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Ecological Risk Assessment of Heavy Metals in Sediment of the Louhajang River, Bangladesh

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Abstract

Heavy metal pollution in sediment of the riverine ecosystem is regarded as a major problem in developing countries like Bangladesh. The present study was conducted to assess the contamination level of heavy metals and their ecological in sediments of the Louhajang River, Bangladesh. Certain indices, including the enrichment factor (EF), pollution load index (PLI), geoaccumulation index (I_{geo}), and contamination factor (C^i_f) were used to assess the ecological risk. The mean concentration of Cr, Ni, Cu, As, Cd, and Pb in sediments was 9.205, 7.676, 17.727, 8.999, 0.083, and 4.597 mg/kg, respectively. Metals in sediment during winter were higher than summer season. Enrichment factor (EF), contamination factor (C^i_f), geoaccumulation index (I_{geo}), and pollution load index (PLI) revealed lower contamination of sediments by the studied heavy metals. The sum of toxic units at different sampling sites was less than 4, indicating a low toxicity of heavy metals in sediments of the study river. Considering the severity of potential ecological risk for single metal, the descending order of pollutants was As > Cu > Cd > Ni > Pb > Cr. In view of the potential ecological risk, sediments of Louhajang River showed low potential ecological risk.

Keywords: Surface sediment; Heavy metal; Ecological risk; Bangladesh

Introduction

Contamination of heavy metals in the environment has attracted a wide concern due to the ever-increasing pollution of soil, sediment and water in many regions of the world, especially in some developing countries like Bangladesh [1-6]. Heavy metals such as nickel, chromium, copper, arsenic, cadmium, and lead are considered as serious contaminants in the aquatic environment because of their persistence and environmental toxicity [7-9]. Both natural and anthropogenic activities are responsible for the huge amount of toxic heavy metals is discharged in the aquatic environment [8,10,11]. Rapid industrial development and urbanization have provoked some serious concerns for the aquatic environment over the last few decades [6,12,13]. Due to sediment contamination by heavy metals in most of the urban rivers, about 80% of the world populations are facing an increasing threat regarding water security [13-15]. Sediment is considered as an essential and dynamic part of the aquatic ecosystems, with the variation of habitats and environments [4,16]. Sediments in the aquatic environment have been broadly used as environmental indicators for the assessment of metal contamination in the natural water [8,17]. In present time, the pollution of aquatic environments has been regarded as a topic of much discussion; and the issue of sediment contamination by heavy metals has received more attention due to the toxic and persistent characters [18-20]. In the riverine ecosystems, sediments can be polluted with various kinds of hazardous substances and heavy metals through several pathways such as disposal of liquid effluents, traffic emissions, terrestrial runoff, brick kilns and leachates carrying chemicals originating from numerous urban, industrial, and agricultural activities [1,21-23].

Numerous indexes have been developed to evaluate the environmental risk of heavy metals in surface sediment based on their total content, bioavailability and poisonousness [15,24]. For example, enrichment factor, geoaccumulation index, and contamination factor of individual heavy metal in sediment are calculated using its total content and sediment quality guideline value [25,26]. The pollution load index (PLI) and potential ecological risk index (PER) have also been developed to assess the combined risk of multiple heavy metals in sediment [24,27]. The pollution load index compares the metal concentrations with baseline values, which helps in assessing the enrichment of heavy metals in sediment [13]. The potential ecological risk index introduces a toxic-response factor

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for a given substance that provides a simple and quantitative value for ecological risk assessment system [28]. Therefore, it is essential to measure the concentrations of heavy metals in sediments of any polluted riverine ecosystem.

In Bangladesh, everyday a huge amount of untreated industrial wastes and toxic substances is being discharged into open water bodies and its adjacent lands [4,13,17]. Besides, a considerable amount of heavy metal enriched suspended solids is coming down from neighboring country like India through the Teesta and the Brahmaputra Rivers and have been accumulated in the riverine environment [13,29,30]. During the last decade, various researches were conducted to assess heavy metals pollution in sediments of the rivers, estuary, and lakes giving special preference to the aquatic environment [8,31-34]. But, no sound research regarding heavy metal pollution in sediment of the Louhajang River has been conducted so far. Therefore, the objectives of this study are to assess the pollution status of the Louhajang River by estimating the levels of heavy metals (Cr, Ni, Cu, As, Cd, and Pb) in surface sediments in two different seasons and to observe the level of ecological risk of heavy metals in sediments.

Materials and Methods

Description of the study area

The area of Tangail district is 334.26km^2 and located at the center point in Bangladesh. Tangail Sadar Upazila is one of the most densely polluted areas in Bangladesh where the density of population is 1,100/km^2 (2011 census). Tangail is an industrial growing site of Bangladesh, which is highly susceptible to environmental pollution over the last decade. There are several types of industrial units including garments, packaging industry, dyeing, brick kiln, metal workshops, battery manufacturing industries, tanneries, pesticide and fertilizer industries, different food processing industries and other factories produce huge volumes of effluents that contain heavy metals. These industries were set up on the bank of Louhajang River. The length and width of this river are 85km and 78m respectively. Now a days, this River is continuously polluted due to discharge of untreated wastes and effluents from these industries and, also household and municipal wastes throw randomly to the River. Tangail District stands on the bank of Louhajang River. This river has great importance on agriculture and household activities and daily uses. Louhajang River branches out from the Jamuna near Gabsain at Bhuapur, Tangail District. Thereafter it splits into two parts before the parts meet up again. It flows near Tangail city, Karotia and Jamurki before joining the Bangshi. The location of the river is shown in the following map (Figure 1). The indiscriminate dumping of domestic, urban, and industrial wastes, combined with the failure of authorities to protect the ecological health of the river has aggravated the situation to the point where this river is dying biologically and hydrologically. The wastes are mixed with sediment and water and continuously polluted by heavy metals cause river pollution. Storage of water in this river relies heavily on rainfall. The water runoff along the ground can pick up soil contaminants including wastes, heavy metals, etc. and deposit as sediments, that become source pollution. For this, we have selected Louhajang River for determining toxic metal contamination with ecological and human health risk.

Sediment sample collection and preparation

Sediment samples were collected by random method from different sampling location in the river. Two seasons summer (August–September, 2017) and winter (February–March, 2017) was selected for sediment sample collection. Composite sediment samples were collected by using standard procedure from every sampling location [35]. Since 15 sampling sites of is the definite number of sites the author worked on and 200g sediment collected at a depth of 0 to 5 cm using a portable Ekman grab sampler. Sediment was taken by means of a percussion hammer corer at a length of 50–80 cm for determining background value of preindustrial sample [36]. A standard procedure was followed for the characterization of sediments [37]. To determine the age and sediment accumulation rates, Lead-210 dating by alpha spectrometry method was applied. Each sediment sample was obtained by mixing sediments randomly collected (3 times) at each sampling point and 15 pairs of composite sediment samples were collected. Sediment samples were then freeze-dried to obtain constant weight. The samples were homogenized by grinding in an agate mortar, sieved through 106mm aperture nylon sieve and stored in labeled glass bottles until chemical analyses.

