Comprehensive Evaluation of Some Toxic Metals in the Surface Water of Louhajang River, Bangladesh

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Comprehensive evaluation of some toxic metals in the surface water of Louhajang River, Bangladesh

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Abstract

Louhajang River, Bangladesh, crosses Tangail, which is a densely industrialized and urbanized city. Louhajang River is an essential water source for domestic, irrigation, and urbanization purposes. This study reports the levels of pH, electrical conductivity (EC), and some toxic heavy metals in 40 water samples collected during summer and winter seasons from Louhajang River. The levels were found to be in the ranges of pH 6.22-7.43 and EC 345-798 mS/cm, Cr 0.18-13.2, Ni 0.02-19.04, Cu 0.96-15.92, As 2.18-12.51, Cd 0.02-2.42, and Pb 0.49-15.74 µg/L. The winter season reported higher levels of the examined parameters than the summer season with significant variation ($p < 0.05$) for all parameters, with the exception of Cd. The metal contents were assessed against local and international standards for drinking, irrigation and aquatic life purposes where different trends were observed. The heavy metal evaluation index (HEI) and the ecological risk index (ERI) reported low to moderate risks. The spatial distribution of metal contents assigned hot spots in some sites along the riverbed, which were attributed to specific manmade sources. The health risk assessment for three population categories, i.e., adult male, adult female, and children, were examined in terms of hazard index (HI) and total cancer risk (TCR) for oral and dermal pathways during both seasons. Cr and Cd recorded HI more than unit in all cases, indicating possible non-cancer risk. TCR values of As for the three examined population categories during both seasons were > 1.0×10^{-6}, indicating possible cancer risk, while that of Pb were < 1.0×10^{-6}. For Ni, about 10-25% of the sampled sites recorded TCR > 1.0×10^{-6}.

Keywords

Toxic metals, water quality, river contamination, heavy metal evaluation index, ecological risk assessment, health risk assessment.

Introduction

Water is an important natural resource for human and the environment (Kabir et al. 2020; Proshad et al. 2020). Rapid urbanization and industrial development in many regions across the world, particularly in developing countries, have resulted in serious concerns about water resources since the last decades (Abbasnia et al. 2018; Shams et al. 2020). Therefore, monitoring and assessing the quality of surface water has become a requirement at a global level (Khosravi et al. 2017; Moghaddama et al. 2018; Mgbenu & Egbueri 2019).

Maintaining hygiene systems do not keep pace with industrialization and urbanization growth in developing countries. The unplanned growth of industrialization and urbanization, followed by deforestation, releasing wastewater into wells, and dumping non-treated solid waste in landfills, leads to the deterioration of all aquatic ecosystems including surface water (Ahmed et al. 2010; Bhuyan et al. 2019).

Due to their multipurpose usage, heavy metals are implemented in almost all anthropogenic activities and hence they are existed in different types of wastes that discharged into different environmental counterparts (Islam et al. 2014; Ram Proshad et al. 2019). Heavy metals have the ability to accumulate in the open environments such as soils and water body (Ali et al. 2019; Ashaiekh et al. 2019). Furthermore, in the rainy season, heavy metals are washed from surface soil and mixed them with the water body. Hence, surface water could be at risk from the input of heavy metals.

Contamination by heavy metals is currently considered one of the most marked threats to water quality. Heavy metals are considered harmful pollutants because of their high environmental toxicity, abundance and persistence in various environmental counterparts including surface water. Heavy metals such as Cr, Ni, Cu, As, Cd and Pb are considered to be systemic toxicants, recognized to trigger multiple organ harm even at trace amounts and to capability of bioaccumulation in the major human body systems (R. Proshad et al. 2019). For instance, high intake of Cr and As throughout consumption of contaminated water and foodstuff may cause genotoxic carcinogen (Khan et al. 2015; Kormoker et al. 2019; Kormoker, Proshad, Islam, Shamsuzzoha, et al. 2020). Ni ingestion can also cause fatal cardiac arrest and hypertension (Knight et al. 1997). In addition, exposure to small concentrations of Cd may affect the physiology and health of wildlife. Pb is considered highly toxic and causes several health problems like damage to the nervous system and immune function, blood pressure, abdominal pain, kidney damage, gliomas, lung cancer, and stomach cancer (Mortada et al. 2001; Järup 2003; Kormoker, Proshad, Islam, Tusher, et al. 2020). Moreover, the contamination of surface water by toxic metals may affect other environmental counterparts as these metals may enter into food chain through the consumption of fish and other aquatic plants (Loska & Wiechuła 2003). Furthermore, surface water contamination by metals can be a serious threat to aquatic organisms.

Toxic metals contamination is currently a serious concern in developing countries such as Bangladesh (Islam et al. 2015; Idriss et al. 2020). Unplanned urbanization and industrialization in Bangladesh have adverse effects on surface water quality and other marine fauna. The release of untreated effluents from different industrialization, urbanization, and agricultural activities into open water bodies and rivers has reached an alarming situation in Bangladesh, which continually raises metal contents and degrades the quality of water (Proshad et al. 2020).

The current study focused on the assessment of some toxic heavy metal contents in the surface water from Louhajang River, Tangail district, central Bangladesh. Due to urbanization and industrialization in nearby areas, Louhajang River has received considerable amounts of urban wastes and hence it may be at risk from exposure to toxic metals (Kormoker et al. 2019). However, so far, no scientific research has been reported concerning toxic metal contamination in the surface water of Louhajang river. The main objective of the present study was therefore to evaluate the contents of some toxic heavy metals (Cr, Ni, Cu, As, Cd, and Pb) in the surface water of Louhajang River. The study demonstrates a comprehensive assessment including comparison with standards, spatial distribution, seasonal variation, and statistical analysis, besides the use of ecological and health risk indices.

Materials and methods

Study area

Louhajang River (Fig. 1) has a length of 85 km and a width of 78 m. Louhajang River crosses Tangail city, which is considered an industrial growing area in Bangladesh. The city includes several industries producing a wide range of materials such as metallic workshop tools, batteries, packing materials, leathers, garments, dyes, bricks, and food (Fig. 1). Despite it has a limited area of about 29.04 km^2, Tangail city is considered a densely populated area with population of 750,000 residents in 2017. The residents have also practiced agricultural activities in some areas in Tangail city and along the banks of Louhajang River. Unfortunately, there is no controllable treatment for wastes dumped from industrial, municipal, and household activities. As a result, Louhajang River may be susceptible to environmental pollution from these discharges. It was reported that (Kormoker et al. 2019) wastes were mixed with sediment and water of Louhajang River, resulting in possible river pollution.

Water sample collection and preparation

Collection of surface water samples were performed from forty sites in Louhajang River, which provide good spatial distribution as shown in Fig. 1. Water samples were collected in during two seasons, i.e., the summer (August–September, 2017) and the winter (January-February, 2018) seasons. The samples were collected in triplicate and stored in high density polyethylene bottles and preserved in acidic media using 2-3 drops of 6 N nitric acid (Federation & Association 2005).

