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## Research Article

# Ecological Risk of the River Halda: A Perspective from Heavy Metal Assessment

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## Abstract

To evaluate the present status of heavy metals in the sediments of river Halda, seven heavy metals, viz. Cd, Cr, Cu, Fe, Mn, Pb, and Zn were assessed by Bangladesh Fisheries Research Institute by collecting data from 4 sampling locations (Khondokia Khal, Katakali, Madari Khal, and Madarsha) and ecological risk impending from these metals were depicted from the study. The concentration of heavy metals in the sediments of the river Halda ranged from 0.89-1.04 for Cd, 24.72-67.30 for Cr, 1.16-7.58 for Cu, 27899.31-60076.37 for Fe, 476.12-1137.20 for Mn, 0.77-8.33 for Pb and 40.33-121.77 mg kg<sup>-1</sup> for Zn, respectively. The sediment geo-accumulation index ( $I_{geo}$ ) values showed contamination only from two heavy metals Cd ( $1.10 \pm 0.16$ ) and Mn ( $1.48 \pm 0.37$ ). The average pollution load index (PLI) ( $0.37 \pm 0.10$ ) showed no marks of pollution in the studied sites, however; the mean degree of contamination factor (CF) showed moderate pollution. In the present study, the highest degree of contamination was observed at Katakali ( $3.79 \pm 2.07$ ) followed by three other sites. The overall degree of contamination of four sampling sites was  $2.68 \pm 1.84$  which indicated a low degree of contamination. The concentration of Cr ranged between 24.72-67.30 mg/kg with the highest ( $56.7 \pm 9.4$ , mg/kg) at Madarsha and the lowest ( $30.2 \pm 5.1$ , mg/kg) at Khondokia Khal. The highest concentration of Fe, Mn, and Zn ( $53926.3 \pm 5338.2$ ,  $980.9 \pm 145.1$ , and  $108.0 \pm 13.5$  mg/kg) exhibited similar results to Cu and Pb; the maximum levels were found at Madarsha and the minimum levels ( $33571.1 \pm 5456.0$ ,  $566.7 \pm 92.8$  and  $56.0 \pm 17.9$  mg/kg) at Khondokia Khal, respectively. The enrichment factor (EF) ( $0.08 \pm 0.02$  to  $4.16 \pm 0$ ) demonstrated none to moderate enrichment of the river. Nevertheless, the ecological risk factor ( $E_i$ ) ( $17.83 \pm 14.29$  to  $759.14 \pm 192.26$ ) and risk index (RI) ( $711.26 \pm 122.55$  to  $1272.04 \pm 175.19$ ) exposed low to serious ecological risk for the river Halda.

## Introduction

Widespread urbanization coupled with industrial development [1] is imposing a severe threat on the harmony of the aquatic environment through different types of pollutants. Of them, heavy metals are considered the most common environmental pollutants [2] and in recent years it has appeared as a great concern for the aquatic environment [3-9], especially in the developing countries where environmental quality maintenance and hygiene structure do not pace up with population growth and rapid urbanization [10]. Albeit, heavy metals (Cd, Co, Cr, Cu, Hg, Ni, Pb, Zn) are essential components of the environment [11,12], biological food chain, and important

for human health [13-18]; they can cause detrimental effects even at low concentrations. Furthermore, heavy metals are non-degradable and can bio-accumulate and bio-magnify in mussels, oysters, shrimps, and fish and can be transferred to humans via the food chain [19-32]. Recently, bioaccumulation and toxicity of heavy metals [33,34] have appeared as a global concern [35] due to having a negative impact on human health, fish, and invertebrates [23-39].

Fluvial natural water bodies are largely liable for the translocation of [40-42] heavy metals that enter the systems through terrestrial runoff, atmospheric deposition, sewage discharge, and others [43,44]. Heavy metals are poorly soluble

in water and are mostly scavenged by fine particles leading to their accumulation in sediments [45,46]. Thus, sediments become the main repository of heavy metals and other chemicals [46,47] and act as the indicator for water pollution in lakes [48] and rivers [49,50]. The sediment matrix provides the best natural hallmarks of recent environmental perturbations. With the change of environmental conditions viz. pH, oxidation-reduction potential ( $E_h$ ), salinity and organic matter, and heavy metals retained in sediments can be remobilized and dissolved into the water again, causing secondary contamination [51–53]. Therefore, the identification and quantification of heavy metals in water and sediments are important environmental issues [54]. Several studies about contamination of heavy metal in soils have been carried out all over the world [55–62].

Heavy metal and metalloid exposure has been increasing in recent years in Bangladesh [38,63,64] from different industries, domestic wastes, and agrochemicals that deteriorate water quality [37,65,66]. To address this issue, several authors have studied the heavy metals in different rivers of Bangladesh [2,10,67–72]. However, very few studies have been conducted to date to assess the heavy metal contamination of the Halda River. In Bangladesh, Halda is one of the most important rivers where Indian major carps shed their eggs naturally during the breeding season making this river a unique heritage of this country [73–76]. But the river is being polluted day by day by different natural and anthropogenic pollutants. Aquatic plants (microalgae and seaweeds) have the highest photosynthetic efficiency, are the highest biomass producers, are resistant to several pollutants, and have the ability to grow on land that is often inappropriate for other uses [77]. The cells of

aquatic plants, such as seaweeds, have bioactive compounds and contain functional groups, including carboxyl, hydroxyl, amino, and sulphate, that can act as metal-binding sites between the adsorbent and adsorbate [78]. The present study was conducted to evaluate the status of heavy metals, their probable ecological risk, and mitigation measures to confirm the unimpeded biodiversity of river Halda.

## Materials and methods

### Sampling sites

The present study was conducted in the Halda river which lies between  $22^{\circ} 25' 13''$ – $22^{\circ} 48' 51.37''$  N and  $91^{\circ} 45' 00''$ – $91^{\circ} 52' 33''$  E [79]. Four sampling points viz. Khondokia Khal, Katakhal, Madari Khal, and Madarsha of the river were selected for sediment sample collection. These “Khal” (local name for canals) are the main discharge routes carrying the pollution loadings towards the Halda river. Sampling and data collection were done on monthly basis for one year (from July 2020 to June, 201). The global positioning system (GPS) coordinates of the sampling sites are furnished in Figure 1 and Table 1.

