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Research paper

Assessment of toxic metals contamination with ecological risk of surface water and sediment of Korotoa river in Bangladesh

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Abstract

Toxic metal contamination is a major problem globally, especially in developing countries. In this study, the levels of toxic metals such as Cr, Ni, Cu, As, Cd and Pb in surface water and sediment of Korotoa River of Bogra City, Bangladesh were investigated. The average concentration of Cr, Ni, Cu, As, Cd and Pb in studied sediment were 1.01, 0.89, 1.98, 6.02, 0.0054 and 0.469 mg/kg, respectively. In the water sample, the mean concentration of Cr, Ni, Cu, As, Cd and Pb were 1.13, 1.33, 3.02, 2.62, 0.75 and 0.81 mg/kg, respectively. A huge amount of municipal wastes, industrial effluents and agricultural runoff from the periphery of Bogra City notably are dumped to this river. Most of the effluents channeled into these rivers are not treated. Considering the sampling sites, the decreasing order of total metal concentration in water samples were Cu > As > Ni > Cr > Pb > Cd and in sediment were As > Cu > Cr> Ni > Pb > Cd. Total average concentrations of Cr, Ni, Cu, As, Cd and Pb in the water samples were higher than WHO guidelines for drinking water quality. This contamination level implied that the condition is much frightening and probably severely affecting the aquatic ecology of the river.

Keywords: *Toxic metal; surface water; sediment; Korotoa River; ecological risk; Bangladesh.*

1. Introduction

In recent years, metal contamination in the aquatic environment has got global attention owing to its environmental toxicity, abundance, and persistence (Sin et al. 2001, Armitage et al. 2007, Yuan et al. 2011). Large quantities of hazardous chemicals especially heavy metals have been released into rivers worldwide due to global rapid population growth and intensive domestic activities as well as expanding industrial and agricultural production (Srebotnjak et al. 2012, Su et al. 2013, Islam et al. 2014). Both natural and anthropogenic activities are responsible for the abundance of heavy metals in the environment (Proshad et al. 2018, Wilson and Pyatt 2007, Khan et al. 2008). However, anthropogenic activities can effortlessly generate heavy metals in sediment and water that pollute the aquatic environment (Sanchez-Chardi et al. 2007). The increasing pollution by heavy metals has significant adverse health effects for invertebrates, fish, and humans (Yi et al. 2011, Martin et al. 2015). In the aquatic environment, sediments have been widely used as environmental indicators for the assessment of metal pollution in the natural water. The principal comportment of metals is a function of the suspended sediment composition and water chemistry in the natural water body (Mohiuddin et al. 2012). Sediment is an essential and dynamic part of the river basin with the variety of habitats and environments (Morillo et al. 2004). The investigation of heavy metals in water and sediments could be used to assess the anthropogenic and industrial impacts and risks posed by waste discharges on the riverine ecosystems (Zheng et al. 2008, Yi et al. 2011, Saleem et al. 2015). Nowadays, heavy metal pollution is the main problem in many developing countries like Bangladesh. The disposal of urban wastes, untreated effluents from various industries and agrochemicals in the open water bodies and rivers has reached the alarming situation in Bangladesh which are continually increasing the metals level and deteriorating water quality (Khadse et al. 2008). High concentration of heavy metals such as chromium (Cr), nickel (Ni), copper (Cu), arsenic (As), cadmium (Cd) and lead (Pb) are discharged into the Korotoa river which pollutes the water and sediments. Therefore, the objectives of this study are to evaluate the water quality parameters of the Korotoa River; to determine the levels of heavy metals in water and sediment, and to assess the heavy metal pollution status in sediments.

2. Materials and methods

2.1. Description of the study area

This study focused on an important river located at the northern part in Bangladesh. The study river was originated in the Himalayas, the mother of numerous rivers. Originating from northern frontier of Bhutan, the Korotoa enters Bangladesh territory through Darjeeling and Jalpaiguri districts of West Bengal in India, and forms for some distance the boundary between Dinajpur and Rangpur districts, Bangladesh. For the present study, the river was selected that flows through the Bogra district (Gokul Union of Bogra Sadar Upazila). The area of Bogra district is about 71.56 km^2 and the total population of this district is about 350,397. Thousands of villages, towns and commercial places like Shibganj, Mohasthangarh, Bogra, and Sherpur have been built on both sides of the Korotoa along its 200 km path. Mohasthangarh, the capital of ancient Pundranagar is still there beside the Korotoa as a witness of history in Bangladesh. Agriculture, aquaculture, and fishing are the primary activities of the people living beside this river. This river receives domestic raw sewage, household waste, and industrial waste from surrounding habitation. During the last decades, natural and human activities have caused a complete deterioration of the river ecosystems.