Measurements of physiochemical properties and heavy metals

The pH and the EC of the collected water samples were measured on-site and before acidification using portable appropriate meters. Standard protocols were applied for the meter calibration. For metal analysis, all chemicals and reagents used in this study were of analytical grade. The water used in this study was Milli-Q grade, which was purified by Elix® Essential 5 UV Water Purification System, US. The Teflon vessels and high-density polyethylene bottles were cleaned, soaked in 5 % (v/v) HNO_3 for more than 24 h, rinsed with water, and allowed to dry at room temperature. Water sample (20 mL) was treated with 5 mL of 69% HNO_3 and 2 mL of 30% H_2O_2 in a closed Teflon vessel, which was afterward digested in a microwave digestion system. Blank samples were prepared in triplicate following the same procedure. Analysis of samples were performed by inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7700 series). Multi-element Standard XSTC-13 (SpexCertiPrep®, USA) solutions were used for system calibration. The calibration curves with R^2 > 0.999 were accepted for concentration calculation. Multi-element solution (1.0 µg/L) supplied by Agilent Technologies, USA, was used as a tuning solution covering a wide range of element masses. Evaluation of all test batches were carried out using an internal quality approach and were validated if they met the defined internal quality controls (IQCs). Measurement of a run included blank, a certified reference material, and samples were done in triplicate to eliminate possible batch-specific error for each experiment according to the (Islam et al. 2015; Proshad et al. 2020).

Statistical analysis

The results of pH, EC, and metal concentrations in the water samples from Lauhajong River collected during the two seasons were statistically analyzed using the statistical package SPSS 20.0 (International Business Machines Corporation, Armonk, NY, USA). Univariate analysis, in terms of minimum, maximum, mean, medium, standard deviation (SD), relative SD (RSD%), kurtosis, and skewness, was carried out to examine the range and the mean as well as the variation and distribution of parameters in each season. Bi-variate analysis was also carried out in terms of correlation matrices to examine possible relationship between parameters. The confidence intervals of 95% (p 0.05) and 90% (p 0.01) were considered for correlation coefficient analysis. Independent sample t-test was carried out for each parameter to recognize the difference between the summer and the winter seasons. In this approach, the p value < 0.05 indicates significant variation between the two seasons regarding the examined parameter. Furthermore, multi-variate analysis in terms of principal component analysis (PCA) were used to construe the potential sources of toxic metals in surface water as suggested by (Liang et al. 2015). Likewise, in order to trace out the principal components (PC), the Eigen values were used as the extraction method. PCA normalized variables with the Bartlett Sphericity and the Kaiser-Meyer-Olkin (KMO) tests were used to obtain significant PCs in the assessment of data suitability. Similarly, cluster analysis (CA), as another multi-variate analysis approach, was also carried out using the Ward's method and a dendrogram was plotted. CA was applied to classify parameters into sub-clusters in order to acquire a full information of the dataset and to gain insight into the distribution of toxic metals (uncovering of both similarities and differences) (Ali et al. 2019; Ashaiekh et al. 2019).

Spatial distribution analysis

The co-ordinates (latitude and longitude) of 40 sampled sites along Lauhajong River were recorded using Global Positioning System (GPS) meter (Garmin eTrex 10) through which a database was created. The database file also contained values of the examined physiochemical parameters (pH and EC), metal concentrations (Cr, Ni, Cu, As, Cd, and Pb), besides ecological indices. The data in the database file was converted into CSV format to generate the spatial distribution maps using ArcGIS 10.5 software version with the help of Inverse Distance Weighted (IDW) data interpolation technique of the spatial analyst tool module. IDW is a deterministic spatial interpolation approach that is used to estimate an unknown value at a location using some known values with corresponding weighted values. This method estimates the unknown cell values in the output surface by averaging the values of all input sample data points that lie within the specified search radius (Kneissl et al. 2011).

Ecological risk assessment indices

The heavy metal evaluation index (HEI), which provides an overall quality of the water with respect to toxic metals, was calculated as described in Equation 1 (Prasanna et al. 2012; Mokarram et al. 2020);

$$HEI = \sum_{i=1}^{n} \frac{H_{Ci}}{H_{max}} \qquad (1)$$

where, $H_{C,i}$ is the measured concentration of constituent i and $H_{max,i}$ is the maximum allowed concentration of constituent i. According to the (WHO 2011), the maximum admissible concentrations of Cr, Ni, Cu, As, Cd, and Pb are 0.05, 0.02, 0.05, 0.04, 0.005, and 0.05 mg/L, respectively. The HEI is classified as follows: < 10 indicates low risk, 10–20 indicates medium risk, and > 20 indicates high risk (Proshad et al. 2020).

The ecological risk index (ERI) from the river water consumption was also computed to assess the ecological impact using Equations 2 and 3 (Ukah et al. 2019; Egbueri 2020a; Egbueri 2020b);

$$ERI = \sum RI = \sum T_i \times PI \qquad (2)$$

$$PI = \frac{C_s}{C_b} \qquad (3)$$

where RI is the potential ecological risk factor of each metal, T_i is the toxic-response factor of heavy metal, PI is the pollution index, C_s is the concentration of heavy metals in the water sample, and C_b is the corresponding background value. It was reported that the toxic-response factors of the examined metals are as follows: 1, 5, 5, 10, 30, and 5 for Cr, Ni, Cu, As, Cd, and Pb, respectively (Ukah et al. 2019; Egbueri 2020b). The ERI is classified as follows: < 150 indicates low ecological risk, 150 < RI < 300 indicates moderate ecological risk, 300 < RI < 600 indicates considerable ecological risk, and ERI > 600 indicates very high ecological risk (Ukah et al. 2019; Egbueri 2020a).

Human health risk assessment indices

Oral intake and dermal contact are possible exposure pathways of toxic metals in water. Hence, the chronic daily intake (CDI), which indicates the daily amount of toxic metals enter into human body, was calculated for the two pathways using Equations 4 and 5 (Idris et al. 2019; R. Proshad et al. 2019; Asiri et al. 2020; Ebrahim et al. 2020; Proshad et al. 2020). The CDI was calculated for adult male, adult female, and children. The full names and values for each population category are descripted in Table 1.

$$CDI_{oral} = \frac{C \times IR \times EF \times ED}{BW \times AT} \times 10^{-3} \qquad (4)$$

$$CDI_{dermal} = \frac{C \times SA \times K_p \times ET \times EF \times ED \times EV}{BW \times AT} \times 10^{-3} \qquad (5)$$

The assessments of health risk in relation to its non-carcinogenic effect based on CDI that is defined by hazard quotient (HQ) for oral intake and dermal contact pathways were calculated using Equation 6 and Equation 7, respectively (Idris et al. 2019; R. Proshad et al. 2019; Asiri et al. 2020; Ebrahim et al. 2020; Idriss et al. 2020; Proshad et al. 2020);

$$HQ_{oral} = \frac{CDI_{oral}}{RfD_{oral}} \qquad (6)$$

$$HQ_{dermal} = \frac{CDI_{dermal}}{RfD_{dermal}} \qquad (7)$$

where the RfD is the reference dose. The values of RfD of each metal is described in Table 1. The RfD_{dermal} values were calculated by multiplication of the RfD_{ing} by the dermal absorption fraction (ABS) as described in Table 1 (USEPA 2009). Thereafter, the hazard index (HI) was calculated using Equation 8. Based on the USEPA (2007) guidelines, it was reported that HI < 1 is assigned for no health risk is expected to occur, whereas HI ≥ 1 is assigned for moderate or high risk for adverse human health effects (Ali et al. 2019; Idris et al. 2019; Kormoker et al. 2019; Ukah et al. 2019; Kormoker, Proshad, Islam, Shamsuzzoha, et al. 2020; Kormoker, Proshad, Islam, Tusher, et al. 2020; Proshad et al. 2020; Shams et al. 2020).

