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RESEARCH ARTICLE

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Assessment of trace element toxicity in surface water of a fish breeding river in Bangladesh: a novel approach for ecological and health risk evaluation

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ABSTRACT

In recent decades, water quality is a great concern for the human being as it is the most important natural resource but water is polluted by increasing population activities. In this study, surface water samples were collected in the wet and the dry season from Halda River, Bangladesh, and 12 physicochemical properties and 10 traces elements (Cd, Cr, Mn, Fe, Co, Cu, Zn, As, Pb, and Hg) were analyzed to assess the metal toxicity, identify the possible sources, and determine possible carcinogenic and noncarcinogenic risk for adults and child residents. During wet season, the mean concentrations of Cd, Cr, Mn, Fe, Co, Cu, Zn, As, Pb, and Hg were 0.032 ± 0.028, 0.004 ± 0.002, 0.010 ± 0.012, 0.38 ± 0.08, 0.010 ± 0.017, 3.33 ± 1.16, 0.062 ± 0.008, 1.34 ± 1.17, 0.030 ± 0.014, and 0.007 ± 0.002 mg/L, respectively, whereas the concentrations were measured as 0.04 ± 0.017, 0.060 ± 0.036, 0.150 ± 0.134, 0.58 ± 0.11, 0.049 ± 0.017, 0.172 ± 0.166, 0.393 ± 0.212, 1.07 ± 0.989, 0.104 ± 0.121, and 0.003 ± 0.002 mg/L, respectively, during the dry season. The levels of trace elements in water samples were much higher than the guideline values for safe limits of drinking water and the protection of freshwater aquatic life, indicated that water from this river is not safe for drinking and/or cooking purposes. Trace element evaluation index indicated that water was found to be high contamination while ecological risk index showed low to very high contamination. All hazard quotient and hazard index (HI) values were higher than the risk threshold of unity. HI values for child were higher than those for adult, indicating that the health of children is at dramatically higher risk than adults. Arsenic for water ingestion and dermal pathways was the primary contributors to total health risk (HI/THI) indicated that As in surface water of the Halda River might pose health risks to residential users.

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1. Introduction

Water is an essential requisite for all life forms on earth (Bytyçi *et al.* 2018), and the main source of water are rivers, lakes, glaciers, rain water, ground water etc. Besides the need of water for drinking, it is also known as the most important irreplaceable natural resource on which the sustainable development of a country depends to a large extent (Yıldız 2017, Pobi *et al.* 2019). However, despite its importance, water is the most poorly managed resource in the world and is facing serious threat as a result of various anthropogenic activities (Reza and Singh 2010, Islam *et al.* 2013). As a means of different activities from human being, indiscriminate growth of industries, increasing population, and urbanization around the world are considered as the principal causes of global water pollution, particularly in the urban riverine environment where rivers are to be considered as the endpoint of effluents discharged by industries (Saha and Paul 2019, Pandey *et al.* 2019, Kumar *et al.* 2020). Owing to the weakness in law enforcement and lack of regular monitoring for most of the developing countries like Bangladesh, different industries discharge untreated industrial effluents into the surrounding surface water which subsequently deteriorates the overall quality of the water body by introducing various water pollutants including toxic trace elements (TEs) (Edokpayi

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Supplemental data for this article can be accessed here.

et al. 2017, Ali *et al*. 2018, Islam 2021). Rivers are the most studied environments, as these are the environments where freshwater is more accessible for population (Alves *et al*. 2014). Rivers play an important role in the acceptance and transport of toxic pollutants by receiving waters of point source (industrial, mining) and nonpoint source (city lives, agriculture, atmospheric precipitation, etc.) pollutants with a view that the dilution strength is sufficient enough (Tenebe *et al*. 2017). Rivers are by far the cheapest form of water supply compared to other sources like groundwater and seawater desalination.

TEs are a group of metals or metalloids which possess a specific density greater than 5 g/cm^3 (Monisha *et al.* 2014). The most commonly detected TEs in the industrial effluents include cadmium (Cd), chromium (Cr), manganese (Mn), iron (Fe), cobalt (Co), copper (Cu), zinc (Zn), arsenic (As), lead (Pb), and mercury (Hg) (Lambert *et al.* 2000). However, TEs are of great concern for the aquatic environment since these elements have persistent behavior and have the propensity to bio accumulate in the food chain (Rahman *et al.* 2013, Islam *et al.* 2015a, Ali *et al*. 2019). TEs contamination in natural water exerts severe toxic effects on aquatic organisms, which ultimately leads to the complete disruption of normal ecosystem function (Islam *et al.* 2015b, Raknuzzaman *et al.* 2016, Ali *et al*. 2018). Moreover, these toxic TEs can enter the human body not only through ingestion of contaminated water and/or aquatic organisms, but also via dermal contact with the contaminated water (Saha and Paul 2019). Human body can be exposed to TEs through several exposure pathways, mostly dependent on pollution mediums such as water, air, food, and soil. However, among these pathways, exposure via drinking of TEs contaminated water is a critical pathway for metals exposure to human body (Muchuweti *et al*. 2006, Ricolfi *et al*. 2020). Exposure to these toxic elements can lead to physical, muscular, and neurological disorders (Ali *et al*. 2019, Saha and Paul 2019), while long-term exposure can also cause deadly diseases like Alzheimer's, Parkinson's, multiple sclerosis, and cancer (Mishra *et al.* 2010). Thus, proper assessment of surface water quality along with the TEs pollution load is of immense importance for the protection of environment and human health.

Bangladesh is a riverine country consisting of more than 230 large and small rivers (Hasan *et al*. 2019). However, almost all the rivers flowing through the country are heavily contaminated with TEs originated from anthropogenic intervention, mainly by indiscriminate disposal of untreated industrial effluents (Islam *et al.* 2016a, Kibria *et al.* 2016, Hasan *et al.* 2019, Ustaoğlu and Islam 2020). Many communities in Chittagong hill tract, Bangladesh for instance, are located along Chittagong a tributary of Halda River, Bangladesh. Some individuals in these communities have turned to the river for their daily domestic water requirements such as washing, cooking, drinking, and feeding of livestock among others when other sources are not really accessible (Emenike *et al.* 2017). Moreover, other major water bodies such as Halda River, Bangladesh at some point which had being initially polluted with industrial effluents and sewage from the upstream section and along the river. With contaminated water having the tendency to mix especially after a flood event, the users have the likelihood of consuming it unknowingly (Tenebe *et al.* 2016). In the same vein, contaminated fish obtained from this river are sold to the public thereby exposing people to possible contamination. Moreover, these TEs, which may have significant contribution to contamination of aquatic ecosystems and health risk assessment, are received little attention when compared to other more often studied elements. Therefore, there is necessity of determining 10 toxic elements for assessing the level of pollution of this important urban river in Bangladesh and the associated health risk to humans. Potential health risks for residential receptors (adult and child) include ingestion of water as drinking purposes and dermal contact while showering, while recreational receptors are incidental ingestion of water and dermal contact while swimming (USEPA 1989, 2020a). Both of the exposure routes can lead to human health risks, and thus their effects have to be investigated and addressed. To date, no scientific research regarding contamination and health risk issues of studied TEs from surface and deep water of Korotoa River has been conducted so far.

Therefore, this study was conducted to determine the contamination levels of 10 TEs in the river water and identify the possible sources of contamination using multivariate statistical tools; to compare the levels of TEs with both international and national water quality guidelines; to evaluate the water quality index for human consumption and ecological risk index (ERI) and to assess cancer and noncancer health risks of TEs for residential adults and children through oral and dermal contact pathways from surface water of Halda River.