### Sample collection, preparations and analysis

A total of 48 surface soil samples were collected for one year from the selected sampling locations of the Halda river in clean polythene covers circumventing all possible contamination. Samples were collected from the river bed with the help of an Ekman dredge (10–50 cm layer of the soil) and the location of each sample was recorded using a handheld GPS (Table 1). At each sampling site, three replicate samples were collected and

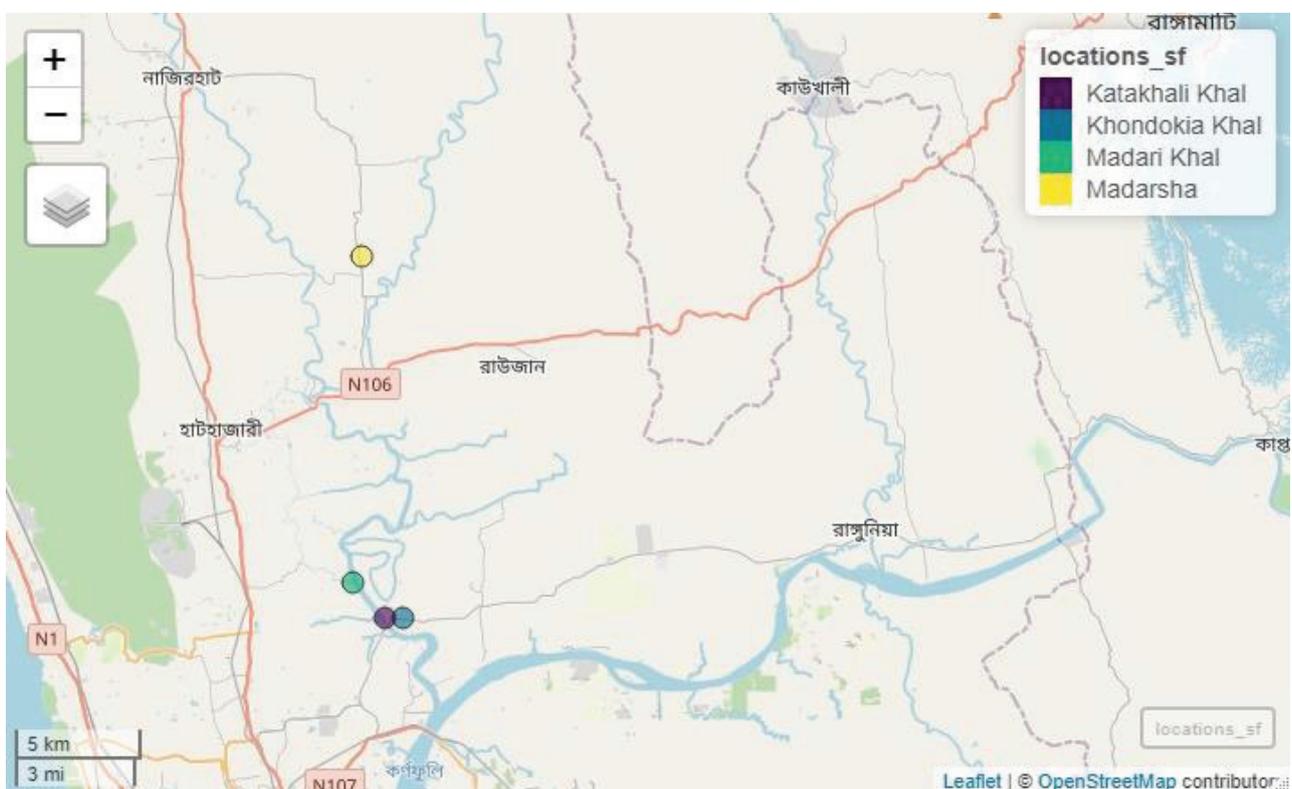


Figure 1: Map of the study area and the location of different sampling sites.



mixed to form a representative sample. Samples from the same locations were further mixed to form only four representative samples for four locations. Seven common heavy metals, viz. Cd, Cr, Cu, Fe, Mn, Pb, and Zn were selected for assessment. In the pretreatment phase, the soil samples were air-dried at room temperature. Plant roots, large stones, debris, organic residues, and visible intrusions present in the samples were removed carefully. Finally, the samples were crushed, ground, and passed through a 0.85 mm plastic sieve and stored at 4°C until the spectrophotometric reading was completed Table 2.

After completion of all pretreatment works, sediment samples were transferred to the soil and water analysis laboratory of the Institute of Water and Flood Management of Bangladesh University of Engineering and Technology (BUET) immediately for analysis. Here, 2 g is equivalent to 1L of the ICP run sample. Concentrations thus can be said as µg/2 g of sample or mg/2kg sample. Concentration is thus multiplied by 0.5 to get the mg/kg value.

Samples were handled carefully. Recommended clean powder-free latex gloves and lab coats were used during the analysis for eluding contamination. Glassware was properly cleaned with chromic acid solution and distilled water. Analytical grade chemicals and reagents were used to get better results. Blank determinations were used to get the correct instrumental readings. The intensity of heavy metal pollution in surface sediments of the river Halda was assessed using several different indices derived utilizing the metal concentration data.

### Sediment quality assessment

**Index of geo-accumulation:** The geo-accumulation index ( $I_{geo}$ ) reduces the interference of human factors in the assessment of soil contamination and is hosted to replace the traditional single factor Nemerow index [81].  $I_{geo}$  was first introduced by Müller [82] and it has been widely applied to sediment geochemistry to assess the degree of heavy metal contaminations in sediments. The  $I_{geo}$  is defined by the following equation:

$$I_{geo} = \log_2 (Cn/1.5 \times Bn) \quad (1)$$

where Cn is the content of elements in the sediment samples and Bn is the geochemical background concentration for the same elements (n). The background values of the studied elements used for the calculation of this index are the same as those used in the calculation of the contamination factors (CFs). Factor 1.5 is the background matrix correction factor due to lithological variations. The  $I_{geo}$  index contains seven classes [83,84] (Table 3).

As  $I_{geo}$  could reduce the effects of parent rocks and protruding artificial effects on soil heavy metal contamination, it is suitable for the evaluation of soil heavy metal contaminations in industrial and mining gathering areas. However, evaluation of  $I_{geo}$  for a single heavy metal contaminant, the index cannot provide a comprehensive description of the contamination status of the study area. Accordingly, an evaluation based on the comprehensive index method is necessary. Therefore, the

**Table 1:** GPS location of selected sampling points of the river Halda.

Places	GPS Point (Longitude and Latitude)
Madarsha	N 22°27'59" E 091°51'47"
Madari Khal	N 22°26'97" E 090°51'56"
Khondokia Khal	N 22°26'13" E 091°52'44"
Katakhal Khal	N 22°26'13" E 091°52'18"

**Table 2:** Wave length used in emission measurements and the instrumental detection limit for measurement by using ICP.