$$HI = HQ_{oral} + HQ_{dermal} \qquad (8)$$

On the other hand, the total carcinogenic risk (TCR) is the summation of cancer risk (CR) from oral ingestion and dermal contact of water including heavy metals, which were calculated using Equations 9–11. The incremental risk of a person developing cancer over a lifetime resulting from exposure to a possible carcinogen is represented by TCR (Rahman et al. 2018).

$$CR_{Ing} = CDI_{Ing} \times CSF \qquad (9)$$

$$CR_{Dermal} = CDI_{Dermal} \times CSF \qquad (10)$$

$$TCR = CR_{Ing} + CR_{Dermal} \qquad (11)$$

CSF is the cancer slop factor as described in Table 1 in the supplementary file. An acceptable value of $\leq 1.0\times10^{-6}$ for TCR assumes that approximately 1 per 1,000,000 develop cancer as a consequence of the exposure to a carcinogen (Adamu et al. 2015; Proshad et al. 2020). The acceptable range of TCR is generally considered 1.0×10^{-6}-1.0×10^{-4} (USEPA 1999, Rahman et al. 2018).

Results and Discussion

Overview of physiochemical properties and heavy metal concentrations

The descriptive statistical results of the physicochemical properties (pH and EC) as well as heavy metal contents (Cr, Ni, Cu, As, Cd, and Pb) in the surface water samples collected throughout the summer and winter seasons from Louhajang River are shown in Table 2. The raw data of the same parameters in each site in the two examined seasons are shown in Table S1. Primarily, wide ranges of all toxic metals and EC within one season, besides high relative standard deviation (RSD) values (27.35-107.39%) for all toxic metals within one season, were observed. These results reflect high levels of variation within one season that might be attributed to different natural and/or anthropogenic factors controlling the levels of the examined properties in the surface water of Louhajang River. In addition, the kurtosis values of EC as well as the concentrations of Cr, Cu, and Pb in both seasons were all < 1.0, representing no significant tailing in the distribution of the levels of the examined properties along the river surface water. Nevertheless, the kurtosis values of pH in the summer season as well as Ni and As concentrations in both seasons were > 1.0, with a positive trend, indicating significant tailing in the distribution toward high levels. Furthermore, the skewness values of Ni and Cd were > 1, with a positive trend, indicating asymmetrical distribution with larger number of high tails than that of low tails. In contrast, the skewness values of other parameters were < 1.0, indicating symmetrical distribution. For the variations between the two examined seasons, the p values were < 0.05 for all parameters, with the exception of Cd, suggesting significant differences. In this context, the levels of all examined parameters were found to be higher during the winter season than those during the summer season. Low levels in summer season might be attributed to dilution of the river water by the rainfall. Notably, this justification means that the rainfall rate in the summer season exceeds the evaporation rate.

It is known that the pH is an extremely important chemical property as most chemical reactions in the aquatic environment are influenced by pH level (Idriss et al. 2020). In the current study, the pH of the surface water of Louhajang River was slightly acidic to slightly alkaline, which varied from 6.22 to 7.31 during the summer season and from 6.55 to 7.43 during the winter seasons. The acidic media in the summer season could be due to the acidic rainfall. The World Health Organization (WHO) did not recommend specific levels for pH for human consumption for drinking purpose. Nevertheless, some local and international regulatory bodies recommend a range of pH of 6.5-8.5 (EPA 2018; Proshad et al. 2020). In another context, the pH scale between 6.22 and 7.43 suggests that water is fit for aquatic life (Garg et al. 2010), while water at low pH is known to be corrosive and may negatively affect the skin and eyes (Li et al. 2017). In conclusion, the observed pH values in this study indicated that the river water is appropriate for human consumption for drinking purpose and aquatic life and accordingly river food production.

EC is also considered an essential property of drinking water (Subramani et al. 2005). EC is the measure of cations and anions concentrations dissolved in water (Islam et al. 2017; Ahmed et al. 2019). In the current study, the EC value of the water varied from 345 to 654 μS/cm (mean 535.47 μS/cm) in the summer season, while it varies from 647 to 798 μS/cm (mean 724.0 μS/cm) in the winter season. The WHO (2011) considered 1500 μS/cm as the maximum permissible limit (MPL). It was also reported that EC more than 2250 mS/cm indicates a high salinity class (Wu & Sun 2016).

For toxic metals contents, the mean values in the summer season were in the following decreasing order: As > Cu > Cr > Cr > Pb > Ni > Cd, while the mean values in the winter season were in the following decreasing order: Cr > Cu > As > Pb > Ni > Cd.

The mean concentration of Cr in the summer season was 4.35 that was lower than 5 μg/L as the health-based guideline value (HB-GV) recommended by the WHO for human consumption purpose (santé et al. 2004), while the mean concentration of Cr in the winter season was 9.04 μg/L that was higher than the WHO's HB-GV. Nevertheless, Cr concentration in present study was lower than the MPLs recommended by Bangladeshi Drinking Water Standard (BDWS) (DoE 1997), Toxicity Reference Value (TRV) (USEPA 1999), Aquatic Life Water Permissible Limits (ALWPL) (CCME 2007), and Irrigation Life Water Permissible Limits (ILWPL) (CCME 2007). In another context, Cr concentration in the present study was compared with other studies conducted in Bangladesh and other countries (Table 3). All examined rivers in Bangladesh reported higher Cr concentration than that reported in Louhajang River, i.e., the present study. In addition, some studies reported lower Cr concentration than that reported in the present study such as Zohreh River, Iran (Ahmadee et al. 2016), To Lich River, Vietnam (Thuong et al. 2013), and Tarim River, China (Xiao et al. 2014). It was reported that Cr is implemented in various industrial activities including tanning, electroplating, ceramics, dyeing, painting, wood processing, paper, and explosives (Eleryan et al. 2020). As mentioned before, the current study area included the industries of textile, battery, and tanneries that could all be possible contamination sources of Cr.

As shown in Table 2, the mean Ni concentrations in both seasons, i.e., winter 5.07 µg/L and summer 3.16 µg/L, were less than the MPLs recommended by BDWS (DoE 1997), WHO (WHO 2004), and TRV (USEPA 2007) for human consumption for drinking purpose.

Additionally, the mean Ni concentrations in both seasons were far below the MPLs recommended by ALWPL (CCME 2007) and ILWPL (CCME 2007) for aquatic life environment and irrigation purpose, respectively. Table 3 shows that the mean Ni concentration in the current study was comparable with that in water samples from Meghna River (Islam et al. 2020), Bangladesh, while they were lower than water samples from Yellow River, China (Gao et al. 2019) and Tarim River, China (Xiao et al. 2014). Notably, mining activities, besides oil and coal combustion, nickel metal refining, and sewage sludge incineration, were all reported considerable sources of Ni contamination (Obasi & Akudinobi 2020).

The range Cu concentrations in both seasons was 0.96-15.92 µg/L, which was lower than the HB-GV recommended by both WHO (2004) and BDWS (DoE 1997). Notably, some samples recorded Cu concentrations higher than TRV (9 µg/L) (USEPA 1999), which is based on short-term exposure and intended to protect against direct gastric irritation that is a concentration-dependent phenomenon. In another context, the range of Cu concentrations in both seasons were lower than the standard levels recommended by ALWPL (CCME 2007) and ILWPL (CCME 2007) for aquatic life environment and irrigation purpose, respectively. Cu may be detrimental to plants as the Cu-enriched liquid dairy waste is used as irrigation water in agricultural land (White & Brown 2010). Excessive amounts of Cu are often detrimental to plants and extremely poisonous to certain microbes (Hasnine et al. 2017).

