



Seasonal Dynamics of Heavy Metal Concentrations in Water, Fish and Sediments from Haor Region of Bangladesh

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ABSTRACT

Heavy metal concentrations in water, fish, and sediments from Bangladesh's haor region were investigated. Fish and sediment samples were taken once a season and evaluated using standard methods, while water samples were collected monthly. Results showed that metals in water (mg/L) were found in order of Mn (0.1694) > Cu (0.0189) > Zn (0.0045) > Pb (0.0040) > Cd (0.0028) within the maximum permissible level of Environment Conservation Rules (ECR). Mean concentrations of metal in fish (mg/kg–dry wt.) found in order of Zn (56.16) > Cu (25.47) > Mn (4.36) > Pb (2.19) > Cd (1.27) that were higher than maximum allowable level of Food and Agricultural Organization (FAO) except Cu. Metal in sediments (mg/kg) found in order of Mn (127.61) > Zn (32.51) > Pb (10.09) > Cu (5.40) > Cd (0.43), and except Cu all metal concentrations were lower than the Environmental Protection Agency's (EPA) probable effect concentrations. In water and sediments, pollution indices revealed a critical pollution threshold for water, and a range of unpolluted to highly polluted for sediments. Sampling sites had low potential ecological risk, despite the fact that metals were showing signs of a negative impact on people' health. Furthermore, bio-concentration factor for fish and water was low to extremely high, but for fish and sediment was low. The level of heavy metal contamination in haor shows the situation is alarming for biota and residents of the region. The relevant authority should control and monitor the aquatic ecology in order to protect it.

Keywords: aquatic environment, heavy metal, health risk, pollution indices, Bangladesh

INTRODUCTION

Because of its environmental toxicity, abundance, and durability in recent years, metal contamination in the aquatic environment has received global attention (Sin et al., 2001; Armitage et al., 2007; Yuan et al., 2011). Due to global rapid population increase and intensive home activities, as well as rising industrial and agricultural production, large quantities of harmful chemicals, particularly heavy metals were released into rivers around the world (Srebotnjak et al., 2012; Su et al., 2013; Islam et al., 2014). Because of the practice of discharging untreated domestic and industrial wastewater into water bodies, which leads to a rise in the quantity of metals in river water, rivers in metropolitan areas have also been linked to water quality issues (Khadse et al., 2008; Venugopal et al., 2009). The aquatic habitat of haor regions, together with its water quality, is thought to be the most important factor in determining the health and illness status of both cultivated and wild fish (Malik et al., 2010; Islam et al., 2017a). Increased population, modernization, commercialization, and agricultural practices have exacerbated the situation in the haor region, particularly in Bangladesh's

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Kishoreganj district (Gupta et al., 2009; Islam et al., 2017a). Heavy metals cannot be destroyed therefore they end up being deposited, assimilated, incorporated, or stored in water, sediments, and aquatic organisms, polluting water bodies (Malik et al., 2010). As a result, heavy metals can bio-accumulate and bio-magnify in the food chain before being digested by humans, posing health hazards (Agah et al., 2009). As a consequence, because fish occupy high trophic levels and are essential food sources, they are frequently utilized as markers of heavy metal contamination in the aquatic ecosystem (Blasco et al., 1999). Thus, determining the amounts and distribution of heavy metals in aquatic environments is critical.