Elements	Wavelength (nm)	The instrumental detection limit (µg/l)
Cd	228.8	0.1
Cr	205.5	0.4
Cu	324.7	0.4
Fe	238.2	0.3
Mn	259.3	0.1
Pb	220.3	1.7
Zn	213.8	0.2

Source: Praveen Sarojam [80]. PerkinElmer, Inc. Shelton, CT 06484 USA.

**Table 3:** Index classification of sediment quality [85].

$I_{geo}$ Values	Class	Sediment quality
≤ 0	0	Unpolluted
0-1	1	Unpolluted to moderately polluted
1-2	2	Moderately polluted
2-3	3	Moderately to strongly polluted
3-4	4	Strongly polluted
4-5	5	Strongly to extremely polluted
≥6	6	Extremely polluted

traditional Nemerow index (IN) was improved by replacing the single factor index with  $I_{geo}$  (Table 4). The following Equation (3) was utilized:

$$IN = \sqrt{(I_{geomax}^2 + I_{geoave}^2)/2} \quad (2)$$

$$I_N = 1.86 \text{ (In the present study).}$$

### Contamination factor, degree of contamination and modified degree of contamination

The Contamination Factor (CF) and Degree of Contamination ( $C_d$ ) are used to assess the pollution load of the sediments with respect to heavy metals [54]. The CF is the ratio obtained by dividing the concentration of each metal in the sediment by the baseline or background value [84]. CF for each metal was determined by the following formula [86]:

$$CF = \frac{\text{Concentration of measured metal}}{\text{Background Concentration of the same metal}} \quad (3)$$

To facilitate pollution control, Hakanson [86] proposed a diagnostic tool named 'degree of contamination' ( $C_d$ ) and it is determined as the sum of the CF for each sample:

$$cd = \sum_{i=1}^n CF \quad (4)$$



The  $C_d$  is aimed at providing a measure of the degree of overall contamination in surface layers in a particular core or sampling site. Hakanson [86] has provided four grade ratings of sediments based on CF and  $C_d$  values (Table 5).

To calculate the degree of contamination, at least five sediment samples are required to provide a mean concentration and to compare with the background value. To avoid this constraint, a generalized index was developed [87]; named the modified degree of contamination (mCd) to assess the overall heavy metal contamination of soil (Table 6). The modified degree of contamination (mCd) was estimated using the following equation:

$$mCd = \frac{\sum_{i=1}^n CF}{n} \tag{5}$$

### Enrichment factor

Enrichment factor (EF) is a convenient method to evaluate the magnitude of anthropogenic heavy metal contaminants [88] in the environment [89]. The EF was calculated using the following equation:

$$EF = \frac{\left(\frac{cM}{cFe}\right)_{\text{sample}}}{\left(\frac{cM}{cFe}\right)_{\text{Earth's crust}}} \tag{6}$$

Where, the (CM/CFe) sample is the proportion of concentration of heavy metal (CM) and iron (CFe) in the sediment sample, and (CM/CFe) Earth's crust is the proportion of heavy metal and iron in the Earth's crust [90]. Iron (Fe) is used for the geochemical normalization to calculate the enrichment factor. Different values of EF, indicates different degrees of enrichment; where  $EF < 1$  = indicates no enrichment;  $EF < 3$  = minor enrichment;  $EF 3-5$  = moderate enrichment;  $EF 5-10$  = moderately severe enrichment;  $EF 10-25$  = severe enrichment;  $EF 25-50$  = very severe enrichment; and  $EF > 50$  = extremely severe enrichment [88,91].

### Pollution load index

Pollution Load Index (PLI) determines the communal effects of various pollutants in sampling sites deposited in soils and sediments [92]. The PLI for each site has been estimated by the multiplications of the  $n^{\text{th}}$  root of the studied heavy metals [93].

$$PLI = (CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n)^{\frac{1}{n}} \tag{7}$$

where CF is the contamination factor and n is the number of metals. The PLI of  $>1$  indicates pollution, whereas  $<1$  indicates no pollution [94]. This index provides a quick assessment for unskilled people to compare the pollution status of different places.

### Ecological risk factor and risk index

The  $E_r^i$  is widely used to assess the ecological risk of heavy

**Table 4:** Improved Nemerow Index [81].

IN Values	Class	Sediment quality
$0 < IN \leq 0.5$	0	Uncontaminated
$0.5 < IN \leq 1.0$	1	Uncontaminated to moderately contaminated
$1.0 < IN \leq 2.0$	2	Moderately contaminated
$2.0 < IN \leq 3.0$	3	Moderately to heavily contaminated
$3.0 < IN \leq 4.0$	4	Heavily contaminated
$4.0 < IN \leq 5.0$	5	Heavy to extremely contaminated
$IN > 5.0$	3	Extremely contaminated

**Table 5:** Sediment classes according to CF and  $C_d$  values [86].

CF/ $C_d$ Values	Class	Sediment quality
$CF > 1$	0	Low CF
$1 \leq CF < 3$	1	Moderate CF
$3 \leq CF < 6$	2	Considerable CF
$CF \geq 6$	3	Very high CF
$C_d < 6$		Low degree of contamination
$6 < C_d < 12$		Moderate degree of contamination
$12 < C_d < 24$		Considerable degree of contamination
$C_d > 24$		High degree of contamination

**Table 6:** Sediment classifications according to mCd [87].

mCd Values	Contamination situation
$mCd < 1.5$	Nil to very low degree of contamination
$1.5 < mCd < 2$	Low degree of contamination
$2 \leq mCd < 4$	Moderate degree of contamination
$4 \leq mCd < 8$	High degree of contamination
$8 \leq mCd < 16$	Very high degree of contamination
$16 \leq mCd < 32$	Extremely high degree of contamination
$mCd \geq 32$	Ultra-high degree of contamination

metals in sediments [95]. The index was calculated by the following equations [86]:

$$ER^i = T_r^i \times C^i \tag{8}$$

$$RI^i = \sum Er^i \tag{9}$$

Where  $E_r^i$  is the potential ecological risk factor for a given contaminant and  $T_r^i$  is the toxic response factor of each element, including Cd = 30, Cr = 2, Cu = 5, Fe = 2.82, Mn = 1, Pb = 1 and Zn = 1 [95-101]. The risk index (RI) is the sum of  $E_r^i$  and represents the potential toxicity response of various heavy metals in sediments. The  $E_r^i$  and RI values [98,101,102] are furnished in Table 7.

### Probable effects level

Probable effects levels (PEL) are guidelines widely accepted to evaluate bio-toxic risks of sediments. Since heavy metals always occur in sediments as complex mixtures, the mean PEL quotient (m-PEL-Q) method has been proposed and used to determine the possible biological effect of combined toxicant

groups by calculating the mean quotients for a range of heavy metals using the following formula:

$$PEL-Q = \frac{\sum_{i=1}^n \left( \frac{C_i}{PEL_i} \right)}{n} \quad (10)$$

Where  $C_i$  is the content of measured element  $i$ ,  $PEL_i$  is the PEL value of element  $i$ , and  $n$  is the number of elements. Several classes of toxicity probability [103] for biota are presented in (Table 8).