Arsenic is called "soft poison" or "death metal" since it gradually kills people after entering the human body (Nawab et al. 2018). As shown in Table 2, the mean As concentrations in winter (6.83 µg/L) and summer (4.88 µg/L) did not exceed all recommended standards for drinking, aquatic life, and irrigation purposes. Compared with other rivers, the current levels of As was found much lower than Buriganga River, Bangladesh (Bhuiyan et al. 2015), Korotoa River, Bangladesh (Islam et al. 2015), and To Lich River, Vietnam (Thuong et al. 2013), while the current levels were found slightly lower than Rupsha River, Bangladesh (Islam et al. 2020), Yellow River, China (Gao et al. 2019), and Tarim River, China (Xiao et al. 2014). In contrast, As levels in the current study were found slightly higher than Meghna River, Bangladesh (Islam et al. 2020), Shitolokkha River, Bangladesh (Islam et al. 2020), Pasur River, Bangladesh (Islam et al. 2020). Fertilizer and pesticide industry, wood industry by exhausting copper arsenate, and tanning in relation to certain chemicals, particularly arsenic sulfide, were all reported possible sources of As (Bhuiyan et al. 2011; Fu et al. 2014).

Table 2 shows that the mean Cd concentrations in both seasons were lower, as for As, than all standards recommended for drinking, aquatic life, and irrigation purposes. However, Table S1 shows that site-2 recorded Cd concentration (2.42 µg/L) higher than the TRV standard (2.0 µg/L) (USEPA 1999), while site-1 recorded marginal Cd concentration (1.96 µg/L). Several industries release Cd to the environment. However, the production of batteries, dyes, and alloys were reported as dominant sources (Hanfi et al. 2020). The mean Cd concentrations in the current study showed comparable mean concentration in Shitolokkha River, Bangladesh (Islam et al. 2020) and Zohreh River, Iran (Ahmadee et al. 2016).

The mean Pb concentration in the summer season was lower than the HB-GV standard (WHO 2011), whereas the mean Pb concentration in the winter season was at greater level compared to the HB-GV standard. Pb in the urban environment is released from both natural and manmade sources. Nevertheless, it was reported that the major manmade sources are industrial activities such as mining, manufacturing, and fossil fuel burning, in addition to different agricultural and domestic applications as well as traffic emissions and weathering of materials (Hanfi et al. 2020). Pb is a non-essential metal and, through the routes of exposure, it may affect the gastrointestinal tract, liver, and central nervous system. Pn often breaks the blood-brain barrier and interferes with an infant's natural brain development (Rajeswari & Sailaja 2014). In aquatic plants, the acute toxicity typically occurs by Pb contamination at a concentration of 100-500 µg/L. Enzymes needed for photosynthesis are inhibited when Pb exceeds 0.5 µg/L in algae (Sadiq et al. 2003). Fish are more prone to Pb than algae. Additionally, high level of Pb can affect the gill function of the fish. In comparison with other metals, lead at low concentrations may pose a life threat in aquatic ecosystems. Pb in the current study shows much lower concentration than that in

Buriganga River, Bangladesh (Bhuiyan et al. 2015) and slightly higher than that in Shitolokkha River, Bangladesh (Islam et al. 2020), Pasur River, Bangladesh (Islam et al. 2020), Rupsha River, Bangladesh (Islam et al. 2020), Korotoa River, Bangladesh (Islam et al. 2015).

Spatio-seasonal distribution of physiochemical properties and heavy metal concentrations

The spatial distribution of the examined physiochemical properties and heavy metal concentrations in the surface water samples throughout the summer as well as the winter seasons collected from Louhajang River are depicted in Fig. 2 and 3, respectively. As observed, while most spots in the riverbed were alkaline in the winter season, few spots in the riverbed area were alkaline in the summer season. It could be observed that the spatial distributions of EC in both seasons were almost similar. In general, the EC in the downstream in both seasons was higher than that in the upstream. This result indicates that the downstream was more influenced by discharge flux (Mao et al. 2019).The distributions of Cr, Cu, and Pb recorded few hot spots in almost the same sites in both seasons; an issue that suggest permanent source during a year for those metals. Notably, the release from the dying industry (Figure 1) could be a source of Cr in the hot spot sites (Fig. 2 and 3). For Ni distribution, the upstream showed more hot spots than the downstream in both seasons. This result may be due to the release from the metal workshops that area located near the upstream. While As recorded few hot spots in the downstream in both seasons, Cd recorded few hot spots in the upstream in both seasons.

Source analysis

The matrices of the Pearson's correlation coefficients of pH, EC, and the examined heavy metal concentrations in the surface water samples collected during the summer and the winter seasons from Louhajang River are shown in Table 4. In principle, inter-metal interactions could provide an indication of the origins and paths of metals in water. The matrix reveals significant positive correlation, at 0.01 level, in both seasons between Cr and Ni, in addition to significant negative correlation, also at 0.01 level, in both seasons between As and Cd. Moreover, significant positive correlations, at 0.05 level, in both seasons were recorded for the combinations of Cr-Cd and Ni-Cd, while significant negative correlation, at 0.05 level, was recorded for the combination of Ni-As. Notably, significant positive correlation in the summer season was observed between pH and EC.

The positive significant relationships suggest that the parameters were interconnected and may derived from the same source in the study area. Accordingly, the positive significant correlations between the three metals Cr, Ni, and Cd indicate that some common pollution trends remain between them (Mao et al. 2019). The absence of a strong positive association between the three metals Cu, As, and Pb, on the other hand, suggests that they were not controlled in the study area by a single factor (Kükrer et al. 2014).

The dendrograms obtained from CA of pH, EC, and the examined heavy metal concentrations during the two seasons are depicted in Fig. 4. It could be observed that the parameters in the summer season were classified into 4 clusters with the following description: cluster-1 included pH and EC, cluster-2 included Cr, Ni, and Cd, cluster-3 included Cu and Pb, and cluster-4 included only As. For the winter season, four clusters could also be observed as follows: cluster-1 included pH, cluster-2 included EC and As, cluster-3 included Cr, Ni, and Cd, and cluster-4 included Cu and Pb. Notably, the nearest parameters in both seasons (at 0.5 distance) were Cr and Ni, indicating that those two metals were controlled by similar contribution sources. Cd was the nearest metal to the Cr-Ni combination, an issue that may suggest similar sources as well.

The rotated component matrix obtained from PCA of pH, EC, and the examined heavy metal concentrations in the two seasons are shown in Table 5. The raw data before and after rotation in the summer and winter seasons are shown in Table S2 and Table S3, respectively. Table 5 shows that three components, out of eight, were extracted in each season. Primarily, this result indicates that the number of contribution factors controlling all examined parameters in both seasons was similar. Interestingly, the cumulative loading values of the extracted components in both seasons were almost similar (64%). The percentages of component-1, -2, and -3 in the summer season were 30%, 18%, and 16%, respectively, while the percentages of component-1, -2, and -3 in the winter season were 30%, 19%, and 15%, respectively. These results strengthen the suggestion of that similar factors controlled the levels of the examined parameters in both seasons. In details, the first principal in both seasons were significantly loaded ($p > 0.5$) by Cr, Ni, and Cd with positive trend as well as by As with negative trend. This result suggests similar sources of contribution by Cr, Ni, and Cd. The second principal in both seasons was significantly loaded by EC, suggesting that this component was probably from natural origin. This is because EC is the measure of dissolved ion concentrations, which is controlled by the major components that are most probably from natural origin. Cu significantly loaded the third principal but with different trends. This result suggests that the source controlling Cu concentration differed than sources controlling the other examined metals.

