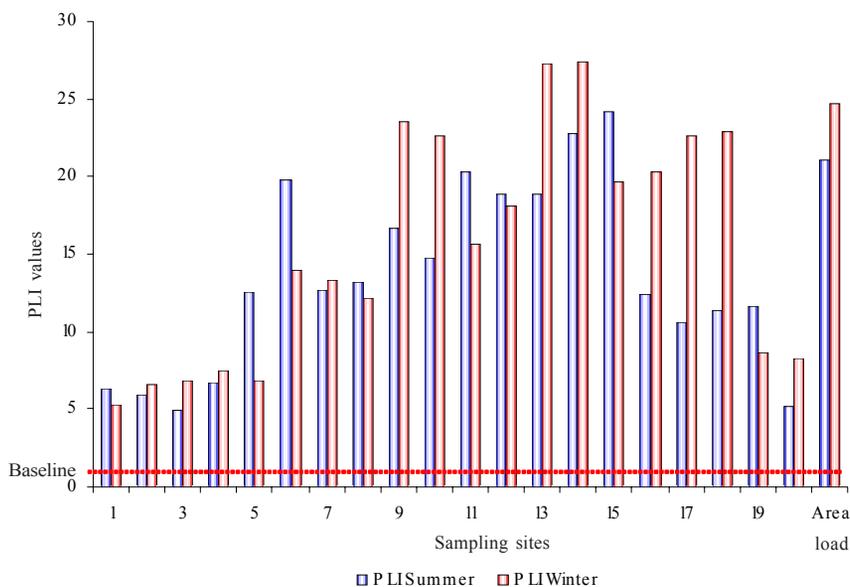


Fig. 4: Variation in PLI values of different sampling sites of Buriganga river Summer and Winter sediment samples

CONCLUSION

Heavy metals like Cd, Pb, Cr and Zn were extremely enriched and to a lesser extent Cu, Ni, Co and As were anthropogenically enriched in the Buriganga river sediments. The overall pollution load was significantly higher in Winter than in Summer season. The Buriganga river near old Dhaka consisting heavy water traffic was severely polluted. Cr, Cd and Pb evolved as main pollutant for both the seasons, might be linked to their point source of pollution, for instance tannery industry of Hazaribagh area for Cr and automobile assembling industries of Zingira as well as leaded urban runoff for Pb. Untreated industrial discharges and domestic waste water of urban households may influence the overall pollution load to Buriganga River. On average 72 % Cr, 92 % Pb, 88 % Zn, 73 % Cu, 63 % Ni and 68 % of total Co were associated with the first three sequential extraction phases, which portion may be readily bioavailable and may associate with frequent negative biological effects. EFC values demonstrated that the Pb and Cd were very severely enriched for both season, which implies that these metals originated from distinct point sources of pollution. Chromium and Zn also severe to very severely enriched in different sites of Buriganga river. The area load index and average PLI values of the river were 21.1, 13.5 in Summer and 24.6, 15.5 in Winter seasons, respectively, which reflects that

the river sediments are in extremely polluted condition. If this situation deteriorate further may lead to a biologically dead river having severe impact on aquatic life and city dwellers.

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