Furthermore, fish are an important element of the human diet, so it's not unexpected that numerous researches on metal pollution in various edible fish species have been conducted (Hulya and Erhan, 2000; Prudente et al., 1997). Specifically, toxicological and environmental researches on fish have sparked interest in determining hazardous components in seafood (Waqar, 2006). Pollutant analysis in live animals is more appealing and promising than pollutant analysis in the abiotic environment, because living organisms offer exact information regarding pollutant bioavailability (Peakall and Burger, 2003). This could help forecast pollutant transfer exposure and its potential health effects on humans. Furthermore, such data is critical for developing accurate risk assessments for seafood safety. Heavy metals have a tendency to accumulate in numerous organs of aquatic species, particularly fish, according to a report, and can then enter the human metabolism through eating, posing major health risks (Puel et al., 1987). Every day, untreated industrial, commercial, and home wastes and effluents are dumped into open water bodies and neighboring areas in Bangladesh. The local community uses the water for a variety of reasons, including fishing. A variety of pollutants from various sources, including heavy metals, have had a significant impact on the haor water. Contamination of this magnitude must be a major concern for the aquatic ecosystems and animals' health, as well as human health. Contaminants, particularly heavy metals, are increasingly polluting the water and sediment quality in this area due to oil spills from a large number of water vessels, sewage from business and domestic sectors, and run-off from agricultural land (Ololade et al., 2007; Islam et al., 2014; Islam et al., 2015; Islam et al., 2017a). As a result, the primary question is whether the concentration of pollutants in the water body poses a threat to humans and aquatic life. However, contamination of toxic metals has now become a global environmental issue in both developed and developing countries (Kabir et al., 2020c). High levels of critical and dangerous metals in sediments and fish have been shown to have a substantial impact on the ecosystem, posing a threat to the ecosystem's occupants (Alhashemi et al., 2012a; Kabir et al., 2020a; Nasrabadi et al., 2015). Furthermore, according to Islam et al. (2021), excessive buildup of toxic metals can cause chronic illnesses in people through food chain interactions, even at low concentrations throughout a lifetime, and can cause a variety of carcinogenic and non-carcinogenic health problems. As a result, it is critical to understand the concentrations of trace elements in water, sediments, and fish that reside on the same sphere, as this provides insight into the levels of trace element contamination in the ecosystem (Kabir et al., 2020a; Kabir et al., 2020b; Alhashemi et al., 2012a). Several numerical water and sediments quality indices such as pollution load index (PLI), potential ecological risk (PER), geo-accumulation index (Igeo) and bio-concentration factor (BCF) were integrated to assess the contamination level of essential and toxic metals in water and sediments segments. Many methods have been used to assess health risks, but the most common are target hazard quotients (THQs), which are based on the concentrations of both essential and toxic metals in edible parts of fish in comparison to the reference dose of metal intake per body weight of consumers, while carcinogenic health risk is assessed by calculating the target cancer risk (TR).

Due to its high contamination, a study on the haor aquatic ecosystem has recently sparked public alarm. Agricultural activities have developed significantly in the wetland and haor areas during the last few decades, negatively impacting wetland ecosystems on both qualitative and quantitative levels (IUCN, 2005; Rahman et al., 2013). Siltation, over-exploitation of natural resources, excessive use of agro-chemicals, and other ecological and man-made disruptions are the reasons of haor aquatic ecosystem degradation, resulting in a shortage of food, fuel, and fodder, as well as deterioration of the aquatic environment and impoverishment (Uddin et al., 2013; Mokaddes et al., 2013). As a result, the dangers of pollution effect are increasing in lockstep. There has been no scientific research on heavy metal issues in the study area to yet. Therefore, this study was carried out to achieve the following objectives as i) to assess the pollution status of the Kishoreganj haor by estimating the levels of heavy metals in water, fish and sediment; and ii) to observe the pollution level, human health risk and bio-concentration factors of fishes in relation to water and sediments.

MATERIALS AND METHODS

The research was carried out in the Baro haor of Nikli upazila in the Kishoreganj district of Bangladesh, which is located between the latitudes of 24°15' and 24°27'N and the longitudes of 90°52' and 91°03'E (Figure 1). With a total area of 2688.59 km², the Kishoreganj district is bordered on the north by Netrokona and Mymensingh districts, on the southwest by Narsingdi district, on the southeast by Brahmanbaria district, on the east by Sunamganj and Habiganj districts, and on the west by Gazipur and Mymensingh districts (Banglapedia, 2013). The study area was divided into five different sampling stations denoted as: St-1 (Bayershuil beel), St-2 (Tegulia beel), St-3 (Singpur beel), St-4 (Neora beel), and St-5 (Bara beel).

During September 2016 to August 2017, water samples were taken from St-1, St-2, St-3, St-4, and St-5 of the haor, which was divided into three seasons: post-monsoon (October to January), pre-monsoon (February to May), and monsoon (June to September). The sampling plastic bottles were cleaned and rinsed with detergent solution before being treated overnight with 5% nitric acid (HNO₃). To collect 500 ml water from each sampling station, the pre-prepared sampling bottles were dipped about 10 cm below the surface water. Following collection, samples were acidified with 10% HNO₃ and transported to the laboratory in an ice bag. To avoid further contamination, the materials were filtered using a 0.45 μm micro-pore membrane filter and kept frozen until analysis. During the study period, the available fish species (Tengra: *Mystus vittatus*) were taken directly from fishermen once a season (post-monsoon, pre-monsoon, and monsoon season) from the above specified sampling stations. Each sampling site collected approximately 200 g of fish samples, which were frozen for preservation and to prevent further contamination until analysis. The sediment samples were taken from the above-mentioned sampling locations about 1 m below the water surface once a season (post-monsoon, pre-monsoon, and monsoon season). Approximately 1000 g sediment samples were collected from each sampling site using a grab sampler, put in polyethylene bags, and transported to the laboratory with an ice box.