Ecological risk assessment

Table S4 shows the HEI values of the examined heavy metals concentrations in the surface water samples collected during the summer and the winter seasons from Louhajang River, Bangladesh. The HEI values in the summer season ranged from 0.22 to 1.19, whereas the HEI values in the winter season ranged from 0.32 to 1.82. Based on the WHO (2011) classification, all water samples were at low risk from exposure to the examined heavy metals. In a previous study on water samples from Kor River, Iran (Mokarram et al. 2020), 15 sites, out of 29 sites, were classified at high risk having HEI values in the range of 20-140, which was attributed to effluents from industries located around the polluted sites. In contrast, another study in Curtin Lake, Malaysia (Prasanna et al. 2012) reported HEI values in the range of 8.57-12.11. Higher values of HEI were attributed to the contribution of Fe, Pb, and Se by some manmade activities.

On the other hand, the spatial distributions of HEI along Louhajang River during the summer and the winter seasons are depicted in Fig. 5. As observed, the spatial distributions of HEI in the two seasons were almost similar. Nevertheless, the HEI values in upstream was higher than the HEI values in the downstream. This result may suggest the influence of one or two phenomenon, i.e., the manmade activities near the upstream banks of the river more influenced on the quality of the river water than the manmade activities near the downstream banks of the river. The other phenomenon is attributed to the natural dilution effect from the upstream to the downstream of the river.

The ERI values in the surface water samples collected from 40 sites along the Louhajang River in the summer and the winter seasons are shown in Table S5. The HEI values in the summer season ranged from 47.32 to 190.31, whereas the HEI values in the winter season ranged from 68.41 to 293.58. Based on the classification proposed by (Taiwo et al. 2019), 80% of the examined sites were at low risk (ERI < 150), while 20% were at moderate risk (ERI 150–300). For the winter season, 25% of the examined sites were at low risk, while 75% were at moderate risk.

The spatial distributions of ERI during the summer and the winter seasons are depicted in Fig. 6. With reference to Fig. 1, it may suggest that the effluents from the industrial area and the metal workshop put the river water in the upstream at moderate risk in both seasons. With reference to Fig. 2, the moderate risk in both seasons was caused by high levels of Cr, Ni, and Cd. Additionally, the agricultural activity and/or aviation activities in the vicinity of the midstream may also put the river water at moderate risk, which was caused by high levels of Cr, Ni, Co, and Pb.

Health risk assessment

The values of CDI_{oral}, CDI_{dermal}, HQ_{oral}, and HQ_{dermal} for the three examined population categories including adult male, adult female, and children possibly exposed to all examined heavy metals are shown in Tables S6-S17, respectively. In addition, the values of HI through oral and dermal pathways for adult male, adult female, and children upon possible exposure to all examined heavy metals are shown in Tables S19-S20, respectively. The minimum and the maximum levels of HI are shown in Table 6. As observed, the minimum HI values for Cr and Pb in both seasons summer and winter for the three examined population categories were more than unit, indicating possible non-cancer risk from exposure to Cr and Pb in the surface water samples of Louhajang River in all sampled sites through both oral ingestion and dermal contact pathways. In contrast, the minimum HI values (Table 6) for Ni, Cu, and Cd for the three examined categories were less than unit, while the maximum values for all examined heavy metals for all population categories were more than unit. This result indicates that adult male, adult female, and children were at possible non-cancer risk from exposure to Ni, Cu, and Cd in water samples through both oral and dermal pathways from some sites along Louhajang River. For Ni, Cu, and Cd, only one to six sampled sites (Tables S18-S20) recorded HI less than unit, while the rest of sampled sites recorded HI more than unit. These results suggest that the majority of sampled sites along Louhajang River may cause non-cancer risks from oral and dermal exposure pathways. Interestingly in As in particular, the majority of sampled sites recorded HI of more than unit for adult male, whereas all sampled sites recorded HI less than unit for adult female and children.

On the other side, during the summer and the winter seasons, the CR_{oral}, CR_{dermal} and TCR from possible exposure to the examined carcinogenic heavy metals through the oral ingestion and dermal contact pathways for adult male, adult female, and children are shown in Tables S21-S29. Among the examined elements in the current study, the carcinogens were Ni, As, and Pb.

The minimum values of TCR from Ni exposure (Table 6) for the three examined population categories were less than 1.0×10^{-6}, indicating acceptable range, while the maximum values were more than 1.0×10^{-6}, indicating possible cancer risk from exposure through oral ingestion and dermal contact pathways. In particular, four, four, and five sampled sites recorded TCR more than 1.0×10^{-6} during the summer season for adult male, adult female, and children, respectively, while seven, eight, and 12 sampled sites recorded TCR more than 1.0×10^{-6} during the winter season for adult male, adult female, and children, respectively. Unfortunately, the TCR values of As (Table 6) for the three examined population categories during both seasons summer and winter were more than 1.0×10^{-6}, indicating possible cancer risk from exposure to As in water samples from all sampled sites along Louhajang River. Fortunately, the TCR values of Pb (Table 6) in all sampled sites during both seasons were $< 1.0\times10^{-6}$, indicating no cancer risk.

Conclusion

This communication reports a comprehensive assessment of some toxic metal contents in the surface water of Louhajang River, Bangladesh, during the summer and the winter seasons. Among several approaches used for assessment is the comparison against several local and international standards, which included the Bangladesh Drinking water standards, the Word Health Organization standards for drinking purpose, the toxicity reference values reported by the United States Environmental Protection Agency, the Canadian Water Quality Guidelines for the Protection of Aquatic Life, and the irrigation water permissible limits. The metal contents in the current study reported different trends against these standards.

Statistical analyses including uni-, bi-, and multi-variates were also applied for assessment. For instance, significant positive correlations in both seasons were recorded for the combinations of Cr-Ni, Cr-Cd, Ni-Cd, signifying related sources of those metals in the river water. Moreover, the CA classified the examined parameters, i.e., pH, EC, and heavy metals contents, into similar clusters in both seasons; an issue that indicates similar conditions controlled those parameters during both seasons. The PCA extracted three principal components, out of eight, suggesting mainly three sources controlling the examined parameters. Notably, the second principal in both seasons was significantly loaded by EC, suggesting that this component was probably from natural origin.

The spatial distribution of pH reported that most spots in the riverbed were alkaline in winter, whereas few spots in the riverbed area were alkaline in summer. In contrast, EC reported similar spatial distribution during the two seasons. In general, the EC in the downstream in both seasons was higher than that in the upstream. This result indicates that the downstream was more influenced by discharge flux. The spatial distributions of Cr, Cu, and Pb recorded few hot spots in almost the same sites in both seasons, indicating permanent sources. Moreover, hot spots were observed for Cr and Ni, which were attributed to the release from dying industry and metallic workshops.