### Equipment used

Eskman dredge (10–50 cm layer of the soil), handheld GPS, 0.85 mm plastic sieve, spectrophotometer, and clean polythene covers.

### Statistical analysis

The data were compiled and processed initially using Microsoft Excel and further investigations were carried out using different statistical software packages. For example, the one-way analysis of variance (ANOVA) was performed by JMP (version 14) software to delineate whether any significant ( $P$

$< 0.05$ ) spatial variation exists in the concentration of heavy metals. Pearson's product-moment correlation matrix (Table 9) was obtained by using GraphPad Prism (version 6). Cluster analysis (CA) was performed to find out the similarity and variation with the influencing factors of studied heavy metals [104]. The dendrogram was prepared to show the similarity among heavy metals and to identify their sources of origin using Past software (version 4). The analytical output of the present study performed through different software is presented as charts and Tables.

## Results and discussion

The concentrations of heavy metals in the sediments of river Halda collected from four sampling locations are presented in Table 9. A one-way ANOVA followed by the Tukey-Kramer test was used to identify the significant differences among the mean concentration of different heavy metals and the results demonstrated significant ( $P < 0.05$ ) spatial differences. The concentration of heavy metals in the sediments of the river Halda found in the present study ranged from 0.89–1.04 for Cd, 24.72–67.30 for Cr, 1.16–7.58 for Cu, 27899.31–60076.37 for Fe, 476.12–1137.20 for Mn, 0.77–8.33 for Pb and 40.33–121.77 mg  $kg^{-1}$  for Zn. The chronological order of the heavy metals found in the present study was: Fe>Mn>Zn>Cr>Pb>Cu>Cd.

The findings in the present study revealed that the concentrations of all studied heavy metals in the sediment were above the permissible limit as set by WHO [105,106] USEPA [107], and DoE [108] (Table 10). In addition to comparing the data of the present study with some other global standards, the results were also compared with some previous works on the same river and another important river system in Bangladesh (Table 11). The results demonstrated that most all heavy metal concentrations were lower than Buriganga, Dhaleshwari, and Shitalakhya as these rivers are severely polluted by municipal and industrial effluents, sewerage, and other non-treated chemicals. However, the present status of the river Halda is not peasant as the condition of the river is gradually exacerbating compared to some recent investigations.

The highest mean concentration of Cd ( $1.0 \pm 0.1$ ) was found at Katakhal Khal, whereas; in the other three sampling locations, it was found beyond the detection limit. The amount of Cd found in the present study was above the acceptable global and country limit presented in Tables 10 and 11. The result was compared with some previous investigations carried out with the sediment samples of different important rivers of Bangladesh and varying results were found. The concentration of Cd in the river Halda was lower than that of the Buriganga

**Table 7:** Ecological risk factor ( $E_r$ ) and risk index (RI) for studied metals in the river Halda.

$E_r$ Values	Ecological Class
$<40$	Low ecological risk
$40 < E_r \leq 80$	Moderate ecological risk
$80 < E_r \leq 160$	Appreciable ecological risk
$160 < E_r \leq 320$	High ecological risk
$E_r > 320$	Serious ecological risk
RI Values	Ecological Class
$<150$	Low ecological risk
$150 \leq RI < 300$	Moderate ecological risk
$300 \leq RI < 600$	Considerable ecological risk
$RI > 600$	High ecological risk

**Table 8:** Probable effects level quotient and ecological classification of the river Halda.

PEL-Q Values	Ecological Class
PEL-Q $< 0.1$	8% probability of toxicity
PEL-Q = 0.11-1.5	21% probability of toxicity
PEL-Q = 1.51-2.3	49% probability of toxicity
PEL-Q $> 2.3$	73% probability of toxicity

**Table 9:** Heavy metal concentration in the sediments of the river Halda.

Sites	Metal Concentrations (mg/kg) (Mean±SD)						
	Cd	Cr	Cu	Fe	Mn	Pb	Zn
Khondokia Khal	*BDL	30.2±5.1 <sup>c</sup>	2.7±0.6 <sup>b</sup>	33571.1±5456.0 <sup>b</sup>	566.7±92.8 <sup>c</sup>	4.3±0.6 <sup>b</sup>	56.0±17.9 <sup>b</sup>
Katakhal Khal	1.0±0.1	38.7±7.0 <sup>bc</sup>	2.0±1.1 <sup>b</sup>	37804.2±2470.9 <sup>b</sup>	635.1±43.6 <sup>bc</sup>	1.4±0.9 <sup>c</sup>	67.5±15.8 <sup>b</sup>
Madari Khal	*BDL	47.3±1.3 <sup>ab</sup>	4.0±0.7 <sup>ab</sup>	48814.1±1760.4 <sup>a</sup>	853.8±76.7 <sup>ab</sup>	2.9±0.8 <sup>bc</sup>	84.9±8.9 <sup>ab</sup>
Madarshah	*BDL	56.7±9.4 <sup>a</sup>	6.4±1.3 <sup>a</sup>	53926.3±5338.2 <sup>a</sup>	980.9±145.1 <sup>a</sup>	7.6±0.9 <sup>a</sup>	108.0±13.5 <sup>a</sup>
*CRV	0.28 ± 0.03	107.0 ± 4.0	29.1 ± 1.1	-	-	29.0 ± 1.6	114.0 ± 6.0

\*BDL= Below Detection Limit; CRV = Certified Reference Values; Levels not connected by the same letters are significantly different.



[2,10,40], Dhaleshwari [110], Turag [112] and Shitalakhya river [114], but higher than Karnafuly [63,111], Meghna [113] and Brahmaputra river [70]. It was also higher than the result of previous studies [115] on heavy metal concentrations of the river Halda (Table 11). This might be associated with the collecting of samples from different sites than the present study. The results indicate that the heavy metal commination of the river Halda is gradually exasperating.

The concentration of Cr ranged between 24.72–67.30 mg/kg with the highest (56.7 ± 9.4, mg/kg) at Madarshah and the lowest (30.2 ± 5.1, mg/kg) at Khondokia Khal. The mean concentration exceeded the limit set by WHO [105,106], USEPA [107], and DoE [108] (Table 10). Mohiuddin, et al. [2] and Islam, et al. [114] found a higher concentration of Cr than the present study in the Buriganga and Shitalakhya rivers, respectively. On the contrary, Ahmed, et al. [110] in Dhaleshwari; Islam, et al. [101] and Ali, et al. [63] in Karnafuly; Banu, et al. [112] in Turag; Hassan, et al. [113] in Meghna; Bhuyan, et al. [70] in Brahmaputra and Bhuyan, et al. [79] in Halda found the lower concentration of Cr than the present study.

