

Assessment of heavy metal contamination in sediment at the newly established tannery industrial Estate in Bangladesh: A case study

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ABSTRACT

This research work was focused to evaluate the concentration of heavy metals (*i.e.*, Cr, Pb, Mn, Cu and Fe) in sediment samples collected from the Dhaleshwari River near the area of a newly established tannery industrial estate, Savar, Bangladesh. The heavy metals in sediment samples were ascertained by the atomic absorption spectrophotometer (AAS). This study revealed that the metal concentration for chromium (Cr), lead (Pb), manganese (Mn), copper (Cu) and iron (Fe) were found to be 14.8–748 (186 ± 241) mg/kg, 2.38–21.1 (8.78 ± 6.15) mg/kg, 1.59–6.29 (3.12 ± 1.38) mg/kg, 0.36–4.75 (1.76 ± 1.47) mg/kg and 3.87–154 (42.7 ± 49.1) mg/kg (dry wt) respectively. The concentration of heavy metals in sediment samples descends with the following order: Cr > Fe > Pb > Mn > Cu. Sediment contamination was assessed on the basis of geo-accumulation index (I_{geo}), contamination factor (CF) and pollution load index (PLI). The average I_{geo} values for Cr, Pb, Mn, Cu and Fe were found to be (-0.44 ± 1.87), (-2.18 ± 1.20), (-8.80 ± 0.63), (-5.80 ± 1.36) and (1.59 ± 1.87) respectively. The I_{geo} values of Cr and Fe for some sampling stations were found to be higher. The PLI value for all the sample stations (S1–S8) were 0.58, 0.66, 0.71, 0.69, 0.88, 0.86, 0.83 and 0.83 correspondingly. The CF values for Cr and Fe in some stations was observed at moderate, considerate and high contamination level. The spatial distribution of heavy metals in the study area was presented by interpolation technique. The results of spatial distribution pattern showed that the high concentrated zones of Cr was found at the dumping zone (~S5) of the leather industrial city. On the other hand, the spatial distribution pattern for the studied metals (Pb, Mn, Cu and Fe) showed that the maximum metals concentrations were found in the southeastern part, which was nearby effluent dumping zone. The ecological risk assessment in sediment samples revealed that there is no significant risk observed by the metal(*oid*)s at this moment. Considering Cr concentration, the modified hazard quotient (mHQ) showed that about 75% of the samples were low to severely polluted by Cr, while 25% of the samples were extremely polluted by Cr as well as $mHQ \leq 3.5$. Based on PMF analysis, two potential sources of heavy metals were identified in the study region: first, biochemical and leather tanning industries (Cr); second, the mixed effect of geogenic sources and atmospheric deposition and traffic emission (Fe and Pb).

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1. Introduction

In the industrial area, waste management is one of the difficult and most challenging issue. Sustainable solution is the best fit for dealing with the waste management. Many developed nations dump the waste effluents of their industrial area into the river after refining them. Unplanned dumping in river caused vital damage to the ecosystem of the surrounding [1]. These wastes from the industries is not only polluting the water but also the sediment in the adjunct river and ultimately affecting the aquatic life. Industrial effluents released on the land as well as dumped into the surface water, which ultimately lead to contamination due to accumulation of toxic metallic components and resulted in a series of well-documented problems in living beings, because they cannot be completely degraded [2]. Hence, industrial effluents offer a wide scope of environmental problems, and health hazards are becoming more complex and critical. Particular emphasis is placed on the status of metals in effluents of tannery industries. To keep this view in mind that tannery industry is a major source of pollution and contributors of metals to the environment. The tannery industries use a variety of chemicals in the tanning process, which are recognized as a major contributor of heavy metals to the environment and poses serious environmental threats. The effluents discharged from tannery industry contain a bulk amount of liquid and solid wastes with substantial quantities of toxic chemicals and heavy metals such as Cr, Cu, Fe, Mn and Pb [3]. All the waste materials of the whole industrial area is dumped into an open landfill and after treating them partially these are released into the river. Due to some internal problems, some of the components of the CETP are not constructed yet. As a result, the effluents are not properly refined, which might be affecting the adjacent area of the dumping station and the river. Areas near landfill have a greater possibility of soil contamination because of the potential pollution source of leachate originating from the nearby sites.

The discharge of effluent is a long term problem in the leather industry of Bangladesh [4]. The increased number of tanneries in Bangladesh might be causing environmental hazards as the effluents used in the tanning process are released into the water resources. Some heavy metals are essential in trace amounts, namely Cu, Fe, Mn, Cr, Cd, Pb etc. But higher concentration of these metals in the ecosystems may lead to an excessive accumulation of metals, becoming toxic to soil and possible danger to human health problem [5–7]. Higher level of metals may frequently react with biological systems by losing one or more electrons and forming metal cations, which have affinity to the nucleophilic sites of vital macromolecules. Several acute and chronic toxic effects of heavy metals affect different body organs. Gastrointestinal and kidney dysfunction, nervous system disorders, skin lesions, vascular damage, immune system dysfunction, birth defects, and cancer are examples of the complications of heavy metals toxic effects. Simultaneous exposure to two or more metals may have cumulative effects [8,9]. A number of cases of health problems related to environmental Cr-poisoning and elevated levels of Pb in the blood of infants have been reported [10]. Pollutants from leather industries discharges into river leach into soil increasing the accumulation of metals in sediments, biota and ultimately humans [11]. Some heavy metals are considered as critical contaminants of aquatic ecosystems, due to their high potential to enter and accumulate in food chains ([12,13]. The term heavy metals refer to any metallic element that has a relatively high density and is toxic or poisonous at low concentration [14]. Toxicity that can last for a long time in nature. Heavy metals cannot be degraded including bio treatment and are very toxic even at low concentration (1.0–10.0 mg/L).

In the industrial areas, waste management is one of the difficult and most challenging issue. Sustainable solution is the best fit for dealing with the waste management. Recently, the biggest tannery industries situated at Hazaribagh, old Dhaka have been shifted to Savar Upozila and Savar Tannery Estate has a great significance due to its location situated in less populated area besides a river (Dhaleshwari River). But unfortunately the common effluents treatment plant (CETP) is still not been completed even after at least eight deadlines were missed. Therefore, the study area might be in a vulnerable situation. On the other hand, almost any heavy metal

and metalloid may be potentially toxic to biota depending upon the dose and duration of exposure. Heavy metal pollutants most common in the environment are Cr, Mn, Ni, Cu, Zn, Cd, and Pb [15]. Thus, it's an important field of studies to assess the pollution status in the newly established tannery industries. However, the specific objectives of this study are to (i) evaluate the heavy metals (i.e., Cr, Pb, Mn, Cu and Fe) concentration in sediment samples collected from the adjacent river of the tannery industries, (ii) find out the spatial distribution of heavy metal in the study area using ArcGIS mapping, (iii) identify the level of vulnerability for toxic metal through geo-accumulation Index (I_{geo}) and pollution load index (PLI), (iv) assessment of the potential ecological risk of the heavy metals contamination through the sediment samples in the study area and (v) source identification of toxic metals in sediment samples using USEPA PMF model.

2. Materials and methods

2.1. Study area

The study area (Dhaleshwari River, Hemayetpur) is located at the Savar Upazilla under Dhaka district, Bangladesh (Fig. 1), which is situated at a distance of about 18 km northeast of the old tannery industrial areas (Hazaribagh Tanneries, Dhaka city). In 2003, the government had stepped up and constructed the BSCIC Tannery Industrial Estate on 200 acres of land at Hemayetpur by moving all tanneries from Hazaribagh in the capital city and 73% industries are shifted by 2020. Flexible road infrastructure, near to the city, large open space are responsible for the developing of the tannery estate. The tannery estate is located just near to the Dhaleshwari River, Savar, Dhaka, Bangladesh. It is situated at 23.8583°N latitude and 90.2667°E longitude with an area of 280.13 km² (Table SI-1).

2.2. Sample collection and analysis

Sampling locations were geographically identified using Global Positioning System (GPS) device (Germin, USA). The sediment samples were collected in 2019 (September) using wooden slub and after each sample collection, the slub was thoroughly cleaned several times with deionised water to avoid cross-contamination and interference. A total 24 sediment samples were collected from eight different stations (Table SI-2) of the newly established tannery industries and the samples were stored in the pre-labeled plastic Ziploc bags and sent to Soil Resource Development Institute (SRDI), Sylhet, Bangladesh. Initially the sediment samples were air-dried for ten days, then vegetables and debris materials removed from the sediment samples. The air dried sediment samples were grinded using a mortar and pestle to get powder form and sieving was done to obtain a homogeneous mass ([16]. [5]). The 2 g of each powder sediment sample was digested following the standard procedure [17]. Briefly, a 2 g of each sediment sample placed in a 50 ml crucible before the addition of 10 ml concentrated HNO₃. The mixture was placed on a hot plate for 30–45 min to allow for oxidation. After cooling, 2.5 ml of concentrated (70%) HClO₄ acid was added and the mixture was reheated on a hot plate until the digest became clear and semi dried. Thereafter, the samples were cooled and filtered through Whatman number 42 filter paper [18]. Finally, the solution was used for elemental analysis using atomic absorption spectrometry (AAS 7000, Shimadzu, Japan) at SRDI, Bangladesh.