2. Materials and method

2.1. Climatic and hydrogeological conditions of the study area

Halda River is located at the south-eastern part of Bangladesh. It originates at the Badnatali Hill Ranges in Ramgarh Upazila in the Chittagong Hill Tracts, flows through Fatikchhari Upazila, Bhujpur Thana, Hathazari Upazila, Raozan Upazila, and Chandgaon Thana of the Chittagong Metropolitan City, Bangladesh, and falls into the Karnaphuli River (Ali *et al.* 2016, Hossain *et al.* 2021). Hydrology of the Halda River presents a complicated interaction of fresh water flow from the upstream, the tides and tidal flows from the Bay of Bengal Tropical Cyclones, storm surge and other meteorological effect from the sea and the physiography of the coastal plains. Hydrological setting, topography, and geology present a general hydrologic environment of the study area. Rainfall, tidal levels, and flows and salinity distribution are major hydrologic elements. Rainfall is resources for agricultural activity but along with dominant tide levels may be create serious drainage problems. The flat deltaic lands are interlaced by an intricate river and tidal channel system which cuts the land into numerous separate areas. The agricultural lands in the study area have been formed by piedmont alluvial deposits transported from the Chittagong Hills by local streams and rivers, some land were formed by beach and tidal flat deposits. Soils in this district are generally younger and coarse textured than those in the remainder of the coastal belt, but show similar variations of sands, silts and occasional clays. During dry season, average rainfall was 9.6 mm, temperature was 21.9 °C, and relative humidity was 76% and during wet season, average rainfall was 1166 mm, temperature was 32.3 °C, and relative humidity was 89%.

2.2. Study area and sample collection

Sampling sites along the Halda River in Bangladesh was selected Madarikhal area near to Modunaghat area. The selection of these points were based upon human activities taking place near the river banks and in the river such as harvesting and farming, geographical proximity of industrial and urban discharges of effluent to the river, proximity of residential areas near the river banks, inflow regions of the river, drainage patterns, and accessibility. In this study, water samples were collected from twelve different stations (S1, S2, S3, S4, S5, S6, S7, S8, S9, S10, S11, and S12) from upstream to downstream of the river (Figure 1). Water samples were collected at a distance of 100 m in Madarikhal area. About 72 composite water samples were collected from 10–15 cm below the water surface. Two seasons, namely wet (August–September, 2019) and dry (January–February, 2020), were selected for water sample collection when Halda River was at high and low

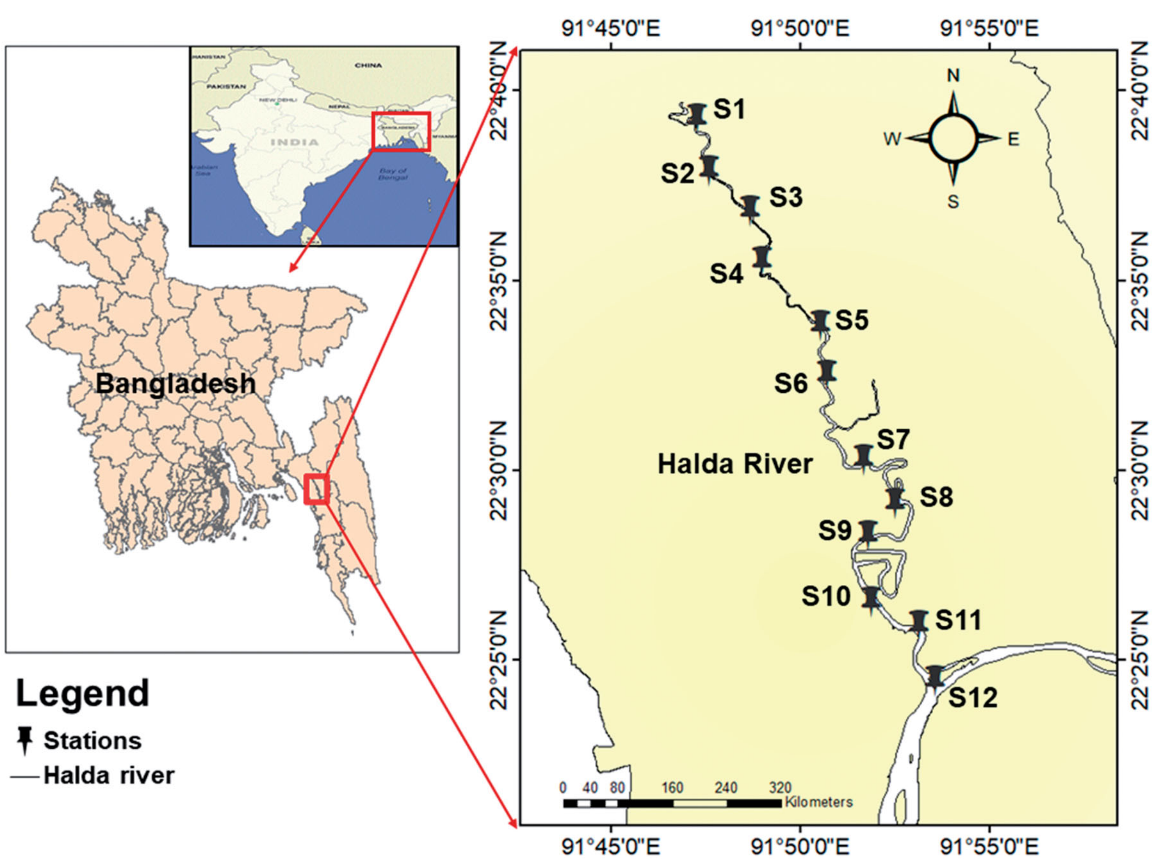

Figure 1. Map of the study area of the Halda River, Bangladesh.

water levels. During wet season, river water levels increase due to heavy rainfall; during dry season, there is no rainfall and river water levels decrease. Considering the water flow in the studied river, dry season exhibited lower than wet season which can cause the variation of TEs concentration in water. The collected samples were placed into pre-cleaned plastic bottles, sealed, labeled, and moved to the laboratory, stored at a temperature of 4 °C for subsequent analysis.

2.3. Major anions and cations measurement

Seven anion such as Fluoride (F^-), Chloride (Cl^-), Nitrate (NO_3^-), Sulfate (SO_4^{2-}), Nitrite (NO_2^-), Bromide (Br^-), Phosphate (PO_4^{3-}) and six major cation Lithium (Li^+), Sodium (Na^+), Potassium (K^+), Magnesium (Mg^{2+}), Calcium (Ca^{2+}), Ammonium (NH_4^-) were analyzed by Ion-chromatography DIONEX-3000 in Chemistry Division of Atomic Energy Center, Dhaka (AECD). Water samples were filtered through Whatman 41 filter paper and then inject into the dionex for anion and cation analysis. Condition of ion analysis by IC is given in Supplementary Table S1.

2.4. Water samples preparation for trace metals assessment

All chemicals used in this study were of analytical grade, while deionized water was used for solution preparation. About 500 ml of river water was filtered using Whatman® 41 filter paper. Water samples were filtered through 0.45 µm filters, cellulose nitrate. Then the water was acidified by adding 1 ml of 65% HNO_3 acid, taken into two individual 300 ml beaker by dividing the water samples into 250 ml, reduced to 25 ml by using hotplate. For metal analysis, 20 ml water sample was treated with 5 ml 69% HNO_3 acid and 2 ml 30% H_2O_2 in a closed Teflon vessel and was digested in a Microwave Digestion System. For arsenic analysis 0.5 ml of potassium iodide, 0.5 ml of ascorbic acid and 3 ml of HNO_3 was added. Further dilution was made as per required for metal analysis by Atomic Absorption Spectrophotometer (AAS).

2.5. Determination of TEs

Atomic Absorption Spectrophotometer is used to determine heavy and trace metal in water sample and each time a triplicate measurement was carried out for the better accuracy and precision. Standard reference material (SRM) obtained from National Institution of Standard and Technology (NIST), USA was used to check the concentration of laboratory prepared standard as minimum three different concentrations for each metal is required for constructing the calibration curve. The deviation was found to be negligible.

2.6. Quality control and quality assurance

Calibration curve for each measured element was prepared using at least three different concentrations level from commercially available sing element standard. Individual calibration curve for each element was used to determine the individual element. It was found that coefficient of variation (R^2 value) for the calibration curve derived from statistical regression analysis was almost 0.9995 for each element that analyzed for the study. For quality assurance, procedural blank was used throughout the sample preparation process to avoid any contamination from the reagent, container etc. sample preparation and experimental results were validated by carrying out all operation in triplicate, moreover SRMs were accounted in each case.