The concentrations of Cu and Pb ranged between (1.16–7.58 and 0.77–8.33 mg/kg) and the highest (6.4 ± 1.3 and 7.6 ± 0.9 mg/kg) concentrations of both of these metals were found at Madarshah and the lowest (2.0 ± 1.1 and 1.4 ± 0.9 mg/kg) at Katakhal Khal, respectively. The concentration of these two heavy metals was above the acceptable limit set by WHO [105]. Furthermore, these were also higher than the results obtained

by Ali, et al. [63], Banu, et al. [112], and Bhuyan, et al. [70] in the Karnafuly, Turag, and Brahmaputra rivers, respectively. But these were lower than Mohiuddin, et al. [2], Ahmed, et al. [110], Islam, et al. [114], and Bhuyan, et al. [115]; who studied the heavy metals of sediment samples of Buriganga, Dhaleshwari, Shitalakhya and Halda rivers, respectively. Nevertheless, the concentration of Pb in the present study was also lower than the amount found by Islam, et al. [111] and Hassan, et al. [113] in the Karnafuly and Meghna rivers, respectively Table 12.

The highest concentration of Fe, Mn, and Zn (53926.3 ± 5338.2, 980.9 ± 145.1, and 108.0 ± 13.5 mg/kg) exhibited similar results to Cu and Pb; the maximum levels were found at Madarshah and the minimum levels (33571.1 ± 5456.0, 566.7 ± 92.8 and 56.0 ± 17.9 mg/kg) at Khondokia Khal, respectively. The concentration of Mn found in the present study (759.0 mg/kg) was substantially lower than the Buriganga (4036 mg/kg) river but it was higher than the level found in some other important rivers in Bangladesh by Islam, et al. [108], Hassan, et al. [113], Bhuyan, et al. [45,115]. On the contrary, the concentration of Zn found in the present study (max 108.0 ± 13.5 mg/kg) was lower than Buriganga [2,10,40] and Shitalakhya [114] but higher than Karnafuly [111], Meghna [113] and Brahmaputra [45] rivers, respectively.

A higher concentration of these metals might be the outcome of discharges from textile and paint industries and domestic sewage waste [10,40,109,113,116,117,118]. Bhuyan and Baker [115] also reported the seasonal fluctuations in the level of the heavy metals in the river Halda.

The  $I_{geo}$  values have been presented in Table 12. In all sampling sites, the  $I_{geo}$  values of all studied heavy metals except Mn configured negative values after calculation, indicating that these sites were not polluted by the heavy metals but Mn (Table 12). The overall  $I_{geo}$  values of all studied heavy metals ranged from -6.01 to 1.86. Muller [85] classification of sediment quality disclosed that all sites were moderately polluted due to contamination with Mn and the orderly arrangements of the sites on the basis of the concentration of metals stand Madarshah>Madarikhal>Katakhal>Khondokia. Mohiuddin, et al. [2] studied the  $I_{geo}$  values for Mn for 11 locations in the Buriganga river and found the values >1.0, indicating moderately polluted sediment quality. Islam, et al. [72] found higher  $I_{geo}$  values for Cd and extremely contaminated sediment quality in Turag, Buriganga, and Shitalakhya. Hasan, et al. [113] studied the sediment quality of the Meghna river and found positive values for Cd, Pb, Ni and Zn indicating unpolluted to moderately polluted sediment. In the Karnafuly river in Bangladesh,  $I_{geo}$  values for As, and Cr exposed unpolluted to extremely polluted status [63]. In the Bortala river in China, the  $I_{geo}$  values of Ni,

**Table 10:** Global standards of different heavy metals compared to the present study.

Global Standards	Heavy Metals					
	Cd	Cr	Cu	Mn	Pb	Zn
WHO (1993)	0.003	0.05	2.00	0.50	0.01	3.00
WHO (2004)	-	0.05	2.00	0.40	-	-
USEPA (2008)	0.005	0.10	-	0.05	-	5.00
DoE (1997) standard	0.005	0.05	-	0.10	0.05	5.00
Present Study	1.0	43.22	3.77	759.0	4.05	79.10

**Table 11:** The concentration of heavy metals in some others rivers in Bangladesh (mg/kg).

Rivers	Heavy Metals						References
	Cd	Cr	Cu	Mn	Pb	Zn	
Buriganga	3.33	177.50	344.20	4036	79.80	502.30	[2,40,109]
Dhaleshwari	2.08	27.39	37.45	-	15.79	-	[110]
Karnofuly	0.24	0.76	1.22	15.30	4.96	16.30	[111]
Turag	1.40	0.44	1.57	-	1.64	1.08	[112]
Meghna	0.23	31.74	-	442.60	9.47	79.02	[113]
Shitalakhya	5.01	74.82	143.70	-	28.36	200.60	[114]
Halda	0.04	8.84	5.90	139.50	8.80	79.58	[115]
Brahmaputra	0.001	0.01	0.12	1.44	0.11	0.01	[70]
Present Study	1.00	43.22	3.77	759.0	4.05	79.10	

**Table 12:** Geo-accumulation indices ( $I_{geo}$ ) of heavy metals for sediments of all studied sites in the river Halda.

Stations	Geo-accumulation indices ( $I_{geo}$ )							Improved Nemerow Index ( $I_N$ )
	Cd	Cr	Cu	Fe	Mn	Pb	Zn	
Khondokia	*BDL	-2.33±0.25	-4.80±0.30	-1.09±0.24	1.10±0.24	-2.86±1.62	-2.00±0.45	1.60
Katakhal	0.73±0.64	-1.97±0.27	-5.35±0.76	-0.91±0.10	1.26±0.10	-3.10±2.74	-1.70±0.32	1.50
Madarikhal	*BDL	-1.67±0.04	-4.23±0.25	-0.54±0.10	1.69±0.13	-3.41±0.37	-1.35±0.16	1.60
Madarshah	*BDL	-1.42±0.23	-3.58±0.31	-0.40±0.14	1.89±0.21	-1.99±0.18	-1.01±0.18	1.53
Mean±SD	1.10±0.16	-1.84±0.40	-4.49±0.40	-0.73±0.32	1.48±0.37	-2.58±1.54	-1.52±0.47	1.56±0.05



Zn, Cr, As, and Cu indicated no pollution [119]. On the contrary, Malvandi [120] found higher  $I_{geo}$  values for As and Se and sediment class ranged from unpolluted to extremely polluted in the Zarrin-Gol River, Iran. Rahman, et al. [121] and Hassan, et al. [113] have opined that a higher concentration of Al and Mn originates lithogenically and is associated with spinning mills and paint industries wastes. The higher  $I_{geo}$  values as a result of the increased concentration of Mn found in the river Halda might be the result of similar lithogenic and anthropogenic effects.