2.3. Quality control

To maintain the analytical integrity (accuracy and precision) of the data, the replicate investigations ($n = 3$) of NIST-1545 by AAS were performed. Relative standard deviations among sample replicates were observed to be <10%, which ensured the precision of analytical data [19]. To decide the lab execution, the boundary of Z-score [20,21] and the proportion of experimental value to the certificate value were determined (Table SI-3). It is seen that the numerical values of Z-scores for different metals are varied from 0.066 to 1.74 for the reference materials. According to the criteria of Z-score (Z-score ≤ 2 : satisfactory performance) the

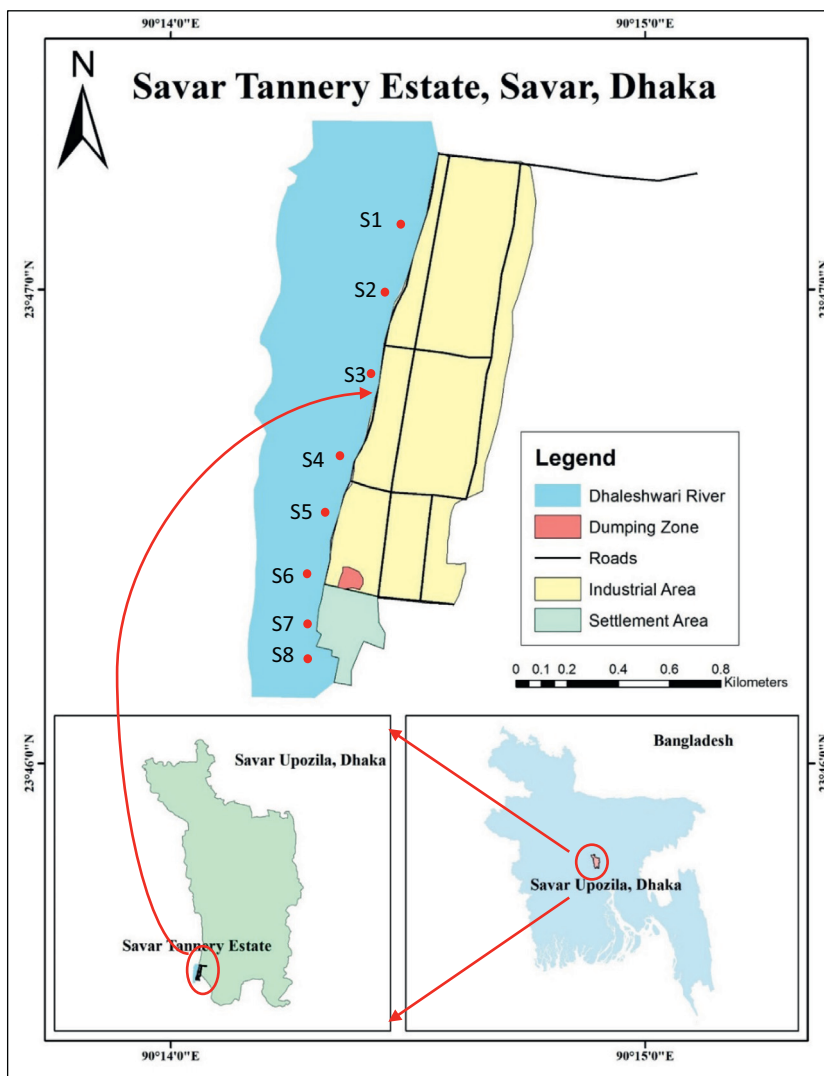


Fig. 1. Map of the study area (nearby Dhaleshwari River), Savar, Dhaka, Bangladesh.

laboratory performance was quite satisfactory as well as the Z-score values for all the elements were within 2. Furthermore, the ratio of experimental and certified values were varied from 0.93 to 1.09, and RE% for the studied metals was below 10% (Table SI-3), which also showed good agreement between measurement values and certified values. Seven procedure blanks were used to calculate the detection limits and the statistical approaches (student-t-test with 99% confidence limit) for Cr, Pb, Zn, Mn and Fe were 0.010, 0.030, 0.010, 0.010 and 0.050 mg/L, respectively.

2.4. Sediment quality guidelines

The acceptance of sediment quality guidelines (SQGs) by numerous specialists for toxicological assessment of sediment related metals has fostered the supervision of aquatic conditions, safety of biota, and the accomplishment of ecological environmental policies and guidelines [22]. However, SQGs including threshold effect level (TEL), probable effect level (PEL), effect range low (ERL), severe effect level (SEL), effects range medium (ERM) and lowest effect level (LEL) were applied to evaluate the possible biotic influence of metal(oid)s estimated in the sediment samples.

2.5. Pollution assessment

The pollution indices act as a potential role in the comprehensive assessment of soil pollution degree [23]. Therefore, five major indices:

geoaccumulation index (I_{geo}), enrichment factors (EFs), modified pollution index (MPI), contamination factor (C_f) and degree of contamination (C_d), pollution load index (PLI), was applied for this study to assess the pollution based on toxic metal concentration in sediment samples.

2.5.1. The geo-accumulation index (I_{geo})

The geo-accumulation index (I_{geo}) was presented by [24]. This index is applied to quantify the metal pollution in the soils and aquatic sediments. The geo-accumulation index (I_{geo}) for sediment samples was calculated using the following equation:

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

where, C_n is concentration of metal measured in sediment samples in the study area, B_n is background value of the corresponding metal, and 1.5 is the background matrix correction due to lithological effects. Muller [24] proposed the geo-accumulation index following seven grades or classes: i) $I_{geo} > 5$ = extremely polluted, (ii) $I_{geo} = 4-5$ = strongly to extremely polluted, (iii) $I_{geo} = 3-4$ = strongly polluted, (iv) $I_{geo} = 2-3$ = moderately to strongly polluted, (v) $I_{geo} = 1-2$ = moderately polluted, (vi) $I_{geo} = 0-1$ = unpolluted to moderately polluted, and (vii) $I_{geo} < 0$ = practically unpolluted.

2.5.2. Contamination factor

The contamination factor is obtained from a ratio between the measured conc. of the heavy metals in sediment of the water body and the pre-industrial reference value for the same metal [25]. The degree of contamination is defined as the sum of all contamination factors. The computing equation for contamination factor (C_f^i) is as follows:

$$C_f^i = C^i / C_n^i \quad (2)$$

where C_f^i is the measured concentration of the heavy metals in sediment and C_n^i is the standard pre-industrial reference level (in mg/kg): 70 for Pb, 1.0 for Cd, 90 for Cr, 50 for Cu, 175 for Zn, 15.0 for As, 68 for Ni, 850 for Mn [25,26].

2.5.3. Pollution load index (PLI)

The pollution load index (PLI) is obtained from the contamination factors (C_f^i). This C_f^i is the quotient obtained by dividing the concentration of each metals [82]. The PLI of the place are calculated by obtaining the n-root from the n-CFs that were obtained for all the metals. Generally the pollution load index (PLI) as developed by Tomlinson et al. [27], which is shown as follows:

$$PLI = \sqrt[n]{C_{f1} \times C_{f2} \times C_{f3} \times \dots \times C_{fn}} \quad (3)$$

where, C_f is the contamination factor and n is the quantity of metals in the study, which are referred in a previous condition (Eq. (3)). The PLI gives unassuming yet sensible intends to evaluating a site quality [27], where an estimation $PLI < 1$ mean perfection; $PLI = 1$ present that only baseline levels of contaminant are available; and $PLI > 1$ would show decline of site quality.

2.5.4. Enrichment factors (EFs)

To assess the degree of heavy metal contamination, we used the following equation based on Mokhtarzadeh et al. [28] to measure enrichment factors (EFs).

$$EF = \frac{\left(\frac{C_i}{C_{ref}}\right)_{Sample}}{\left(\frac{C_i}{C_{ref}}\right)_{background}} \quad (4)$$

where C_i is the target element's concentration, and C_{ref} is the reference element's concentration. Arsenic was chosen as the reference element because of the low variation coefficient (CV) in the samples [28]. Background metal concentrations for EF calculation were used as 27 for Cu, 20 for Pb, 35 for Cr, 68 for Zn, and 3 for As [21,29]. The classification of EF: $EF < 1$ indicates no enrichment, $EF < 3$ is minor enrichment, $EF = 3-5$ is moderate enrichment, $EF = 5-10$ is moderately severe enrichment, $EF = 10-25$ is severe enrichment, $EF = 25-50$ is very severe enrichment, and $EF > 50$ is extremely severe enrichment.

2.6. Potential ecological risk index (PERI)

The assessment of the potential ecological risk of the heavy metals contamination was proposed as a diagnostic tool for water pollution control purposes as a result of the increasing content of heavy metals in sediments and their subsequent release into the water, which could threaten ecological health [83]. Hakanson [25] developed a method to assess the potential ecological risk index for aquatic pollution control purposes, i.e. to sort out which lakes or rivers and substances should be given special attention. According to this method, the potential ecological risk factor (E_r^i) of single element and the potential ecological risk index (RI) of multi-element can be computed by the following equations [18]:

$$E_r^i = T_r^i / C_f^i \quad (4)$$

$$RI = \sum_{i=1}^n E_r^i \quad (5)$$

where C_f^i is the contamination factor for the element of "i"; T_r^i is the toxic-response factor for the given element of "i", which accounts for the toxic requirement and the sensitivity requirement. The toxic response factors for Pb, Cr, Cu, and Mn were 5, 2, 5, and 1 respectively [25,30]. Using eqs. 2, 4 and 5, the potential ecological risk assessment was calculated.

2.7. Toxic risk index

The toxic risk index developed by Zhang et al. [31] was applied to provide a more comprehensive of their risk to the biota in the aquatic environment. Two threshold values for SQGs (TEL and PEL standard) were used to calculate TRI following Eq. (6).

$$TRI_i = \sqrt{\frac{(C_i/TEL_i)^2 / (C_i/PEL_i)^2}{2}} \quad (6)$$

where the concentration of i th metal is denoted as C_i , and the probable effect level and threshold effect level for the i th metals is denoted as PEL and TEL. To interpret the TRI values, ' $TRI \leq 5$ ' is considered as no toxic risk, ' $5 < TRI \leq 10$ ' as low toxic risk, ' $10 < TRI \leq 15$ ' as moderate toxic risk, ' $15 < TRI \leq 20$ ' as considerable toxic risk, and ' $TRI > 20$ ' as very high toxic risk.

2.7.1. Modified hazard quotient (mHQ)

The modified hazard quotient is a new technique for calculating the degree of risk that each metal poses to living organisms in a specific area [32]. Different researchers have established its validity, reliability, and accuracy [33–36]. As previously described, this new technique allows for contamination detection by comparing metal concentrations in sediment (mg/kg) with adverse ecological impact distributions at slightly different threshold effect level (TEL), probable effect level (PEL), and severe effect level (SEL), which are summarized in Table 1. However, the mHQ index is calculated using the following eq. (7):

$$mHQ = \sqrt[2]{\left[C_i \left(\frac{1}{TEL_i} + \frac{1}{PEL_i} + \frac{1}{SEL_i} \right) \right]} \quad (7)$$

where, C_i indicates the measured metal concentration of the studied sample. The mHQ values were grouped into eight classes [32]: (i) $mHQ > 3.5$ (extreme severity of contamination), $3.0 \leq mHQ < 3.5$ (very high severity of contamination), $2.5 \leq mHQ < 3.0$ (high severity of contamination), $2.0 \leq mHQ < 2.5$ (considerable severity of contamination), $1.5 \leq mHQ < 2.0$ (moderate severity of contamination), $1.0 \leq mHQ < 1.5$ (low severity of contamination), $0.5 \leq mHQ < 1.0$ (very low severity of contamination), and $mHQ < 0.5$ (nil to very low severity of contamination).