The Bangladesh Institute of Nuclear Agriculture (BINA) in Mymensingh, Bangladesh, studied heavy metal concentrations in water. Using a pipette, 50 mL water was poured to a beaker, followed by 2 mL pure HNO₃ and the beaker was placed on the hot-plate for digestion. Following adequate digestion, the sample was placed in a 50-ml volumetric flask and filled to the mark with distilled water. Finally, a filter paper (Whatman Qualitative 1) was used to filter the sample and it was stored in a container (APHA, 1998). The contents of lead (Pb), copper (Cu), cadmium (Cd), zinc (Zn), and manganese (Mn) in water samples were

determined using an atomic absorption spectrophotometer (AAS) as defined by the American Public Health Association (APHA) (1998). The Bangladesh Institute of Nuclear Agriculture examined heavy metal content in fish (BINA). For instrumental analysis, tissue samples from each fish were weighed, dried, and microwave digestion was performed (Damodharan and Reddy, 2013). The residues were diluted to 25 mL with 2.5 percent HNO₃ after digestion. Metal analyses of samples (Cr, Cu, Pb, and Zn) were performed using a UNICAM-929 atomic absorption spectrophotometer (AAS) in accordance with the AOAC 19th Edition 2012 BY ICP-OES technique, with a lower detection limit of 0.05 ppm. Heavy metal concentrations in sediments were measured at the Bangladesh Institute of Nuclear Agriculture's Soil Laboratory (BINA). The sediment samples were air dried, sieved using 230 mesh (600 mesh) stainless screens to remove bigger particles and stones, then digested with a 4:1 nitric acid (HNO₃) and perchloric acid mixture (HClO₄). The volume was increased to 10 ml with 0.1N HNO₃ and examined by AAS according to the APHA technique (1998). The importance of qualitative examination of samples in determining the quality cannot be overstated. For greater accuracy and precision, all reagents and chemicals utilized in this investigation were analytical grade (Merck, India). During the sample analysis, sample triplicates and repeated experiments according to the requirements were done for quality control. To ensure the quality of the data, each analytical step was accompanied by a quality assurance program. Every stage of the laboratory analysis process was meticulously documented. These records were preserved for data management and to identify missing steps and values, as well as to serve as recall points for repeat analyses.

The heavy metal pollution index (HPI) proposed by Prasad and Bose (2001) was used to investigate the pollution status of water in this study. This index is frequently used to assess the quality of water for drinking and irrigation reasons, as well as to evaluate the general state of water quality in respect to heavy metals (Nasrabadi, 2015; Nasrabadi et al., 2013). Prasad and Bose (2001) used unit weightage (W_i) as a number inversely proportional to the suggested standard (S_i) of the associated parameter while calculating the HPI. The HPI model is defined as follows:

$$HPI = \frac{\sum_{i=1}^n W_i Q_i}{\sum_{i=1}^n W_i} \quad (1)$$

Where, Q_i is the sub-index of the *i*th parameter, W_i is the unit weightage of the *i*th parameter and *n* is the number of parameters considered. The sub-index (Q_i) of the parameter is calculated by:

$$Q_i = \sum_{i=1}^n \frac{\{Mi(-)Ii\}}{(Si-Ii)} \times 100 \quad (2)$$

Where, M_i is the monitored value of heavy metal of *i*th parameter. The sign (–) indicates numerical difference of the two values, ignoring the algebraic sign. The weightage (W_i) was calculated as the inverse of MAC, with S_i representing the WHO drinking water standard in ppb and I_i representing the guidance value for the selected element in ppb, and MAC representing the highest allowable concentration/upper permissible concentration (Table 1).

