Assessment of Trace Metal Contamination in Water and Sediment of Some Rivers in Bangladesh

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ABSTRACT
Trace metal contamination is a major problem globally, especially in developing countries. In this study, the levels of trace metals (Cr, Ni, Cu, As, Cd, and Pb) in water and sediment of some peripheral rivers of Dhaka City, Bangladesh, were investigated. Huge amount of municipal wastes, industrial effluents and agricultural runoff from the periphery of Dhaka City notably are dumped to these rivers. Most of the effluents channeled into these rivers are not treated. Sediment and water samples were collected from selected stations along the various rivers in winter and summer seasons and analyzed for the trace metals of concern. Considering the sampling sites, the decreasing order of total metal concentration in water samples were Cr > Cu > As > Ni > Pb > Cd and in sediment were Cr > Pb > Ni > Cu > As > Cd. Total concentrations of Cr, As and Pb in the water samples were higher than WHO guidelines for drinking water quality for some sites. Geoaccumulation index (I_{geo}) revealed high values of Cd for all the stations. The extent of metal pollution in the rivers around Dhaka City implied that the condition is much frightening and probably severely affecting the aquatic ecology of the rivers.

Keywords: Bangladesh, metal pollution, river, sediment

INTRODUCTION

Trace metal contamination of water bodies ranks among the major environmental problems globally, with many issues reported from developing countries in recent times (Wu *et al*., 2011). The sources of trace metals in these countries are varied, but notably include landfill, mining, tanning, textile and various cottage industries (Bhuiyan *et al*., 2011). The present study observed the situation of rivers around Dhaka City, the capital of Bangladesh. The city is surrounded by some important rivers notably Turag, Buriganga and Shitalakha. These rivers contribute important socio-economic functions; for instance, they provide water for drinking, washing, fishing, transportation and carrying merchandise across the city. Trace metals can enter the river system through many possible pathways, including disposal of liquid industrial effluents, traffic emissions, brick kilns, terrestrial runoff and leachates carrying chemicals originating from numerous urban, industrial and agricultural activities, as well as atmospheric deposition (Ahmad *et al*., 2010). Sediments act as adsorptive sink for metals since they can scavenge some elements, thus, much higher metal concentration is found in sediment than in water column. Sediment, therefore, is an appropriate matrix to monitor the contamination of trace metals in the aquatic environment (Kalantzi *et al*., 2013). Sediment is regarded as a possible source of contaminants into the water column due to remobilization, desorption, degradation of sorptive substances and redox reactions.

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Few studies have been conducted so far especially pointing out on the deterioration of some physicochemical properties of water and sediment of the peripheral rivers of Dhaka City, Bangladesh (Ahmad *et al*., 2010; Mohiuddin *et al*., 2011). The present study reports on trace metals in water and sediment in three major rivers of Dhaka City. We conducted an extensive monitoring of trace metals: Cr, Ni, Cu, As, Cd and Pb in water and sediment in different seasons. The study provided an understanding on the extent of contamination of these metals and their spatial and seasonal distribution in aquatic environment of the rivers.

MATERIALS AND METHODS

Study area and sampling locations

This study focused on three major rivers Turag, Buriganga and Shitalakha around Dhaka City, Bangladesh (Fig. 1). Sampling was done in winter and summer seasons. Heavy rainfall occurred in summer season and caused higher water flow in river. The metropolitan area of Dhaka is about 380 km^2 and is located at the center of Bangladesh. The greater Dhaka City is one of the most densely populated cities in the world, home to approximately twelve million people of which less than 25% are served by sewage treatment facility (Ahmad *et al*., 2010).

Sample collection and preparation

Eighteen pairs of water and sediment samples were collected in March 2012 (winter) and September 2012 (summer). For dissolved metals analyses, water samples were filtered immediately after collection using ADVANTEC® 0.45 µm size sterile syringe filter. In addition, unfiltered samples were collected for total metal analyses. The samples were then transferred into acid cleaned 100 mL polypropylene bottles. The river bed sediment samples (about 200 g) were taken at a depth of 0 to 5 cm using a portable Ekman grab sampler. The lower particle size fraction was homogenized by grinding in an agate mortar, sieved through 106 µm aperture and stored in labeled glass bottles until chemical analyses were carried out.