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 electrical conductivity (EC) determination, 5.0g of sediment was taken in 50mL polypropylene tubes. Then, 30mL of distilled water was added to the tube. The lid was closed properly and was shaken for 5min. After that, EC was measured using an EC meter (Horiba D-52). Percent N and C of sediment was measured using 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 followed by USDA system. About 50g of oven dried soil was taken in a dispersion cup and 10mL of 5% calgon solution was added to the samples and allowed to soak for 15 minutes. Then 90mL 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 litre 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. The percentage of sand, silt and clay were calculated as follows:

%(Silt + Clay) = (Corrected hydrometer reading at 40 seconds/ Oven dry weight of sediment) × 100

%(Clay) = (Corrected hydrometer reading after 2 hours/ Oven dry weight of sediment) × 100

Sand (%) = 100 - %(Silt + Clay)

Silt (%) = %(Silt + Clay) - % Clay

Heavy metal assessment from sediment

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% HNO_3 for more than 24h, then rinsed with Milli-Q water and dried. For metal analysis, 20mL water sample and 0.5g of sediment sample was treated with 5mL 69% HNO_3 acid and 2mL 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 = 0.45μm) Toyo Roshi Kaisha, Ltd., Japan and stored in 50mL polypropylene tubes (Nalgene, New York). Afterwards, the vessels were cleaned with Milli-Q water and dried with air. Finally, blank digestion with 5mL 69% HNO_3 following the said digestion procedures were carried out to clean up the digestion vessels. For heavy metals, samples were analyzed by using inductively coupled plasma mass spectrometry (ICPMS, Agilent 7700 series). Multi-element Standard XSTC-13 (SpexCertiPrep®, USA) solutions was used to prepare calibration curve. The calibration curves with $R_2 > 0.999$ were accepted for concentration calculation. Multi element solution (Agilent Technologies, USA) 1.0μg/L was used as 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, a run included blank, certified reference materials (CRM) and samples were analyzed in duplicate to eliminate any batch-specific error.

Sediment quantitative ecological assessment

Enrichment factor (EF): Enrichment factor (EF) is considered as an effective tool to evaluate the magnitude of contaminants in the environment [38,39]. The EF for each element was calculated to evaluate anthropogenic influences on heavy metals in sediments using the following formula [40]:

$$EF = (C_M/C_{Al})_{sample} / (C_M/C_{Al})_{background} \qquad (1)$$

where, $(C_M/C_{Al})_{sample}$ is the ratio of concentration of heavy metal (C_M) to that of aluminium (C_{Al}) in the sediment sample, and (C_M/C_{Al}) background is the same reference ratio in the background sample. The background values of Cr, Ni, Cu, As, Cd, and Pb in sediments were 41, 39, 35, 8.5, 0.92, and 23 mg/kg, respectively and the background value of Al was taken from another study [17]. Generally, an EF value of about 1 suggests that a given metal may be entirely from crustal materials or natural weathering processes [41]. 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, respectively [42].

Contamination factor: The contamination factor (CF) is the ratio obtained by dividing the concentration of each metal in the sediment by the baseline or background value.

$$C_f^i = \frac{C_{heavymetal}}{C_{background}} \qquad (2)$$

The contamination factor being classified into four grades for monitoring the pollution of one single metal over a period of time [28]. The contamination levels may be classified based on their intensities on a scale ranging from 1 to 6: low degree ($C_f^i < 1$), moderate degree ($1 \le C_f^i < 3$), considerable degree ($3 \le C_f^i < 6$), and very high degree ($C_f^i \ge 6$) [43] (Table 5). Thus, the values can monitor the enrichment of one given metal in sediments over a period of time.

Geoaccumulation index (I_{geo}): 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 sediment contamination [44]. In order to characterize the level of pollution in the sediment, geoaccumulation index (I_{geo}) values were calculated using the equation:

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

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 background sample [45]. 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: $I_{geo} \le 0$ – practically uncontaminated; $0 \le I_{geo} \le 1$ – uncontaminated to moderately contaminated; $1 \le I_{geo} \le 2$ – moderately contaminated; $2 \le I_{geo} \le 3$ – moderately to heavily contaminated; $3 \le I_{geo} \le 4$ – heavily contaminated; $4 \le I_{geo} \le 5$ – heavily to extremely contaminated; and $5 < I_{geo}$ – extremely contaminated.

Pollution load index (PLI): Pollution load index act as an integrated approach which assess sediment quality of heavy metals. According to Suresh et al. 2011 [46] pollution load index may be assessed from six hazardous elements (Cr, Ni, Cu, As, Cd, and Pb). Pollution load index can be determined for six toxic metals like Cr, Ni, Cu, As, Cd, and Pb. Pollution load index was measured by using the following formula [47]:

$$PLI= (CF_1 \times CF_2 \times CF_3 \times \ldots \times CF_n)^{1/n} \qquad (4)$$

Where, CF is contamination factor for each single metal and n is number of heavy metals. The overall toxicity status of heavy metals in sediment may be assessed from Pollution load index (PLI) calculation. PLI is the share of conclusion of six heavy metals.

Potential ecological risk (PER): The degrees of hazardous elements contamination in sediments are determined by PER index. Luo et al. 2007 [43] proposed equations, which were used to calculate PER and are as follows:

$$C_f^i = \frac{C^i}{C_n^i} C_d = \sum_{i=1}^{n} C_f^i \qquad (5)$$

$$E_r^i = T_r^i \times C_f^i PER = \sum_{i=1}^{m} E_r^i \qquad (6)$$

where, C_f^i is the single element contamination factor, C^i is the content of the element in samples and C_n^i is the background value of the element. The background value of Cr, Ni, Cu, As, Cd, and Pb in sediments were 90, 68, 45, 13, 0.3, and 20 mg/kg, respectively [48]. The sum of C_f^i for all metals represent the integrated pollution degree (C_d) of the environment. E_r^i is the potential ecological risk index and is the biological toxic factor of an individual element. The toxic-response factors for Cr, Ni, Cu, As, Cd, and Pb were 2, 6, 5, 10, 30, and 5, respectively [13,28,49]. PER is the comprehensive potential ecological risk index, which is the sum of E_r^i. Sensitivity of the biological community is represented by it to the toxic substance and indicates the potential ecological risk caused by the overall contamination.

Toxic unit analysis

The sum of toxic units (ΣTUs) is considered as potential acute toxicity of hazardous elements in sediment samples. Toxic unit analysis is stated as the ratio of the assessed concentration of hazardous elements in sediment to probable effect level (PELs) [50]. A moderate to serious toxicity of hazardous elements remain in sediment when the sum of toxic units for all sediment samples is more than 4 [21]. The TU for each metal was calculated using the following formula:

$$TU= (C_M/PEL) \qquad (7)$$

Where, C_M is the concentration of heavy metal (C_M) in sediment and PEL is the probable effect levels value of heavy metals in sediment.

$$\Sigma Tus = TU_{metal1} \times TU_{metal2} \times TU_{metal3} \times TU_{metal4} \times \ldots\ldots\ldots \times TU_{metaln} \qquad (8)$$

where, ΣTus is the product of toxic units for heavy metals in sediments.