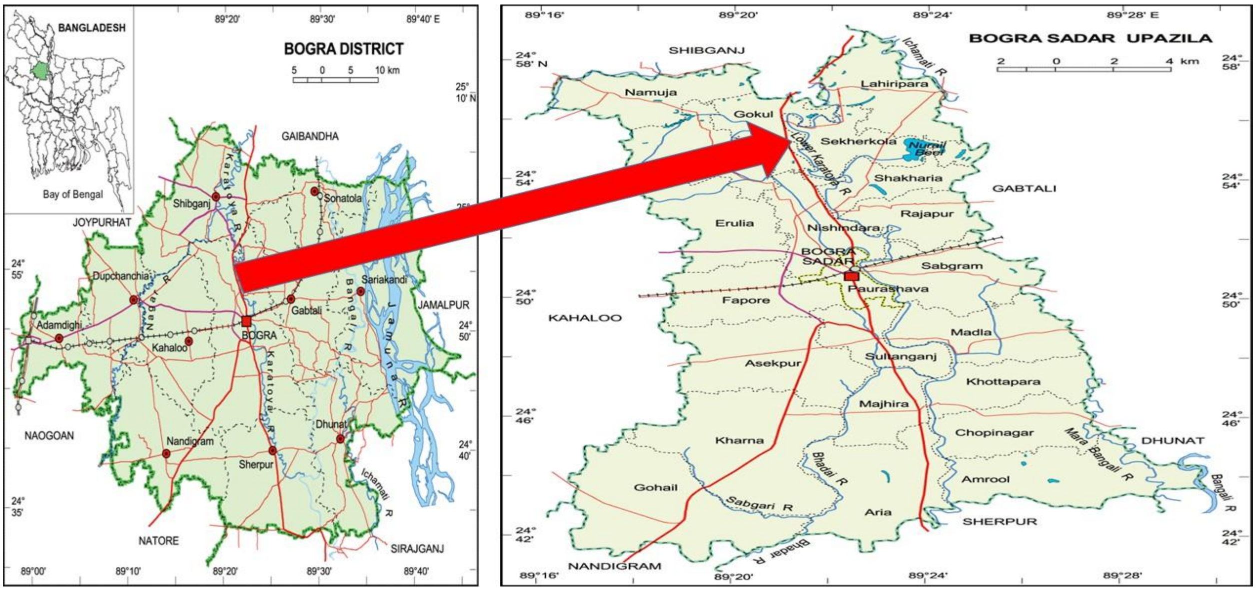

Fig. 1: Map Showing the Study Area of Surface Water and Sediment Sample Collected from Korotoa River in Bangladesh