2.8. Positive matrix factorization (PMF) method

As a receptor method for source apportionment [40,41], PMF has been widely accepted worldwide. The initial matrix data ($X_{n \times m}$) was decomposed into two matrices, including source contribution matrix ($G_{n \times p}$) and the source profile matrix F ($G_{p \times m}$) [41]. It can be expressed as follows:

$$x_{ij} = \sum_{k=1}^p g_{ik} f_{kj} + e_{ij} \quad (8)$$

where, i is the series of the samples, j is the heavy metals determined, p is the number of potential sources, x_{ij} is the concentration of the j th heavy metal in the i th sample (mg/kg); g_{ik} is the contribution of the k th source in the i th sample; f_{kj} is the amount of the j th heavy metal from the k th source factor; p represents the number of source factors; and e_{ij} is the residual.

Table 1

Concentration of the studied heavy metals in sediment sample collected from the leather tannery village, Savar, Dhaka, Bangladesh.

	Concentration (mg/kg) as dry wt basis				
	Cr	Pb	Fe	Cu	Mn
Experimental Data					
Mean (n = 24)	186	8.78	42.7	1.76	3.12
SD (1σ)	242	6.15	49.1	1.47	1.38
RSD (%)	129	70.0	114	83.8	44.21
Median	64.9	7.90	14.5	1.02	2.65
Min.	14.9	2.38	3.87	0.36	1.59
Max.	778	21.1	154.4	4.75	6.29
SQG Threshold values					
LEL [37]	26.0	31.0	20,000	16.0	460
SEL [38]	110	250	40,000	110	1100
TEL [38]	52.3	30.2	–	18.7	–
PEL [38]	160	112	–	108	–
ERL [39]	81.0	46.7	–	34.0	–
ERM [39]	370	218	–	270	–
TRV [61]	26.2	31.0	–	16.0	–
Impact (%) on ecology					
<LEL	25.0	100	63.0	100	100
LEL-SEL	38.0	–	25.0	–	–
>SEL	38.0	–	13.0	–	–
<TEL	46.0	–	63.0	–	–
TEL-PEL	17.0	–	38.0	–	–
>PEL	38.0	–	13.0	–	–
<ERL	58.0	–	83.0	–	–
ERL-ERM	29.0	–	17.0	–	–
>ERM	13.0	–	–	–	–

The concentration and uncertainty of each heavy metal are input into the PMF Model. The uncertainty (u) of the heavy metals was calculated as follows:

$$u_{ij} = \frac{5}{6} \times MDL, x_{ij} \leq MDL \quad (9)$$

$$u_{ij} = \sqrt{(\sigma \times x_{ij})^2 + (MDL)^2}, x_{ij} > MDL \quad (10)$$

where x_{ij} is the concentration of heavy metals, MDL is the method detection limit, and σ is the relative standard deviation of the concentration of PTEs.

Factor contributions and profiles can be determined by minimizing the objective function as follows: [41].

$$Q = \sum_{i=1}^n \sum_{j=1}^m \left[\frac{x_{ij} - \sum_{k=1}^p g_{ik} f_{kj}}{u_{ij}} \right]^2 \quad (11)$$

where u_{ij} is the uncertainty of the j th heavy metal in the i th sample. Q_{true} and Q_{robust} are two set data of Q used in the PMF model. The best simulated run was identified by the lowest Q_{robust} . The determination of the number of the factors is critical to the results of the PMF model. In this study, we adopted the PMF method is that this method deploys the uncertainty matrix, which allows us each weight for all variables to explain the factorization issue [42].

2.9. Statistical analysis

All the samples were taken in duplicate and the repeatability was checked by paired Student's t -test at a 95% confidence level. The data were summarized using mean values, standard deviations (Stdev), the median, ranges, geometrical mean (GM) and the percentage of relative standard deviation (%RSD). Subsequently, the analysis of variance (ANOVA) tests at a significance level of 95% were used to evaluate the impact of

different variables on the contamination in the study area using SPSS 17 Software (IBM SPSS Inc., USA).

3. Result and discussion

3.1. Heavy metals concentration

Tannery effluents have been identified as one of the major threats to environment. Therefore, sediment samples from the nearby tannery industries (tannery village) were analyzed to determine the concentration of heavy metals, which is presented in Table 1. The average analysis of data depicted an order of heavy metals accumulation in sediment (mg/kg as dry wt basis) that was Cr (186 ± 541) > Fe (42.7 ± 49.1) > Pb (8.78 ± 6.2) > Mn (3.12 ± 1.4) > Cu (1.76 ± 1.5) respectively. The metals (i.e., Cr, Pb, Mn, Cu and Fe) concentrations for each sampling points could be found in supplementary section (Table SI-2). The data showed that Cr got extreme concentration in the sediments, while Cu was minimally accumulated (Fig. 2). The percentage of relative standard deviation (%RSD) for the studied heavy metals distribution in sediments at different sampling points showed that the abundance of Cr, Fe, Cu, Pb and Mn were varied a wide range (%RSD: 44.21–129.85%), which is consistent with the ANOVA test at a 95% confidence level ($F_{cal} > F_{critic} = 3.73 > 2.64$; $p = 0.012$; $df = 35$) (Table SI-4). Therefore, it has been suggested that the sources of these metals in sediments were mainly anthropogenic [45] as well as the studied heavy metals were distributed homogeneously. The concentrations of the studied heavy metals in the sediment samples (mg/kg) were also compared with the threshold values of the sediment quality guidelines (SQGs): Probable Effect Level (PEL), Threshold Effect Level (TEL), Severe Effect Level (SEL), Effect Range Low (ERL), Lowest Effect Level (LEL), and the Effects Range Medium (ERM),

3.1.1. Chromium (Cr)

The average Cr concentration in sediment samples in the study area was found to be 186 ± 541 mg/kg ranging from 14.9 to 778 mg/kg. This study revealed that the average Cr concentration in the sediment samples in the study area was found to be higher than the upper continental crust [43] value (92 mg/kg), the average sediment (90 mg/kg) value ([26]) and the soil background value (62.5 mg/kg) in China [44] (Fig. 2a). Subsequently it was observed that the average Cr concentration in the sediment samples was higher than the several different rivers sediments in Bangladesh: Turag River [46,47], Brahmaputra River [48]; and also lower than the reported results of the river sediments in several countries in the world [26,49–56] (Table 2). It might be happened due to the reason that there is a big tannery industry is established in the study area [60], which is continuously releasing effluents enriched with Cr. However, the average Cr concentration in sediment samples was observed to be higher than all the threshold values (LEL, SEL, TEL, PEL, ERL, ERM and TRV) for the SQGs ([37–39,61]). It was noticed that 38%, 13% and 38% samples were higher than SEL [37], ERM [39] and PEL [38], while 38%, 17% and 29% samples were fallen between LEL-SEL, TEL-PEL and ERL-ERM respectively. Therefore, it could be suggested that the present levels of Cr in the study area have significant impact on ecology by damaging bacterial DNA [62]. Cr is also treated as harmful for the lungs, heart, kidney and might have impact on human health to create cancer [63,64].

3.1.2. Lead (Pb)

The toxicity level of Pb is so hazardous, even at a low concentration, it can pose a significant threat to the ecosystem [65]. The average Pb concentration at different sampling points was significantly different following a sequence: S7 > S6 > S8 > S5 > S1 > S2 > S3 > S4 respectively (Fig. 2b), and the average value was 8.78 ± 6.2 mg/kg ranging from 2.38 to 21.09 mg/kg. The coefficient of variance (CV) for Pb concentration in different sampling points exhibited the high ($51\% < CV \leq 100$) variability [66], which indicating the sources of Pb in the study area might be mainly anthropogenic. Reversely, the average Pb concentration was 3 times lower than the Pb in surface soils (32 mg/kg) of the worldwide average

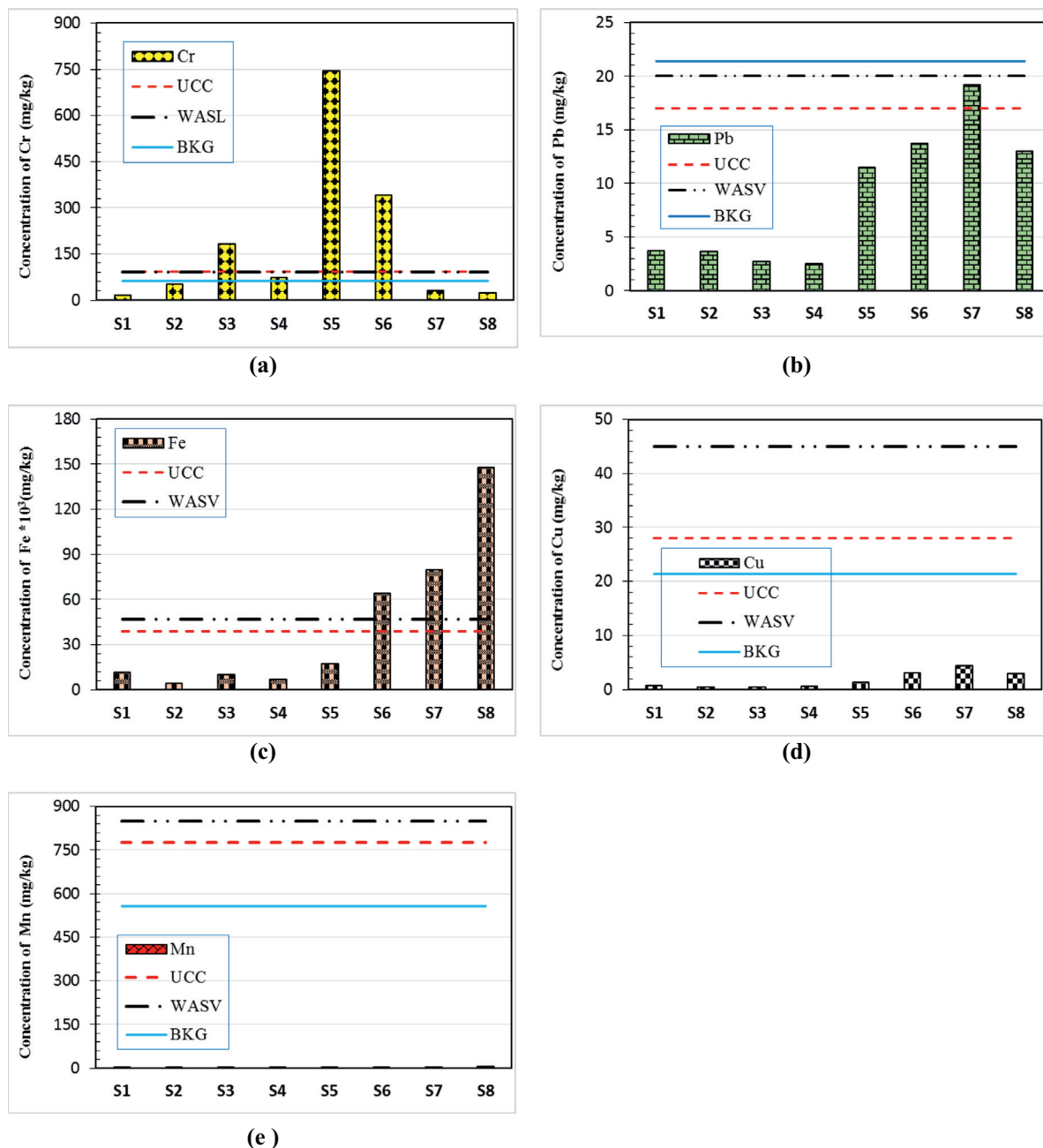


Fig. 2. Concentration (mg/kg) of (a) Cr, (b) Pb, (c) Mn, (d) Cu and (e) Fe in sediments samples in comparison with the recommended values ([26,43,44]. NB.: UCC: Upper continental crust values [43], WASL: World average soil values [26] and BKG: Background values for soil ([44].