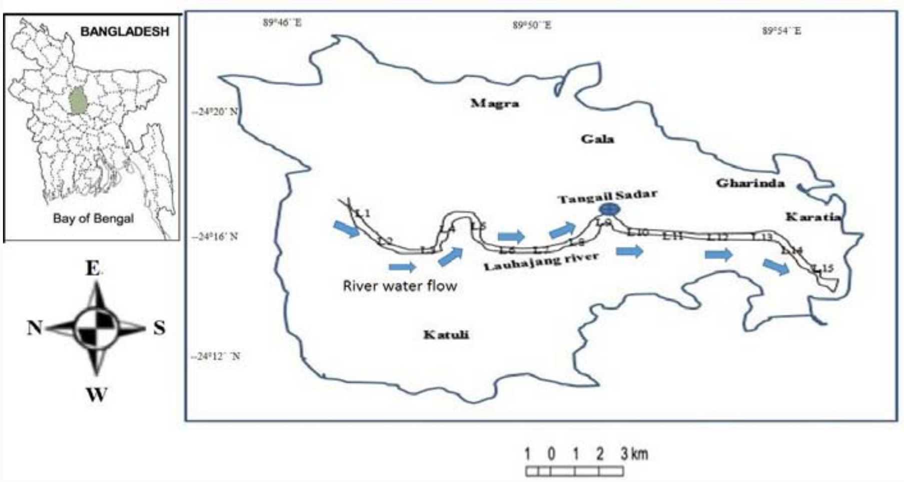

Figure 1: Map of the study area of Louhajang River, Bangladesh.

Table 1: Physicochemical properties of sediment collected from Louhajang River, Bangladesh.

Sites									Sediment texture (%)						
	pH		EC(dS/m)		%N		%OC		Sand		Silt		Clay		Textural class*
	Summer	Winter	Summer	Winter	Summer	Winter	Summer	Winter	Summer	Winter	Summer	Winter	Summer	Winter	
L1	6.48	6.53	0.41	0.89	0.12	0.18	0.37	0.76	25.6	37.6	52.6	54.2	21.8	8.2	Silt loam
L2	7.18	7.12	0.32	1.07	0.09	0.13	0.23	0.69	31.9	35.4	56.1	50.8	12	13.8	Silt loam
L3	7.1	7.23	0.56	1.18	0.16	0.15	0.16	1.21	22.1	42.7	57.8	39.8	20.1	17.5	Silt loam
L4	7.34	7.47	0.96	1.16	0.11	0.17	2.14	1.97	35.4	23.8	48.7	49.2	15.9	26	loam
L5	7.33	7.26	0.89	1.19	0.15	0.24	1.85	3.26	39.2	29.8	42.5	50.9	18.3	11.3	loam
L6	6.91	7.13	0.45	1.24	0.07	0.15	0.13	0.35	26.7	40.7	52.9	51.9	20.4	8.4	Silt loam
L7	7.09	7.19	0.31	1.53	0.16	0.25	0.42	0.78	22.3	30.7	58.9	53.8	18.8	15.5	Silt loam
L8	7.2	6.99	0.34	0.83	0.18	0.23	0.26	0.49	30.8	28.7	58.7	55.8	10.5	15.5	Silt loam
L9	7.18	7.31	0.29	0.75	0.15	0.24	1.02	1.46	42.4	31.9	51.7	61.2	5.9	6.9	Silt loam
L10	7.39	7.47	0.66	0.87	0.08	0.13	0.31	0.66	29.3	31.2	61.9	58.5	8.8	10.3	Silt loam
L11	7.46	7.74	0.19	0.84	0.1	0.18	0.18	0.31	45.6	49.5	36.9	42.1	17.5	8.4	loam
L12	7.22	7.39	0.42	1.02	0.21	0.2	0.26	0.83	22.3	27.6	59.2	52.6	18.5	19.8	Silt loam
L13	6.87	6.59	0.65	1.28	0.13	0.18	1.34	2.45	44.6	39.8	50.6	54.7	4.8	5.5	Silt loam
L14	7.62	7.81	0.91	1.34	0.12	0.16	2.31	2.56	23.6	26.2	51.6	56.3	24.8	17.5	Silt loam
L15	7.04	6.95	0.41	0.98	0.09	0.14	0.13	0.26	29.8	35.5	54.2	58.8	16	5.7	Silt loam

*Note: According to the United States Department of Agriculture soil classification system.

Statistical analysis

The data were statistically analyzed by using the statistical package SPSS 20.0 (International Business Machines Corporation [IBM] Armonk, NY, USA). The means and standard deviations of the metal concentrations in sediment were calculated. A Pearson correlation analysis was used to assess the inter element relationship in sediment. Multivariate methods in terms of principal component analysis (PCA) were used to interpret the potential sources of hazardous element in sediment. Microsoft Excel 2013 was used for other calculations.

Results and Discussion

Physicochemical properties and heavy metals in sediment

The physicochemical properties of sediments in the study sites are presented in Table 1. The pH of the sediments was slightly acidic for all the sites except L4, L10, L11, and L14 which showed slight alkalinity. Due to the variation in topography, hydrology and geology within the catchment areas, as well as differences in precipitation and local climate, the chemical properties such as pH, alkalinity, and concentration of heavy metals may differ substantially between streams even within small distances [6,13,17]. The lower pH at most of the sites of the studied river might be due to discharge of the acidic effluent from nearby industries. Electrical conductivity in sediments ranged from 0.19 to 0.96 (dS/m) during summer and 0.75 to 1.53 (dS/m) during winter. Total nitrogen content was ranged from 0.07 to 0.21% during summer and 0.13 to 0.25% during winter (Table 1). The composition of the organic carbon in the riverine sediments is varying due to its origin in the aquatic environment [14]. Phytoplankton and zooplankton are the most abundant sources of the organic material in the sediments [17,51]. The composition of the organic carbon in sediments was ranged from 0.13 to 2.31% and 0.26 to 3.26% during summer and winter season, respectively (Table 1). The highest percentage of organic carbon might be attributed to the high amount of drainage water at L5 site. The high rate of organic growth together

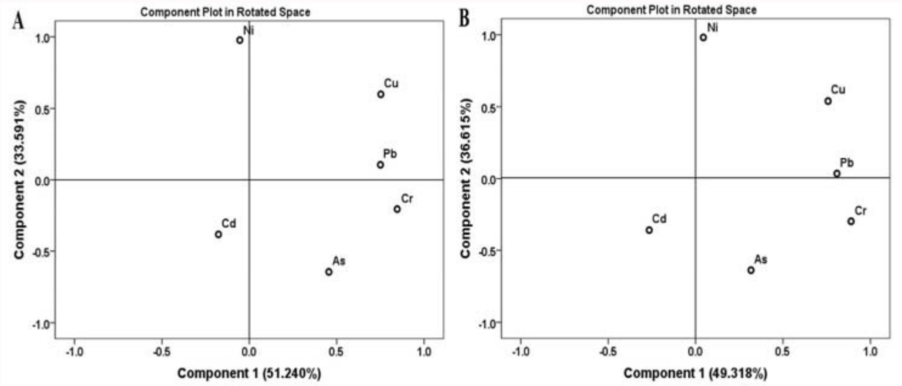

Figure 2: Principal component analysis (PCA) of heavy metals in sediments (A in summer and B in winter season) collected from Louhajang River, Tangail, Bangladesh. Considering the highest component loading, first PC exhibited elevated loadings of Cr, Cu, As, Cd, and Pb. Second PC exhibited elevated loadings of Ni.