2.2. Sample collection preparation and digestion

A total of 20 samples (water and sediment) were collected during April 2016 and samples were collected from ten different stations (S1–S10) (Fig. 1). Unfiltered water samples were collected from the center of the river for total metal analyses. The samples were then transferred into acid cleaned 100 mL polypropylene bottles. One mL of ultrapure nitric acid was added to each polypropylene bottle to achieve a pH of ∼1 (Cenci and Martin 2004). At each point, composite sediment samples were collected using a standard protocol (US EPA 2001). The river bed sediment samples were taken at a depth of 0 to 5 cm using a portable Ekman grab sampler. Three composite samples of mass approximately 200 g were collected at each station. The upper 2 cm of each sample was taken from the center of the catcher with an acid-washed plastic spatula to avoid any contamination from the metallic parts of the sampler. For considering the pre-industrial sample, sediment was taken by means of a percussion hammer corer (50–80 cm in length) for metal analysis (Schottler and Engstrom 2006). Composite sediment samples were collected into polyethylene airtight bags in the field and transported to the central laboratory of Patuakhali Science and Technology University, Bangladesh for pretreatment. The samples were dried in an oven at 45◦C for 48 h to gain constant weight. The dried samples were then ground using mortar and pestle and sieved through 106 µm aperture. The lower particle size fraction was homogenized by grinding in an agate mortar and stored in labeled glass bottles until chemical analyses were carried out. All chemicals were analytical grade reagents and Milli-Q (ElixUV5 and MilliQ, Millipore, USA) water was used for solution preparation. The Teflon vessel and polypropylene containers were cleaned, soaked in 5% HNO3for more than 24 h, then rinsed with Milli-Q water and dried. For metal analysis, 20 mL water sample and 0.5 g of sediment sample was treated with 5 mL 69% HNO_3 acid and 2 mL 30% H_2O_2 in a closed Teflon vessel and was digested in a Microwave Digestion System. The digested solution was then filtered by using syringe filter (DISMIC®– 25HP PTFE, pore size = 0.45 µm) Toyo Roshi Kaisha, Ltd., Japan and stored in 50 mL polypropylene tubes (Nalgene, New York). Afterward, the vessels were cleaned by Milli-Q water and dried with air.

2.3. Instrumental analysis and quality assurance

For heavy metals, samples were analyzed by using inductively coupled plasma mass spectrometry (ICP-MS). Multi-element Standard XSTC-13 (SpexCertiPrep®, USA) solutions were used to prepare the calibration curve. The calibration curves with R2> 0.999 were accepted for concentration calculation. Multi element solution (Agilent Technologies, USA) 1.0 µg/L was used as a tuning solution covering a wide range of masses of elements. All test batches were evaluated using an internal quality approach and validated if they satisfied the defined internal quality controls (IQCs). For each experiment, arun included blank, certified reference materials (CRM) and samples were analyzed in duplicate to eliminate any batch-specific error.

2.4. Analytical methods for chemical parameters

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, pH 7 and pH 9 standards. For EC determination, 5.0 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 min. After that, EC was measured using an EC meter (Horiba D-52). Percent N and C of sediment were measured using an elemental analyzer (model type: vario EL III, Elen-emtar, Germany). The textural classes for different soil samples were then determined by plotting the results on a triangular diagram designed by Marshall (1947) followed the USDA system. About fifty grams of oven dried soil was taken in a dispersion cup and 10 mL of 5% calgon solution was added to the samples and allowed to soak for 15 minutes. Then 90 mL distilled water was added to the cup. The suspension was then stirred with an electrical stirrer for 10 minutes. The content of the dispersion cup was then transferred to a liter sedimentation cylinder and distilled water was added to make the volume up to the mark. A cork was placed on the mouth of the cylinder and the cylinder was inverted several times until the whole sediment mass appeared in the suspension. The cylinder was set upright and the hydrometer readings were taken at 40 seconds and 2 hours of sedimentation. The corrections of hydrometer readings were made as the hydrometer was calibrated at 68°F.

2.5. Ecological risk assessment

Enrichment factor (EF) is considered as an effective tool to evaluate the magnitude of contaminants in the environment (Proshad et al. 2018, Franco-Uria et al. 2009). The EF for each element was calculated to evaluate anthropogenic influences on heavy metals in sediments using the following formula (Islam et al. 2018, Selvaraj et al. 2004):

$$EF = (C_M/C_{Al})_{Sample} / (C_M/C_{Al})_{Background} \quad (1)$$

Where, $(C_M/C_{Al})_{Sample}$ is the ratio of concentration of heavy metal (C_M) to that of aluminium (C_{Al}) in the soil sample, and $(C_M/C_{Al})_{Background}$ is the same reference ratio in the background sample. Generally, an EF value of about 1 suggests that a given metal may be entirely from crustal materials or natural weathering processes (Proshad et al. 2018, Zhang and Liu 2002). 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 the evidence of minor, moderate, severe, and very severe modification (Birch and Olmos 2008). In this study, (Al) was used as the reference element for geochemical normalization because of the following reasons: (1) Al is associated with fine solid surfaces; (2) its geochemistry is similar to that of many trace metals and (3) its natural concentration tends to be uniform.
The contamination factor (CF) is the ratio obtained by dividing the concentration of each metal in the soil by the baseline or background value (hazardous elements in the pre-industrial soil of the study area).