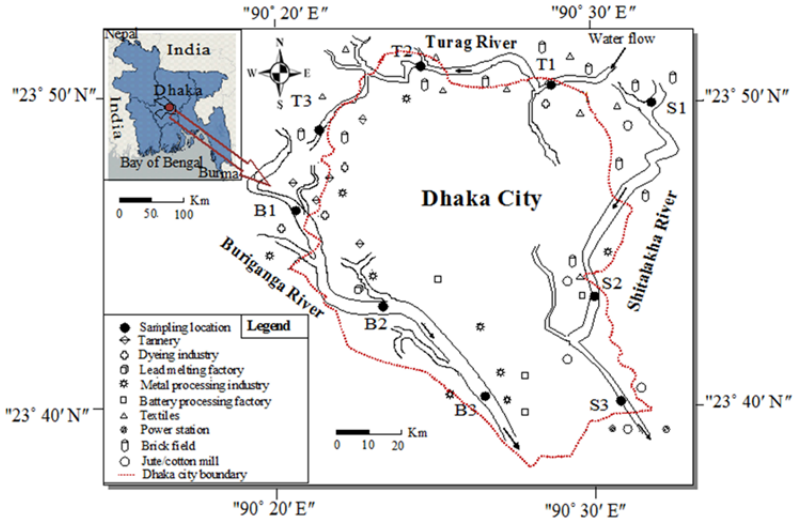

Fig. 1 - Map showing sampling locations in Bangladesh.
The locations of industries were plotted by the authors during the sampling campaign.

Analytical methods for physicochemical parameters

The physicochemical parameters such as pH, EC and temperature were measured immediately after sample collection. The pH of sediments was measured in 1:2.5 sediment to water ratio. The suspension was allowed to stand overnight prior to pH determination. The pH was measured using a pH meter with the calibration of pH 4 and pH 7 standards. For EC determination, 5 g of sediment was taken in 50 mL polypropylene tubes. Then, 30 mL of distilled water was added to the tube. The lid was closed properly and was shaken for 5 minutes. After that, EC was measured using an EC meter (D-52, Horiba, Japan). Percent N and C of sediment was measured using elemental analyzer (Vario EL III, Elenemtar, Germany) at Yokohama National University, Japan. For N and C determination, sediment samples were weighed in tin or silver vessels and loaded in the integrated carousel. In a fully automatic process, the transfer of the sample through the ball valve into the combustion tube was performed. Each sample was individually flushed with carrier gas to remove atmospheric nitrogen, resulting in a zero blank sampling process. The catalytic combustion was carried out at a permanent temperature of up to 1,200°C. The element concentration from the detector signal, and the sample weight on the basis of stored calibration curves were measured.

Metal extraction and analysis

Samples were digested in a Berghof-MWS2 model microwave digestion system (Berghof speedwave, Germany). Digestion reagents that were used included 5 mL 69% HNO_3 (Kanto Chemical Co, Japan) and 2 mL 30% H_2O_2 (Wako Chemical Co, Japan). The weighed samples (20 mL water and 0.2 g sediment) were then placed into a Teflon vessel (DAP-60 K, Berghof, Germany) with the digestion reagents. After digestion, samples were transferred into a Teflon beaker and total volume was made up to 25 mL for water samples and 50 mL for sediment samples with MilliQ water (Elix UV5 and MilliQ, Millipore, USA). The digested solution was then filtered using a syringe filter (DISMIC® - 25HP PTFE, pore size = 0.45 μm) from Toyo Roshi Kaisha, Ltd., Japan, and stored in 50 mL polypropylene tubes (Nalgene, USA). For trace metals, samples were analyzed using inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7700 series, USA).