2.7. Trace element evaluation index (TEI)

The TEI provides an overall quality of the water with respect to heavy metals (Edet and Offiong 2002) and was calculated as:

$$\mathrm{TEI} = \sum_{i=1}^{n} Tc/T\mathrm{mac} \tag{1}$$

Where Tc is the monitored value of the ith parameter and Tmac is the maximum admissible concentration of the ith parameter. The maximum admissible concentration for Cd, Cr, Mn, Fe, Co, Cu, Zn, As, Pb, and Hg are 0.003, 0.05, 0.5, 0.3, 0.05, 2.0, 3.0, 0.01, 0.01, and 0.006 mg/L, respectively (WHO 2011). The TEI is classified as: low (less than 10), medium (between 10 and 20) and high (more than 20).

2.8. Ecological risk assessment

The ERI for the water was calculated using the functions described in Equations 3 and 4 (Taiwo *et al.* 2019):

$$\mathrm{ERI} = \sum \mathrm{RI} = \sum \mathrm{Ti} \times \mathrm{PI} \tag{2}$$

$$\mathrm{PI} = C_s/C_b \tag{3}$$

Where RI is the potential ecological risk factor of each heavy metal; *Ti* is the toxic-response factor of heavy metal; PI is the pollution index; C_s is the concentration of heavy metals in the sample; and C_b is the corresponding background values. The corresponding background value are 0.01, 0.05, 0.1, 1, 0.5, 1, 5, 0.05, 0.05, and 0.05 for Cd, Cr, Mn, Fe, Co, Cu, Zn, As, Pb, and Hg, respectively (DoE

1997). The toxic-response factor of each studied trace metals are 30, 2, 1, 2.7, 2.3, 5, 1, 10, 5, and 40 for Cd, Cr, Mn, Fe, Co, Cu, Zn, As, Pb, and Hg, respectively (Taiwo *et al.* 2019). The ERI value <150 indicates low ecological risk, $150 < \mathrm{RI} < 300$ indicates moderate ecological risk, $300 < \mathrm{RI} < 600$ indicates considerable ecological risk, and ERI > 600 indicates very high ecological risk (Taiwo *et al.* 2019, Proshad *et al.* 2020).

2.9. Human health risk assessment

Noncarcinogenic health risks of TEs were quantified using the hazard quotients (HQs). The carcinogenic risks (CRs) were estimated only for TEs having carcinogenic slope factors (CSF) (Varol 2019). Noncarcinogenic and carcinogenic health risks of TEs via ingestion and dermal absorption of dam lake water were separately calculated for residential and recreational receptors (adult and child) using the following equations (USEPA 2020a, 2020b).

Noncarcinogenic risks for residential receptors:

$$HQ_{\mathrm{ingestion}} = \frac{C_W \times IRW_{\mathrm{res}} \times EF_{\mathrm{res}} \times ED \times EV \times ET_{\mathrm{res}}}{BW \times AT_{\mathrm{res}} \times RfD_o \times 10^3} \tag{4}$$

$$HQ_{\mathrm{dermal}} = \frac{C_W \times SA \times K_P \times ET_{\mathrm{res}} \times EV \times EF_{\mathrm{res}} \times ED}{BW \times AT_{\mathrm{res}} \times RfD_o \times \ GIABS \ \times 10^6} \tag{5}$$

Also, hazard index (HI) was calculated in order to account for the total non-CRs of all individual TEs combined in each exposure pathway. Total HI (THI) was calculated by summing the HIs in each exposure pathway.

$$HI = \Sigma Hqs \tag{6}$$

$$THI = HI_{\mathrm{ingestion}} + HI_{\mathrm{dermal}} \tag{7}$$

HQ, HI, and THI values > 1 indicates that there may be a potential for adverse noncarcinogenic health effects to occur, while HQ, HI, and THI values < 1 indicates that noncarcinogenic health effects are not expected (USEPA 2004).

Carcinogenic risks for residential receptors:

$$CR_{\mathrm{ingestion}} = \frac{C_W \times IFW_{\mathrm{res}} \times CSF_0}{AT \times 10^3} \tag{8}$$

$$IFW_{\mathrm{res}} = \frac{EF_{\mathrm{res}} \times ED_a \times IRW_{\mathrm{res}-a}}{BW_a} + \frac{EF_{\mathrm{res}} \times ED_c \times IRW_{\mathrm{res}-c}}{BW_c} \tag{9}$$

$$CR_{\mathrm{dermal}} = \frac{C_W \times K_p \times \ 0.001 \times ET_{\mathrm{event-res}} \times DFW_{\mathrm{res}} \times CSF_0}{AT \times \ GIABS \times 10^3} \tag{10}$$

$$ET_{\mathrm{event-res}} = \frac{ET_{\mathrm{res}-a} \times ED_a + \ ET_{\mathrm{res}-c} \times ED_c}{ED} \tag{11}$$

$$DFW_{\mathrm{res}} = \frac{EV_a \times EF_{\mathrm{res}} \times ED_a \times SA_a}{BW_a} + \frac{EF_{\mathrm{res}} \times EV_c \times IRW_{\mathrm{res}-c} \times ED_c}{BW_c} \tag{12}$$

Total cancer risk (TCR) was obtained by the sum of cancer risks (CRs) calculated for two pathways (ingestion and dermal exposure).

$$TCR = \ CR_{\mathrm{ingestion}} + \ CR_{\mathrm{dermal}} \tag{13}$$

The exposure of TEs for CR and TCR values between 10^{-4} and 10^{-6} indicates that CR is considered an acceptable range. The CR and TCR values $<10^{-6}$ indicates that CR is considered to be negligible, while these values $> 10^{-4}$ indicates that CR is considered as unacceptable level (USEPA 1991a, 1991b).

For human health risk assessment, all definitions, units, and values associated with these equations are presented in Supplementary Table S2, while Supplementary Table S3 shows the values of oral reference dose (RfDo), gastrointestinal absorption (GIABS), dermal permeability constant (K_p), and oral slope factor (CSFo). In this study, the author used Regional Screening Level calculator developed by USEPA (2020c, 2020d) to confirm HQs and CRs values of TEs from river water for residential and recreational receptors.

2.10. Data treatment and statistical analyses

The data were statistically analyzed by using the statistical package software SPSS 20.0 (International Business Machines Corporation [IBM] Armonk, NY). The means and standard deviations of the metal concentrations in water were calculated. For calculations, when the concentration of a TE was below the LOD, the concentration was assumed to be half of the respective LOD and if more than 60% of the water samples were lower than the LOD, these values below the LOD were replaced with by zero. One-way ANOVA was applied to determine differences in TE concentrations among sampling sites ($p < 0.05$, Duncan's test). Also, Student's *t*-test was used to determine significant differences in TE concentrations between seasons and between surface water samples and deep water samples ($p < 0.05$). Multivariate methods in terms of principal component analysis (PCA) were used to interpret the potential sources of heavy metal in water (Liang *et al.* 2015, Proshad *et al.* 2020). The Eigen values were used as the extraction method to find out the principal components (PC) during PCA analysis.

3. Result and discussion

3.1. Variation in physicochemical parameter in water samples

Mean values of physical–chemical parameters of surface water samples from Halda River, Bangladesh are shown in Table 1. The pH of the aquatic systems is an important indicator of the water quality and the extent pollution in the watershed areas. It is the indicator of acidic or alkaline condition of water status. The standard for any purpose in-terms of pH is 6.5–8.5, in that respect; the mean value of pH in this study is 6.02 which indicates slightly acidic water. The pH value represents the acidity or alkalinity of the water (Ravikumar *et al*. 2013). A pH value ranged between 6 and 8.5 indicates a productive water body (Garg *et al*. 2010), whereas the water with a low pH value is considered to be corrosive and can adversely affect the skin and eyes (Li *et al*. 2017). APHA (2012) stated that industrial or municipal waste materials played a significant role in increasing or decreasing pH of the adjacent water body. Moreover, activities like bathing, washing, and latrines along water bodies related to fluctuations of water pH (Islam *et al*. 2018a). Biochemical and chemical reactions are influenced by the pH (Manjare *et al*. 2010). Ali *et al*. (2016) studied the water quality of Karnaphuli River, one of the major coastal rivers in Bangladesh, and also found similar result of this study.

Electrical conductivity (EC) is usually used to indicate the total concentration of ionized constituents of water (Huq and Alam 2005). The most influential water quality guideline on crop productivity is the water salinity hazard as measured by EC (Zaman *et al*. 2018). The primary effect of high EC water on crop productivity is the inability of the plant to compete with ions in the soil solution for water (physiological drought). The higher the EC, the less water is available to plants, even though the soil may appear wet. Because plants can only transpire "pure" water, usable plant water in the soil solution decreases dramatically as EC increases. In this study, EC ranged between 31.3 and 33.7 μS/cm (Table 1) which indicates no threat for water quality.