$$C^i_f = C_{heavy\ metal} / C_{background} \quad (2)$$

The contamination levels may be classified based on their intensities on a scale ranging from 1 to 6: low degree (C^i_f <1), moderate degree ($1 \leq C^i_f < 3$), considerable degree ($3 \leq C^i_f < 6$), and very high degree ($C^i_f \geq 6$) (Table 4). Thus, the values can monitor the enrichment of one given metal in sediments over a period. 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 contamination (Proshad et al. 2018, 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,

$$I_{geo} = \log_2 (C_n/1.5B_n) \quad (3)$$

Where, C_n is the measured concentration of metal n in the soil and B_n is the geochemical background value of element n in the background sample (Yu et al. 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.

2.6. Statistical analysis

The data were statistically analyzed using the statistical package, SPSS 20.0 (SPSS, USA). The means of the metal concentrations in water and sediments were calculated. Other calculations were performed by Microsoft Excel 2013.

3. Results and discussion

3.1. Physiochemical properties of sediment and surface water

The physiochemical parameters of the sediment and surface water such as pH, EC, and texture of sediment were assessed (Table 1). The physiochemical parameters are very important because they have a significant effect on the water and sediment quality. Furthermore, aquatic life also suffers due to the degradation of water quality. The average pH was 7.01 and 5.63 for sediment and water, respectively (Table 1). Salinity is a measure of the salt content of the water. The average electrical conductivity was 0.28 and 0.28 dS/m, respectively. Salinity level was negligible for the present study area. Average sand, silt, and clay were found 656.7, 191.5 and 151.8 mg/kg, respectively.

3.2. Toxic metal concentration in sediment

Heavy metal concentrations of sediments are presented in Table 2 and Figure 2. The mean concentration of Cr, Ni, Cu, As, Cd and Pb were 1.01, 0.89, 1.99, 6.02, 0.01 and 0.47 mg/kg, respectively. The average concentration of heavy metals in sediments were in the decreasing order of As > Cu > Cr > Ni > Pb > Cd. Chromium concentration in sediment was higher than other metals as a consequence of direct discharging of untreated wastes from petroleum, fertilizers and textile industries (Facetti et al. 1998).

Table 1: Physiochemical Properties of Sediment and Surface Water Collected from Korotoa River in Bangladesh

Sample Id	pH of sediment sample	EC of sediment sample (dS/m)	pH of water sample	EC of water sample (dS/m)	Texture of sediment			
					Sand (mg/kg)	Silt (mg/kg)	Clay (mg/kg)	Textural class
S1	7.62	0.12	4.78	0.26	620	183	197	Sandy loam
S2	6.64	0.18	5.46	0.25	515	252	233	Sandy clay loam
S3	8.05	0.89	5.81	0.29	513	310	177	Loam
S4	6.92	0.14	4.83	0.28	666	150	184	Sandy loam
S5	6.61	0.13	6.78	0.36	527	251	222	Sandy clay loam
S6	7.1	0.09	6.73	0.22	572	238	190	Sandy clay loam
S7	7.07	0.21	6.9	0.19	664	241	95	Sandy loam
S8	7.44	0.72	4.44	0.29	912	52	36	Sand
S9	6.46	0.17	6.36	0.23	692	176	132	Sandy loam
S10	6.26	0.21	4.25	0.42	886	62	52	Sand

However, high level of Cr for site S10 (3.385) indicates its higher input which might be originated from the urban and industrial wastes (Mohiuddin et al. 2012). The highest level of Ni and Cu was found in S7 (1.34 mg/kg) and S10 (4.30 mg/kg), respectively. The mean concentration of As in sediment was observed 6.02 mg/kg in which was less than the average shale value (ASV) (13 mg/kg) (Turekian and Wedepohl 1961). As concentration in sediments might be attributed to the anthropogenic activities such as treatment from the fertilizers and arsenical pesticides industries (Fu et al. 2014, Ahmed et al. 2016) treating of wood by exhausting copper arsenate (Pravin et al. 2012, Baeyens et al. 2007) and tanning in relation to some chemicals especially arsenic sulfide. Cd concentration was negligible in the present study area (Table 2). The comparison between the present study with previous studies conducted in Bangladesh and other countries (Table 3) and it was observed that most of the study were found a higher amount of toxic metals (Ahmad et al. 2010, Datta and Subramanian 1998, Rahman et al. 2014, Gupta et al. 2009, Liu et al. 2009, Akcay et al. 2003, Karbassi et al. 2008, Raphael et al. 2011). Pearson's correlation coefficient matrix for analyzed heavy metals from sediment parameters was calculated to see if some of the parameters interrelated with each other and the results are presented in Table 5. The elements in sediments showed correlation with each other where Cr showed significant positive correlation with Cu (0.72*) and Cd (0.93**) as well as significant negative correlation with As (-0.66*) and Cu showed significant positive correlation with Cd (0.74*). High correlations between specific heavy metals in water may reflect similar levels of contamination and/or release from the same sources of pollution, mutual dependence and identical behavior during their transport in the river system (Li et al. 2009, Chen et al. 2012).