Assessment of metal pollution in sediment

The degree of contamination from the trace metals could be assessed by determining the geoaccumulation index (I_{geo}). The index of geoaccumulation (I_{geo}) has been widely applied to the assessment of soil and sediment contamination (Santos Bermejo *et al.*, 2003). In order to characterize the level of pollution in the sediment, geoaccumulation index (I_{geo}) values were calculated using the equation

$$I_{geo} = \log_2[C_n/1.5B_n] \quad (1)$$

where C_n is the measured concentration of metal n in the sediment and B_n is the geochemical background value of element n in the upper continental crust (UCC) (Rudnick and Gao, 2003; Yu *et al*., 2011; Rahman and Ishiga, 2012). The factor 1.5 is introduced to minimize the possible variations in the background values which may be attributed to lithogenic effects. Geoaccumulation index (I_{geo}) values were interpreted as: $I_{geo} \leq 0$ – practically uncontaminated; $0 \leq I_{geo} \leq 1$ – uncontaminated to moderately contaminated; $1 \leq I_{geo} \leq 2$ – moderately contaminated; $2 \leq I_{geo} \leq 3$ – moderately to

heavily contaminated; $3 \leq I_{geo} \leq 4$ – heavily contaminated; $4 \leq I_{geo} \leq 5$ – heavily to extremely contaminated; and $5 < I_{geo}$ – extremely contaminated.

Statistical analysis

The data were statistically analyzed using the statistical package, SPSS 16.0 (SPSS, USA). A Pearson bivariate correlation was used to evaluate the interelement relationship in sediment. Other calculations were performed by Microsoft Excel 2010.

RESULTS AND DISCUSSION

Concentration of trace metals in water

The physicochemical parameters and concentrations of total and dissolved metals in water from three peripheral rivers of Dhaka City, Bangladesh are presented in Table 1 and Figs. 2 and 3.

Among the trace metals (Cr, Ni, Cu, As, Cd and Pb) analyzed, the total concentration of Cr in water was the highest and Cd was the lowest. The total metal concentration in water followed a decreasing order of site-B1 > site-B2 > site-B3 > site-T3 > site-T2 > site-S3 > site-S2 > site-S1 > site-T1. The highest total Cr concentration was obtained at B1 site (96 μg/L) during winter and (82 μg/L) during summer season, presumably as a result of the effects from tannery and dyeing industries of this site. Chromium in water supplies is generally found in the hexavalent form which is highly toxic and in higher concentration to be carcinogenic. The present study revealed that Cr at all sites in water samples for both seasons were much higher than WHO (5 μg/L) recommended values (Table 2). High concentrations of nickel (now considered to be a human carcinogen) as both soluble and sparingly soluble compounds may cause changes in muscle, brain, lungs, liver, kidney along with causing cancer (WHO, 2004). Copper in aqueous systems received attention mostly because of its toxic effects on biota. The highest value of total Cu was observed at T3 site (42 μg/L) during winter and B3 site (20 μg/L) during summer, which were close to the densely populated urban area, might be attributed to domestic sewage and runoff from extensively farmed areas (Koukal *et al*., 2004).

Arsenic forms a variety of inorganic and organic compounds of different toxicity reflecting the physicochemical properties of arsenic at different valence. Among the sampling sites, the highest total As concentration was observed at B1 site (24 μg/L) during winter and (16 μg/L) during summer season, which was higher than the WHO prescribed value for drinking water (Table 2). However, in river water, higher amount of As comes mostly from sediments (Mitamura *et al*., 2008) deposited by the river systems from the upland Himalayan catchments as well as the northern part of Bangladesh. Slightly higher concentrations of total and dissolved Cd were found at B3 site compared to other sites, due to the presence of metal processing, battery factories and paint industries (Caruso and Bishop, 2009). Operations involving the removal of cadmium paints by scraping or blasting might pose a significant increase in winter compared to summer. These kinds of pattern indicate the accumulation of Cd in water during low flow condition of river in winter season.

Table 1 - Physicochemical properties of water during two seasons.

Sites	pH		EC (mS/m)		Temperature (°C)	
	Winter	Summer	Winter	Summer	Winter	Summer
T1	8.2	7.8	62.6	13.6	23.1	31.2
T2	7.6	7.1	56.4	13.6	23.5	31.5
T3	7.9	7.6	53.1	24.3	23.8	30.7
B1	6.5	6.8	57.3	71.8	25.2	32.1
B2	5.7	6.5	54.1	13.3	25.9	31.9
B3	6.5	6.9	53.9	14.8	24.7	31.6
S1	7.4	7.3	65.1	11.7	24.8	29.7
S2	8.1	7.7	55.0	25.5	23.6	30.9
S3	7.8	7.5	59.0	17.6	24.3	31.3