Total dissolved solids indicate the amount of chemical substances dissolved in the water. At increasing levels, palatability decreases. Levels in excess of 1000 mg/L may produce a bad taste. Water used for irrigation can vary greatly in quality depending upon type and quantity of dissolved salts. TDS consists of oxygen-demanding wastes, disease-causing agents, which can cause immense harm to public health. The presence of synthetic organic chemicals (fuels, detergents, paints, solvents, etc.) imparts objectionable and offensive tastes, odors and colors to fish and aquatic plants even when they are present in low concentrations (Sawyer *et al*. 1994, Leo and Dekkar 2000). Dissolved ions affect the pH of water, which in turn may influence the health of aquatic species. In our study, TDS level is below 20.30 mg/L which is far below the maximum tolerable limit set by WHO.

3.2. Anion and cation in Halda river water samples

Water samples were analyzed for common inorganic anions fluoride, chloride, nitrite, sulfate, and major cation sodium, ammonium, potassium, magnesium, and calcium by ion exchange chromatography. The concentration of anions in this study follows the decreasing order of chloride > sulfate > nitrate > fluoride. Fluoride was found in only five samples in which highest value is 0.29 ppm which is also below the safe limit set by WHO. The range of chloride, sulfate, and nitrite are 6.30–8.93, 3.475–4.255, 0.29–9.535, respectively (Table 1), and all the values are within the range of international standard and consider Halda river water is not significantly polluted by these anions. In cation test major cation like sodium, ammonium, potassium, magnesium, and calcium were analyzed in DIONEX. In all of the water samples, lithium was below its LOD value. Hardness of water is due to alkaline earth

Table 1. Physico-chemical parameters in surface water samples from the Halda river in Bangladesh (mean ± SD, $n = 3$).

Sites	Fl^- (ppm)	Cl^- (ppm)	SO_4^{2-} (ppm)	NO_2^- (ppm)	Na^+	NH_4^+ (mg/L)	K^+ (ppm)	Mg^{++} (ppm)	Ca^{++} (ppm)	Ec (μS/cm)	pH	TDS (mg/L)
SW1	0.09 ± 0.01	7.23 ± 0.47	3.915 ± 0.26	1.51 ± 0.38	8.40 ± 0.28	0.39 ± 0.02	3.35 ± 0.14	3.33 ± 0.12	6.31 ± 0.2	33.4 ± 0.21	6.42 ± 0.05	20.1 ± 3.1
SW2		6.68 ± 1.00	3.545 ± 0.5	0.37 ± 0.04	8.96 ± 0.18	0.45 ± 0.05	3.42 ± 0.09	3.53 ± 0.07	6.7 ± 0.07	33.1 ± 3.2	6.22 ± 0.02	19.8 ± 2.2
SW3	0.28 ± 0.03	7.52 ± 0.11	3.94 ± 0.11	0.72 ± 0.01	9.56 ± 0.04	0.36 ± 0.05	3.57 ± 0.12	3.81 ± 0.11	8.31 ± 0.2	33.7 ± 4.1	6.16 ± 0.21	20.3 ± 2.2
SW4	0.29 ± 0.00	7.51 ± 0.04	3.995 ± 0.04	0.22 ± 0.01	9.03 ± 0.16	0.47 ± 0.04	3.38 ± 0.13	3.53 ± 0.11	6.51 ± 0.11	33.5 ± 2.7	6.05 ± 0.5	20.1 ± 4.3
SW5		7.92 ± 0.01	4.205 ± 0.01	0.45 ± 0.01	10.3 ± 0.28	0.36 ± 0.05	3.78 ± 0.13	4.07 ± 0.07	9.06 ± 0.13	32.5 ± 5.2	5.84 ± 0.04	19.94 ± 4.0
SW6		8.93 ± 0	4.18 ± 0	0.50 ± 0.02	10.6 ± 0.4	0.29 ± 0.01	3.88 ± 0.10	4.02 ± 0.07	7.92 ± 0.35	33.6 ± 3.6	5.87 ± 0.01	20.2 ± 2.6
SW7		8.915 ± 0.01	4.245 ± 0.01	1.26 ± 0.02	10.8 ± 0.2	0.28 ± 0.01	3.95 ± 0.03	4.05 ± 0.18	7.69 ± 0.24	32.8 ± 5.1	6.21 ± 0.02	20.1 ± 6.2
SW8		8.875 ± 0.08	4.255 ± 0.08	0.33 ± 0.01	10.8 ± 0.13	0.45 ± 0.02	3.97 ± 0.13	4.3 ± 0.06	8.45 ± 0.45	33.6 ± 5.2	6.14 ± 0.02	20.1 ± 2.2
SW9	0.07 ± 0.02	7.665 ± 0.02	4.16 ± 0	9.53 ± 0.02	10.7 ± 0.16	0.37 ± 0.03	3.85 ± 0.08	4.03 ± 0.11	7.5 ± 0.21	32.6 ± 1.9	5.98 ± 0.01	19.5 ± 1.8
SW10		6.30 ± 2.5	3.475 ± 0.36	0.29 ± 0.01	9.9 ± 0.04	0.41 ± 0.01	3.88 ± 0.33	4.05 ± 0.06	7.5 ± 0.3	31.3±.3.6	5.98 ± 0.05	18.7 ± 2.8
SW11		8.025 ± 0.08	4.205 ± 0.04	0.45 ± 0.01	10.6 ± 0.1	0.38 ± 0.03	4.03 ± 0.13	3.83 ± 0.13	3.83 ± 0.13	33.6 ± 0.66	5.98 ± 0.04	20.2 ± 2.5
SW12	0.09 ± 0.007	7.845 ± 0.01	4.225 ± 0.01	0.52 ± 0.01	10.2 ± 0.24	0.36 ± 0.07	3.41 ± 0.05	4.1 ± 0.14	4.1 ± 0.14	32.8 ± 4.2	5.51 ± 0.04	20.1 ± 3.3

metals such as Mg and Ca ions (Abbasi 1998). Suitable hardness for fish growth is about 15 mg/L, less than 11 mg/L requires liming for higher fish production, so water having less than 5 mg/L $CaCO_3$ cause pain, slow growth rate, and ultimately death of fish (Boyd 1981). In our study TDS value is between 18.74 and 20.30 mg/L which is within the safe range set by WHO. The highest and lowest value of ammonium ion recorded in 0.47 mg/L in station 4 and 0.28 mg/L in station 7, respectively. Sodium and potassium ions mean concentrations between the range of 8.40–10.8 ppm and 3.35–4.03 ppm was reported during the present study.

3.3. Concentration of TEs in water samples

Mean values of TEs of water samples from Halda River, Bangladesh are shown in Table 2. A wide range of TEs concentrations were observed among the sampling sites and seasons. Factors such as salinity, TDS, pH, Temp., geomorphological setup, and terrestrial runoff might have played a role in the variation of TEs in the water samples of the study river (Islam *et al*. 2015c, Proshad *et al*. 2020). The average concentration of studied TEs in water samples followed the decreasing order of Cu > As > Fe > Zn > Cd > Pb > Mn > Co > Cr > Hg during wet season and As > Fe > Zn > Cu > Mn > Pb > Cr > Co > Cd > Hg during dry season (Table 2).

The mean concentration of Cd was observed in surface water samples (0.032 mg/L in wet season and 0.04 mg/L in dry season) (Table 2). Slightly higher Cd level was found in dry season which might be due to the variation in water capacity of the river where low water flow in dry season resulted the precipitation of the TEs in water; thereby increasing its concentration (Islam *et al*. 2014). The dissolved Cr concentration varies from 0.001 to 0.006 mg/L during wet season and 0.019–0.167 mg/L during dry season (Table 2). The highest Cr concentration was obtained at SW4 site for both the season, presumably as a result of the effects from tannery and dyeing industries (Arias-Barreiro *et al*. 2010). The chromium enrichment of surface water can have been caused by two reasons: (1) natural: concentration of Cr-bearing minerals; and (2) anthropogenic: industrial activities such as tanneries and textile factories which are discharging Cr-based oxidants (chromate, dichromate, etc.) (Facetti *et al*. 1998). Consequently, the waste discharged from such industries is responsible for elevated Cr level in river water (Mohiuddin *et al*. 2011, Islam *et al*. 2014).