Table 2: Concentration of heavy metals (mg/kg dw) in sediments collected from Louhajang River, Bangladesh.

Sites	Cr		Ni		Cu		As		Cd		Pb	
	Summer	Winter	Summer	Winter	Summer	Winter	Summer	Winter	Summer	Winter	Summer	Winter
L1	3.708	6.674	2.188	3.501	9.041	12.658	5.015	6.519	0.098	0.166	0.575	0.833
L2	4.024	7.243	3.11	4.976	13.224	18.513	9.236	12.007	0.071	0.121	3.215	4.662
L3	2.937	5.286	2.951	4.721	8.133	11.386	8.181	10.635	0.059	0.099	2.122	3.077
L4	5.464	9.835	5.443	8.708	7.036	9.851	9.231	12.001	0.032	0.055	2.658	3.854
L5	11.667	21	3.021	4.833	18.779	26.291	9.314	12.108	0.069	0.118	4.592	6.659
L6	11.395	20.511	3.152	5.044	20.602	28.843	10.122	13.158	0.049	0.083	4.841	7.019
L7	7.916	14.248	3.291	5.266	17.802	24.922	11.521	14.977	0.035	0.059	4.397	6.376
L8	15.326	27.588	3.644	5.831	13.103	18.344	8.576	11.149	0.037	0.063	8.792	12.748
L9	9.186	16.535	10.287	16.459	21.621	30.27	5.695	7.404	0.041	0.07	5.822	8.442
L10	4.589	8.259	17.624	28.198	19.652	27.513	2.987	3.883	0.035	0.06	4.713	6.834
L11	3.785	6.813	5.119	8.19	9.419	13.187	2.58	3.355	0.043	0.073	3.337	4.839
L12	3.691	6.643	17.195	27.512	20.617	28.864	3.515	4.57	0.035	0.059	2.944	4.269
L13	5.191	9.344	3.064	4.902	12.658	17.721	7.021	9.127	0.137	0.233	0.805	1.167
L14	5.634	10.141	4.354	6.966	18.513	25.918	12.931	16.81	0.1	0.169	4.501	6.527
L15	4.111	7.4	4.131	6.61	11.386	15.941	11.453	14.889	0.082	0.139	2.971	4.308
Mean±SD	6.575±3.707	11.835±6.674	5.905±5.047	9.448±8.075	14.773±5.103	20.682±7.144	7.825±3.247	10.173±4.221	0.061±0.031	0.105±0.052	3.752±2.032	5.441±2.947

with the organic detritus introduced by the drainage system can be considered the main source of organic carbon [52]. According to the USDA soil texture classification, the textural analysis revealed that the sediments of the studied river were loam and silt loam (Table 1).

The concentrations of heavy metals in sediment are presented in Table 2. A wide range of values for metal concentrations was observed among the sampling sites. Factors such as salinity, geomorphological setup and land runoff might have played a vital role in the variation of metals [17]. Metals concentrations in sediment during winter season were slightly higher than summer due to the lower water flow during season winter which could support to accumulate the heavy metals in sediment [8,30]. The concentrations of heavy metals at L8 and L9 sites were much higher than others sites indicated that downstream river activities and urbanization drove heavy metals contamination in surface sediment [24]. The urban activities (industrial discharges, municipal waste water, household garbage, and urban runoff) of Tangail district are the main causes of higher metal input at P7 and P8 sites. The average concentration of heavy metals in sediments were in the decreasing order of Cu> Cr > As > Ni> Pb> Cd.

Among the sites in the current study, the highest level of Cr was observed in sediment collected from L8 site (15.326 and 27.588 mg/kg, summer and winter season, respectively) (Table 2). An elevated concentration of Cr was observed at sites (L5–L6). The chromium enrichment of sediment could have been caused by two reasons: (1) natural: concentration of Cr-bearing minerals; and (2) anthropogenic: industrial activities such as tanneries and textile factories which are discharging Cr based oxidants (chromate, dichromate, etc.) [4]. Hence, the waste discharged from such industries was most probably responsible for elevated Cr level in the exposed sediment [30]. The mean concentration of Ni in sediment was found as 5.905 and 9.448 mg/kg in summer and winter season, respectively (Table 2). The highest concentration of Ni was observed at L10 site (17.624 and 28.198 mg/kg, winter and summer season, respectively). Slightly higher level of Ni was observed at the site near to the district urban area, which indicates the higher input of Ni in sediment that might be originated from urban and industrial wastes [13,30].

Table 3: Comparison of metals in sediment (mg/kg dw) with some reference values and some reported values.

Study location	Cr	Ni	Cu	As	Cd	Pb	References
Louhajang River, Bangladesh	9.205 (2.937-27.588)	7.676 (2.951-28.198)	17.727 (7.036-30.270)	8.999 (2.580-16.810)	0.083 (0.032-0.233)	4.597 (0.575-12.748)	Present study*
Buriganga River, Bangladesh	297 (17–841)	240 (62–539)	280 (62–712)	21 (7.6–67)	7.7 (2.0–19)	731 (43–3312)	[13]
Jamuna River, Bangladesh	110	33	28	-	-	19	[59]
Buriganga River, Bangladesh	178	200	28	NA	3.3	70	[1]
Bangshi River, Bangladesh	98	26	31	1.9	0.61	60	[60]
Ganges River, India	1.8–6.4	NA	0.98–4.4	NA	0.14–1.4	4.3–8.4	[61]
Gomti River, India	8.15	16	5	NA	2.4	40	[62]
Okumeshi River, Nigeria	0.87	1.21	NA	NA	1.32	0.45	[63]
TRV (Toxicity reference value)	26	16	16	6	0.6	31	[64]
ASV (Average shale value)	90	68	45	13	0.3	20	[65]
LEL (Lowest effect level)	26	16	16	6	0.6	31	[66,67]
TEL (Threshold effect level)	37	18	36	5.9	0.59	35	[67]
CUC (Continental upper crust)	92	47	28	5	0.09	17	[68]
SEL (Severe effect Level)	110	75	110	33	10	250	[66,67]
PEL (Probable effect level)	90	36	197	17	3.5	91	[67]