Pollution load index (PLI) assesses shared contamination weight at different locations using different metals in soils and sediments, and provides an assessment of the overall toxicity grade of every sampling site (El-Gohary et al., 2012). The PLI was determined for all sampling sites using the following equation (Eq. 3) proposed by Tomlinson et al. (1980) as the *n*th root of the product of the contents multiplications:

$$PLI = \sqrt[n]{(CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n)} \quad (3)$$

A PLI value of 0 indicates perfection, while a value of 1 indicates only baseline levels of pollutants and above 1 denotes gradual degradation of site quality (Tomlinson et al., 1980). This empirical index offers a clear and comparative means of weighing the extent of heavy metal emissions, where $PLI > 1$ indicates that pollution occurs and $PLI < 1$ indicates that metal pollution does not occur (Varol, 2011).

The potential ecological risk (PER) of heavy metal pollution is measured using the potential ecological risk index (E_r^i) (Hakanson, 1980), which considers both the contamination factor (C_f^i) and the toxic response factor (T_f^i) (Roy et al., 2019). The following formulas (Eq. 4, 5, 6) can be used to measure the possible risk index:

$$E_r^i = T_f^i \times C_f^i R_i \quad (4)$$

$$C_f^i = \frac{C_m^i}{C_n^i} \quad (5)$$

$$PER = \sum_{i=1}^n E_r^i \quad (6)$$

In these equations, C_f^i is the accumulating coefficient of metal (i) and T_f^i is the toxic-response factor of metal (i). The toxic-response factors for Pb, Cu, Zn, Cd and Mn were 5, 5, 1, 30 and 1, respectively (Hakanson, 1980). Whereas, C_m^i is the concentration of heavy metals in sediments samples, and C_n^i is the pre-industrial history values of sediments. PER is the comprehensive potential ecological risk index, which is the sum of E_r^i . The biological community's sensitivity to the toxic material is reflected by it, and it shows the possible ecological danger posed by the overall pollution (Roy et al., 2019). Kabir et al. (2020b) identified four levels of heavy metal contamination in sediments ecological pollution for example $E_r^i < 40$ or $RI < 150$: low, $40 \leq E_r^i < 80$ or $150 \leq RI < 300$: moderate, $80 \leq E_r^i < 600$ or $300 \leq RI < 600$: considerable, and $160 \leq E_r^i < 320$ or $600 \leq RI$: very high ecological risk for the sediments. The geo-accumulation index (I_{geo}) suggested by Muller (1969) may be used to determine the degree of contamination from trace metals in sediments by means of the following equation (Nasrabadi et al., 2010):

$$I_{geo} = \log_2 \left(\frac{C_n}{1.5} \times B_n \right) \quad (7)$$

Where C_n represents the measured concentration of the inspected metal (n), and B_n represents the metal's geochemical background concentration (n). The factor 1.5 is used to account for possible lithological impact variations in background values (Kabir et al., 2020b; Fazeli et al., 2019).

The study used the risk assessment methods and parameters stated by Wongsasuluk et al. (2014). Siriwong (2006) recorded estimates of human exposure to the environment in terms of frequency, magnitude, and length as average daily dose (ADD), which can be calculated manually using following equation (Eq. 8) and the parameters in Table 1.

$$ADD = \frac{C * IR * ED * EF}{BW * AT} \quad (8)$$

The non-carcinogenic health risks were calculated using each metal's reference dose (Table 1) through the following equation (Eq. 9):

$$\text{Hazard Quotient (HQ)} = \frac{ADD}{RfD} \quad (9)$$

For the computation of the risk of mixtures, the individual hazard quotients (HQs) are summed to form HI (hazard index): $HI = \sum HQ$. HI or $HQ > 1$ indicates that non-carcinogenic effects on health are an unacceptable risk, while HI or $HQ < 1$ indicates that the risk is reasonable (Lim et al., 2008). Table 2 indicates the key exposure variables that were taken into account in the risk evaluation calculations.

Bio-concentration factors were analyzed based on metal concentration in water, fish and sediment. The BCF analysis was performed to determine the accumulated level of Pb, Cu, Zn, Cd and Mn in water, fish and sediments samples. The BCF of heavy metals in samples were calculated using Eq. 10 suggested by Potipat et al. (2015) as:

$$BCF = C_{\text{fish}}/C_{\text{ambient medium}} \quad (10)$$

Where C_{fish} is the heavy metals concentration in fish, and $C_{\text{ambient medium}}$ for fish is the heavy metals concentration in water and sediment.

The data was compiled and formatted appropriately before being subjected to statistical analysis. The data was presented and interpreted using the Statistical Package for Social Sciences (SPSS version 16.0). To analyze particular correlations among the factors studied, Pearson's correlation matrix was used.