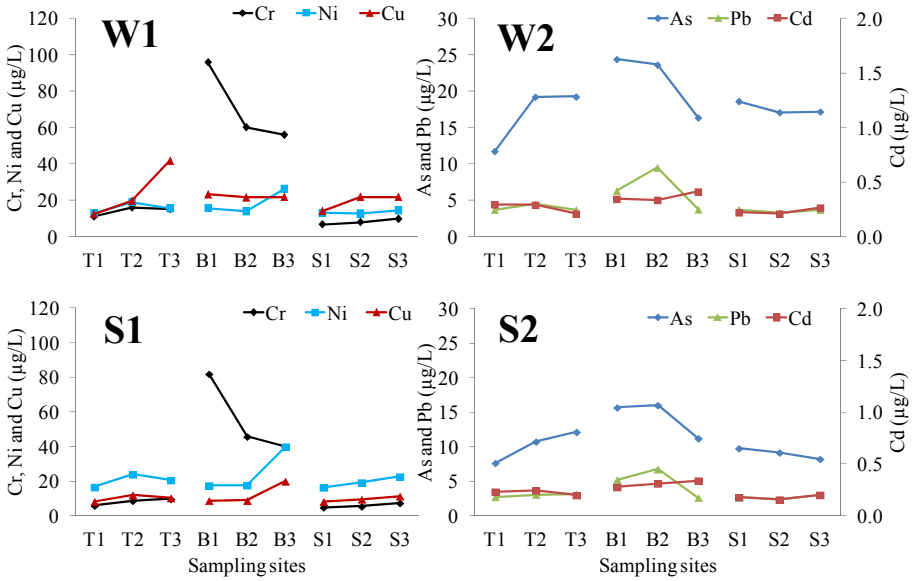

Fig. 2 - Total metal concentration in water during winter (W) and summer (S).

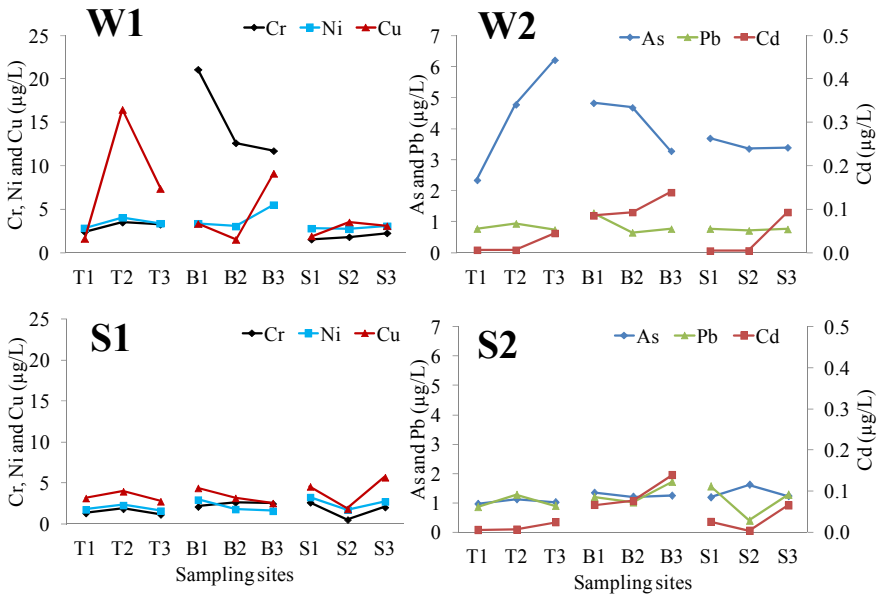

Fig. 3 - Dissolved metal concentration in water during winter (W) and summer (S).

Table 2 - Maximum permitted trace metal concentrations (μg/L) for drinking water quality.

Water quality guidelines	Cr	Ni	Cu	As	Cd	Pb
DWSB[a]	50	100	1000	50	5	50
TRV[b]	11	52	9	150	2.2	2.5
WHO (2004)	5	70	2000	10	3	10

[a]Drinking water standard for Bangladesh proposed through ECR (Department of Environment, Government of the People's Republic of Bangladesh (1997)).
[b]TRV (Toxicity Reference Value) for fresh water proposed by USEPA (1999).