Arsenic forms a variety of inorganic and organic compounds of different toxicity reflecting the physicochemical properties of arsenic in water environment at different valences. In the surface water, average concentration of As was higher in wet season (1.34 ± 1.17 mg/L) than that in dry season (1.07 ± 0.989 mg/L). In both season, relatively high As level was observed at stations 8, 9, 10, 11, and 12 which might be due to some activities from Chittagong Metropolitan area in Bangladesh such as treatment of agricultural land with fertilizers and arsenical pesticides (Renner 2004, Fu *et al*. 2014, Shao *et al*. 2016), treating of wood by using copper arsenate (Baeyens *et al*. 2007, Pravin *et al*. 2012, Ali *et al*. 2019) and tanning in relation to some chemicals especially arsenic sulfide (Asaduzzaman *et al*. 2002, Bhuiyan *et al*. 2011). Considerable amount of As comes mostly from the upland Himalayan catchments which is connected with the study river at the northern part of Bangladesh (Mitamura *et al*. 2008). The average concentration of Pb in surface water was (0.104 and 0.00 mg/L during dry and wet season, respectively). The average concentration of Cu was observed for surface water 3.33 and 0.172 mg/L during wet and dry season, respectively (Table 2). Interestingly an elevated level of Cu was observed during wet season at station 8 (3.853 mg/L) and at station 9 (4.555 mg/L) of studied river which might be due to the fact that these sites are located at the downstream of the river and extensive discharging of domestic sewage and urban runoff from extensively farmed areas (Koukal *et al*. 2004, Wu *et al*. 2008, Islam *et al*. 2014).

Chromium, Mn, Co, Zn, and Pb concentrations in water samples were statistically higher in dry season than wet season ($p < 0.05$), whereas the concentrations of Cu and Hg were higher in wet season than dry season ($p < 0.05$). Other TEs in water samples did not show statistically differences between dry and wet seasons ($p > 0.05$) (Table 2). Average level of trace element toxicity load (TETL) in water samples was higher in wet season (5.20 ± 1.88 mg/L) than dry season (2.62 ± 0.947 mg/L). Trace element toxicity load indicate the content in water body and indicates the necessary elimination percentage of toxic metals from the water to make it safe for human use. It evaluates toxic metals level found in water that affect human health. TEs in water samples were seasonally varied, where dry season exhibited slightly higher than wet season. Lower level of TE concentration in water samples during wet season can be associated with the dilution effect of high water level due to high inflow discharges and high precipitation (Mohiuddin *et al*. 2012, Islam *et al*. 2014). Several studies (e.g., Ali *et al*. 2016, 2018) also reported that the heavy metal

Table 2. Mean concentrations of trace elements (mg/L) in surface water samples from the Halda River in Bangladesh ($n = 3$).

Sites	Cd	Cr	Mn	Fe	Co	Cu	Zn	As	Pb	Hg	TETL
Wet season											
SW1	0.036	0.002	0.007	0.453	0.064	2.858	0.073	0.277	0.026	0.008	3.804
SW2	0.005	0.001	0.008	0.253	0.004	3.793	0.053	0.525	0.020	0.007	4.669
SW3	0.006	0.005	0.009	0.315	0.005	3.862	0.073	0.762	0.026	0.003	5.066
SW4	0.006	0.006	0.007	0.312	0.006	1.859	0.057	0.490	0.046	0.006	2.794
SW5	0.050	0.005	0.006	0.327	0.004	2.419	0.069	0.082	0.008	0.005	2.976
SW6	0.049	0.005	0.005	0.380	0.006	1.720	0.045	0.453	0.017	0.003	2.683
SW7	0.070	0.005	0.048	0.381	0.005	5.765	0.059	0.505	0.032	0.007	6.877
SW8	0.005	0.003	0.005	0.534	0.004	3.853	0.065	3.168	0.031	0.009	7.677
SW9	0.005	0.002	0.007	0.403	0.006	4.555	0.061	2.768	0.035	0.010	7.851
SW10	0.079	0.005	0.003	0.305	0.006	3.034	0.062	1.735	0.019	0.007	5.255
SW11	0.066	0.005	0.008	0.419	0.006	3.627	0.063	2.710	0.052	0.007	6.962
SW12	0.005	0.001	0.006	0.447	0.006	2.606	0.061	2.643	0.052	0.009	5.837
Mean ± SD	0.032 ± 0.028	0.004 ± 0.002	0.010 ± 0.012	0.38 ± 0.08	0.010 ± 0.017	3.33 ± 1.16*	0.062 ± 0.008	1.34 ± 1.17	0.030 ± 0.014	0.007 ± 0.002*	5.20 ± 1.88*
Dry season											
SW1	0.040	0.057	0.047	0.769	0.037	0.137	0.300	0.310	0.400	0.003	2.099
SW2	0.037	0.057	0.200	0.480	0.030	0.157	0.490	0.592	0.020	0.002	2.063
SW3	0.026	0.065	0.060	0.412	0.056	0.053	0.457	0.662	0.027	0.003	1.820
SW4	0.050	0.167	0.267	0.473	0.040	0.070	0.590	0.523	0.027	0.003	2.210
SW5	0.027	0.047	0.057	0.688	0.060	0.094	0.200	0.066	0.200	0.003	1.440
SW6	0.057	0.053	0.353	0.566	0.060	0.150	0.413	0.190	0.267	0.002	2.111
SW7	0.027	0.042	0.067	0.494	0.070	0.300	0.620	0.048	0.043	0.006	1.716
SW8	0.053	0.019	0.420	0.701	0.023	0.080	0.073	2.168	0.057	0.005	3.600
SW9	0.057	0.052	0.050	0.554	0.027	0.015	0.133	2.102	0.043	0.003	3.035
SW10	0.071	0.045	0.189	0.670	0.061	0.500	0.700	1.140	0.067	0.003	3.445
SW11	0.020	0.053	0.047	0.626	0.073	0.030	0.172	2.477	0.068	0.001	3.567
SW12	0.020	0.060	0.043	0.552	0.050	0.480	0.570	2.623	0.030	0.006	4.435
Mean ± SD	0.04 ± 0.017	0.060 ± 0.036*	0.150 ± 0.134*	0.58 ± 0.11	0.049 ± 0.017*	0.172 ± 0.166	0.393 ± 0.212*	1.07 ± 0.989	0.104 ± 0.121*	0.003 ± 0.002	2.62 ± 0.947

TETL: trace element toxicity load.
*Indicates statistically significant difference of elements between two seasons ($p < 0.05$).

concentrations in the surface water of varied seasonally, while wet season exhibited lower concentrations than dry season.

When compared with water quality guidelines for drinking water, maximum concentrations of all TEs in water samples from Halda River, Bangladesh were much higher than the drinking water guideline values set by WHO (2011) (Table 3). The mean concentrations of all TEs in surface waters greatly exceeded the limit for safe drinking water guideline values set by European Community (EC 1998) (Table 3) indicated that water from this river is not safe for drinking and/or cooking purposes. Also, average concentrations of all TEs in water samples were much higher than maximum contaminant level (MCL) values for drinking water established by USEPA (2020f) (Table 3). However, the concentrations of all studied TEs in water samples exceeded maximum contaminant level goal (MCLG) values set by USEPA (2020f), indicated that the water from studied river pose significant risk to the surrounding ecosystems. When compared with water quality guidelines for the protection of freshwater aquatic life, mean concentrations of TEs in water samples were exceeded the criterion maximum concentrations (CMC) and criterion continuous concentrations (CCC) established by USEPA (2020e, 2020g) (Table 3). In Bangladesh, the most concern elements are As, Cd, and Pb, which are in water samples greatly exceeded the international standard value as well as other studies in Bangladesh and other countries, which can be due to the effect from point and non-point sources; such as leaded gasoline, municipal runoffs and atmospheric deposition (Mohiuddin *et al.* 2012, Shikazono *et al.* 2012), chemical manufacturing and steel works in urban area of Chittagong district. As shown in Table 3, concentrations of TEs in water samples from Halda River were much higher than those in other rivers in Bangladesh like Turag River (Mokaddes *et al.* 2013) and Buriganga River (Mokaddes *et al.* 2013) but lower than Karnafuly River (Islam *et al.* 2013). Compared with other countries, the TEs concentrations were generally higher than some other countries, such as India, China, Malaysia, Iran, and Spain (Li *et al.* 2008, Carafa *et al.* 2011, Rajaei *et al.* 2012, Gao *et al.* 2016, Wang *et al.* 2017a). This indicated that the pollution level of TEs in Halda River in Bangladesh was in high level of contamination.