RESULTS AND DISCUSSION

The mean concentrations of Pb, Cu, Zn, Cd and Mn in water were found 0.0053, 0.0225, 0.0059, 0.0034 and 0.188 mg/L, respectively in pre-monsoon; 0.0028, 0.0123, 0.0026, 0.002 and 0.1243 mg/L, respectively in monsoon; and 0.0042, 0.0219, 0.005, 0.0029 and 0.196 mg/L, respectively in post-monsoon season. The Pb, Cu, Zn, Cd and Hg concentrations in the Shitalakhya River water were found higher in all seasons (Kabir et al., 2020a) when compared with present study. The concentration heavy metals was higher in pre-monsoon followed by post-monsoon and monsoon, because of higher level of pollutants in the haor water due to lack of heavy rainfall for dilution of pollutants (Ahmad et al., 2010). Moreover, the mean concentrations were observed in decreasing order of $Mn > Cu > Cd > Zn > Pb$, whereas all the metal concentrations were found lower than the ECR (1997) standard (Table 3). The concentration of heavy metals were varied from stations to stations which might be due to river water flow, locations of industries, municipal and commercial drainage system and agricultural runoff (Hassan et al., 2015). Rehnuma et al. (2016) found that Pb, Cd, Cu, Mn and Zn concentrations in the Bangshi River ranged from 0.005 to 0.016, 0.0005 to 0.0009, 0.04 to 0.06, 0.78 to 1.39 and 1.30 to 1.94 mg/L, respectively during the wet season and 0.011 to 0.021, 0.0011 to 0.0019, 0.07 to 0.13, 1.32 to 2.05 and 2.06 to 3.05 mg/L, respectively during the dry season, which is more or less similar to the present study. The level of Pb, Cd and Zn of the Turag River were found 0.0021, 0.0136 and 0.0191 mg/L, respectively (Mokaddes et al., 2013), which was little bit higher than the present study, except Pb. Ahmad et al. (2010) found that the concentrations of Pb (0.0654 mg/L), Cu (0.1630 mg/L), and Cd (0.0093 mg/L) in Buriganga River were higher than the present study. Ahmed et al. (2009) reported that the concentration of Pb (50.05 mg/L), Cd (6.49 mg/L) and Cu (154.69 mg/L)

from Shitalakhya River which were several times higher than the present study. Likewise, the concentrations of Cu and Zn were found several times higher in Mokesh beel than the present study (Barmon et al., 2018). Khan et al. (1998) reported that heavy metals concentration in Dhaleshwari River were Pb (0.221 mg/L) and Cd (0.054 mg/L). In relation to the toxicity reference value (TRV) proposed by USEPA (1999) for fresh water, the concentration of Pb, Zn and Cd were exceeded the TRV in great extent whereas Cu found lower than the TRV value (Table 3). The HPI for all heavy metals was calculated for each sampling site, and it was observed that the HPI for all examined metals in three seasons ranged from 28.61 to 206.51, with the mean HPI of 109.89 indicating the critical pollution index value for drinking water (HPI<100) (Figure 2). The HPI values of a river system in southern Caspian Sea basin indicates relatively better (Nasrabadi, 2015). According to HPI study, the Shitalakhya river water was tainted with heavy metals, however the contamination level was not significant (Kabir et al., 2020a). In Harike wetland, the HPI for all metals was higher above the proposed critical limit of 100 for drinking water (Brraich and Jangu, 2015). Pearson correlation coefficients (r) was determined to reveal the relationships among the heavy metals in different seasons (Table 4), which may provide notable information on the sources and pathways of these metals in the study area. The coefficient determined for all the seasons showed significant positive correlation between the heavy metals which clearly supported the fact that these heavy metals Cr-Cu in pre-monsoon and Cu-Cd in post-monsoon seasons were strongly correlated which demonstrated the similar pollution sources and pathways into the aquatic environment of the study area (Armah et al., 2010; Hassan et al., 2015).