3.3. Toxic metal concentration in water

The results of heavy metal concentrations in surface waters are shown in Table 4. The average concentration of Cr, Ni, Cu, As, Cd and Pb in water was 1.14, 1.33, 3.02, 2.62, 0.75 and 0.81 mg/kg, respectively. The studied metal followed the decreasing order of Cu > As > Ni > Cr > Pb > Cd. The concentration of Cr and Ni in water for each sampling site was much higher than the WHO standard level for drinking water (Table 4). For Cu, 50% of samples exceed WHO standard. The average concentration of As in water samples was 252 times higher than the WHO reference value which causes serious contamination of As in the study area water. Considering the toxicity reference values (TRV) proposed by USEPA (1999) almost all the heavy metals especially Cr, Ni, As and Pb greatly exceeded the limit for safe water, indicated that water from this river is not safe for drinking and/or cooking. Considering Correlation for heavy metals among water samples (Table 5), Cr showed significant positive correlation with Pb (0.63*) and Ni with Cu (0.82**). Other relations were insignificant.

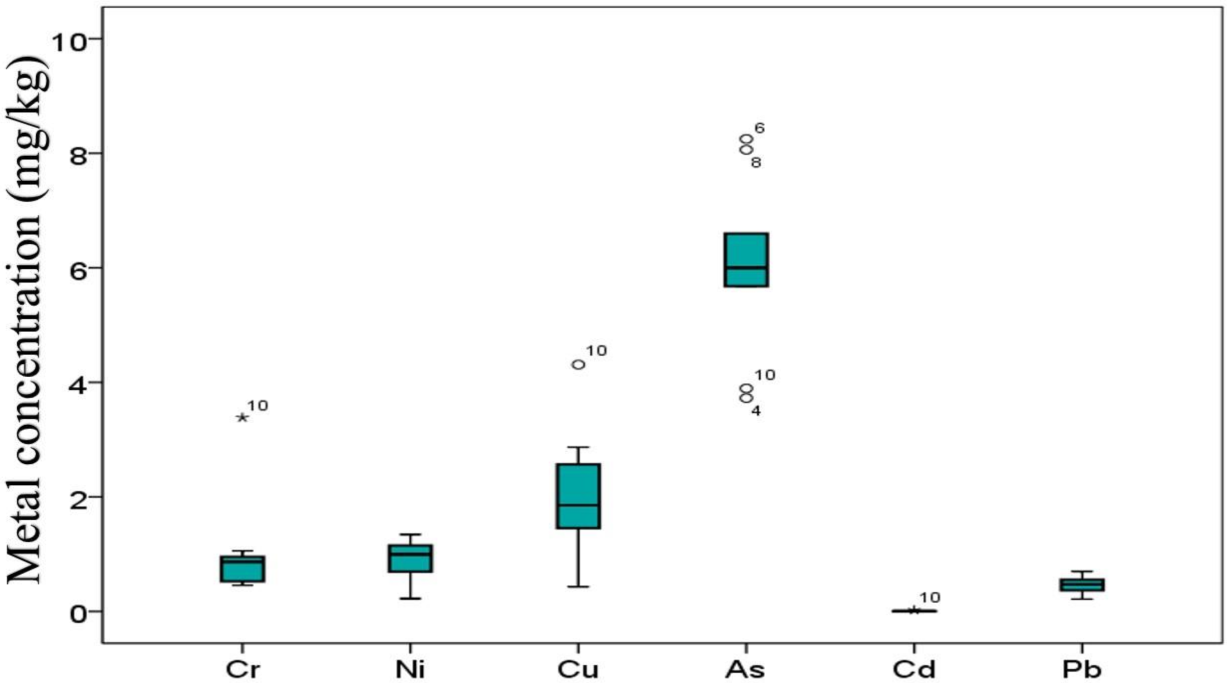

Fig. 2: Heavy Metal (Cr, Ni, Cu As, Cd and Pb) Concentrations (mg/kg) of Sediment Collected from Korotoa River in Bangladesh