3.4. Multivariate statistical analysis for source analysis of TEs

In this study, statistical analyses were performed to reveal the associations among TEs in surface waters of Halda River and to identify the factors involved in

Table 3. Comparison of trace element concentrations (mg/L) of the Halda river water with other rivers of Bangladesh.

Locations		Cd	Cr	Mn	Fe	Co	Cu	Zn	As	Pb	Hg	References
Halda River, Bangladesh (wet season)	Mean ± SD	0.032 ± 0.028	0.004 ± 0.002	0.010 ± 0.012	0.58 ± 0.11	0.010 ± 0.017	3.33 ± 1.16	0.062 ± 0.008	1.34 ± 1.17	0.030 ± 0.014	0.007 ± 0.002	Present study
Halda River, Bangladesh (dry season)	Mean ± SD	0.04 ± 0.017	0.060 ± 0.036	0.150 ± 0.134	0.58 ± 0.11	0.049 ± 0.017	0.172 ± 0.166	0.393 ± 0.212	1.07 ± 0.989	0.104 ± 0.121	0.003 ± 0.002	Present study
Turag River, Bangladesh	Mean	0.01	NA	0.06	NA	NA	0.004	0.02	NA	0.002	NA	Mokaddes *et al.* (2013)
Buriganga, Bangladesh	Mean	0.059	0.114	0.157	NA	NA		0.332	NA	0.112	NA	Mokaddes *et al.* (2013)
Damodar River, India	Range	4.5-14.9	NA	140-3020	114-1085	NA	44835	6.5-58.5	NA	NA	NA	Mohanta *et al.* (2020)
Kelantan River, Malaysia	Mean	30	NA	160	1951	NA	72	242	NA	NA	NA	Wang *et al.* (2017a)
Red Sea, Saudi Arabia	Mean ± SD	0.17 ± 0.04	1.36 ± 0.37	1.25 ± 0.29	NA	NA	7.85 ± 1.52	3.58 ± 0.94	0.28 ± 0.13	0.56 ± 0.13	NA	Mortuza and Al-Misned (2017)
Karnafuly River, Bangladesh	Mean	0.01	0.25	0.12	NA	NA	0.05	0.28		0.14	NA	Islam *et al.* (2013)
Danjiangkou Reservoir, China	Mean	0.0117	0.0063	0.00569	0.0191	0.00108	0.0133	0.00202	0.0111	0.0106	Nd	Li *et al.* (2008)
Three Gorges Reservoir, China	Mean	0.00102	NA	NA	NA	NA	0.00894	0.01304	0.00233	0.01121	0.00003	Gao *et al.* (2016)
Chah nimeh reservoir-1, Iran	Mean	0.0106	0.0624	NA	0.312	0.0037	0.0225	NA	0.0095	0.0291	NA	Rajaei *et al.* (2012)
Catalan River, Spain	Mean	0.0012	0.0024	NA	NA	NA	0.0013	0.0019	0.0029	0.0022	NA	Carafa *et al.* (2011)
Freshwater quality criteria for protection of aquatic life												
USEPA	CMC, acute	0.0018	0.016	NA	NA	NA	NA	0.12	0.34	0.082	0.0014	USEPA (2020g)
USEPA	CCC, chronic	0.00072	0.011	NA	NA	NA	NA	0.12	0.15	0.0032	0.00077	USEPA (2020g)
Drinking water quality criteria												
DWSB		0.005	0.05	0.1	0.3-1.0	NA	1	5	0.05	0.05	0.001	DoE (1997)
European Community		0.005	0.05	0.05	0.2	NA	2	NA	0.01	0.01	0.001	EC (1998)
WHO		0.003	0.05	0.5	0.3	0.05	2	3	0.01	0.01	0.006	WHO (2011)
USEPA	MCLG	0.005	0.1	NA	NA	NA	1.3	NA	0	0	0.002	USEPA (2020f)
USEPA	MCL	0.005	0.1	NA	NA	NA	NA	NA	0.01	0.015	0.002	USEPA (2020f)

CMC: criterion maximum concentration; CCC: criterion continuous concentration; MCLG: maximum contaminant level goal; MCL: maximum contaminant level; Nd: not detected; NA: data not available.

controlling the distribution of TEs. Moreover, inter-metal interactions may also illustrate the sources of the TEs present in the particulate media (Proshad *et al.* 2019). Spearman correlation and PCA were implemented for inter-variable relationships of the studied elements and potential sources identification of studied TEs in water samples of Halda River, Bangladesh (Dong *et al.* 2015, Kumar *et al.* 2017) as presented in Supplementary Table S4. Three PCs were extracted from 10 elements in water samples. Three PCs together contributed 67.4 and 61.2% of the total variance during wet and dry season, respectively. Water samples during wet season, Hg, Pb, and As had high loadings in PC1, and can explain 27.7% of the variance (Figure 2A) whereas Pb and Fe had high loadings in PC1, and can explain 25.0% of the variance (Figure 2B). Mercury, Pb, and As in PC1 were mainly from anthropogenic input such as vehicular exhaust and Pb melting was the major contributor of Pb to the water environment while industrial discharge was the main source of Hg (Kumar *et al.* 2016, Islam *et al.* 2016b). On the other hand application of arsenic-based pesticides/herbicides during intensive agricultural activities besides the agricultural fields of the study area (Wang *et al.* 2019). This was also supported by the significant correlation among Hg, Pb, and As during wet season and Pb and Fe during dry season (Supplementary Table S4). Iron is the major elements in crust (Wang *et al.* 2017b). The weathering of calcite and chlorite in loess can provide an amount of Fe to the river water of Korotoa River (Yokoo *et al.* 2004).

Principal component 2 (PC2) explained 21.9% of the variance with high loadings of Zn, Co and Fe in the PC2 during wet season (Figure 2A), whereas, PC2 explained 18.8% of the variance with high loadings of Zn, Co, Hg, Cu, and As during dry season (Figure 2B) which were mainly from weathering and/or leaching of loess and the sorption and desorption behavior of these elements might be similar. Some characteristics in loess, such as fine particles of hydrous oxides and fine grain size, can accelerate the sorption of these elements in the surface water of river (Currell *et al.* 2011, Díaz *et al.* 2016). This was also supported by the high correlation among these elements (Supplementary Table S4). The PC3 was responsible for 17.7% of the variance with high loadings of Cd, Cr, Cu, and Mn during wet season (Figure 2A) whereas, 17.3% of the variance with high loadings of Cd, Cr, and Mn during dry season (Figure 2B). Cadmium and Cr in PC3 were mainly from the industrial effluents and domestic sewage (Islam *et al.* 2015a, 2015c, 2018b) where Mn in the PC3 was siderophile element, which was mainly from parent material weathering and pedogenic process. In the study area of Bangladesh, water movement in river, environmental condition as well as industrial operation greatly varied between wet and dry season which could be the reason for variation of metal source between two seasons. Overall, this class of parameters is typical of sources linked to anthropogenic inputs from industrial activities such as discharge of untreated chemical wastes, automobile wastes, and paints (Bhutiani *et al.* 2017, Egbueri 2018, Mgbenu and Egbueri 2019).