To quantify the soil heavy metal contamination in the study area, the improved Nemerow index (IN), which depicts the combined effects of all heavy metals were assessed. The  $I_N$  ranged from 1.50 to 1.60 in all four sampling sites which indicates the study area is moderately contaminated and these results also comply with the results of  $I_{geo}$ . Akbor, et al. [122] found severe contamination in some sampling sites of the river Buriganga in Bangladesh in terms of  $I_N$ . Guan, et al. [79] also found extreme contamination in all sampling sites in the mining gathering area in Tianjin, China. The reasons for getting higher values in their studies are quite obvious, they conducted their studies in heavily industrialized areas and the anthropogenic load was much higher in those areas compared to the present study.

Table 13 shows the results of contamination factors (CF) and pollution load index (PLI), degree of contamination, and modified degree of contamination ( $mC_d$ ) of heavy metals in sediment samples collected from the river Halda. The overall CF value for Cadmium (Cd) was  $>3.0$  indicating considerable contamination of this metal, however; the CF values for all other heavy metals exhibited "low contamination". The CF values found in the present study in the river Halda was lowered compared to the values found in the river Meghna [113] and Buriganga [10,40,69]. The river Buriganga is polluted by the thousands of industrial and sewerage lines that dispose of huge volumes of toxic wastes into the river [123] every day. The Meghna river is also polluting different sites from industries that are situated on the banks of this river or very close to the river system. The dominant industries in this area are shipyard, cement, paper, jute, super board, oil, sugar, food processing, salt, and chemical industries. The river receives wastewater directly from these industries and also domestic and agro-chemical wastes contribute to heavy metal pollution in water and sediment [110]. In many other investigations around the world where the CF has been calculated, higher values of CF values were found. For example, CF value from 1.3 to 5.5 was found in the Balok river [96]; 0.14 to 6.08 was found in the Dikrong river [124]; 0.44 to 2.47 was found in the Yauri river [125] and 1.1 to 14.6 was found in the Tamaki estuary [87].

The PLI represents the number of times by which the metal content in the sediment exceeds the background concentration and gives a summative indication of the overall level of heavy metal toxicity in a particular sample [43]. The PLI of all sampling sites presented in Table 13 was calculated according to Tomlinson, et al. [93] and the values ranged from 0.29 to 0.51 with the overall value for all four sampling sites ( $0.37 \pm 0.10$ ) considered to be unpolluted. Individual PLI of all sites was also  $<1$  that must be classified as unpolluted. The order of PLI of four sampling sites from higher to lower was Khondokia  $>$  Katakhal  $>$  Madarikhah  $>$  Madarshah. The PLI values found in the present study in the river Halda were lower than in some previous studies, for instance; Ali, et al. [63] found higher PLI values in the Karnafuly river. Mohiuddin, et al. [126] reported PLI values of 4.9–24.2 and 5.2–27.4 in summer and winter samples of the Buriganga river which was manifold higher than the present study. Ahmed, et al. [110] stated that 100% of sampling points of the Buriganga river had  $PLI > 1$ , which indicated a polluted condition. In another study, Islam, et al. [127] also reported similar results. Furthermore, Abdullah, et al. [96] and Varol [84] found higher PLI values than the present study in the Balok and Tigris rivers, respectively. The reasons for these higher PLI values might be associated with the direct disposal of untreated effluents in the river from different industrial and agro-chemical sources.

In the present study, the highest degree of contamination was observed at Katakhal ( $3.79 \pm 2.07$ ) followed by three other sites. The overall degree of contamination of four sampling sites was  $2.68 \pm 1.84$  which indicated a low degree of contamination. Similarly,  $mC_d$  for the seven analyzed elements was found  $<1.5$  in the present study, indicating nil to a lower degree of contaminations. Both of these parameters ( $C_d$  and  $mC_d$ ) exhibited a lower range of values in the present study compared to some previous investigations at home and abroad. Sikder, et al. [69] and Akbor, et al. [122] found a higher degree of contamination than the present study in the Buriganga river. Sivakumar, et al. [128] in Tamil Nadu, India, and Abraham and Parker [87] in New Zealand found higher  $mCd$  values than the present study. However, they conducted their study in the coastal areas where all suspended and dissolved solids are washed out by the river and depositions are higher. Considering the average  $mCd$ , it should be noted that compared to geo-chemical background values, cadmium is highly enriched than other elements in the sampling sites. This localized enrichment is considered to be linked with the application of phosphate fertilizers to arable soils. Acidification of soils and lakes may also result in enhanced mobilization of cadmium from soil and sediments [129].

**Table 13:** Metal contamination factors (CF) and pollution load index (PLI), degree of contamination, and modified degree of contamination ( $mC_d$ ) in the sediment of the river Halda.

Stations	Contamination Factor							PLI	Degree of Contamination	$mC_d$
	Cd	Cr	Cu	Fe	Mn	Pb	Zn			
Khondokia	*BDL	0.30±0.05	0.05±0.01	0.71±0.12	0.63±0.10	0.14±0.13	0.39±0.12	0.29	1.52±0.30	0.22±0.04
Katakhal	2.14±1.87	0.39±0.07	0.04±0.02	0.80±0.05	0.71±0.05	0.05±0.05	0.47±0.11	0.34	3.79±2.07	0.54±0.30
Madarikhah	*BDL	0.47±0.01	0.08±0.01	1.03±0.04	0.95±0.09	0.14±0.04	0.59±0.06	0.35	2.24±0.15	0.32±0.02
Madarshah	*BDL	0.57±0.09	0.13±0.03	1.14±0.11	1.09±0.16	0.38±0.05	0.75±0.09	0.51	2.92±0.31	0.42±0.04
Overall (Mean±SD)	3.33±0	0.43±0.11	0.06±0.05	0.92±0.20	0.74±0.38	0.15±0.17	0.46±0.31	0.37±0.10	2.68±1.84	0.37±0.18



The values of enrichment factor (EF) of studied heavy metals found in the sediments of the river Halda have been furnished in Table 14. The results showed that the mean EF values for cadmium (Cd) were >4, suggesting a moderate enrichment of the river. However, the EF values for other metals studied at all sites showed “minor enrichment” (Table 14). In some previous investigations where the EF has been calculated, for instance; the mean EF values of Cr, Ni, and Zn in the Luanhe river and the mean EF values of As, Ni, and Cu in the Bortala river [130] were >1.5. Abdullah, et al. [96] and Varol [84] opined that the heavy metals resulting in higher EF entirely originate from the natural processes or crustal material. This might affect the EF values of the present study as well.