The HEI and ERI, as ecological indices used for overall assessment of collection of heavy metals, were also applied in this study. Based on the WHO classification of HEI, the water was at low risk from exposure to heavy metals. Nevertheless, the ERI reported low to moderate risks.

The HI, as a health risk assessment index, reported possible non-cancerogenic risk from exposure of adult male, adult female, and children through oral intake and dermal contact pathways to Cr and Pb in the river water from all sampled sites. Moreover, the majority of sampled sites put adult male were at risk from exposure to As, while no possible risk was reported for adult female and children. TCR, as another health risk assessment index, was applied as well. As showed possible cancer risk for the three examined population categories during both seasons, while Pb showed no possible cancer risk. Additionally, Ni reported that some sampled sites were at cancer risk.

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Ethical Approval

Not applicable.

Consent to Participate

Not applicable.

Consent to Publish

Not applicable.

Authors Contributions

Ram Proshad, Tapos Kormoker, Abu Sayed Shuvo and Maksudul Islam collected water samples during winter and summer season. Md. Saiful Islam and Dan Zhang designd the total experiment. Abubakr Mustafa Idris and Md. Saiful Islam analyzed the data. Ram Proshad, Tapos Kormoker, Md. Saiful Islam wrote the whole manuscript whereas Dan Zhang, Md Nazirul Islam Sarker and Sujan Khadka revised and improved the whole manuscript. All authors reviewed and approved this manuscript.

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Competing Interests

The authors declare that they do not have any competing interests that could have appeared to influence the work reported in this paper.

Availability of data and materials

Not applicable.

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Figures

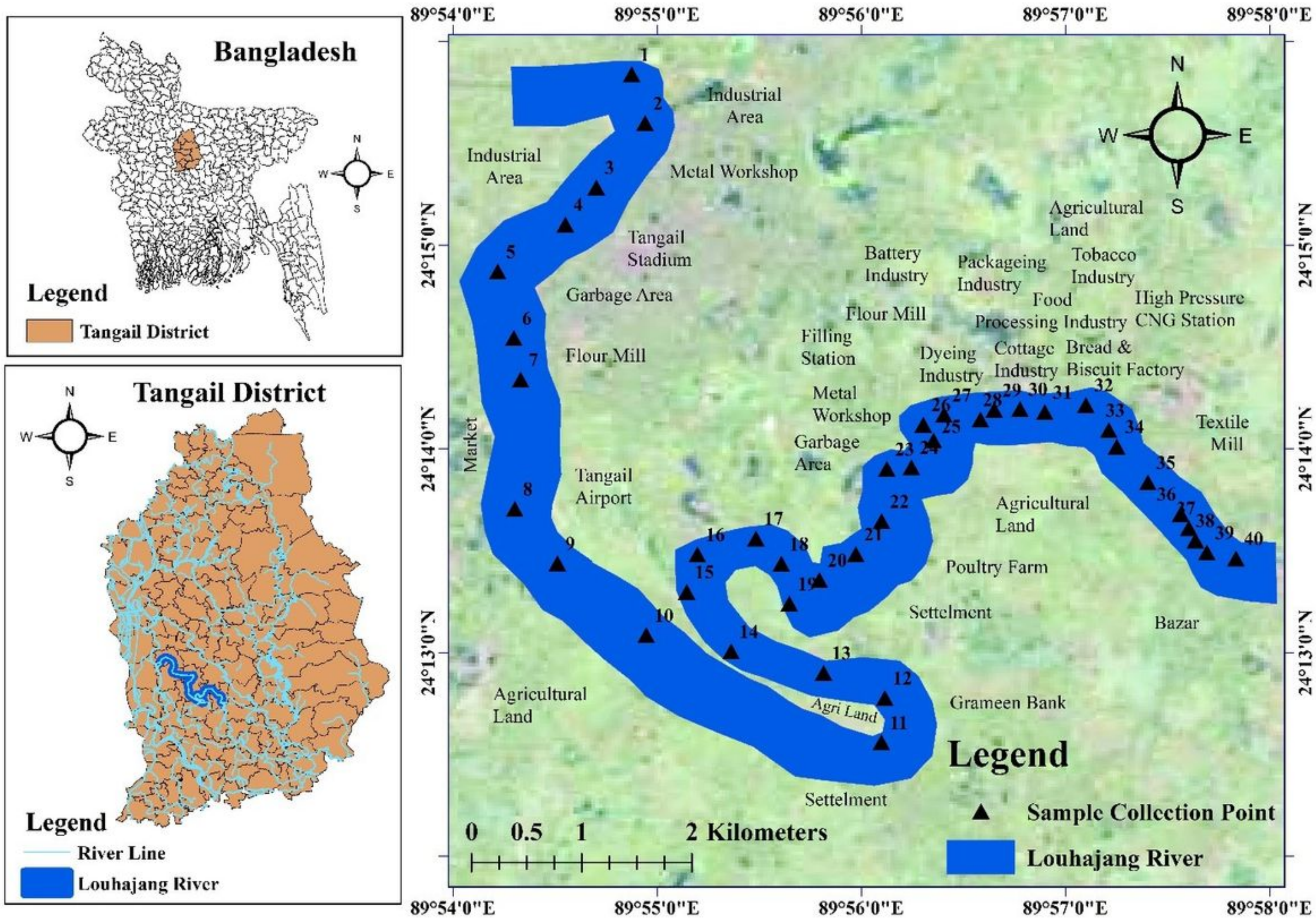

Figure 1

Sampling sites and the major manmade activities along Louhajang River, Bangladesh. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