Table 4: Pearson correlation coefficient matrix for heavy metals in the sediments of Louhajang River, Bangladesh.

	pH	EC	N	OC	Sand	Silt	Clay	Cr	Ni	Cu	As	Cd	Pb
Summer													
pH	1												
EC	0.33	1											
N	0.02	-0.08	1										
OC	0.39	0.82**	0.04	1									
Sand	0.14	-0.21	-0.19	0.27	1								
Silt	-0.2	-0.14	0.22	-0.39	-0.70**	1							
Clay	0.02	0.18	0.009	0.06	-0.60*	-0.14	1						
Cr	0.04	-0.005	0.21	0.11	0.13	-0.02	-0.15	1					
Ni	0.33	-0.01	0.17	-0.13	-0.08	0.36	-0.28	-0.2	1				
Cu	0.23	0.008	0.15	0.05	-0.12	0.24	-0.09	0.39	0.5	1			
As	0.06	0.32	-0.1	0.35	-0.31	0.09	0.33	0.29	0.60*	0.01	1		
Cd	-0.37	0.25	-0.18	0.29	0.15	-0.18	-0.005	-0.26	-0.45	-0.21	0.23	1	
Pb	0.44	-0.14	0.18	-0.03	0.001	0.17	-0.19	0.80**	0.14	0.17	0.17	-0.54	1
Winter													
pH	1												
EC	0.02	1											
N	-0.05	0.04	1										
OC	0.1	0.41	0.27	1									
Sand	-0.2	-0.15	-0.29	-0.37	1								
Silt	-0.23	-0.17	0.16	0.02	-0.52*	1							
Clay	0.44	0.3	0.04	0.18	-0.58*	-0.32	1						
Cr	0.08	-0.001	0.58*	0.12	-0.28	0.28	-0.05	1					
Ni	0.38	-0.39	-0.05	-0.16	-0.31	0.31	0.12	-0.2	1				
Cu	0.25	0.09	0.3	0.11	-0.35	0.54*	-0.16	0.39	0.5	1			
As	-0.03	0.66**	0.001	0.27	-0.31	0.13	0.2-	0.29	-0.60*	0.01	1		
Cd	-0.5	0.25	-0.24	0.42	0.22	0.14	-0.42	-0.25	-0.45	-0.21	0.23	1	
Pb	0.32	-0.22	0.4	-0.13	-0.36	0.33	0.08	0.80**	0.14	0.47	0.17	-0.54	1

*Correlation is significant at the 0.05 level (two-tailed); **Correlation is significant at the 0.01 level (two-tailed).

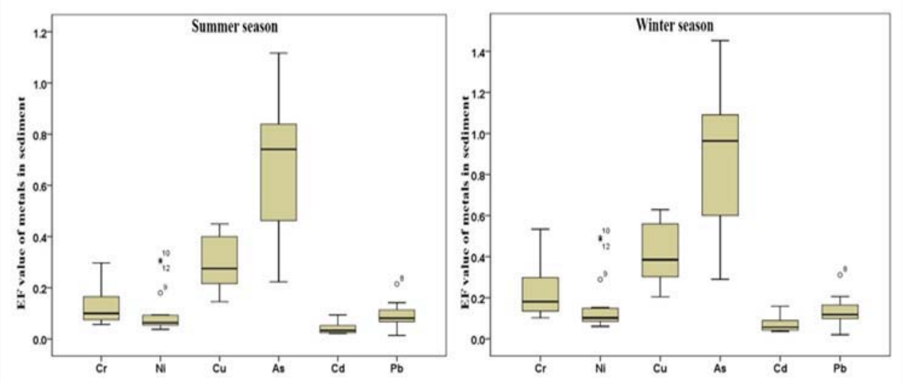

Figure 3: Enrichment factor value of metals in sediment collected from Louhajang River, Bangladesh.

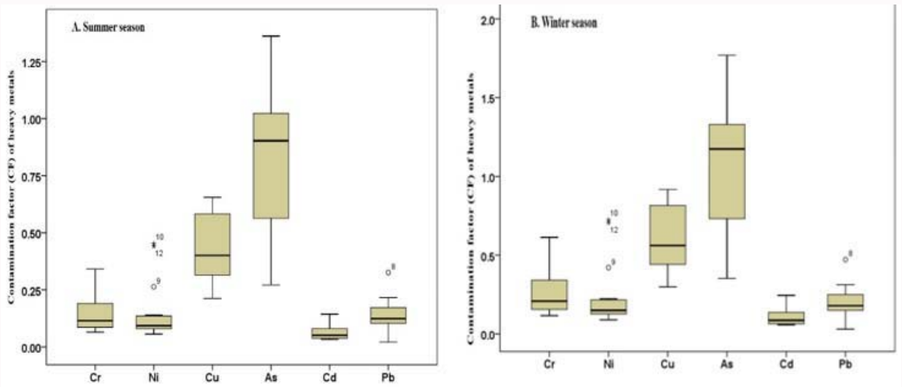

Figure 4: Contamination factors of heavy metals in sediment collected from Louhajang River, Bangladesh.

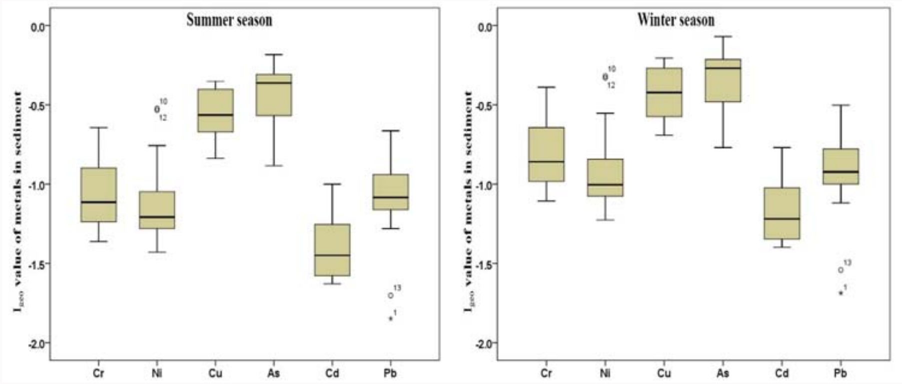

Figure 5: Geoaccumulation index (I_{geo}) of heavy metals in sediment collected from Louhajang River, Bangladesh.

The highest concentration of Cu was observed at L9 site (21.621 and 30.270 mg/kg, summer and winter season, respectively), where an elevated level of Cu was found at sites L5, L6, L12, and L14 (Table 2). Higher level of Cu indicates its higher input in the sites (L5, L6, L12, and L14), which is originated from anthropogenic activities such as vehicle and coal combustion emissions, car lubricants, and natural phenomenon such as metal contents of rocks and parent materials, processes of soil formation [4,24,48,53]. In the present investigation, the mean concentration of As in sediment was observed 7.825 and 10.173 mg/kg during summer and winter season, respectively (Table 2). The highest concentration of As was observed at L14 site followed by L7 site. Recently, the anthropogenic activities such as treatment of agricultural land with arsenical pesticides [53], treating of wood using chromated copper arsenate [6], burning of coal in thermal plants power stations, and sediment excavation that alters the hydraulic regime and/or arsenic source material increased the rate of discharge into freshwater habitat [54].