The mean concentrations of Pb, Cu, Zn, Cd and Mn in fish were found 2.57, 25.91, 61.49, 1.37 and 4.44 mg/kg–dry wt., respectively in pre-monsoon; 1.61, 15.04, 34.75, 0.76 and 2.61 mg/kg–dry wt., respectively in monsoon; and 2.37, 33.80, 69.57, 1.61 and 5.75 mg/kg–dry wt. basis, respectively in post-monsoon season (Table 5). The mean concentrations were observed in decreasing order of Zn>Cu>Mn>Pb>Cd, whereas all the metals concentrations were found higher than the permissible level set by FAO (1984) standard, except Cu (Table 5). The fishes were highly contaminated with heavy metals where Zn concentration was the highest and Cd concentration was the lowest while Pb, Cu and Mn were moderate in concentration when compared to other metals of the study. The highest concentration of Cu, Zn, Cd and Mn were found in the post-monsoon season whereas the highest Pb was found in pre-monsoon season, and all the concentrations indicated that there were no significant variations among seasons (Table 5). It could be due to dry season when the heavy metals concentrations in water were much higher than wet season. As a result, the fish absorbed more heavy metals throughout the dry season, and their concentrations increased at the start of the wet season. In general, heavy metal bioaccumulation is influenced by the concentration of exposed metal, the length of exposure, the mode of metal uptake, environmental circumstances, and intrinsic characteristics such as fish age, eating patterns, and so on. (Timbrell, 2002). Metals can be taken up by organisms directly or through food particles in water, and then these metals are bound into various areas of the organism's body, potentially causing harm (Gupta et al., 2009). Ahmad et al. (2010) studied the heavy metal concentration in fish from the Dhaleshwari River and found seasonal variation of Cd (0.52 to 0.8 mg/kg), Pb (7.03 to 12.18 mg/kg) and Cu (7.55 to 11.50 mg/kg), which are almost similar to the present study. Almost a similar study was conducted by Rehnuma et al. (2016) in fishes of the Bangshi River, found Pb, Cd, Cu, Mn and Zn were 0.21, 0.02, 0.65, 3.45 and 5.81 mg/kg, respectively during the wet season, and 0.18, 0.023, 0.77, 2.80 and 5.67 mg/kg, respectively during the dry season. The concentration of Zn in fish (*Liza parse*) was always higher than other metals in the Passur river (Islam et al., 2017b). Moreover, only Zn was found 25.42, 18.30, and 29.34 mg/kg

during the pre-monsoon, monsoon and post-monsoon season, respectively from fishes of the Shitalakhya River while the concentrations of Pb, Cu, and Cr in fishes were found less than the lower detection limit (≤ 0.05) (Kabir et al., 2020a). Pearson correlation coefficients (r) revealed the relationships among the heavy metals in different seasons (Table 6), which may provide notable information about the sources and pathways of heavy metals. The coefficient determined for all the seasons showed significant positive correlation between the heavy metals which supported the fact that the metals Zn-Pb in pre-monsoon and Mn-Pb in monsoon seasons were strongly correlated which indicated pollution sources and pathways in the aquatic environment (Armah et al., 2010; Hassan et al., 2015).

Human health risk assessment of heavy metals connected to the exposure duration and consumption of fish (depends on the quantity and individual body weight). Average daily exposure doses (Table 7) and HQ (Table 8) for heavy metals in different seasons in fish species was determined using the concentration of Pb, Cu, Zn, Cd and Mn in the fish species, respectively. The study showed that values of HQ for the heavy metals were higher than 1 for child and adult which indicate the adverse human health effect. Comparatively the higher value of HQ for Pb and Cu were found for child whereas Cd, Zn and Mn were found for adult. The highest HQ values of all studied heavy metals were found for child and adult in post-monsoon season except Mn for adult. The highest HI value for Cd was found for child and adult in pre-monsoon, monsoon and post-monsoon season, especially in post-monsoon season (Figure 3). However, the HI values of all studied heavy metals were above the safe level of consumption (>1) for child and adult, indicating the potential non-carcinogenic adverse health effect for residents. Similar study was found that Cd, Cu, Mn, Pb and Zn HQ values above 1 suggesting that the significant non-carcinogenic adverse health effect for human beings (Asare et al., 2018). However, the measured HI for studied heavy metal that shows the fish consumption might be pose significant adverse health risk to humans.