(Kabata-Pendias and Pendias, 2001) and 2 times lower than the upper continental crust (UCC; [43]) and the average sediment value [26]. The average Pb level was found to be lower than the value observed in many international river sediments: China [56], Russia [55], Turkey [58], Angola [54], Germany [67], Ireland (Jones and Jordan [57], Malaysia [50], Congo [49]. It could be possibly originated from the use of gasoline additives, pesticides, as well as chemical manure [68], sand extraction [69], mining [22] (Table 2). On the other hand, the average Pb concentration for 100% sediment samples was found to be lower than the severe effect level (SEL) threshold values (Table 1). It's noteworthy to mention that even at low level, Pb is toxic as well as its' naturally non-biodegradable [60,64,70]. Therefore, like as other fundamental divalent metals (Mn^{2+} and Zn^{2+}), Pb^{2+} might have changed the compliance of nucleic acids,

proteins, restraint of chemical movement of bacteria as well as alterations of the osmotic balance of the bacterial cells [62], which might have impact on ecology [18].

3.1.3. Manganese (Mn)

This study revealed that the average Mn concentration in the sediments samples was found to be 3.12 ± 1.4 mg/kg (Table 1). The highest Mn concentration was found in the sampling point of S8 and the lowest Mn concentration was found in the sampling point of S5. The sequence for the average Mn concentration in the sediment samples collected from the different sampling points were found to be the following order: $S8 > S1 > S7 > S4 > S6 > S3 > S2 > S5$ respectively (Fig. 2c). On the other hand, the coefficient of variance for Mn concentration in different sampling points exhibited the

Table 2

Comparison of heavy metal concentration in sediment sample for this study with literature value and recommended values in the world.

	Metals concentration (mg/kg) as dry wt.					Reference
	Cr	Pb	Fe	Cu	Mn	
<i>Recommended values</i>						
UCC	92.0	17.0	39,000	28.0	775	[43]
BKG	62.5	21.4		21.4	557	[44]
ASV	90.0	20.0	47,200	45.0	850	[26]
Leather Village, Dhaka	186	8.78	42.70	1.76	3.12	This study
Amur River, Russia	36.3	19.2	25,400	7.43	768	[55]
Elbe River, Germany	386	122	33,400	206	1230	[48]
Liffey River, Ireland	–	–	26,900	220	496	[57]
Catumbela River, Angola	26.0	19.0	26,800	–	620	[54]
Lubumbashi River, Congo	86.8	1549	45,418	14,822	2920	[49]
Buyukmelen River, Turkey	169	12.1	45,500	30.6	1007	[58]
Godavari-Krishna River, India	129	–	63,500	91.0	800	[53]
Kumho River, Korea	99.7	149	–	125	–	[51]
Linggi River, Malaysia	33.3	30.5	17,710	14.6	–	[50]
Pearl River, China	71.4	97.1	19,600	54.5	908	[31]
Buriganga River, Bangladesh	788	25.0	25,000	45.0	508	[59]
Turag River, Bangladesh	70.0	–	29,000	–	768	[46,47]
Brahmaputra River, Bangladesh	96.2	19.5	35,500	17.2	752	[48]

moderate ($21\% < CV \leq 50$) variability [66], which indicating the sources of Mn in the study area might be mainly natural and anthropogenic. Reversely, the average Mn concentration was observed to be much lower than the background value of Chinese soil [44], the upper continental crust value [43] and the average world soil value (Kabata-Pendias and Pendias, 2001). Subsequently the average Mn concentration was lower than the reported sediment values by the international scientists in the world ([49,50,54–59,67] (Table 2). The average Mn concentration in 100% of the sediment samples for this study was observed to be lower the SEL [37] threshold value, which indicated the lower contamination of Mn in the study area.

3.1.4. Copper (Cu)

The average Cu concentration in the sediment samples was found to be 1.76 mg/kg ranging from 0.36 to 4.75 mg/kg. The highest Cu concentration was at the sampling point of S7, while the lowest concentration was found to be at S3 (Figs. 1 and 2). This study revealed that the average Cu concentration in the sediment samples was much lower than the background value of Cu in Chinese soil [44], the upper continental crust value [43] and Cu in average world soil value [71] (Fig. 2d). Subsequently the findings for this study was lower than the Cu concentration in sediments in many countries in the world: Bangladesh [59], China [56], Russia [55], Turkey [58], Angola [54], Germany [67], Ireland (Jones and Jordan, 1979), Malaysia [50], Congo [49] (Table 2). It is noteworthy to mention that copper is usually emitted in the environment through vehicle exhausts (Xia and Gao, 2011) and smelting from burning furnace (Yang et al., 2003), coal burning (Aksu et al., 1998a). As the study area is situated far away from urban area and in a village, therefore, the study area is till now free Cu contamination. On the other hand, the average Cu concentration in the sediment samples was observed that 100% of sediment samples were lower than LEL [37]. Therefore, it has been suggested that there might be less impact of Cu on ecology.

3.1.5. Iron (Fe)

Among the analyzed elements, the concentration of Fe in the sediments was the second highest among all the elements studied following the sequential order: Cr > Fe > Pb > Mn > Cu respectively. The concentration of Fe in the sediment samples were ranged from 3870 to 154,380 mg/kg with an average value of 42,699 mg/kg (Table 1). The statistical analysis for the average values in different sampling points exhibited the very high ($CV < 100$) variability [66], which indicating the sources of Fe in the study area might be mainly anthropogenic. It might be happened due to the reason that iron is abundant in nature and many iron related construction works are on-going at the new leather processing industries. On the other hand, the average Fe concentration in the sediment samples were

little bit higher than UCC (39,000 mg/kg), while little bit higher than the world soil average value (47,200 mg/kg) (Table 2). The finding for this study was in line with the reported results for sediments collected from the different countries in the world ([49,50,54–59,67]. On the other hand, the average Fe concentration in 100% of the sediment samples were exceeded the LEL [37] and SEL [37] threshold values (Table 1).

3.2. Special distribution

Using ArcGIS software (Version 10.2, Esri, California, USA), the spatial distributions for Pb, Cr, Mn, Ni, Cu, Zn, As and Cd in river sediment samples of Dhaleshwari River attached with Savar Tannery City were obtained based on their concentrations (Fig. 3). The results of spatial distribution pattern showed that the high concentrated zones of Cr was found at the dumping zone (~ S5) of leather industrial city (Fig. 3a). This study also showed that the average Cr concentration in sediment samples was observed to be higher than all the threshold values (LEL, SEL, TEF, PEL, ERL, ERM and TRV) for the sediment quality guidelines ([37–39,61]. It might be happened due to the reason that chromium-based tanning process (based on basic chromium sulfate) is predominantly followed in the tannery industries and leather industry produces chromium-based waste both in liquid and solid form such as chromium sludge, chrome-tanned leather shavings (CTLs), and chrome leather trimmings. These wastes are unavoidable due to adaptability of chromium, and possess a serious threat to the environment. Among heavy metals presents in tannery waste, chromium is one of the most common pollutant. However, the spatial distribution pattern for the studied metals (Pb, Mn, Cu and Fe) showed that the maximum metals concentrations were found in the southeastern part (Figs. 3b–e) of the Dhaleshwari River, which is attached with the Savar Tannery City, Bangladesh (Fig. 1).

3.3. Pollution assessment

3.3.1. The geo-accumulation index (I_{geo})

The geoaccumulation index (I_{geo}) has been broadly applied in European trace metal studies since the late 1960s [24] and this approach is greatly applied in this study for the assessment of heavy metal contamination level in sediment samples. The average range of I_{geo} values for different sediment samples were found to be -3.14 to 2.46 , -3.58 to -0.65 , -9.57 to -7.78 , -0.76 to 4.39 for Cr, Pb, Mn, Cu and Fe respectively (Fig. 4). The detailed data for each sampling points and each metal can be found in supplementary section (Table SI-5). The I_{geo} values of the studied heavy metals are shown in Fig. 4. This study revealed that the I_{geo} for Cr in S5, S3 and S6 were 3.14, 0.59 and 1.34, which means these samples belong to class 1, 3 and 2 respectively (Table SI-5). In S1, S3, S5, S6, S7 and S8 sample, the

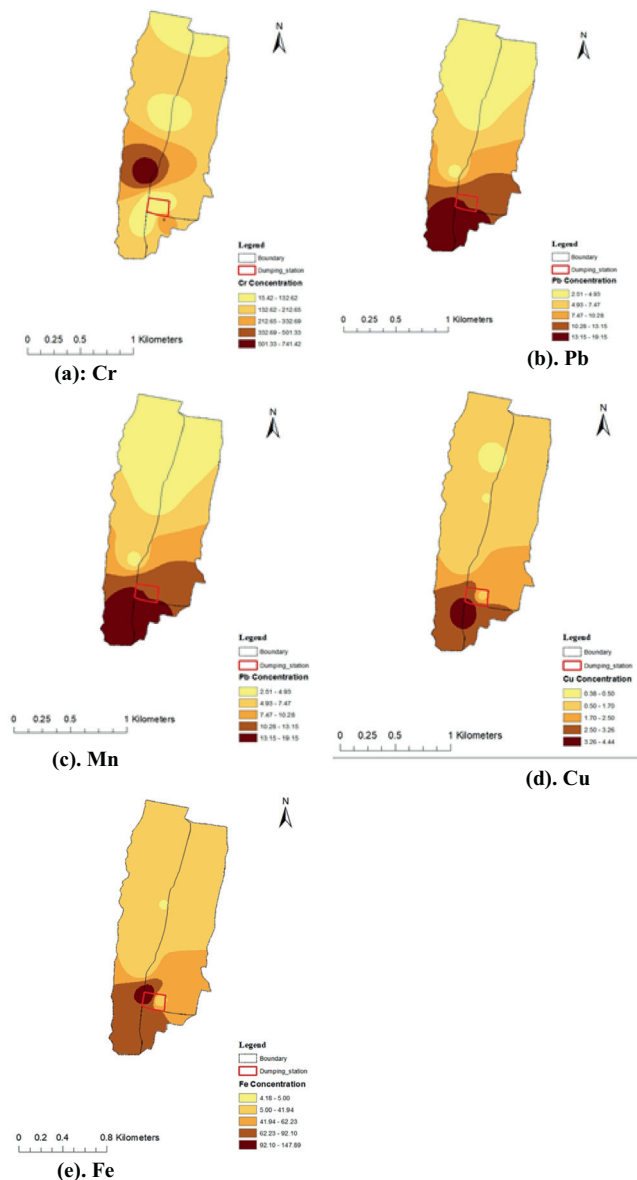


Fig. 3. Special distribution of metals: (a) Cr, (b) Pb, (c) Mn, (d) Cu and (e) Fe in the study area (Savar, Dhaka, Bangladesh).