The mean concentration of Cd was 0.061mg/kg in summer and 0.105mg/kg in winter (Table 2). Among the sampling sites, the highest level of Cd was observed at L13 site (0.137 and 0.233 mg/kg) during winter and summer season, respectively. Higher Cd concentration in sediment of Louhajang River might be related to industrial activity, atmospheric emission, and Cd plated items. 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 contaminants in the sediment [13,14].

Table 5: Indices and grades of potential ecological risk of heavy metal pollution [43].

Contamination factor (C_r^i)	Contamination degree of individual metal	Degree of contamination (C_d)	Contamination degree of the environment		Grade of ecological risk of individual metal	Risk index (PER)	
<1	Low	C_d <5	Low contamination	<40	Low risk	RI<65	Low risk
1≤<3	Moderate	5≤ C_d <10	Moderate contamination	40≤<80	Moderate risk	65≤RI < 130	Moderate risk
3≤<6	Considerable	10≤ C_d <20	Considerable contamination	80≤<160	Considerable risk	130 ≤RI < 260	Considerable risk
≥6	High	C_d ≥20	High contamination	160≤<320	High risk	RI ≥ 260	Very high risk
				≥320	Very high risk		

Table 6: Contamination factors, degree of contamination, contamination level, and pollution load of heavy metals in sediments collected from Louhajang River, Bangladesh.

Sites	Season	Contamination factors ()						Degree of contamination (C_d)	Contamination level
		Cr	Ni	Cu	AS	Cd	Pb		
L1	Summer	0.082	0.056	0.274	0.528	0.103	0.021	1.065	Low contamination
	Winter	0.148	0.09	0.384	0.686	0.175	0.031	1.514	Low contamination
L2	Summer	0.089	0.08	0.401	0.972	0.075	0.119	1.736	Low contamination
	Winter	0.161	0.128	0.561	1.264	0.127	0.173	2.413	Low contamination
L3	Summer	0.065	0.076	0.246	0.861	0.062	0.079	1.389	Low contamination
	Winter	0.117	0.121	0.345	1.119	0.105	0.114	1.922	Low contamination
L4	Summer	0.121	0.14	0.213	0.972	0.034	0.098	1.578	Low contamination
	Winter	0.219	0.223	0.299	1.263	0.058	0.143	2.204	Low contamination
L5	Summer	0.259	0.077	0.569	0.98	0.073	0.17	2.129	Low contamination
	Winter	0.467	0.124	0.797	1.275	0.124	0.247	3.032	Low contamination
L6	Summer	0.253	0.081	0.624	1.065	0.051	0.179	2.254	Low contamination
	Winter	0.456	0.129	0.874	1.385	0.087	0.26	3.191	Low contamination
L7	Summer	0.176	0.084	0.539	1.213	0.037	0.163	2.212	Low contamination
	Winter	0.317	0.135	0.755	1.577	0.062	0.236	3.082	Low contamination
L8	Summer	0.341	0.093	0.397	0.903	0.039	0.326	2.099	Low contamination
	Winter	0.613	0.15	0.556	1.174	0.066	0.472	3.031	Low contamination
L9	Summer	0.204	0.264	0.655	0.599	0.044	0.216	1.982	Low contamination
	Winter	0.367	0.422	0.917	0.779	0.074	0.313	2.873	Low contamination
L10	Summer	0.102	0.452	0.596	0.314	0.037	0.175	1.676	Low contamination
	Winter	0.184	0.723	0.834	0.409	0.064	0.253	2.466	Low contamination
L11	Summer	0.084	0.131	0.285	0.272	0.045	0.124	0.941	Low contamination
	Winter	0.151	0.21	0.4	0.353	0.076	0.179	1.37	Low contamination
L12	Summer	0.082	0.441	0.625	0.37	0.036	0.109	1.663	Low contamination
	Winter	0.148	0.705	0.875	0.481	0.062	0.158	2.429	Low contamination
L13	Summer	0.115	0.079	0.384	0.739	0.144	0.03	1.49	Low contamination
	Winter	0.208	0.126	0.537	0.961	0.245	0.043	2.119	Low contamination
L14	Summer	0.125	0.112	0.561	1.361	0.105	0.167	1.065	Low contamination
	Winter	0.225	0.179	0.785	1.769	0.178	0.242	3.379	Low contamination
L15	Summer	0.091	0.106	0.345	1.206	0.086	0.11	1.944	Low contamination
	Winter	0.164	0.169	0.483	1.567	0.147	0.16	2.69	Low contamination

Among the sampling sites, the highest level of Pb was observed at L8 site (8.792 and 12.748 mg/kg) during winter and summer season, respectively (Table 2). The highest level of Pb in sediments at L8 site can be due to the effect from point and non-point sources; such as municipal runoffs, atmospheric deposition and leaded gasoline, chemical manufacturing and steel works in urban area of Tangail district [23,30,52].

To predict the metal pollution in sediment of the studied river in Bangladesh, the available data for a comparative analysis with background and toxicological reference values and some studied river sediment values are presented in Table 3. It was noted that the average concentration of As and Cd in the sediment samples exceeded the geochemical background, i.e., average worldwide shale standard and continental upper crust value. The mean concentrations of Cu, As, and Cd were also higher than those of the U.S. Environmental Protection Agency's (USEPA) toxicity reference values, lowest effect levels and threshold effect level (Table 3), which are expected to be frequently associated with the adverse biological effects. The mean concentrations of heavy metals in sediment of the studied river were higher than some other study rivers such as Ganges, Gomoti, and Okumeshi rivers from other countries (Table 3). The results indicated that the levels of heavy metals found in sediment of the studied river might create an adverse effect on the aquatic ecosystem, especially since it receives urban wastewaters from the nearby district.

Statistical analyses were performed in order to elucidate the relations among the metals and physicochemical properties in sediment of the studied river. Inter-metal interactions may illustrate the sources and pathways of the metals present in the particulate media [20]. Pearson's correlation coefficients for the investigated metals and the physicochemical properties are depicted in Table 4. During summer season, a clear pattern of significant positive correlation was found among physicochemical properties and metals in the sediment samples: OC-EC, As-Ni, and Pb-Cr; negative correlation was found among Silt-Sand and Clay-Sand. However, in winter season, significant positive correlation was found among Cr-N, Cu-Silt, As-EC, and Pb-Cr; negative correlation was found among Silt-Sand, Clay-Sand, and As-Ni. High positive correlations between specific heavy metals in sediment may reflect similar levels of pollution and/or release from the common sources of pollution, mutual dependence and identical behavior during their transport [8, 22].