In the study area, total concentration of Pb in water samples ranged from 2.9 to 8.1 μg/L and dissolved concentration of Pb ranged from 0.40 to 1.72 μg/L. Higher concentration of Pb found in the water of Buriganga River might be due to the acidic drainage from the industrial wastes and Pb mineralization. Lead can be carried in water, either dissolved or as waterborne particles. However, few compounds of Pb dissolve readily in water, though most of this Pb is then precipitated and becomes incorporated in the sediments at the base of the watercourse.

A notable increase in pH of Turag and Shitalakha river water, which causes a large amount of metals precipitation, resulted from the formation of their insoluble higher oxides. Similar result for metals in river water was observed by Vicente-Martorell *et al*. (2009) and Aktar *et al*. (2010). The lower levels of trace metals during summer might be due to the dilution effect of water. However, seasonal industrial operation along with domestic, municipal and agricultural wastes might have direct effect on the variations of metal concentrations in water.

Concentration of trace metals in sediment

The physicochemical parameters and seasonal distribution of trace metals viz. Cr, Ni, Cu, As, Cd, and Pb in sediment samples are presented in Table 3 and Fig. 4. The sites generally have pH ranging from 6.33 to 7.40 which was neutral or slightly acidic except at T1 site where pH was 8.68 indicating slightly alkaline condition (Table 3). The composition of the organic carbon in sediment samples were varied among the sites due to its origin in the aquatic environment. The organic carbon in sediments ranged from 0.18 to 3.31% (Table 3). The highest percentage of organic carbon might be attributed to the high amount of drainage water at B3 site. A wide range of values for metal concentrations were observed among the sampling sites. Factors such as salinity, geomorphological setup, and land runoff might have played a role in the variation of metals. Among the trace metals, Cr was the highest in concentration and Cd posted the lowest value. The highest concentration of Cr, 2471 and 2039 mg/kg-dw, was obtained at B1 site during winter and summer season, respectively (Fig. 4). The main leather tanning zone with about 270 tanneries were situated on the banks of Buriganga River on an area of 4 km^2 of site B1. The chromium enrichment of sediment could have been caused by two reasons: i) natural: concentration of Cr-bearing minerals; and ii) anthropogenic: industrial activities such as tanneries and textile factories discharging chromates and dichromate used as oxidants.

Table 3 - Physicochemical properties of sediment during two seasons.

Sites	pH		EC (mS/m)		%N		%C		C/N ratio	
	Win.	Sum.	Win.	Sum.	Win.	Sum.	Win.	Sum.	Win.	Sum.
T1	6.92	8.68	27.5	40.4	0.09	0.16	0.18	1.83	2.03	11.78
T2	6.93	7.16	39.8	5.4	0.17	0.15	2.79	1.55	16.69	10.66
T3	6.92	6.69	29.8	6.5	0.11	0.13	0.93	1.20	8.42	9.17
B1	6.33	6.48	40.8	25.7	0.17	0.23	1.41	1.55	8.52	6.70
B2	6.33	6.92	27.1	3.3	0.09	0.14	0.74	1.02	8.03	7.29
B3	7.17	6.68	12.5	24.3	0.31	0.05	3.31	0.51	10.67	9.81
S1	6.43	6.94	9.7	6.2	0.09	0.07	0.84	0.59	9.06	8.25
S2	6.48	6.91	16.7	10.4	0.11	0.13	0.89	1.14	8.38	8.50
S3	7.40	7.24	39.2	14.1	0.09	0.18	0.66	1.64	7.59	9.01

Note: Win. = winter and Sum. = summer.