3.5. Trace element evaluation index (TEI)

Trace element evaluation index (TEI) is a method of estimating the quality of water with a focus on TEs in

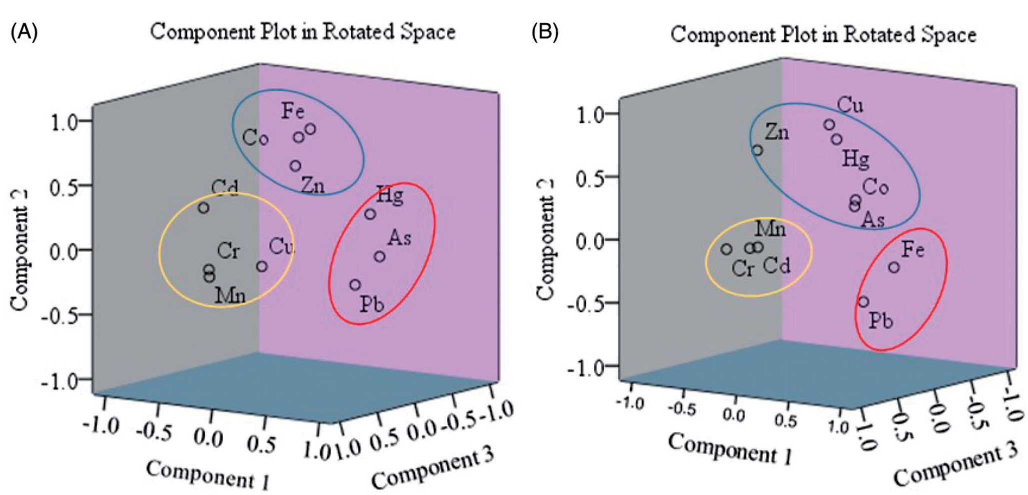

Figure 2. Principal component analysis (PCA) for dissolved trace elements in surface water during wet season (A) and dry season (B) of Halda River, Bangladesh.

Table 4. Ecological risk index (ERI) of studied metal in Halda river (wet and dry season) in Bangladesh.

Risk index of single element											
Cd	Cr	Mn	Fe	Co	Cu	Zn	As	Pb	Hg	ERI	Risk level
Wet season											
106.8	0.933	0.065	1.224	0.294	14.29	0.015	55.33	2.633	6.613	188.2	Moderate
16.20	0.573	0.077	0.683	0.020	18.96	0.011	105.0	1.967	5.227	148.7	Low
18.70	1.960	0.085	0.851	0.022	19.31	0.015	152.3	2.567	2.773	198.6	Moderate
18.80	2.333	0.065	0.842	0.027	9.293	0.011	98.00	4.567	4.773	138.7	Low
149.0	1.960	0.063	0.884	0.021	12.10	0.014	16.47	0.800	4.107	185.4	Moderate
146.5	1.880	0.054	1.027	0.029	8.599	0.009	90.67	1.700	2.160	252.6	Moderate
210.0	2.093	0.480	1.030	0.021	28.82	0.012	101.0	3.200	5.867	352.5	Moderate
15.50	1.387	0.052	1.441	0.018	19.26	0.013	633.7	3.133	6.907	681.4	Very high
14.60	0.760	0.072	1.088	0.025	22.78	0.012	553.7	3.500	7.653	604.2	Very high
238.0	1.947	0.035	0.824	0.027	15.17	0.012	347.0	1.900	5.227	610.1	Very high
199.0	1.987	0.076	1.130	0.025	18.14	0.013	542.0	5.167	5.973	773.5	Very high
15.60	0.480	0.058	1.207	0.027	13.03	0.012	528.7	5.167	6.987	571.2	Considerable
Dry season											
120.0	22.67	0.467	2.076	0.169	0.683	0.060	62.00	40.00	2.400	250.5	Moderate
110.0	22.67	2.000	1.295	0.138	0.783	0.098	118.3	2.000	1.600	258.9	Moderate
79.00	25.87	0.600	1.113	0.256	0.267	0.091	132.3	2.667	2.133	244.3	Moderate
150.0	66.67	2.667	1.278	0.184	0.350	0.118	104.7	2.667	2.400	331.0	Moderate
80.00	18.67	0.567	1.859	0.276	0.468	0.040	13.13	20.00	2.133	137.1	Low
170.0	21.33	3.533	1.528	0.276	0.750	0.083	38.00	26.67	1.707	263.9	Low
80.00	16.80	0.667	1.334	0.322	1.500	0.124	9.533	4.333	4.773	119.4	Low
160.0	7.467	4.200	1.894	0.107	0.400	0.015	433.7	5.667	4.267	617.7	Very high
171.0	20.80	0.500	1.495	0.123	0.073	0.027	420.3	4.333	2.187	620.9	Very high
212.0	18.13	1.887	1.809	0.279	2.500	0.140	228.0	6.667	2.080	473.5	Considerable
60.00	21.33	0.467	1.690	0.337	0.150	0.034	495.3	6.800	1.067	587.2	Considerable
60.00	24.00	0.433	1.491	0.230	2.400	0.114	524.7	3.000	5.067	621.4	Very high

water body. It provides an overall assessment of the water quality with regard to the contamination level of TEs. In this study, TEI was calculated for Cd, Cr, Mn, Fe, Co, Cu, Zn, As, Pb, and Hg and is presented in Supplementary Table S5. The ranges of TEI for the studied elements during wet and dry season were 28.97–327 and 23.36–277.6, respectively. The values of TEI in the present study were classified in terms of pollution levels as low, medium and high. According to TEI classification (Edet and Offiong 2002), TE pollution in the waters of Halda River was found to be high index of TEs evaluation (more than 20).

3.6. ERI of TEs

The calculated ERI of TEs for Halda river water ranged from 138.7 to 773.5 in wet season and 119.4 to 621.4 in dry season (Table 4). According to the ERI classification reported by Taiwo *et al*. (2019), 20.8% of the total samples were found to pose low ecological risks, 37.5% of the total samples were found to moderate ecological risk, 12.5% of the total samples were found to considerable ecological risk and 29.2% of the total samples were found to very high ecological risks for both the seasons (Table 4). In respect to the risk index for single element, As contributed the most ecological risk of water samples during wet and dry seasons. On the other hands, the mean risk index of single metal was found in the descending order of: As > Cd > Cu > Hg > Pb > Cr > Fe > Mn > Co > Zn during wet season, whereas As > Cd > Cr > Pb > Hg > Fe > Mn > Co > Cu > Zn during dry season.

3.7. Assessment of health risks

The potential non-carcinogenic toxic effects on human health is caused by TEs are usually characterized by calculating HQ and HI. The HQs and HI values for TEs from ingestion and dermal exposure pathways relating to the residential receptors for surface waters are presented in Table 5. Concerning water ingestion exposure pathway, As in river water showed the highest HQ values for residential receptors (Table 5). Arsenic in waters during wet season contributed 95% of the total health risk, HI (sum of all individual element HQ values), for residential adult and child receptors (Table 5) while this element during dry season contributed 92% of the HI for adults and child inhabitants (Table 5). In this study, all HQ values for As were slightly lower than the risk threshold of unity (HQ $<$ 1.0). The result showed that As pollution was the worst in river water and was the potential pollutant in river water especially for children, so we can hypothesize negative effects on populations, especially on the most sensitive subjects. However, high As HQ values for residential receptors via water ingestion pathway are reported in the literature (Li and Zhang 2010, Giri and Singh 2014, Xiao *et al*. 2019). Water samples during

Table 5. Noncarcinogenic (HQ, HI, and THI) and carcinogenic (CR and TCR) risks from trace elements in surface water of the Halda river for residential receptors.