Table 2: Heavy Metal Concentration in Sediment (mg/kg) Collected from Korotoa River in Bangladesh

Sample Id	Cr	Ni	Cu	As	Cd	Pb
S1	0.613	1.118	2.868	5.736	0.005	0.553
S2	0.896	1.200	2.150	6.267	0.003	0.481
S3	0.940	1.150	1.781	5.680	0.004	0.697
S4	1.059	0.223	0.430	3.725	0.004	0.369
S5	0.455	0.460	1.454	6.597	0.005	0.465
S6	0.524	0.950	1.557	8.249	0.005	0.215
S7	0.952	1.343	2.566	5.811	0.002	0.521
S8	0.477	0.724	0.822	8.062	0.001	0.408
S9	0.836	1.043	1.929	6.189	0.003	0.658
S10	3.385	0.694	4.309	3.894	0.022	0.329
Average	1.01	0.89	1.99	6.02	0.01	0.47

Table 3: Comparison of Metal Concentration in Sediment (mg/kg, dw) with Different International Guidelines and Other Studies in the World

River Location	Cr	Ni	Cu	AS	Cd	Pb	References
Korotoa River (Bangladesh)	1.01	0.89	1.99	6.02	0.01	0.47	Present study*
Buriganga River (Bangladesh)	178	200	28	NA	3.3	70	Ahmad et al. (2010)
Jamuna River (Bangladesh)	110	33	28	NA	NA	19	Datta and Subramanian (1998)
Bangshi River (Bangladesh)	98	26	31	1.93	0.61	60	Rahman et al. (2014)
River Ganges (India)	1.8–6.4	NA	0.98–4.4	NA	0.14–1.4	4.3–8.4	Gupta et al. (2009)
Yellow River (China)	41–128	NA	30–10-2	14–48	NA	26–78	Liu et al. (2009)
Gediz River (Turkey)	170–220	101–129	108–152	NA	NA	105–140	Akcay et al. (2003)
Shur River (Iran)	NA	NA	9174	NA	6.85	162	Karbassi et al. (2008)
Okumeshi River (Nigeria)	0.87	1.21	NA	NA	1.31	0.45	Raphael et al. (2011)
ASV	90	68	45	13	0.3	20	Turekian and Wedepohl (1961)
TRV	26	16	16	6	0.6	31	USEPA (1999)
LEL	26	16	16	6	0.6	31	Persuad et al. (1993)
SEL	110	75	110	33	10	250	Persuad et al. (1993)

Table 4: Heavy Metal Concentration in Surface Water (mg/kg) Collected from Korotoa River in Bangladesh

Sample Id	Cr	Ni	Cu	As	Cd	Pb
S1	0.893	1.554	4.889	2.377	0.018	0.928
S2	1.364	1.766	3.644	2.606	0.014	0.789
S3	1.544	1.797	3.265	2.445	0.018	1.184
S4	1.799	0.272	0.803	1.574	0.015	0.591
S5	0.620	0.565	2.466	2.687	0.022	0.781
S6	0.688	1.289	2.605	3.438	0.019	0.347
S7	1.525	2.139	5.109	2.499	0.008	0.896
S8	0.568	1.015	1.115	3.363	6.634	0.660
S9	1.257	1.606	3.325	2.612	0.007	1.120
S10	12.871	0.679	1.705	1.617	0.235	1.399
Average	2.31	1.26	2.89	2.52	0.69	0.86
WHO (2004)	0.005	0.07	2	0.01	0.003	0.01

Table 5: Pearson Correlation Coefficient Matrix for Heavy Metals in Sediment and Surface Water Collected from Korotoa River in Bangladesh