Source analysis of heavy metals in sediment

To identify the hypothetical sources of heavy metals (natural or anthropogenic) in sediment of different sites of Louhajang River, a principal component analysis (PCA) was performed following the standard procedure stated in the literature [38], which showed clustering of the variables into different groups, where variables belonging to one group are highly correlated with each other [6,55]. The principal component analysis was performed on the dimensionless standardized form of data set and is presented in Figure 2. The Varimax rotation was used to maximize the sum of the variance of the factor coefficients. Two principal components were obtained, and those accounted for 84.831% (summer season) and 85.934% (winter season) of all the total variation. In the PCA analysis, first two PCs were computed, and the variances explained by them were 51.240% and 33.591% for sediment collected during summer season and 49.318% and 36.615% for sediment collected during winter season, respectively (Figure 2). Overall, the PCA revealed two major groups of the metals in sediments for both season. PC1 is strongly correlated with Cr, Cu, As, Cd, Pb and PC2 is also strongly correlated with Ni. The source of PC1 and PC2 can be considered as a mixed source of anthropogenic inputs, particularly from industrial effluents and agricultural activities in the study area.

Assessment of metal pollution in sediment

An enrichment factor (EF) has been applied by several researchers, for the assessment of heavy metals pollution in sediment [56]. Enrichment factor (EF) is a standardization method widely used to categorize the metal fractions that is associated with sediments [17]. The calculated values of EF for each of the studied metals are presented in Figure 3. Taking as a whole, the mean EF values of all the studied metals suggested their enrichments in surface sediments of the Louhajang River in Bangladesh. The mean enrichment factor values of Cr, Ni, Cu, As, Cd, and Pb were 0.127, 0.103, 0.307, 0.675, 0.042, and 0.091 during summer and 0.229, 0.165, 0.429, 0.878, 0.071, and 0.132 during winter season, respectively. The EF values of heavy metals for all sampling sites were less than 1 suggested that these metals might be delivered from non-crustal materials, or non-natural weathering processes [13]. As a whole, the enrichment factor of all the studied metals for all sampling sites were in the descending order of As>Cu>Cr>Pb>Ni>Cd. The value of contamination factor (CF) for all metals showed low degree of contamination (CF <1), whereas As showed very moderate degree of contamination during winter season (Figure 4). Overall, the CF for all metals were the descending order of As > Cu > Cr > Ni > Pb > Cd. The mean CF values of Cr, Ni, Cu, As, Cd, and Pb were 0.145, 0.151, 0.447, 0.823, 0.064, and 0.139 during summer and 0.263, 0.242, 0.626, 1.070, 0.110, and 0.201 during winter season, respectively.

The values of geoaccumulation index (I_{geo}) of the studied heavy metals are presented in Figure 5. Among the studied metals, the I_{geo} values showed the decreasing order of As>Cu>Cr>Pb>Ni>Cd. The I_{geo} values for the studied metals indicated practically uncontaminated. Pollution load index (PLI) value equal to zero indicates perfection; value of one indicates the presence of only baseline level of pollutants and values above one indicates progressive deterioration of the site and estuarine quality [30,46]. The pollution load index values of heavy metals in sediments are summarized in Figure 6. The PLI values were ranged from 0.11 to 0.24 with an average 0.178 during summer and 0.16 to 0.37 with an average 0.281 during winter confirming that the sediment of the studied river was contaminated (PLI <1). Comparing both season, PLI values of winter slightly higher than summer season, which was consistent with the metal concentration in sediment for both seasons. The PLI can provide some understanding to the inhabitants about the quality of the sediment [8]. In addition, it also provides valuable information about the pollution status of the study area that helps the decision-makers to take any decision [57]. For both seasons, higher pollution load index values were perceived in sampling sites L9 to L14, which might be due to the effects of various industries and urban activities at these sites.

Ecological risk assessment

A methodology was developed by Hakanson [28] to evaluate ecological risks for aquatic pollution control. The methodology was developed based on the assumption that the sensitivity of the aquatic system depends on its productivity. Hakanson [28] defines four categories of C^i_f, four categories of C_d, five categories of E^i_r, and four categories of PER, as shown in Table 5. The contamination factor (C^i_f) for individual metal and degree of contamination (C_d) are presented in Table 6. The assessment of integrated contamination degree of sediments is based on the degree of contamination (C_d). The ranges of C_d were 0.941–3.379 with an average 2.097, indicating low contamination of the sediment environment [13]. The results of potential ecological risk factor and the potential ecological risk index (PER) are presented in Table 7. The order of PER in sediments were in the following descending order of As > Cu > Cd > Ni > Pb > Cr and the potential ecological risk for single metal was low risk group. Combining the potential ecological risk index of individual metal (Table 7) with its grade classifications, each single metal showed low potential ecological risk. However, As in the present study showed higher ecological risk. Generally, treatment of agricultural land with arsenical pesticides and treating of wood using chromated copper arsenate are the main sources of As in sediment [53]. The potential ecological risk index (PER) in the sampling sites were 7.069–29.689, indicates low risk.

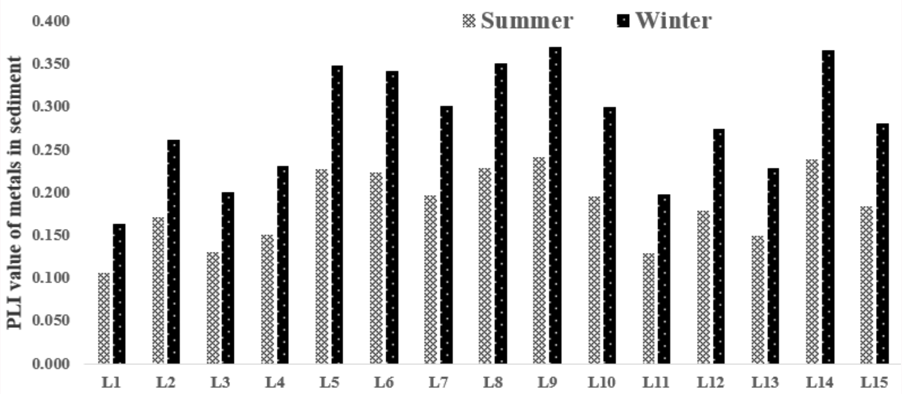

Figure 6: Pollution load index (PLI) of heavy metals in sediment collected from Louhajang River, Bangladesh.

Table 7: Potential ecological risk factors (E_r^i) and potential ecological risk indexes (PER) of heavy metals in sediments collected from Louhajang River, Bangladesh.