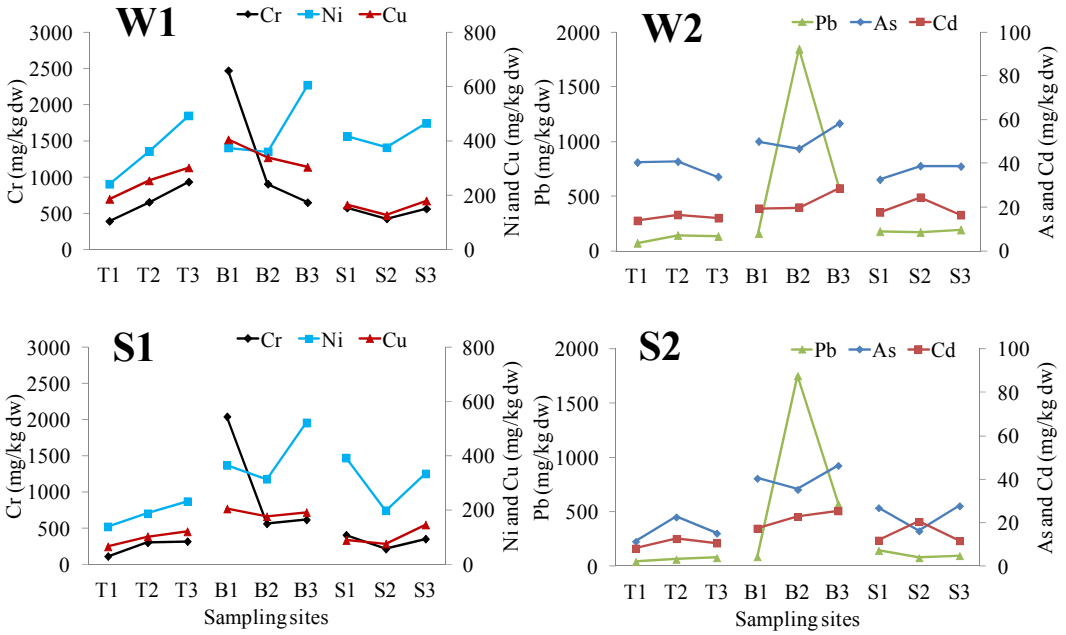

Fig. 4 - Concentration of trace metals in sediment during winter (W) and summer (S).

Hence, the discharging of waste from tanneries was most probably responsible for the high Cr concentration at B1 site. This study has provided the evidence that effluents discharged from the tanneries, dyeing and auxiliary industries and urban sewage system were the main sources of Cr in the river systems of Hazaribagh area of southwestern Dhaka City, Bangladesh (Ahmad *et al*., 2010). In this study, Ni concentration in sediment showed higher value at downstream site for each river (i.e. T3, B3 and S3). This might be the geochemical trap for Ni bonded in the fine-grained sediment; depositional acceleration of clay mineral and accumulation of Ni adsorbed on clay particles.

Buriganga River sediment samples contained excessive Cu and sampling site B1 recorded the highest Cu content, 2471 and 2039 mg/kg dw, during winter and summer, respectively. The seasonal variation of Cu could be brought about by the organic-Cu compounds formed. Higher level of Cu indicates its higher input in the sites (B1 to B3),

which might have originated from urban and industrial wastes. With respect to Cd, B3 site showed the highest value (29 and 26 mg/kg) in both seasons compared with other sites. Higher Cd concentration in sediment of Buriganga River might be related to industrial activity, atmospheric emission, leachates from defused Ni-Cd batteries and Cd plated items (Mohiuddin *et al*., 2011). Slightly higher Cd levels during winter might be attributed to the variation in water capacity of the river, where water input to the river is generally limited in winter, resulting in the precipitation of pollutants in the sediment. Arsenic and Cd might reflect the possible formation of complexes with organic matter and thereby slight natural variability of these two metals in surface sediment. There was a large variation observed for Pb in sediment samples in the studied rivers, which might be related to Pb sources and sediment characteristics. The highest concentration of Pb was obtained at B2 site (Fig. 4), attributed to a nearby Pb melting factory. Casting lead and lead product manufacturing are established at the old Dhaka and Keraniganj areas on the southern side of Dhaka City (adjacent to B2 site). In sediment, Pb concentrations in most of the sites were slightly higher in winter compared to summer which indicates that there could be some changes in organic profile due to resuspension/deposition of sediment. Additionally, Pb manifested a little dispersion during both seasons which may be attributed to anthropogenic intrusions.

In order to predict the metal pollution in sediment of the selected rivers, the available data for a comparative analysis with background reference values and other Bangladeshi river sediment values are summarized in Table 4. It is evident that the average total concentration of all the trace metals in sediment samples exceeded the geochemical background (shale standard and upper continental crust). It is apparent from Table 4 that the concentrations of all metals were very much varied from the average shale values, indicating that these elements did not originate from lithogenic sources. The anthropogenic activities might have contribution to the enrichment of these metals in sediment. When compared with toxicological reference levels, severe metal pollution was observed for the studied river sediment. The mean concentrations of all the analyzed trace metals were higher than those of the toxicity reference values, lowest effect levels and severe effect level (Table 4). The mean concentrations of total metals in sediment of the studied rivers were several times higher than those of the sediment of the other Bangladeshi rivers Padma and Jamuna. The results indicate that the levels of trace metals found in the sediment of the rivers might create an adverse effect on the aquatic ecosystem associated with the rivers, especially since it receives urban wastewaters.