	Noncarcinogenic risks for adult			Noncarcinogenic risks for child			Carcinogenic risks		
Elements	HQ ingestion	HQ dermal	THI	HQ ingestion	HQ dermal	THI	CR ingestion	CR dermal	TCR
Wet season									
Cd	8.41E − 04	4.69E − 07	8.41E − 04	1.59E − 03	7.01E − 07	1.59E − 03			
Cr	3.35E − 05	1.49E − 05	4.84E − 05	6.33E − 05	2.23E − 05	8.57E − 05	2.68E − 08	1.14E − 08	3.82E − 08
Mn	1.08E − 05	1.51E − 06	1.23E − 05	2.05E − 05	2.25E − 06	2.27E − 05			
Fe	2.19E − 05	1.22E − 07	2.20E − 05	4.15E − 05	1.83E − 07	4.17E − 05			
Co	8.85E − 04	1.98E − 06	8.87E − 04	1.68E − 03	2.95E − 06	1.68E − 03			
Cu	2.19E − 03	1.22E − 05	2.20E − 03	4.15E − 03	1.83E − 05	4.17E − 03			
Zn	5.42E − 06	1.82E − 08	5.44E − 06	1.03E − 05	2.71E − 08	1.03E − 05			
As	1.18E − 01	6.58E − 04	1.19E − 01	2.23E − 01	9.84E − 04	2.24E − 01	2.83E − 05	1.51E − 07	2.85E − 05
Pb	5.69E − 04	3.18E − 07	5.70E − 04	1.08E − 03	4.75E − 07	1.08E − 03	3.61E − 09	1.93E − 11	3.63E − 09
Hg	8.82E − 04	4.92E − 06	8.87E − 04	1.67E − 03	7.35E − 06	1.68E − 03			
HI	*1.23E−01*	*6.95E−04*	*1.24E−01*	*2.34E−01*	*1.04E−03*	*2.35E−01*	*2.84E−05*	*1.63E−07*	*2.85E−05*
Dry season									
Cd	1.06E − 03	5.93E − 07	1.06E − 03	2.01E − 03	8.86E − 07	2.01E − 03			
Cr	5.24E − 04	2.34E − 04	7.58E − 04	9.92E − 04	3.50E − 04	1.34E − 03	4.19E − 07	1.79E − 07	5.98E − 07
Mn	1.65E − 04	2.30E − 05	1.87E − 04	3.11E − 04	3.43E − 05	3.46E − 04			
Fe	2.19E − 05	1.22E − 07	2.20E − 05	4.15E − 05	1.83E − 07	4.17E − 05			
Co	4.29E − 03	9.58E − 06	4.30E − 03	8.12E − 03	1.43E − 05	8.14E − 03			
Cu	1.13E − 04	6.32E − 07	1.14E − 04	2.14E − 04	9.45E − 07	2.15E − 04			
Zn	3.45E − 05	1.16E − 07	3.46E − 05	6.54E − 05	1.73E − 07	6.55E − 05			
As	9.44E − 02	5.27E − 04	9.49E − 02	1.79E − 01	7.87E − 04	1.79E − 01	2.27E − 05	1.21E − 07	2.28E − 05
Pb	1.96E − 03	1.09E − 06	1.96E − 03	3.70E − 03	1.63E − 06	3.71E − 03	1.24E − 08	6.63E − 11	1.25E − 08
Hg	4.36E − 04	2.44E − 06	4.39E − 04	8.26E − 04	3.64E − 06	8.30E − 04			
HI	*1.03E−01*	*7.98E−04*	*1.04E−01*	*1.95E−01*	*1.19E−03*	*1.96E−01*	*2.31E−05*	*3.00E−07*	*2.34E−05*

wet season, Cu was the second element having highest HQ values for adult and child receptors while in dry season, Pb was the second element having highest HQ values for adult and child receptors (Table 5). The HQ value of Cu during wet season was lower than the risk threshold (HQ <1), this element in surface water contributed 2% of the HI for adult receptors, whereas in dry season water Pb contributed 2% of the HI for adult receptors. Other elements like Cd, Co, Pb, and Hg during wet season and Cd, Co, Cr, Hg, Ni, Mn, Cu, and Hg during dry season samples of Halda River contributing a considerable level to the HI from ingestion exposure pathway, indicating that it can cause serious adverse effects on health to local people. As a whole, the HQ values for both adults and child inhabitants from ingestion pathway in waters during wet season follow the descending order of As > Cu > Co > Hg > Cd > Pb > Cr > Fe > Mn > Zn, while the HQ values for both adults and child inhabitants from ingestion pathway in waters during dry season follow the descending order of As > Pb > Co > Cd > Cr > Hg > Mn > Cu > Zn > Fe. From both wet and dry seasons, the HQ and HI values from both pathways for children were higher than adults (Table 5), indicating that children are more vulnerable than adults when exposed to the same environment.

Concerning dermal exposure pathway, As in water samples during both seasons was the highest HQ values for both adults and child receptors (Table 5). Arsenic in wet season contributed 95% of the HI for both adults and child receptors while this element contributed 66% of the HI for both adults and child receptors during dry season (Table 5). Arsenic HQ values for residential receptors in this study were higher than those reported in the literature (Li and Zhang 2010, Giri and Singh 2014). Chromium was the second element having the highest HQ values for adult and child receptors, while Cd and Zn were elements with the lowest HQ values (Table 5). In this study, all HI and THI values of TEs were lower than the risk threshold of unity (Table 5). Also, adverse effect of water ingestion pathway on the health of residential receptors was more than dermal contact pathway. It is noteworthy that the HI values for ingestion and dermal contact pathways for residential child were higher than those for residential adult, indicating that children were more sensitive than adults when exposed to TEs in water of Halda River, that is in agreement with the results of previous studies (Giri and Singh 2014, Saha *et al*. 2017, Wang *et al*. 2017a, Saleem *et al*. 2019, Xiao *et al*. 2019).

Three elements such as As, Pb, and Cr having CSF, were used to assess CRs in this study. The CR values of As, Pb, and Cr in water during both seasons via ingestion and dermal contact pathways for residential adult receptors were lower than the target risk of 1×10^{-4} (Table 5). The CR values of As in water during wet and dry season for ingestion pathway for residents were, 2.83×10^{-5}, and 2.27×10^{5}, respectively (Table 5). Data of the present study revealed that As was the main contributor to the TCR ($CR_{ingestion} + CR_{dermal}$) via ingestion and dermal contact pathways for residents (Table 5). The TCR values of As in waters for resident and recreator were higher than the acceptable range of 1×10^{-6}; this shows that this element would pose a CR for residential receptors at the study area in Bangladesh. Also, the TCR values of As in water for residential receptors were (2.85×10^{-5} during wet season and 2.28×10^{-5} during dry season) higher than the acceptable range of 1×10^{-6}; that is in agreement with the results of high CRs of As for residents in other surface water bodies such as Three Gorges Reservoir (Gao *et al*. 2016), Wen-Rui Tang River (Qu *et al*. 2018), ZhiXi Reservoir (Wang *et al*. 2018), and Han River (Li and Zhang 2010) in China and Bangshi River (Saha *et al*. 2017) in Bangladesh.

The exposure of As from drinking water in long time may cause potentially carcinogenic effects and hypertension, skin lesions, diabetes, neuropathy, etc. (Yu *et al*. 2007, He and Charlet 2013). Therefore, local residents especially the sensitive children should be paid special attention to samples with high As, especially for surface waters of Halda River in Bangladesh. In addition, corresponding measures should be taken to the removal of As from surface waters of Halda River. Except As, Cu, Co, Cd, Pb, and Hg were other elements of concern with relatively high values of HQ (Table 5). In the future, the monitoring program of As and other potential elements in surface waters of Halda River and effective health education for As pollution is necessary. In the future, more detailed work should be carried out in these areas with high concentrations of potentially toxic TEs.

4. Conclusions

Due to increase of poorly planned urban areas and industrial activities in developing countries like Bangladesh, the quality of rivers waters is highly being affected by various contaminants like toxic TEs. In this study, the geochemistry characteristics of 10 potentially toxic TEs in surface water collected from an urban river (Halda) were studied. In the present investigation, concentrations of the studied TEs were higher than the safe recommended values which suggested that the Halda River of Bangladesh is polluted by TEs and might create an adverse effect on this riverine ecosystem. TEI indicated that Halda River water was found to be high index of TEs contamination. ERI showed low to very high contamination of water by the studied toxic elements. Health risk assessment indicated that THI values of As, Cu, Cd, Pb, and Hg in surface water for ingestion and dermal pathways for residential adult and child receptors were close to the risk threshold of unity indicated potential health risk. The values of HQ, HI, and THI for children were higher than adults, indicating that children are more susceptible to adverse effects of TEs in water of Halda River. The TCR values of As in waters for residents were higher than the acceptable range of 1×10^{-6}; this showed that this element would pose a CR for residential receptors. The subjects most at risk are children, for whom HQ values reached near to the maximum safety limit. The daily use of metals contaminated drinking water can lead to the development of serious diseases. In this study, drinking pathway was investigated, so the results did not consider many other exposure routes such as food and/or soil. Moreover, it is necessary to consider the high sensitivity of subjects such as infants and pregnant women and extreme hygienic health, nutritional, and climatic conditions around the study area.

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Disclosure statement

The authors declare no conflict of interest.

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