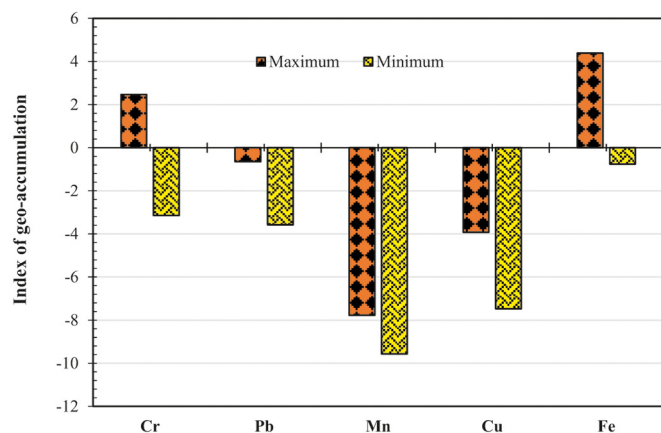


Fig. 4. Maximum and minimum geo-accumulation value for each heavy metal in soil sample.

I_{geo} for Fe was individually 0.70, 0.49, 1.29, 3.18, 3.50 and 4.39. The other I_{geo} of Pb, Mn and Fe were less than 0 indicating that the sediments was not polluted. At the sampling point of S3 the I_{geo} of Cr and at S1 and S3, the I_{geo} of Fe were falling into the 1st category means from unpolluted to moderately polluted. The sediment quality for Cr in S6 and Fe in S5 were moderately polluted. The quality soil was from moderately to strongly polluted in S5 for Cr. The sediment quality for Fe in S6 and S7 were strongly polluted. The quality of sediment was from strongly to extremely polluted in S8 for Fe. The I_{geo} for Cr and Pb in the study area ranged from -3.14 to 2.46 and -3.58 to -0.65 with a mean concentration of -0.44 ± 1.87 and -2.18 ± 1.20 respectively. The I_{geo} concentration was given and the maximum and minimum I_{geo} for all the heavy metals were listed in Fig. 4. The figure indicates the positive maximum values of Cr and Fe. On the other hand, the I_{geo} values for Cr in 62.5%, 25% and 12.5% of the sampling points are falling into class 1, class 2 and class 3 indicating the contamination status of sediments are (1) uncontaminated, (2) uncontaminated to moderately contaminated, (3) moderately to heavily contaminated respectively [24]. Subsequently, I_{geo} values for Fe in 50%, 25% and 25% of the sampling points are class 1, class 2 and class 4 respectively. However, 100% I_{geo} values for Pb, Mn and Cu are falling in class 0, which indicating that the geoaccumulation index for all the sampling points are practically uncontaminated.

3.3.2. Contamination factor (CF) and pollution load index (PLI)

[25] proposed two important indices: (i) contamination factor (CF) and (ii) pollution load Index (PLI) to measure the metal pollution in sediment. However, those indices have been widely used by many scientists [27,29,72] to evaluate the contamination status of heavy metals in sediment samples. The contamination level in S3 sample is moderate, S6 sample is considerate and S5 sample is high for Cr and for Fe the level of contamination is moderate in S1, S3 and S4 sample, is considerate in S5 sample and high in S6, S7 and S8 sample. The calculated results for the contamination factor (CF) are given in Table 3. This investigation showed that CF values of metal analyzed ranged from 0.17 to 8.27, 0.13 to 0.96, 0.00 to 0.01, 0.01 to 0.10 and 0.66 to 0.86 for Cr, Pb, Fe, Cu and Mn respectively (Table SI-6). Analysis of the data depicted an order of mean CF values in sediment samples: Fe (9.06 ± 10.86) > Cr (2.07 ± 2.80) > Pb (0.46 ± 0.34) > Cu (0.04 ± 0.03) and Mn (0.1 ± 0.0) respectively. This study revealed that 75% and 25% of the sediments samples for Cr levels were belonged to low ($CF < 1$) and moderate ($1 \leq CF < 3$) contamination factor. While, CF values for Fe in 25%, 37.5%, 12.5% and 25% of the sampling points are falling into class 1 (low contamination), class 2 (moderate contamination), class 3 (considerable contamination) and class 4 (very high contamination) respectively (Fig. 5) (Hakanson [25]).

The pollution load index (PLI) was used to apply for the determination of the level of heavy metal pollution in the particular studied site [27, 81]. This index is a quick tool in order to compare the pollution status of different places. The list as introduced gives an essential and relative method for assessing an area or estuarine class: an assessment of zero shows faultlessness, an assessment worth of one that solitary benchmark levels of toxins are available. Conversely, the qualities more than one would show dynamic crumbling of the site and estuarine quality. However, this study revealed that the average PLI value for 8 different sampling points was found to be 0.76 ± 0.11 ranging from 0.58 to 0.88 (Table SI-6). It was observed that the pollution load index values in 100% of the sediments samples were lower than the base line of pollution background levels (Fig. SI-1), which indicated that the study area has low level of pollution at this moment considering the studied heavy metals. However, a continuous monitoring should be needed to assess more accurately the pollution load index due to the contamination of heavy metals in indoor dust samples [70].

3.4. Ecological risk

The results of evaluation on potential ecological risk factor (E_r^i) and the potential ecological risk index (RI) are summarized in Table 3. This study revealed that the potential ecological risk factor (E_r^i) for Cr, Pb, Cu and

Table 3Potential ecological risk factors (E_i^p) and potential ecological risk indexes (RI) of heavy metals in sediments from Dhalaibeel and Bangshi River.

ID	Contamination factor (CF)				Pollution Load Index	Potential ecological risk factor (PERF)				RI	Pollution level
	Cr	Pb	Cu	Mn		Cr	Pb	Cu	Mn		
S1	0.17	0.16	0.03	0.002	0.58	0.34	0.82	0.13	0.002	1.29	Low
S2	0.52	0.05	0.01	0.002	0.66	1.19	0.26	0.05	0.002	1.49	Low
S3	2.22	0.04	0.01	0.003	0.71	4.53	0.19	0.04	0.003	4.76	Low
S4	0.81	0.04	0.01	0.003	0.69	1.64	0.18	0.06	0.003	1.89	Low
S5	8.21	0.05	0.02	0.005	0.88	16.5	0.27	0.08	0.005	16.9	Low
S6	3.78	0.19	0.06	0.003	0.86	7.59	0.98	0.31	0.003	8.89	Low
S7	0.31	0.27	0.09	0.005	0.83	0.72	1.37	0.44	0.005	2.54	Low
S8	0.26	0.19	0.06	0.007	0.84	0.51	0.93	0.29	0.007	1.74	Low

Mn were ranged from 0.34–16.54, 0.18–1.37, 0.04–0.44 and 0.002–0.007 respectively with an average value of 4.13, 0.626, 0.176 and 0.004. The order of potential ecological risk factor of heavy metal in sediments of tannery village was $Cr > Pb > Cu > Mn$ (Table 3). It was observed that the potential ecological risk factors (E_i^p) of Cr, Pb, Cu and Mn were all lower than 40, which belong to low ecological risk [25]. All the sampling sites were at low risk level where the RI values were much lower than 150 [25]. The results indicated that there was low potential ecological risk for the sediment samples collected from the newly established leather tanning industries. However, the order of potential ecological risk index for the studied heavy metal in sediments of tannery village was found to be in the order of $S5 > S6 > S3 > S7 > S4 > S8 > S2 > S1$ respectively.

3.5. Toxic risk index (TRI)

An alternative calculation known as the toxic risk index (TRI) was approved to provide a more detailed to estimate the potential toxicity of the individual metal (oid) in the ecosystem. This study revealed by using the above-mentioned index (Eq. 6) that the average TRI value for Cr, Pb and Cu was 2.16, 2.62 and 4.08 respectively. Considering the average value for this study, it has been suggested that there is no risk observed by the metal(oid)s as well as the TRI values were below 5 ($TRI \leq 5$: no risk; [31]). However, attention should be directed at Cr because of the heightened TRI values in S-5, S-3 and S-6. It could be happened due to the reasons that Cr concentration in these sampling points was high due to direct dumping leather waste.

3.6. Modified hazard quotient (mHQ)

Recently a pollution index related to the level of contamination was established by Benson et al. [33], and it is known as the modified hazard quotient (mHQ). The mHQ evaluates levels of pollution by depicting each metal

(oid) concentration found in the sediments with the threshold edge of adverse environmental dispersions like the SEL, PEL and TEL. The assessment of mHQ is of most extreme significance since it assesses the threat of individual metal(oid)s to the biota and the aquatic environment [22]. The mHQ was calculated following the Eq. (7), and the result of mHQ for specific metal contributions was shown in Table 4. This study for Cr level in sediments revealed that 25% of the samples' mHQ values were in range of $1 > mHQ \geq 0.5$ (Very Low Severity of Pollution), while 25%, 12.5%, 12.5% and of the sediments' mHQ values were in $1.5 > mHQ \geq 1$ (Low Severity of Pollution), $2 > mHQ \geq 1.5$ (Moderate Severity of Pollution) and $2.5 > mHQ \geq 2$ (Considerable Severity of Pollution) respectively (Table 4). It was observed that 75% of samples were low to severely polluted by Cr, while 25% of the samples were extremely polluted by Cr as well as $mHQ \leq 3.5$ (Extreme Severity of Pollution). Therefore, it has been suggested that the study area was at severe ecological risk by Cr, and the associated environment with the floral and faunal community was at significant risk. Further insight to Pb contamination, it was observed that 50% of the sediments were in very low severity of pollution ($1 > mHQ \geq 0.5$), which 50% of the sediments were in Nil to very Low Severity of Pollution ($0.5 > mHQ$) considering mHQ values. On the other hand, 83% and 100% of the sediment samples indicated no concern for ecology and the environment by Cu and Mn (Table 4).