			Sediment			
	Cr	Ni	Cu	As	Cd	Pb
Cr	1					
Ni	-0.14	1				
Cu	0.72*	0.41	1			
As	-0.66*	0.28	-0.36	1		
Cd	0.93**	-0.23	0.74*	-0.52	1	
Pb	-0.23	0.47	0.04	-0.12	-0.35	1
			Water			
Cr	1					
Ni	-0.31	1				
Cu	-0.27	0.82**	1			
As	-0.59	0.33	0.12	1		
Cd	-0.13	-0.15	-0.44	0.47	1	
Pb	0.63*	0.18	0.21	-0.53	-0.22	1

3.4. Assessment of toxic metal pollution

Enrichment factor (EF) is a good methodology to differentiate the metal source of anthropogenic origin from those occurring naturally in the environment (Zhang et al. 2009). Enrichment factor (EF) is considered as an effective tool to evaluate the magnitude of contaminants in the environment. Enrichment factor in the present study sediment was considered indicative of human influence to the evidence of minor contaminants in the environment. The enrichment factor of the present study was presented in Figure 3. From that figure, it was cleared that the enrichment factor for the heavy metal in sediment were less than 1.5 and the level of pollution was low. Generally, studies have observed that low enrichment values indicate a great contribution for the crusted source to the sediment, while high EFs indicate a significant contribution from anthropogenic sources (Yadao and Rajamani 2006). The contamination factors (CF) for individual metal were presented in Table 6. The contamination levels of six heavy metals were less than 1 indicating the low degree of contamination except As (6.33) in the present study area sediment. I_{geo} values of the present study are presented in Figure 4. For all heavy metals in the studied samples for different sampling sites, the I_{geo} values indicated the decreasing order of As>Cu>Ni>Cr>Pb>Cd. The mean of I_{geo} values for all the studied metals for all sampling sites indicating the sediments were slowly contaminated with heavy metals.

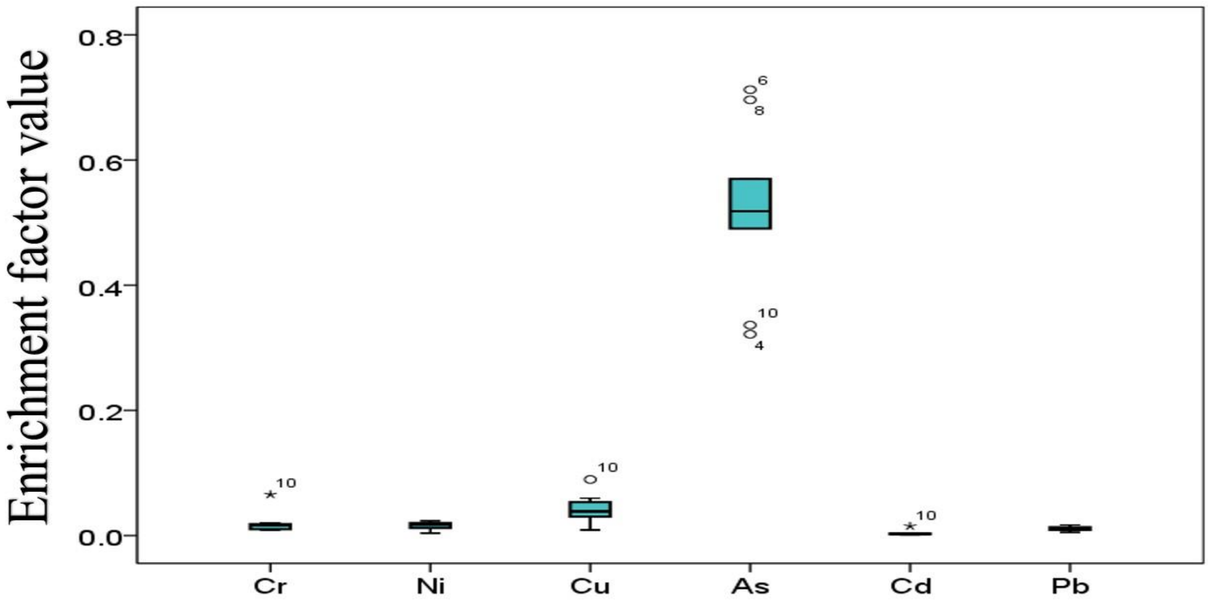

Fig. 3: Enrichment Factor of Sediment Collected from Korotoa River in Bangladesh

Table 6: Contamination Factor value (CF) of Sediment Collected from Korotoa River in Bangladesh