Sites	Season	Potential ecological risk factors (E_r^i)						Risk index (PER)	Pollution degree
		Cr	Ni	Cu	AS	Cd	Pb		
L1	Summer	0.164	0.336	1.37	5.28	3.09	0.105	10.345	Low risk
	Winter	0.296	0.54	1.92	6.86	5.25	0.155	15.021	Low risk
L2	Summer	0.178	0.48	2.005	9.72	2.25	0.595	15.228	Low risk
	Winter	0.322	0.768	2.805	12.64	3.81	0.865	21.21	Low risk
L3	Summer	0.13	0.456	1.23	8.61	1.86	0.395	12.681	Low risk
	Winter	0.234	0.726	1.725	11.19	3.15	0.57	17.595	Low risk
L4	Summer	0.242	0.336	1.37	9.72	1.02	0.49	13.178	Low risk
	Winter	0.438	1.338	1.495	12.63	1.74	0.715	18.356	Low risk
L5	Summer	0.518	0.462	2.845	9.8	2.19	0.85	16.665	Low risk
	Winter	0.934	0.744	3.985	12.75	3.72	1.235	23.368	Low risk
L6	Summer	0.506	0.486	3.12	10.65	1.53	0.895	17.187	Low risk
	Winter	0.912	0.774	4.37	13.85	2.61	1.3	23.816	Low risk
L7	Summer	0.352	0.504	2.695	12.13	1.11	0.815	17.606	Low risk
	Winter	0.634	0.81	3.775	15.77	1.86	1.18	24.029	Low risk
L8	Summer	0.682	0.558	1.985	9.03	1.17	1.63	15.055	Low risk
	Winter	1.226	0.9	2.78	11.74	1.98	2.36	20.986	Low risk
L9	Summer	0.408	1.584	3.275	5.99	1.32	1.08	13.657	Low risk
	Winter	0.734	2.532	4.585	7.79	2.22	1.565	19.426	Low risk
L10	Summer	0.204	2.712	2.98	3.14	1.11	0.875	11.021	Low risk
	Winter	0.368	4.338	4.17	4.09	1.92	1.265	16.151	Low risk
L11	Summer	0.168	0.786	1.425	2.72	1.35	0.62	7.069	Low risk
	Winter	0.302	1.26	2	3.53	2.28	0.895	10.267	Low risk
L12	Summer	0.164	2.646	3.125	3.7	1.08	0.545	11.26	Low risk
	Winter	0.296	4.23	4.375	4.81	1.86	0.79	16.361	Low risk
L13	Summer	0.23	0.474	1.92	7.39	4.32	0.15	14.484	Low risk
	Winter	0.416	0.756	2.685	9.61	7.35	0.215	21.032	Low risk
L14	Summer	0.25	0.672	2.805	13.61	3.15	0.835	21.322	Low risk
	Winter	0.45	1.074	3.925	17.69	5.34	1.21	29.689	Low risk
L15	Summer	0.182	0.636	1.725	12.06	2.58	0.55	17.733	Low risk
	Winter	0.328	1.014	2.415	15.67	4.41	0.8	24.637	Low risk

Toxic unit analysis

Potential acute toxicity of pollutants in sediment samples can be estimated as the sum of the toxic units, defined as the ratio of the determined concentration to probable effect levels (PELs) value [50]. Toxic unit (TU) and sum of toxic units (∑Tus) for heavy metals in surface sediments of Louhajang River are presented in Figure 7. Toxic

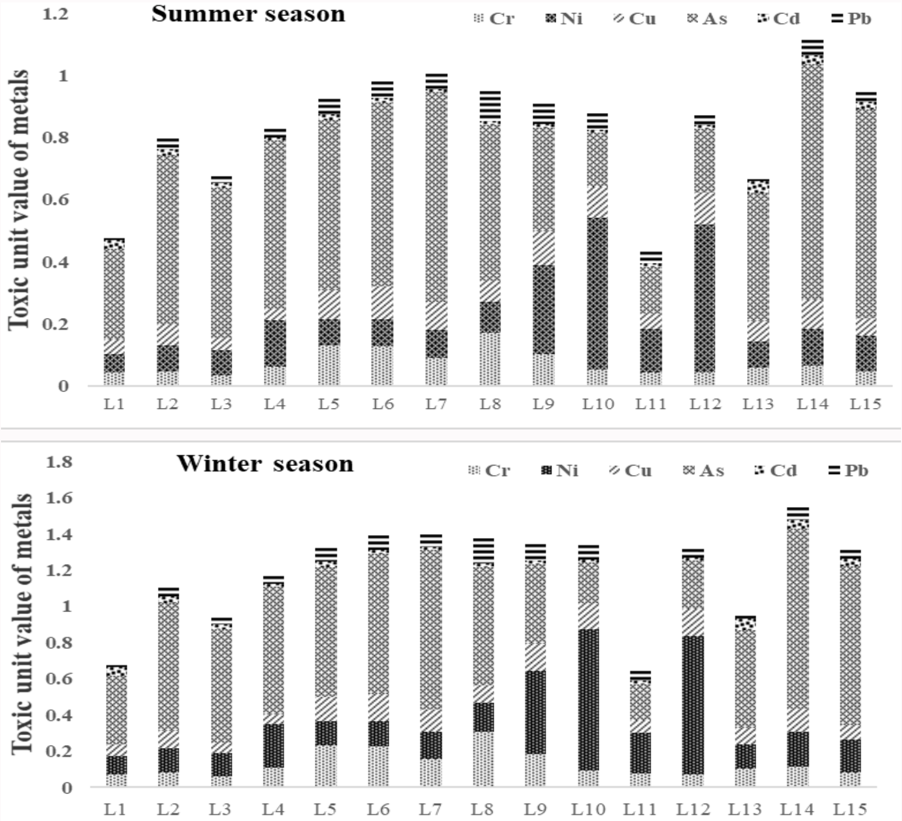

Figure 7: Toxic unit values of heavy metals in sediment collected from Louhajang River, Bangladesh.

units of heavy metals in the studied river decreased in the order of As > Ni > Cr > Cu > Pb > Cd. The mean toxic unit values of Cr, Ni, Cu, As, Cd, and Pb in sediments were 0.073, 0.164, 0.075, 0.460, 0.017, and 0.041 during summer, whereas they were 0.131, 0.262, 0.105, 0.598, 0.029, and 0.059 during winter season. The sum of toxic units at different sampling sites was less than 4, indicating a low toxicity of heavy metals to sediment-dwelling fauna in the study river [58]. The sum of toxic units for the studied heavy metals for sampling sites L7, and L14 was higher than the other sites, which was not in the similar trends of metal concentrations in sediments (Figure 7 and Table 2).

Conclusions

The study conclude that the average concentration of heavy metals in sediments of Louhajang River were in the decreasing order of Cu> Cr > As > Ni> Pb> Cd. The concentrations Cr, Ni, Cd, and Pb were lower than TRV, ASV, LEL, TEL, CUC, SEL, and PEL standard value indicating low contamination of these metals in sediment. Principal component analysis revealed a mixed source of anthropogenic inputs, particularly from industrial effluents and agricultural activities in the study area. On the basis of ecological risk, enrichment factor, contamination factor, geoaccumulation index, pollution load index showed lower contamination of sediment by the studied heavy metals. Louhajang River sediment showed low potential ecological risk for all the studied metals. This study suggested that more attention should be directed to the comprehensive risk assessment of heavy metals of this riverine aquatic environment.

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