3.7. Source identification by PMF

Source identification by PMF in recent times, PMF attracts much attention to the researcher community as an appropriate receptor tool, which may calculate the contribution and source of contaminants in the ecosystem [42,73]. For precision purpose, the value of Q was decreasing to control the residual matrix E . However, different factors (2–3) were examined, and the system was run 10 times to get the optimal results. The optimal result from this PMF method verified two factors, which stranded in the minimum Q value, where the residuals varied between 0.13 and 17.41. The coefficient of determination (r^2) between the detected and the projected value varied from 0.1 (Cr) to 0.35 (Mn), as displayed in the supplementary Table SI-7, suggesting a strong correlation except for Mn. Thus, tested toxic metals were well distributed by the PMF method, and the outcomes were reliable (Table SI-8).

Factor 1 was extremely loaded with Cr (186.16%), which was the key element to show the prime source of pollution in this factor. In most of the sediment samples, Cr was concentrated greater than the geochemical background values and even surpassed the environmental quality guidelines (Table SI-9). Earlier cited work reported that Cr was originated from anthropogenic inputs, especially tannery industrial inputs and sewage water system [59]. In contrast, Lv et al. [74] revealed that Cr was derived from parent source rock materials due to geogenic weathering process. Besides, the leather tanning industrial complex in the study area releases a huge amount of Cr in the river [75]. Thus, Factor 1 most likely has dominant human point sources such as severe discharge of untreated tannery and industrial plant material.

Factor 2 was predominated by Fe and Pb with weak loadings of 25.29% and 6.63%, respectively. This factor was controlled by a mixed geogenic and anthropogenic sources. Fe was mainly derived from geogenic parent material of weathering. Two hot spot areas were observed, with one

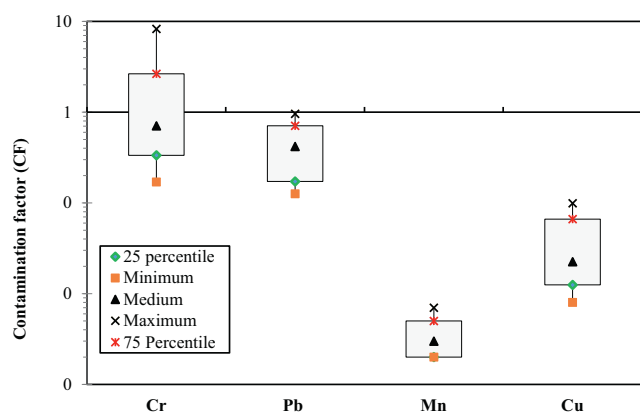


Fig. 5. The Box-whisker plot of the contamination factor (CF) for the studied metals in sediment samples.

Table 4Grading of ecological risk of some metals using modified Hazard Quotients (*mHQ*) in sediment samples in leather village.

Ele-ment	0.5 > <i>mHQ</i> (Nil to Very Low Severity of Pollution)	1 > <i>mHQ</i> ≥ 0.5 (Very Low Severity of Pollution)	1.5 > <i>mHQ</i> ≥ 1 (Low Severity of Pollution)	2 > <i>mHQ</i> ≥ 1.5 (Moderate Severity of Pollution)	2.5 > <i>mHQ</i> ≥ 2 (Considerable Severity of Pollution)	3 > <i>mHQ</i> ≥ 2.5 (Very High Severity of Pollution)	3.5 > <i>mHQ</i> ≥ 3 (High Severity of Pollution)	<i>mHQ</i> ≤ 3.5 (Extreme Severity of Pollution).
Cr	(0), 0.0%	(6) 25%	(6), 25%	(3), 13%	(3), 13%	(0), 0.0%	(0), 0.0%	(6), 25%
Pb	(12) 5%	(12) 50%	(0), 0.0%	(0), 0.0%	(0), 0.0%	(0), 0.0%	(0), 0.0%	(0), 0.0%
Fe	(7) 29%	(8) 33%	(6), 25%	(3) 12.5%	(0), 0.0%	(0), 0.0%	(0), 0.0%	(0), 0.0%
Cu	(20) 83%	(4) 17%	(0), 0.0%	(0), 0.0%	(0), 0.0%	(0), 0.0%	(0), 0.0%	(0), 0.0%
Mn	(24) 100%	(0), 0.0%	(0), 0.0%	(0), 0.0%	(0), 0.0%	(0), 0.0%	(0), 0.0%	(0), 0.0%

being located in the northern part of the river and the other located in the north-central part of the river. These hot spot sites were classified by an elevated concentration of Fe than Pb and were mostly inherited in urban areas. Lead is mostly derived from emission from traffic, fossil fuels, vehicle tires, brake pad (Zheng et al., 2002; [76,77]). Fuel combustion also increase Pb content in soil [78]. Besides, Pb may be concentrated from atmospheric deposition, which is considered as a significant contributor to soil pollution [79]. The present findings are similar with the earlier results in the study region [46,47,75,80]. Hence, it can be concluded that factor 2 is associated with a mixed of natural metals and traffic emission within the river system.

4. Conclusion

Sediment samples from the nearby tannery industries (tannery village) were analyzed to determine the concentration of heavy metals. The average analysis of data depicted an order of heavy metals accumulation in sediment that was Cr (186 ± 541) > Fe (42.7 ± 49.1) > Pb (8.78 ± 6.2) > Mn (3.12 ± 1.4) > Cu (1.76 ± 1.5) respectively. The data showed that Cr got extreme concentration in the sediments, while Cu was minimally accumulated. The percentage of relative standard deviation (%RSD) for the studied heavy metals distribution in sediments at different sampling points showed that the abundance of Cr, Fe, Cu, Pb and Mn were varied a wide range (%RSD: 44.2–129%), which is consistent with the ANOVA test at a 95% confidence level. The results of spatial distribution pattern showed that the high concentrated zones of Cr was found at the dumping zone (~S5) of leather industrial city. On the other hand, the spatial distribution pattern for the studied metals (Pb, Mn, Cu and Fe) showed that the maximum metals concentrations were found in the southeastern parts, which were connected with the effluent discharging channel.

The pollution assessment in sediment samples was conducted following several pollution assessment indices: geo-accumulation index, contamination factor and pollution load index; which suggested that the study area had low level of pollution at this moment considering the studied heavy metals. Nevertheless, the ecological risk assessment in sediment samples revealed that there is no risk observed by the metal(*oid*)s as well as the *TRI* values were below 5 (*TRI* ≤ 5: no risk). The modified hazard quotient (*mHQ*) showed that about 75% of the samples were low to severely polluted by Cr, while 25% of the samples were extremely polluted by Cr as well as *mHQ* ≤ 3.5. Subsequently, sources of metals identification through the positive matrix factorization (PMF) modeling revealed that Cr (186%) inherited from leather tanning and chemical industrial activities while Fe (25.3%), and Pb (6.63%), contributed from the mixed effects of geogenic sources and traffic emission and atmospheric deposition.

Declaration of Competing Interest

The authors are declaring that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.enceco.2021.10.001>.

References

- [1] N. Saha, M.S. Rahman, Multivariate statistical analysis of metal contamination in surface water around Dhaka export processing industrial zone, Bangladesh, *Environ. Nanotechnol. Monit. Manag.* 10 (2018) 206–211, <https://doi.org/10.1016/j.enmm.2018.07.007>.
- [2] M. Malarkodi, R. Krishnasamy, R. Kumaraperumal, T. Chitdeshwari, Characterization of heavy metal contaminated soil of Coimbatore District in Tamil Nadu, *J. Agron.* 6 (1) (2007) 147–151.
- [3] M.A.H. Bhuiyan, N.I. Suruvi, S.B. Dampare, M.A. Islam, S.B. Quraishi, S. Ganyaglo, S. Suzuki, Investigation of the possible sources of heavy metal contamination in lagoon and canal water in the tannery industrial area in Dhaka, Bangladesh, *Environ. Monit. Assess.* 175 (1–4) (2010) 633–649.
- [4] M.G. Rasul, I. Faisal, M.M.K. Khan, Environmental pollution generated from process industries in Bangladesh, *Int. J. Environ. Pollut.* 28 (1–2) (2006) 144–161, <https://doi.org/10.1504/IJEP.2006.010881>.
- [5] M.S. Hossain, M.K. Ahmed, S. Sarker, M.S. Rahman, Seasonal variations of trace metals from water and sediment samples in the northern bay of Bengal, *Ecotoxicol. Environ. Saf.* 193 (2020) 110347.
- [6] M.R. Islam, J.S. Islam, G.M. Zatzman, M.S. Rahman, M.A.H. Mughal, The Greening of Pharmaceutical Engineering: Theories and Solutions, vol. 2, Wiley, USA, 2016 370.
- [7] W. Mertz, Trace Elements in Human and Animal Nutrition, 5th ed. Academic Press, Maryland, U.S.A, 1987 480.
- [8] M. Balali-Mood, K. Naseri, Z. Tahergorabi, M.R. Khazdair, M. Sadeghi, Toxic mechanisms of five heavy metals: mercury, lead, chromium, cadmium, and arsenic, *Front. Pharmacol.* (13 April 2021) (2021) <https://doi.org/10.3389/fphar.2021.643972>.
- [9] N. Saha, M.S. Rahman, M.B. Ahmed, J.L. Zhou, Industrial metal pollution in water and probabilistic assessment of human health risk, *J. Environ. Manag.* 185 (2017) 70–78, <https://doi.org/10.1016/j.jenvman.2016.10.023>.
- [10] S. Singh, W.G.L. Aalbersberg, R.J. Morrison, Nutrient Pollution in Laucala Bay, Fiji Islands, *Water Air Soil Pollut.* 204 (2009) 363.
- [11] P.J. Gibbs, A.G. Miskiewicz, Heavy metals in fish near major primary treatment sewage outfall, *Mar. Pollut. Bull.* 30 (10) (1995) 667–674.
- [12] G.M.R. Islam, M.R. Habib, J.L. Waid, M.S. Rahman, J. Kabir, S. Akter, Y.N. Jolly, Heavy metal contamination of freshwater prawn (*Macrobrachium rosenbergii*) and prawn feed in Bangladesh: a market-based study to highlight probable health risks, *Chemosphere* 170 (2017) 282–289, <https://doi.org/10.1016/j.chemosphere.2016.11.163>.
- [13] E.A.A. Olojo, K.B. Olurin, G. Mbaka, A.D. Oluwemimo, Histoathology of the gill and liver tissues of the African catfish *Clarias gariepinus* exposed to lead, *Afr. J. Biotechnol.* 4 (1) (2005) 117–122.
- [14] Lenntech, Water Treatment and Air Purification, Water Treatment, Publish by Lenntech, Rotterdamseweg, Netherlands, 2004.
- [15] T. Khan, S. Muhammad, B. Khan, H. Khan, Investigating the levels of selected heavy metals in surface water of Shah Alam river (a tributary of river Kabul, Khyber Pakhtunkhwa), *J. Himalayan Earth Sci.* 44 (2) (2011) 71–79.
- [16] M.S. Rahman, M.D.H. Khan, Y.N. Jolly, J. Kabir, S. Akter, A. Salam, Assessing risk to human health for heavy metal contamination through street dust in the southeast Asian megacity: Dhaka, Bangladesh, *Sci. Total Environ.* 660 (2019) 1610–1622, <https://doi.org/10.1016/j.scitotenv.2018.12.425>.
- [17] APHA, Standard Methods for the Examination of Water and Wastewater, 21st edition American Public Health Association/American Water Works Association/Water Environment Federation, Washington DC, 2005.
- [18] M.S. Rahman, N. Saha, A.H. Molla, Potential ecological risk assessment of heavy metal contamination in sediment and water body around Dhaka export processing zone, Bangladesh, *Environ. Earth Sci.* 71 (5) (2014) 2293–2308, <https://doi.org/10.1007/s12665-013-2631-5>.
- [19] A.M. Idris, Between-bottle homogeneity test of new certified reference materials employing wavelength dispersive X-ray fluorescence spectrometry, *BMC Chem.* 2019 (13) (2019) 23.
- [20] P. Bode, Instrumental and Organizational Aspects of a Neutron Activation Analysis Laboratory, Interfaculty Reactor Institute, Delft University of Technology, Delft, 1996 147.