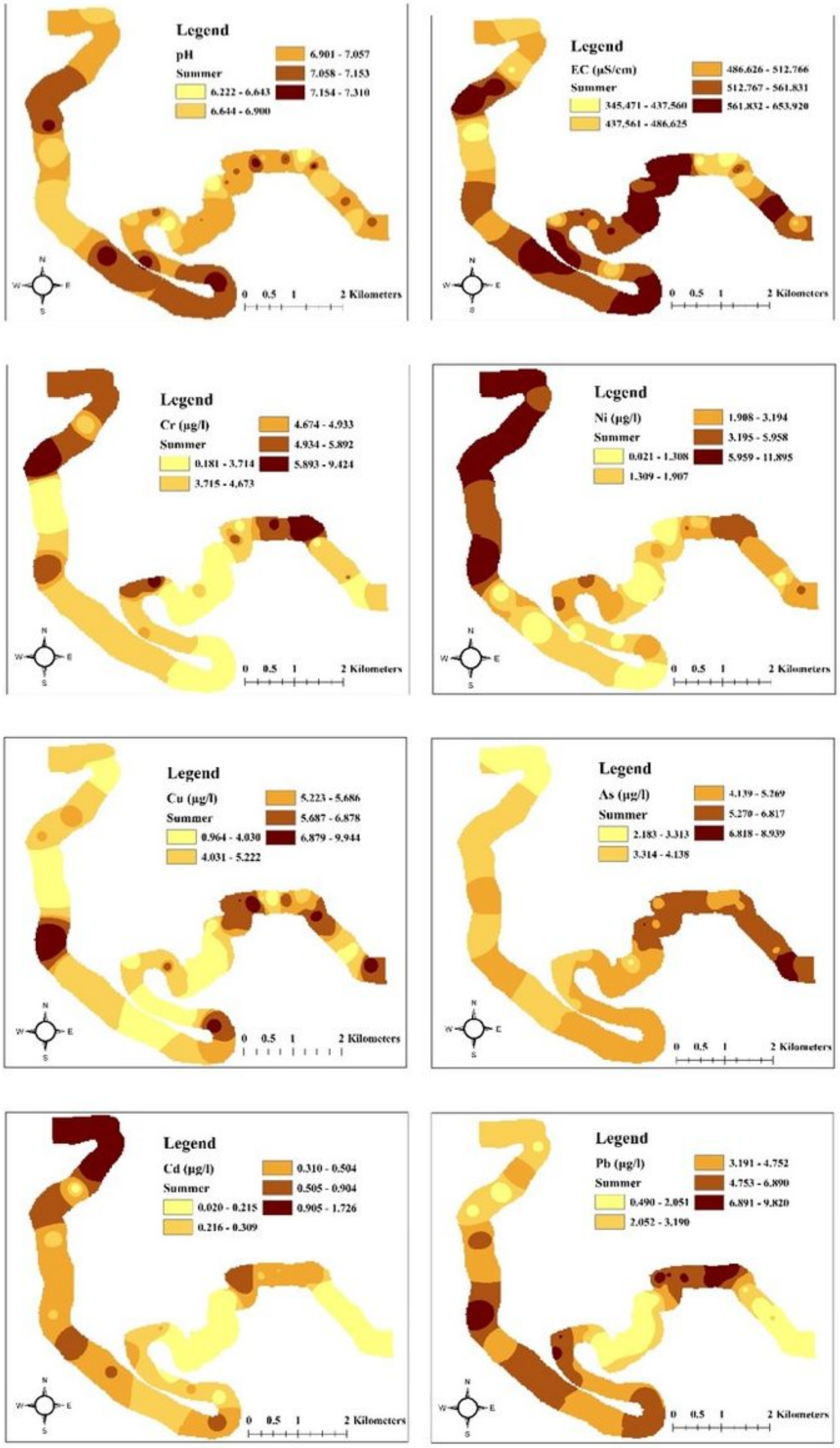

Figure 2

Spatial distribution of pH, EC, and the examined toxic metal concentrations in the surface water samples collected during the summer season from Louhajang River, Bangladesh. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

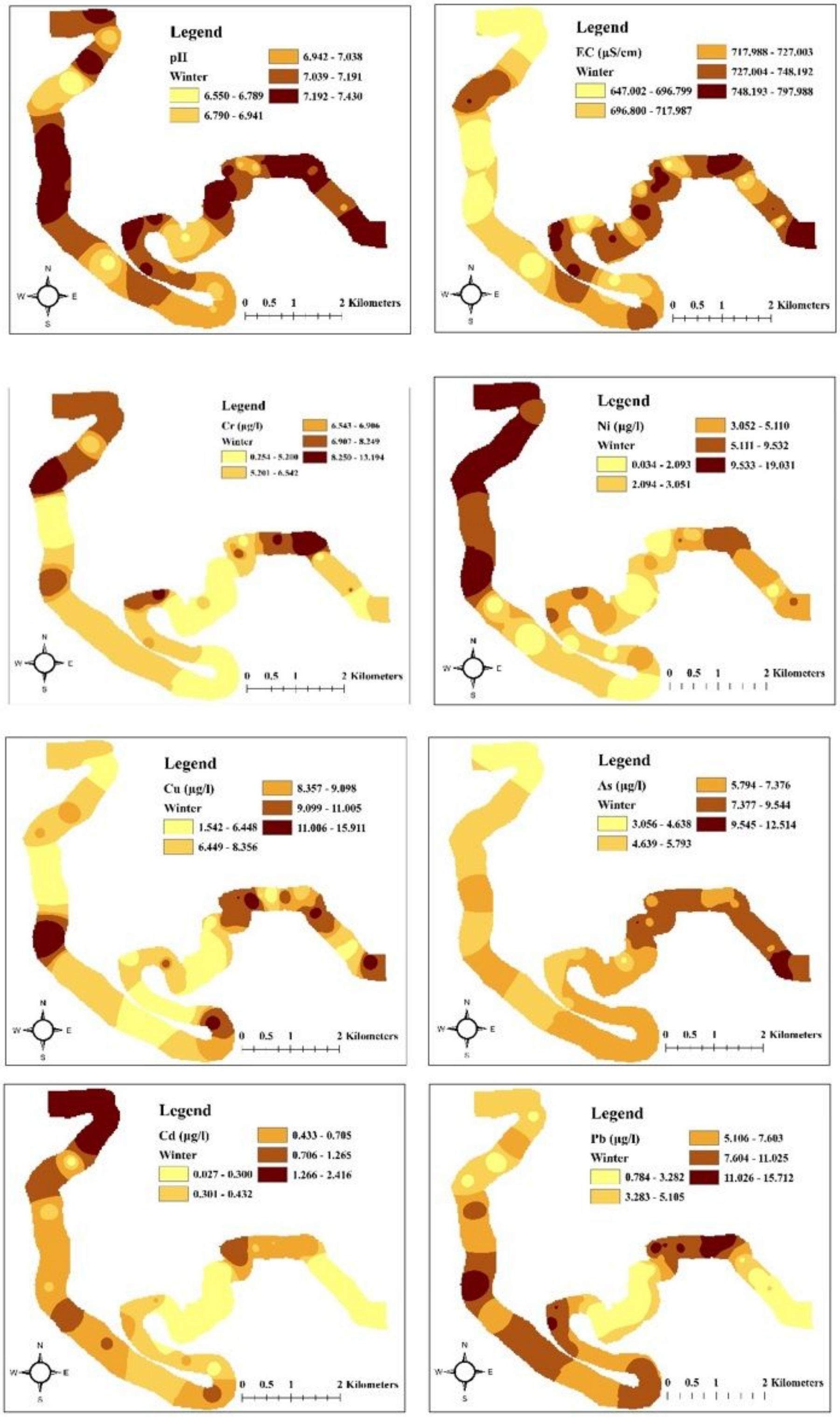

Figure 3

Spatial distribution of pH, EC, and the examined toxic metal concentrations in the surface water samples collected during the winter season from Louhajang River, Bangladesh. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