To assess the ecological risk of the studied elements to the river Halda, the potential ecological risk indices ( $E_r^i$  and RI), were measured and are summarized in Table 15. The order of potential ecological risk factor ( $E_r^i$ ) of heavy metals in sediments of the river Halda was Mn > Cr > Zn > Cu > Pb > Cd. Except for Fe, the mean Mn concentration of four sites poses serious ecological risk whereas Cr and Zn pose appreciable to moderate ecological risk, respectively. The mean potential ecological risk coefficient of Cd, Cu, and Pb was lower than 40, which belongs to low ecological risk. Also, the values of RI at all sites were >600 which indicated high ecological risk. In brief, the  $E_r^i$  and RI indices for the studied elements in the surface sediment of the river Halda pose a potential ecological risk. Rahman, et al. [131] found lower  $E_r^i$  and RI indices in an adjacent area of Dhaka Export Processing Zones than in the present study. The reasons might be that they conducted their study in a floodplain area

and in a river located at Savar Upazila which receives lower content of municipal effluent surges than the capital Dhaka. But in another study, Islam, et al. [132] found significantly higher  $E_r^i$  and RI indices in the Burignaga river than in the river Halda. Malvandi, et al. [120] found lower indices values in the Zarrin-Gol River in Iran. Solaiman, et al. [98] found higher  $E_r^i$  for Cd in Egypt whereas other studied heavy metals exhibited lower values than the present study. Sivakumar, et al. [128] found lower RI than the present study. The reasons behind this discrepancy of  $E_r^i$  and RI indices with local and international studies might be associated with the receiving of different types of contaminants from different anthropogenic sources where variation among metallic elements normally exists.

The mean Probable effects level (PEL) was calculated for the four sampling sites based on the metals Cd, Cr, Cu, Fe, Mn, Pb, and Zn to evaluate the potential risk to aquatic lives [103]. The PEL ranged from 0.04 to 1.09, and the mean probable effects level quotient (PEL-Q) ranged from 0.26 to 0.45 with an overall value of  $0.35 \pm 0.08$  (Table 16). The results indicated that the combination of heavy metals may have a 21 % probability of being toxic (Table 8). Probable effects level (PEL) and effects level quotient (PEL-Q) of heavy metals of any native river were not reported in the literature, therefore; it was not possible to compare the results of the present study with the previous ones. However, Li, et al. [133] found a similar probability of toxicity on the Weihai coast, China. Soliman, et al. [98] also found 30% probability of being toxic on the Mediterranean coast, Egypt.

Heavy metals in sediments normally originate from

**Table 14:** The values of enrichment factor (EF) of studied heavy metals for sediments in the river Halda.

Stations	Enrichment Factors (EF)						
	Cd	Cr	Cu	Fe	Mn	Pb	Zn
Khondokia	*BDL	0.42±0.01	0.08±0.00	1.00±0.00	0.89±0.02	0.21±0.20	0.54±0.09
Katakhal	4.17±0.28	0.48±0.06	0.05±0.03	1.00±0.00	0.88±0.01	0.06±0.06	0.58±0.10
Madarikhal	*BDL	0.46±0.01	0.08±0.02	1.00±0.00	0.92±0.05	0.14±0.04	0.57±0.04
Madarshah	*BDL	0.49±0.03	0.11±0.02	1.00±0.00	0.95±0.06	0.34±0.07	0.66±0.05
Mean±SD	4.16±0	0.47±0.03	0.08±0.02	1.0±0.0	0.91±0.03	0.22±0.12	0.59±0.05

**Table 15:** Potential ecological risk factors ( $E_r^i$ ) and risk index (RI) for studied heavy metals in the river Halda.

Stations	$E_r^i$							RI	Risk grade
	Cd	Cr	Cu	Fe	Mn	Pb	Zn		
Khondokia	*BDL	60.47±10.23	13.68±2.87	94670.37±15385.84	566.68±92.83	14.47±12.71	55.97±17.90	711.26±122.55	Low
Katakhal	19.30±16.87	77.46±14.08	10.12±5.57	106607.96±6968.00	635.13±43.61	4.57±4.96	67.46±15.84	814.03±85.90	Low
Madarikhal	*BDL	94.53±2.54	20.23±3.40	137655.67±4964.23	853.85±76.66	14.45±3.89	84.93±8.91	1068.00±84.18	Low
Madarshah	*BDL	113.48±18.73	31.83±6.54	152072.06±15053.86	980.91±145.10	37.85±4.70	107.96±13.52	1272.04±175.19	Low
Mean±SD	5.79±12.25	80.43±18.68	16.48±7.83	116121.54±22269.59	712.20±161.01	14.21±12.63	71.98±18.72	901.10±206.10	Low
$E_r^i$ grade									

**Table 16:** Probable effects level and effects level quotient of heavy metals in the river Halda.

Stations	PEL							PEL-Q
	Cd	Cr	Cu	Fe	Mn	Pb	Zn	
Khondokia	0.00±0.00	0.19±0.03	0.03±0.01	0.84±0.14	0.52±0.08	0.03±0.02	0.21±0.07	0.26±0.05
Katakhal	0.15±0.13	0.24±0.04	0.02±0.01	0.95±0.06	0.58±0.04	0.01±0.01	0.25±0.06	0.31±0.05
Madarikhal	0.00±0.00	0.30±0.01	0.04±0.01	1.22±0.04	0.89±0.13	0.03±0.01	0.31±0.03	0.40±0.02
Madarshah	0.00±0.00	0.35±0.06	0.06±0.01	1.35±0.13	0.89±0.13	0.07±0.01	0.40±0.05	0.45±0.05
Mean±SD	0.23±0.03	0.27±0.07	0.04±0.02	1.09±0.23	0.72±0.20	0.04±0.02	0.29±0.09	0.35±0.08



different natural and anthropogenic sources [134]. It is well established that organic matter and grain size are two main factors influencing the heavy metal regimes in the sediments [135]. The connotation among metals in sediment affords crucial information on sources and pathways of heavy metals in the aquatic milieu. The result of correlations between heavy metals conceded with the results of PCA and CA endorsed some new relations between parameters, viz. strong, moderately strong, and very strong, indicating that their sources of origin are similar, especially from industrial effluents, municipal wastes, and agricultural inputs. With this view, a correlation matrix was applied to discover relationships among studied elements and to determine a possible common metal source in the river Halda. According to the Pearson correlation matrix, (95% confidence level,  $P=0.05$ ), a significant correlation was found among some metals studied (Table 17). Cr displayed close relationships with Cu, Fe, Mn, and Zn. Similarly, Cu showed a close relation with Fe and Mn. Iron showed a close relationship between Mn, Pb, and Zn. Mn showed a close relationship with Zn and Pb showed a close relationship with Zn proposing a common source of these metals. These highly significant positive correlations between heavy metals suggest the possibility of common sources of origin which may be anthropogenic [136]. On the other hand, the rest of the elemental pairs showed no significant correlation with each other which could be an indication of separate source input or sources of