- [21] M. Rahman, M.A. Islam, R.A. Khan, Characterization of chemical elements in common spaces of Bangladesh for dietary intake and possible health risk assessment by INAA and AAS techniques, *J. Radioanal. Nucl. Chem.* 318 (2018) 1347–1357.
- [22] P.C. Emenike, I.T. Tenebe, J.B. Neris, D.O. Omole, O. Afolayan, C.U. Okeke, I.K. Emenike, An integrated assessment of land-use change impact, seasonal variation of pollution indices and human health risk of selected toxic elements in sediments of river Atuwara, Nigeria, *Environ. Pollut.* 265 (2020) 114795.
- [23] R. Mazurek, J. Kowalska, M. Gąsior, P. Zdrożny, A. Józefowska, T. Zaleski, K. Orłowska, Assessment of heavy metals contamination in surface layers of Roztoż National Park forest soils (SE Poland) by indices of pollution, *Chemosphere* 168 (2017) 839–850, <https://doi.org/10.1016/j.chemosphere.2016.10.126>.
- [24] G. Müller, Index of geoaccumulation in sediments of the Rhine River, *Geojournal* 2 (1969) 108–118, <https://ci.nii.ac.jp/naid/10030367619/>.
- [25] L. Hakanson, An ecological risk index for aquatic pollution control. A sedimentological approach, *Water Res.* 14 (8) (1980) 975–1001.
- [26] K.K. Turekian, K.H. Wedepohl, Distribution of the elements in some major units of the earth's crust, *Geol. Soc. Am. Bull.* 72 (1961) 175–192.
- [27] D.C. Tomlinson, J.G. Wilson, C.R. Harris, D.W. Jeffrey, Problems in the assessment of heavy-metal levels in estuaries and the formation of a pollution index, *Helgol. Mar. Res.* 33 (1980) 566–575.
- [28] Z. Mokhtarzadeh, B. Keshavarzi, F. Moore, F.A. Marsan, E. Padoan, Potentially toxic elements in the Middle East oldest oil refinery zone soils: source apportionment, speciation, bioaccessibility and human health risk assessment, *Environ. Sci. Pollut. Res.* 27 (32) (2020) 40573–40591, <https://doi.org/10.1007/s11356-020-09895-7>.
- [29] N. Saha, M.S. Rahman, Y.N. Jolly, A. Rahman, M.A. Sattar, M.A. Hai, Spatial distribution and contamination assessment of six heavy metals in soils and their transfer into mature tobacco plants in Kushia district, Bangladesh, *Environ. Sci. Pollut. Res.* 23 (4) (2016) 3414–3426, <https://doi.org/10.1007/s11356-015-5575-3>.
- [30] Z.Q. Xu, S.J. Ni, X.G. Tuo, Calculation of heavy metals toxicity coefficient in the evaluation of potential ecological risk index, *Environ. Sci. Technol.* 31 (2008) 112–115.
- [31] G. Zhang, J. Bai, Q. Zhao, Q. Lu, J. Jia, X. Wen, Heavy metals in wetland soils along a wetland-forming chronosequence in the Yellow River Delta of China: levels, sources and toxic risks, *Ecol. Indic.* 69 (2016) 331e339, <https://doi.org/10.1016/j.ecolind.2016.04.042>.
- [32] S.S. Barjoe, S.Z.M. Abadi, M.R. Elmi, V.T. Varaoon, M. Nikbakht, Evaluation of trace elements pollution in deposited dust on residential areas and agricultural lands around Pb/Zn mineral areas using modified pollution indices, *J. Environ. Health Sci. Eng.* (2021) 1–17, <https://doi.org/10.1007/s40201-021-00643-8>.
- [33] N.U. Benson, A.E. Adedapo, O.H. Fred-Ahmadu, A.B. Williams, E.D. Udosen, O.O. Ayejuyo, A.A. Olajire, A new method for assessment of sediment-associated contamination risks using multivariate statistical approach, *MethodsX* 5 (2018) 268–276, <https://doi.org/10.1016/j.mex.2018.03.005>.
- [34] Ç.S. Eker, Distinct contamination indices for evaluating potentially toxic element levels in stream sediments: a case study of the Harşit stream (NE Turkey), *Arab. J. Geosci.* 13 (22) (2020) 1–18, <https://doi.org/10.1007/s12517-020-06178-w>.
- [35] F. Ustaoglu, M.S. Islam, Potential toxic elements in sediment of some rivers at Giresun, Northeast Turkey: a preliminary assessment for ecotoxicological status and health risk, *Ecol. Indic.* 113 (2020) 106237, <https://doi.org/10.1016/j.ecolind.2020.106237>.
- [36] Q. Yuan, P. Wang, C. Wang, J. Chen, X. Wang, S. Liu, T. Feng, Metals and metalloids distribution, source identification, and ecological risks in riverbed sediments of the Jinsha River, China, *J. Geochem. Explor.* 205 (2019) 106334, <https://doi.org/10.1016/j.jgexplo.2019.106334>.
- [37] D. Persuad, R. Jaagumagi, A. Hayton, Guidelines for the protection and management of aquatic sediment quality in Ontario, Ontario Ministry of the Environment, Canada, 1993.
- [38] D.D. MacDonald, C.G. Ingersoll, T.A. Berger, Development and evaluation of consensus-based sediment quality guidelines for freshwater ecosystems, *Arch. Environ. Contam. Toxicol.* 39 (1) (2000) 20–31.
- [39] E.R. Long, D.D. MacDonald, S.L. Smith, F.D. Calder, Incidence of adverse biological effects within ranges of chemical concentrations in marine and estuarine sediments, *Environ. Manag.* 19 (1995) 81–97.
- [40] M.A.H. Bhuiyan, S.C. Karmaker, M. Bodrud-Doza, M.A. Rakib, B.B. Saha, Enrichment, sources and ecological risk mapping of heavy metals in agricultural soils of Dhaka district employing SOM, PMF and GIS methods, *Chem.* 263 (2021) 128339.
- [41] USEPA, The United States Environmental Protection Agency, EPA positive matrix factorization (PMF) 5.0 fundamentals and user guide, 2014 https://www.epa.gov/sites/production/files/2015-02/documents/pmf_5.0_user_guide.pdf.
- [42] A.R.M.T. Islam, M. Hasanuzzaman, H.M.T. Islam, et al., Quantifying source apportionment, co-occurrence and Ecotoxicological risk of metals from up-mid-Downstream River segments, Bangladesh, *Environ. Toxicol. Chem.* 39 (10) (2020) 2041–2054, <https://doi.org/10.1002/etc.4814>.
- [43] R.L. Rudnick, S. Gao, Composition of the continental crust, *Treat. Geochem.* 4 (2014) 1–64 2nd ed.
- [44] CNEMC, China National Environmental Monitoring Center. The Background Values of Elements in Chinese Soils. 330–493 (Environmental Science Press of China, 1990), 1990.
- [45] M.S. Rahman, M.B. Hossain, S.M.O.F. Babu, M. Rahman, A.S.S. Ahmed, Y.N. Jolly, T.R. Choudhury, B.A. Begum, J. Kabir, S. Akter, Source of metal contamination in sediment, their ecological risk, and phytoremediation ability of the studied mangrove plants in ship breaking area, Bangladesh, *Mar. Pollut. Bull.* 141 (2019) 137–146.
- [46] R. Khan, M.S. Islam, A.R.M. Tareq, K. Naher, A.R.M.T. Islam, et al., Distribution, sources and ecological risk of trace elements and polycyclic aromatic hydrocarbons in sediments from a polluted urban river in Central Bangladesh, *Environ. Nanotechnol. Monit. Manag.* 14 (2020) 100318, <https://doi.org/10.1016/j.enmm.2020.100318>.
- [47] R. Khan, M.S. Islam, A.R.M. Tareq, K. Naher, A.R.M.T. Islam, M.A. Habib, M.A.B. Siddique, M.A. Islam, S. Das, M.B. Rashid, A.K.M.A. Ullah, M.M.H. Miah, S.U. Masrura, M. Bodrud-Doza, M.R. Sarker, A.B.M. Badruzzaman, Elemental and polycyclic aromatic hydrocarbons distributions in the sediments of an urban river: influence of anthropogenic runoff, *Environ. Nanotechnol. Monit. Manag.* 14 (2020) 100318, <https://doi.org/10.1016/j.enmm.2020.100318>.
- [48] E. Garzanti, S. Ando, C. France-Lanord, G. Vezzoli, P. Censi, V. Galy, Y. Najman, Mineralogical and chemical variability of fluvial sediments 1. Bedload sand (ganga-Brahmaputra, Bangladesh), *Earth Planet. Sci. Lett.* 299 (2010) 368–381.
- [49] E.K. Atibu, N. Devarajan, A. Laffite, G. Giuliani, J.A. Salumu, R.C. Muteb, C.K. Mulaji, J. Otamanga, V. Elongo, P.T. Mpiana, J. Pote, Assessment of trace metal and rare earth elements contamination in rivers around abandoned and active mine areas. The case of Lubumbashi River and Tshamilemba Canal, Katanga, Democratic Republic of the Congo, *Geochemistry* 76 (3) (2016) 353–362.
- [50] M.S. Elias, S. Ibrahim, K. Samudung, S.A. Rahman, Y.M. Wo, J.A.D. Daung, Multivariate analysis for source identification of pollution in sediment of Linggi River, Malaysia, *Environ. Monit. Assess.* 190 (2018) 257.
- [51] Y. Kim, B. Kim, K. Kim, Distribution and speciation of heavy metals and their sources in Kumho river sediment, Korea, *Environ. Earth Sci.* 60 (2010) 943–952.
- [52] B.J. Mathis, T.F. Cummings, Selected metals in sediments, water, and biota in the Illinois river, *J. Water Pollut. Control Fed.* 45 (7) (1973) 1573–1583.
- [53] J.N. Pattan, G. Parthiban, C.P. Babu, N.H. Khadge, A.L. Paropkari, V.N. Kodagali, A note on geochemistry of surface sediments from Krishna-Godavari Basin, East Cost of India, *J. Geol. Soc. India* 71 (2008) 107–114.
- [54] M.M.V.G. Silva, M.M.S.C. Pinto, P.C.S. Carvalho, Major, trace and REE geochemistry of recent sediments from lower Catumbela River (Angola), *J. Afr. Earth Sci.* 115 (2016) 203–217.
- [55] O.A. Sorokina, N.V. Zarubina, Chemical composition of the bottom sediments in the middle reaches of the Amur River, Russian J. Pacific Geol. 5 (5) (2011) 469–476.
- [56] C. Zhang, L. Wang, Multi-element geochemistry of sediments from the Pearl River system, China, *Appl. Geochem.* 16 (2001) 1251–1259.
- [57] G.B. Jones, M.B. Jordan, The distribution of organic material and trace metals in sediments from the river Liffey estuary, Dublin, *Estuar. Coast. Mar. Sci.* 8 (1979) 37–47.
- [58] R. Pehlivan, The effect of weathering in the Buyukmelen river basin on the geochemistry of suspended and bed sediments and the hydrogeochemical characteristics of river water, Duzce, Turkey, *J. Asian Earth Sci.* 39 (2010) 62–75.
- [59] U. Tamim, R. Khan, Y.N. Jolly, K. Fatema, S. Das, K. Naher, M.A. Islam, S.M.A. Islam, S.M. Hossain, Elemental distribution of metals in urban river sediments near an industrial effluent source, *Chemosphere* 155 (2016) 509–518.
- [60] M.S. Rahman, M. Ullah, Y.N. Jolly, S. Akhter, J. Kabir, B.A. Begum, A. Salam, Elemental analysis in surface soil and dust of roadside academic institutions in Dhaka city, Bangladesh and their impact on human health, *Environ. Chem. Ecotoxicol.* 3 (2021) 197–208.
- [61] US EPA, U.S. Environmental Protection Agency, Screening level ecological risk assessment protocol for hazardous waste combustion facilities. Vol. 3, Appendix E: Toxicity reference values. EPA 530-D99-001C, 1999.
- [62] M.O. Fashola, V.M. Ngole-Jeme, O.O. Babalola, Heavy Metal Pollution from Gold Mines: Environmental Effects and Bacterial Strategies for Resistance, *Int. J. Environ. Res. Public Health* 13 (2016) 1047.
- [63] Y. Gao, H. Ji, Microscopic morphology and seasonal variation of health effect arising from heavy metals in PM_{2.5} and PM₁₀: one-year measurement in a densely populated area of urban Beijing, *Atmos. Res.* 212 (2018) 213–226.
- [64] M.S. Rahman, S.S. Bhuiyan, Z. Ahmed, N. Saha, B.A. Begum, Characterization and source apportionment of elemental species in PM_{2.5} with especial emphasis on seasonal variation in the capital city “Dhaka”, Bangladesh, *Urban Clim.* 36 (march 2021) (2021) 100804, <https://doi.org/10.1016/j.uclim.2021.100804>.
- [65] R.C. Patra, A.K. Rautray, D. Swarup, Oxidative stress in lead and cadmium toxicity and its amelioration, *Vet. Med. Int.* 2011 (2011) <https://doi.org/10.4061/2011/457327>.
- [66] J.E. Zhang, J.L. Liu, Y. Ouyang, B.W. Liao, B.L. Zhao, Removal of nutrients and heavy metals from wastewater with mangrove Sonneratia apetala Buch-ham, *Ecol. Eng.* 36 (2010) 807–812, <https://doi.org/10.1016/j.ecoleng.2010.02.008>.
- [67] L. Brüggemann, Metals in sediments and suspended matter of the river Elbe, *Sci. Total Environ.* 159 (1995) 53–65.
- [68] J.S. Aboud, N. Nandini, Impact assessment of heavy metals pollution of Vartur lake, Bangalore, *J. Appl. Nat. Sci.* 1 (2009) 53e61.
- [69] Z. Madzin, M.F. Shai-in, F.M. Kusin, Comparing heavy metal mobility in active and abandoned mining sites at bestari jaya, selangor, *Procedia Environ. Sci.* 30 (2015) 232e237.
- [70] M.S. Rahman, Y.N. Jolly, S. Akter, N.A. Kamal, R. Rahman, T.R. Choudhury, B.A. Begum, Sources of toxic elements in indoor dust sample at export processing zone (EPZ) area: Dhaka, Bangladesh; and their impact on human health, *Environ. Sci. Pollut. Res.* (2021) <https://doi.org/10.1007/s11356-021-13167-3> accepted on Feb., 22, 2021.
- [71] A. Kabata-Pendias, H. Pendias, Trace Elements in Soils and Plants, CRC Press, Boca Raton, FL, USA, 2001.
- [72] G. Abraham, R. Parker, Assessment of heavy metal enrichment factors and the degree of contamination in marine sediments from Tamaki Estuary, Auckland, New Zealand, *Environ. Monit. Assess.* 136 (1–3) (2008) 227–238 <https://doi.org/10.1007/s10661-007-9678-2>.
- [73] J. Tao, L. Zhang, J. Cao, L. Zhong, D. Chen, Y. Yang, D. Chen, L. Chen, Z. Zhang, Y. Wu, Y. Xia, S. Ye, R. Zhang, Source apportionment of PM_{2.5} at urban and suburban areas of the Pearl River Delta region, south China – with emphasis on ship emissions, *Sci. Total Environ.* 574 (2017) 1559–1570.
- [74] J.S. Lv, Y. Liu, Z.L. Zhang, J.R. Dai, Factorial kriging and stepwise regression approach to identify environmental factors influencing spatial multi-scale variability of heavy metals in soils, *J. Hazard. Mater.* 261 (13) (2013) 387e397, <https://doi.org/10.1016/j.jhazmat.2013.07.065>.
- [75] M.A.H. Bhuiyan, S.B. Dampare, M.A. Islam, S. Suzuki, Source apportionment and pollution evaluation of heavy metals in water and sediments of Buriganga River, Bangladesh, using multivariate analysis and pollution evaluation indices, *Environ. Monit. Assess.* 187 (2015) 4075, <https://doi.org/10.1007/s10661-014-4075-0>.