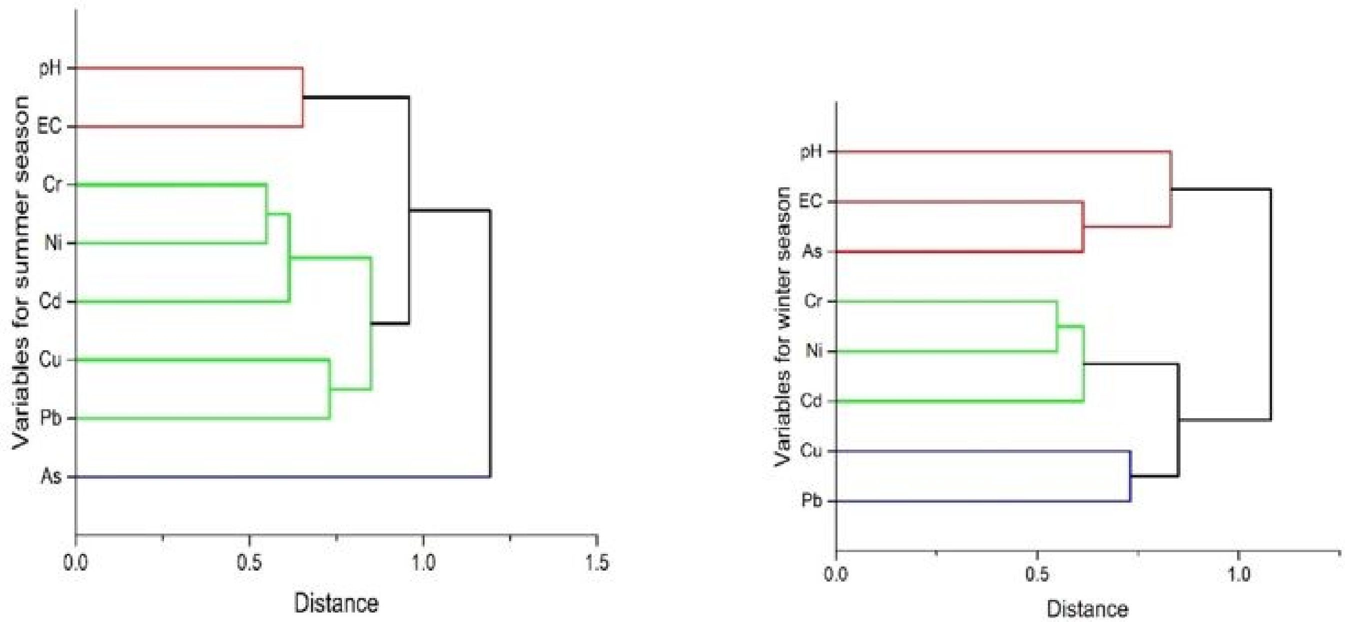

Figure 4

Dendrograms obtained from cluster analysis of pH, EC, and the examined toxic metal concentrations in the surface water samples from Louhajang River in the summer and the winter seasons

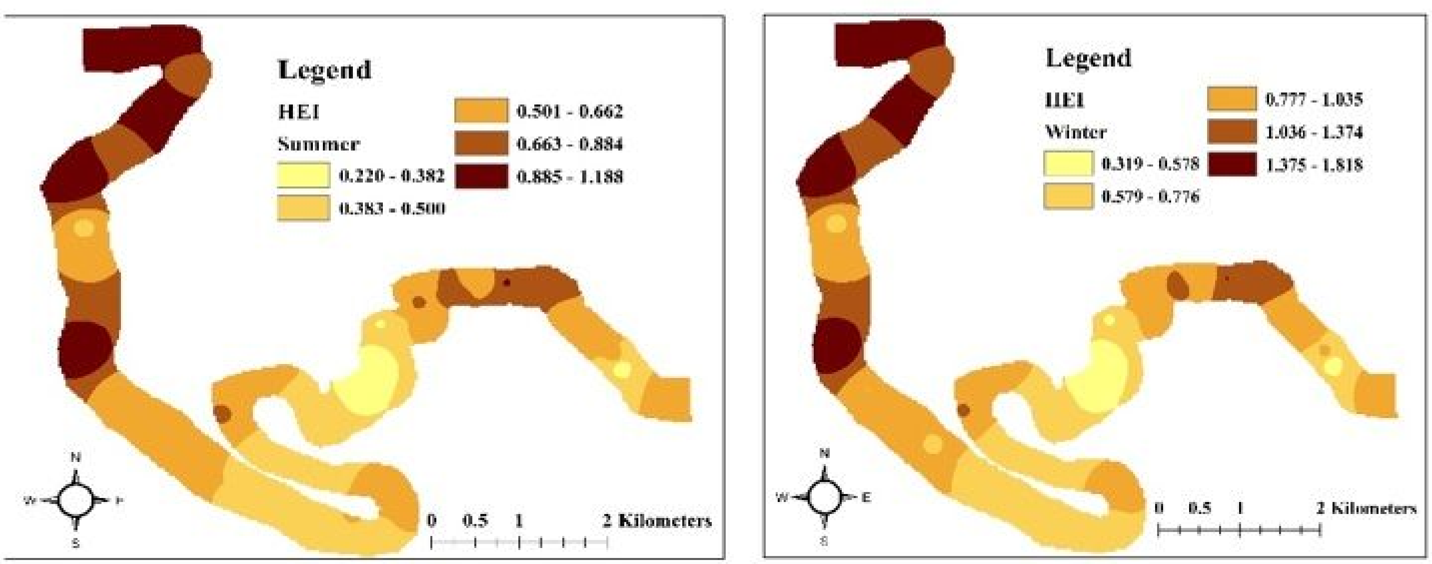

Figure 5

Spatial distribution of heavy metal evaluation index (HEI) in the surface water samples collected during the summer and the winter seasons from Louhajang River, Bangladesh. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

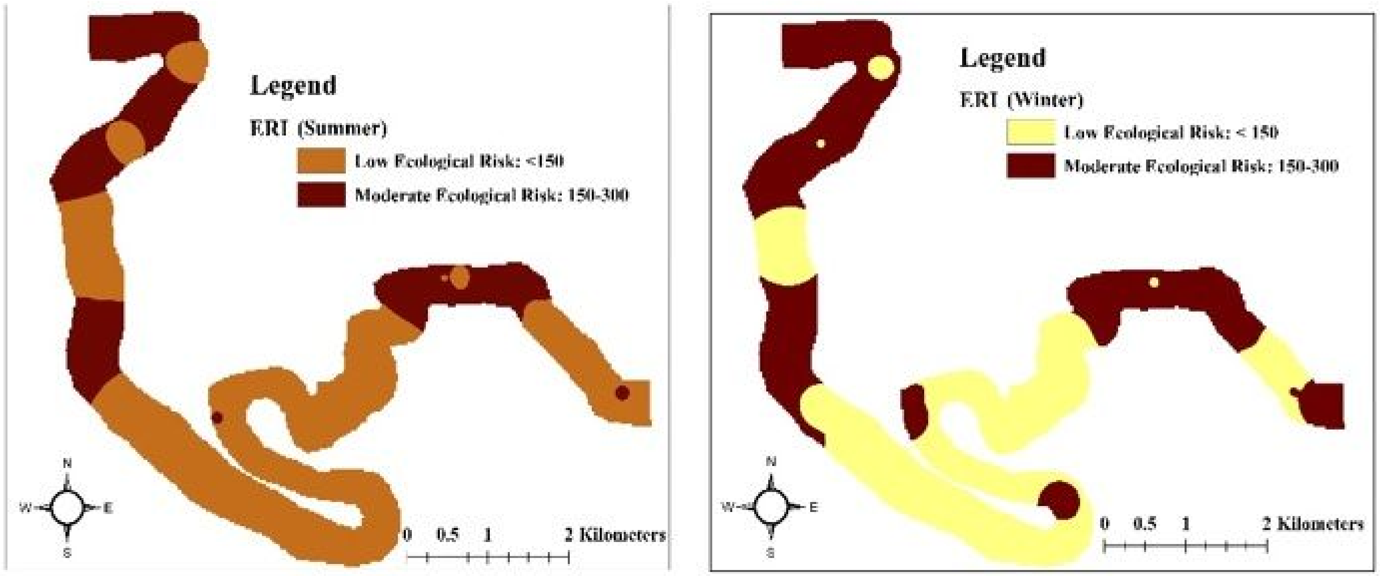

Figure 6

Spatial distribution of ecological risk index (ERI) in the surface water samples collected during the summer and the winter seasons from Louhajang River, Bangladesh. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

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