Statistical analyses were performed in order to elucidate the associations among metals in sediment. Inter-metal interactions may illustrate the sources and pathways of the metals present in the particulate media. Pearson's correlation coefficients for the investigated metals and the physicochemical properties are depicted in Table 5. A clear pattern of strong association was found among the metal pairs in the sediment samples: Cr-Cu, Cr-As, Ni-Cu, Ni-As, Ni-Cd and As-Cd. Content of Pb was not correlated with any of the studied metals. Higher correlation coefficient between the metals indicated common sources, mutual dependence and identical behavior during their transport.

Table 4 - Comparison of metal concentration in sediment with some reference values and some reported values in Bangladeshi river sediment.

Metal	Present study [Mean, (Range)] (mg/kg dw)	Sediment quality guideline values (mg/kg dw)					Other Bangladeshi rivers (mg/kg dw)	
		ASV[a]	UCC[b]	TRV[c]	LEL[d]	SEL[d]	Padma[e]	Jamuna[e]
Cr	695 (112 – 2,471)	90	92	26	26	110	97	110
Ni	355 (139 – 606)	68	47	16	16	75	28	33
Cu	191 (65 – 405)	45	28	16	16	110	25	28
As	35 (12 – 58)	13	5	6	6	33	–	–
Cd	17 (8 – 29)	0.30	0.09	0.6	0.6	10	–	–
Pb	356 (45 – 1,846)	20	17	31	31	250	17	19

[a]ASV: Average shale value proposed by Turekian and Wedepohl (1961).
[b]UCC: Upper continental crust values proposed by Rudnick and Gao (2003).
[c]TRV: Toxicity reference value proposed by USEPA (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]Datta and Subramanian (1998).

Table 5 - Pearson correlation coefficient matrix for heavy metals and physicochemical characteristics in sediments (n = 18).

	Cr	Ni	Cu	As	Cd	Pb	pH	EC	%C
Cr	1								
Ni	0.299	1							
Cu	0.712**	0.560*	1						
As	0.542*	0.759**	0.794**	1					
Cd	0.219	0.610**	0.401	0.702**	1				
Pb	0.037	0.168	0.346	0.363	0.456	1			
pH	–0.496*	–0.335	–0.450	–0.480*	–0.448	–0.237	1		
EC	0.411	0.104	0.461	0.288	–0.122	–0.160	0.224	1	
%C	0.081	0.129	0.214	0.161	0.146	–0.110	0.286	0.089	1

* Correlation is significant at 0.05 level (2-tailed).
** Correlation is significant at 0.01 level (2-tailed).

In the present study, Cr, Ni, Cu, As and Cd metals reflected their similar origins or the processes that controlled their behavior in sediments. Metal and physicochemical associations show pairs Cr/pH and As/pH are correlated with each other, whereas the rest are not significantly correlated (Table 5). The correlation analysis result indicated that the flocculation or co-precipitation of Cr, Cu and As is influenced by pH (Balachandran *et al*., 2006). Aprile and Bouvy (2008) have recently shown that limnological processes are natural factors controlling such spatial variations of the heavy metals in sediments. The presence of pH in this correlation is most likely due to its association with Cr, Cu and As (Table 5). Significant correlation was observed between water and sediment samples of both the seasons for the metals except Pb (Table 6).

Table 6 - Correlation of metals between water (total metal) and sediment samples.

Metal	water-sediment (winter, n = 18)	water-sediment (summer, n = 18)
Cr	0.704**	0.576*
Ni	0.949**	0.880**
Cu	0.897**	0.884**
As	0.968**	0.862**
Cd	0.949**	0.892**
Pb	0.453 (0.712**)§	0.401 (0.550*)§

* Correlation is significant at 0.05 level (2-tailed).
** Correlation is significant at 0.01 level (2-tailed).
§ The numbers in parentheses show the correlation coefficients excluding the site B2.