- [76] W. Cheng, S. Le, Z. Bian, Y. Zhao, Y. Li, Y. Gan, Geographic distribution of heavy metals and identification of their sources in soils near large, open-pit coal mines using positive matrix factorization, *J. Hazard. Mater.* 387 (2020) 121666, <https://doi.org/10.1016/j.jhazmat.2019.121666>.
- [77] M.S. Islam, M.K. Ahmed, M. Raknuzzaman, M.H. Al-Mamun, M.K. Islam, Heavy metal pollution in surface water and sediment: A preliminary assessment of an urban river in a developing country, *Ecol. Indic.* 48 (2015) 282–291.
- [78] S. Kumar, A.R.M.T. Islam, M. Hasanuzzaman, S. Roquia, R. Khan, M.S. Islam, Preliminary assessment of heavy metals in surface water and sediment in Nakuvadra-Rakiraki River, Fiji using indexical and chemometric approaches, *J. Environ. Manag.* 298 (2021) 113517.
- [79] S. Kumar, A.R.M.T. Islam, H.M.T. Islam, M. Hasanuzzaman, V. Ongoma, R. Khan, J. Mallick, Water resources pollution associated with risks of heavy metals from Vatukoula Goldmine region, Fiji, *J. Environ. Manag.* 293 (2021) 112868, <https://doi.org/10.1016/j.jenvman.2021.112868>.
- [80] P.G. Whitehead, G. Bussia, R. Peters, M.A. Hossain, L. Softley, S. Shawal, L. Jind, P.N. Rampleye, R. Holdship, R. Hope, G. Alabaster, Modelling heavy metals in the Buriganga river system, Dhaka, Bangladesh: impacts of tannery pollution control, *Sci. Total Environ.* 697 (2019) 134090, <https://doi.org/10.1016/j.scitotenv.2019.134090>.
- [81] J.P. Brady, G.A. Ayoko, W.N. Martens, A. Goonetilleke, Development of a hybrid pollution index for heavy metals in marine and estuarine sediments, *Environ. Monit. Assess.* 187 (5) (2015) 1–14, <https://doi.org/10.1007/s10661-015-4563-x>.
- [82] L. Derek, D.L. Tomlinson, J. Wilson, C.R. Harris, D.W. Jeffrey, Problems in assessment of heavy metals in estuaries and the formation of pollution index, *Helgoländer Meeresuntersuchungen* 33 (1) (1980) 566–575, <https://doi.org/10.1007/BF02414780>.
- [83] J. Hu, B. Lin, M. Yuan, Z. Lao, K. Wu, Y. Zeng, H. Fan, Trace metal pollution and ecological risk assessment in agricultural soil in Dexing Pb/Zn mining area, China, *Environ. Geochem. Health* 41 (2) (2019) 967–980, <https://doi.org/10.1007/s10653-018-0193-x>.