Sample Id	Cr	Ni	Cu	As	Cd	Pb
S1	0.0136	0.0287	0.0869	0.6038	0.0053	0.0205
S2	0.0199	0.0308	0.0652	0.6597	0.0032	0.0178
S3	0.0209	0.0295	0.0540	0.5979	0.0042	0.0258
S4	0.0235	0.0057	0.0130	0.3921	0.0042	0.0137
S5	0.0101	0.0118	0.0441	0.6944	0.0053	0.0172
S6	0.0116	0.0244	0.0472	0.8683	0.0053	0.0080
S7	0.0212	0.0344	0.0778	0.6117	0.0021	0.0193
S8	0.0106	0.0186	0.0249	0.8486	0.0011	0.0151
S9	0.0186	0.0267	0.0585	0.6515	0.0032	0.0244
S10	0.0752	0.0178	0.1306	0.4099	0.0232	0.0122
Total	0.2252	0.2284	0.6022	6.3379	0.0571	0.174

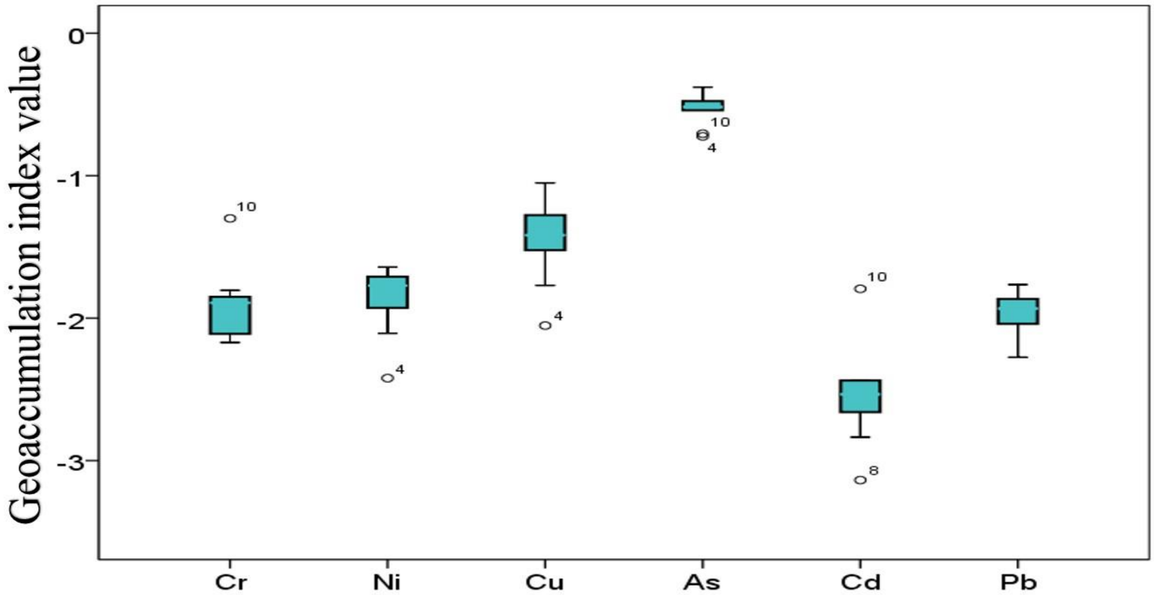

Fig. 4: I_{geo} Value of Sediment Collected from Korotoa River in Bangladesh

4. Conclusions

Heavy metal pollution may be a threat for the Korotoa River basin, Bangladesh. In this study, the concentrations of Cr, Ni, Cu, As, Cd and Pb of sediment were low and the contamination level was also low. But the toxic metal concentration in water in Korotoa River was higher than WHO reference value and Cr, Ni, As, Cd and Pb contamination were severe here. This study suggested that the point sources of heavy metals in the water and sediments should be closely monitored; improvement of conditions and industrial effluent and domestic sewage discharge should be reduced.

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