**Table 18:** Factor loadings on elements in sediments from the river Halda ( $n = 12$ ).

Element	PC1	PC2
Cd	0.979	-0.464
Cr	0.966	-0.074
Cu	0.920	0.133
Mn	0.981	-0.046
Pb	0.532	0.881
Zn	0.960	-0.021
Eigen value	4.911	1.018
% variance explained	82	17
Cumulative % variance	81.86	98.84

pollution. In contrast, no positive correlations were observed between Cd and other metals, suggesting that Cd pollution might be from a different source than other metals. Bhuyan and Bakar [79] and Hossain, et al. [137] also found a similar type of association between different heavy metals in the river Halda. Hassan, et al. [113] and Akbor, et al. [122] studied the metal-to-metal correlation of the Buriganga river and found a positive correlation among most of the metals except for very few metals with no significant correlation Figure 2.

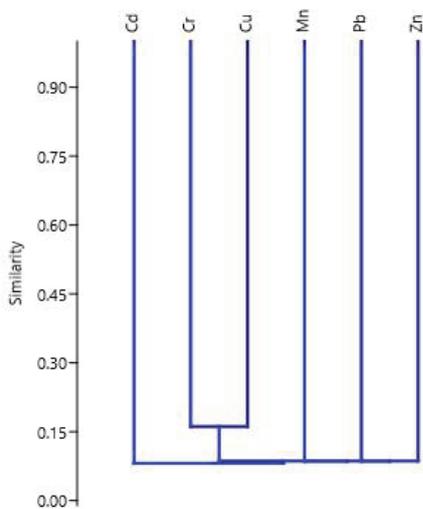
The principal component analysis (PCA) disclosed that all heavy metals could be grouped into two components (Eigen values >1) which explains 98.84% of the total variance (Table 18). The first component (PC1) accounted for about 82% of the total variance and exhibited high loading values for Cd, Cr, Cu, Mn, and Zn, whereas the second component (PC2) accounted for 17% of the total variance and exhibited high loading value only for Pb. Together with Pearson's Correlation Matrix (significant positive correlations between pairs of variables), we speculated that elements with loading values in the PC1 (Cd, Cr, Cu, Mn, and Zn) were from the lithogenic (natural) origin [138]. Probable sources are industrial discharges, municipal waste, household garbage, and urban runoff [122]. On the contrary, in the PC2, Pb presented a high loading value also implying its natural or anthropogenic origin. Pb mainly transfers as a reducible element in surface sediments and is strongly allied with Fe and Mn oxides acting as the natural accumulators of the metal in sediments [139]. Bhuyan and Bakar [115] found almost similar results in the river Halda with the existence of two principal components. On the other hand, Bhuyan, et al. [79] found three principal components in the river Halda, and Akbor, et al. [122] found four principal components in the river Buriganga while analyzing the heavy metals of sediment samples from the respective rivers. Soliman, et al. [98] also found two principal components while analyzing the heavy metals in sediments from the Mediterranean coast, Egypt, and Li, et al. [133] found three components in the Weihai coast, China. The difference that was visible in terms of the nos. of components and loading values of different elements might be due to spatial variations and procedures applied for heavy metal detection and analysis.

### Conclusion and recommendations

The results of this investigation disclose vital information about metal contamination in sediments of the river Halda. The

**Table 17:** Pearson's Correlation Matrix of heavy metals of Halda river.

	Cd	Cr	Cu	Fe	Mn	Pb	Zn
Cd							
Cr	-0.019						
Cu	-0.578*	0.757*					
Fe	-0.218	0.966*	0.814*				
Mn	-0.239	0.961*	0.847*	0.984*			
Pb	-0.408	0.544	0.733*	0.575*	0.594*		
Zn	-0.092	0.939*	0.738*	0.944*	0.918*	0.649*	



**Figure 2:** Bray-Curtis similarity index of heavy metals found in the sediments of the river Halda.



distribution order of heavy metal concentration in sediments was Fe>Mn>Zn >Cr>Pb>Cu and >Cd (mg/kg), respectively and the heavy metals were above the certified reference values set by WHO [106] and USEPA [140]. Albeit, the seven elements showed different distribution characters, lithogenic or anthropogenic contributions were the main sources of these metals. The adverse impact of heavy metals on aquatic biota was assessed using different indices. The average  $I_{geo}$  values displayed pollution only from two heavy metals Cd ( $1.10 \pm 0.16$ ) and Mn ( $1.48 \pm 0.37$ ). The average PLI ( $0.37 \pm 0.10$ ) exhibited no marks of pollution among the studied sites, however; the average CF values showed a moderate degree of pollution. The average EF ( $0.08 \pm 0.02$  to  $4.16 \pm 0$ ) demonstrated none to moderate enrichment of the river. Nevertheless, average  $E_r^i$  ( $17.83 \pm 14.29$  to  $759.14 \pm 192.26$ ) and RI ( $711.26 \pm 122.55$  to  $1272.04 \pm 175.19$ ) exposed low to serious ecological risk for the river Halda.

Multivariate statistical analysis demonstrated significant correlations between the studied heavy metals indicating similar sources and/or similar lithogenic/anthropogenic processes regulating the occurrence of these metals. In light of the findings of the present study, the following recommendations can be made:

- No industrial effluents should be allowed to dispose of in the river Halda without prior treatment
- No mining and dumping sites should be allowed on the bank of the river Halda
- Canals carrying municipal sewerage discharge to the river Halda should be stopped permanently
- The construction of a dam on the upper stretch of river Halda for water management and irrigation should not be allowed
- No project should be allowed to collect water from the river Halda for municipal use
- No mechanized boats should be allowed to run through the river Halda
- The natural navigation route of the river Halda should be maintained
- The river should be allowed only for research purposes and further research should be conducted on this aspect to know the future status and trends of heavy metals

Halda is an important river in Bangladesh. This study will support understanding of the present status of metal pollution in the river Halda and could be used as a useful tool for the academicians, researchers, and authorities of the Govt. of Bangladesh to formulate future management strategies to conserve and restore the river as it is considered as the only natural breeding ground of Indian major carps and declared as the Bangabandhu Fisheries Heritage.

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