Assessment of sediment contamination

The I_{geo} values showed the following decreasing order: Cd > Pb > Cr > Ni > Cu > As for the studied river (Fig. 5). The highest I_{geo} values obtained for Cd were 6.4 to 7.7 (> 5.0) and indicated extreme contamination of sediment in the study area. This might have happened due to higher concentration in sediment and lower geochemical background values resulting to higher I_{geo} values of Cd. The highest values of Cd might be due to contributions from atmospheric emission, leachates from defused batteries and Cd-plated items. This is of particular importance as the concentrations of Cd could be detrimental to the majority of benthic organisms. The I_{geo} values for Cr among the locations ranged from 0.9 to 4.0, indicating unpolluted to heavily polluted areas. The range of I_{geo} values for Ni, Cu and As were 1.4 to 3.0, 1.3 to 2.9 and 1.7 to 2.8, respectively, indicating moderate to heavy contamination. On the other hand, the I_{geo} values for Pb (1.2 to 6.1), indicated moderately to extremely contaminated sediment quality. Chromium and Pb showed outliers for I_{geo} value, because of the extreme concentration of Cr at sampling site B1 (tanning activity) and Pb at site B2 (lead melting activity).

The I_{geo} values for Cr and Pb indicated moderately to heavily contaminated sediment, whereas for Ni and Cu was moderately contaminated sediment.

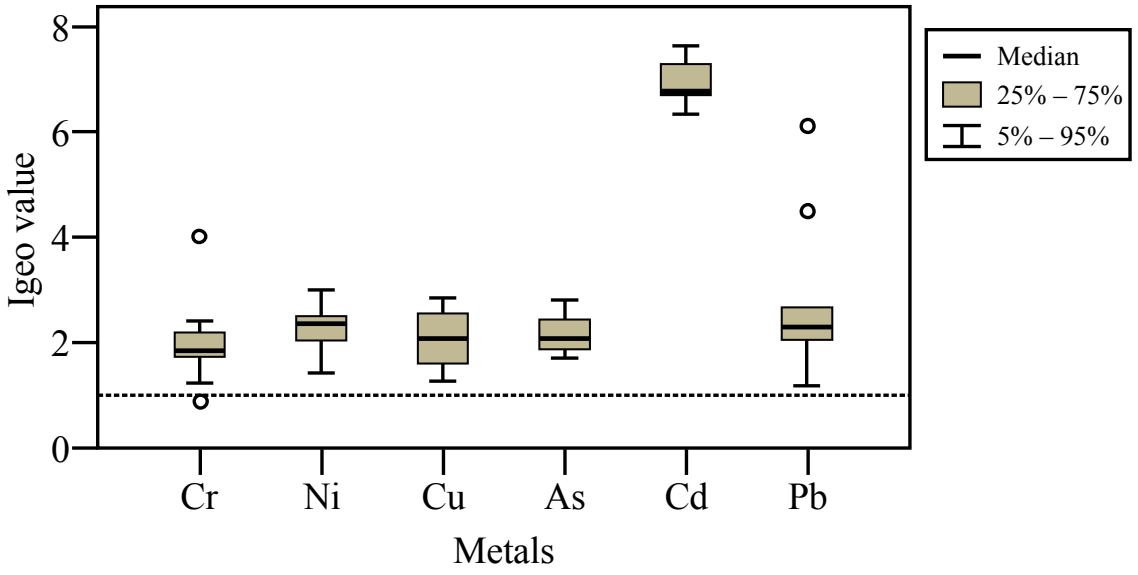

Fig. 5 - Geoaccumulation index (I_{geo}) values of trace metals in sediment.
Boxes represent a range of 25th to 75th percentile, solid lines in boxes are median values, error bars represent 5th and 95th percentile and circle symbols represent outliers.

CONCLUSIONS

This study has shown that the sediment of the rivers was heavily polluted with trace metals. Significantly higher levels of metals were observed during winter compared to summer. As some of the selected metals exceeded the safe levels, the water from contaminated sites should not be used without treatment. This study confirmed the seriousness of Cd and Pb levels especially in sediment. Finally, it is concluded that further detailed assessment of these two vital metals are highly recommended for the study area.

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