The concentrations of Pb, Cu, Zn, Cd and Mn in sediments were 12.45, 6.07, 37.48, 0.51 and 144.54 mg/kg, respectively in pre-monsoon; 6.19, 4.58, 25.43, 0.32 and 121.48 mg/kg, respectively in monsoon; and 11.63, 5.55, 34.63, 0.46 and 116.80 mg/kg, respectively in post-monsoon season. The mean concentrations of heavy metals Pb, Cu, Zn, Cd and Mn were observed 10.09, 5.40, 32.51, 0.43 and 127.61 mg/kg, respectively (Table 9). These metals were decreased in order of $Zn > Cu > Mn > Pb > Cd$, whereas all the metals were found lower than the EPA (1977) standard, except Cu (Table 10). However, heavy metal concentrations in Shitalakhya River sediments were several times higher than the present where Pb (64.22 mg/kg), Cd (3.23 mg/kg) and Cu (44.05 mg/kg) (Ahmed et al., 2009). The variation of heavy metal from locations to locations may be correlated with the flow of the rivers and location of industries and their waste disposal system (Alam et al., 2004). A moderately polluted sediments were observed for Pb 46.91 mg/kg from Shitalakhya River, Narayanganj (Monirul et al., 2014) that exceed the standard level, this might be presence of large scale of industries in study area which discharge a huge amount of wastewater into the river (Table 9). In the Meghna river sediments, Pb, Cu, Zn, and Fe contents were also varied seasonally and spatially from 6.34 to 20.46, 1.39 to 28.06, 81.30 to 98.90 and 2274.28 to 34.62.10 mg/kg, respectively, and the Fe content in all sediment samples were above the EPA (1977) standard, whereas the content of Cu and Zn fall in the criteria of moderately polluted range (Akter et al., 2019). The highly polluted concentration was found for Cu (163 mg/kg) and Cd (3.31 mg/kg) in Buriganga River (Ahmad et al., 2010). Heavy metals status in sediment from southern part of Bangladesh were observed moderately to highly polluted for Pb (44.706 mg/kg), Cd (3.84 mg/kg), Cu (61.1 mg/kg), Zn (188.3 mg/kg) and Mn (23.96 mg/kg) (Tareq et al., 2017). Pearson correlation coefficients (r) revealed that the relationships among the heavy metals in

different seasons (Table 11), which provided the sources and pathways of these metals in the study area. The coefficient determined for all the seasons showed significant positive correlation which supported the fact that these heavy metals Mn-Cd in monsoon, Cd-Zn in post-monsoon and Mn-Zn in post-monsoon seasons were strongly correlated, indicated similar pollution sources and pathways in the aquatic environment (Armah et al., 2010; Hassan et al., 2015).

The PLI values for the heavy metals analyzed ranged from 0.05 to 0.56, with a mean value of 0.273 indicating that the haor sediments were not contaminated. The highest PLI (0.56) was found at St-3 during the pre-monsoon season, suggesting that the studied heavy metals had adequately polluted the St-3 sediments. In this study, the PLI values of all sampling stations in pre-monsoon monsoon, and post-monsoon seasons were < 1 which indicated the lower level of pollution (Table 12). The PLI value is significant for understanding the quality of a component of their environment and represents the trend overtime and area (Kabir et al., 2020b). The PLI of the sediments at all seasons followed the order of pre-monsoon $>$ post-monsoon $>$ monsoon, according to the investigation. The PER of heavy metals in haor sediments was found to be in the order of Cd (64.00) $>$ Pb (2.52) $>$ Cu (0.035) $>$ Zn (0.25) $>$ Mn (0.25). (0.17). The Cd (87.00 to 42.00) was the most abundant and important metal in all stations over the three seasons, with significant risk factors for the water body, while the majority of the metals, such as Cu, Pb, Mn, and Zn, were found to be in low in terms of risk factors. The PER ranged from 42.38 to 90.41, with the highest value at St-1 during the post-monsoon season and the lowest value at St-3 during the monsoon season. The haor sediments had a mean PER of 66.97, suggesting that the haor presents low potential ecological risks from heavy metal pollution (Table 13). The highest PER value was found in the post-monsoon season, possibly due to a lack of heavy rainfall for pollutant mixing, followed by the pre-monsoon and monsoon seasons (Kabir et al., 2020b). The mean Igeo values of the studied five metals followed the descending order of Cd (0.85) $>$ Zn (0.13) $>$ Pb (0.12) $>$ Cu (0.08) $>$ Mn (0.05). The Cd (1.16) had the greatest Igeo value in the sediment at St-5 and St-1 during the post-monsoon and post-monsoon seasons, respectively, whereas Mn (0.04) had the lowest at St-4 and St-5 during the monsoon season and at St-1 and St-5 during the post-monsoon season (Figure 4). Except for Cd, which showed moderately contaminated enrichment throughout the pre-monsoon and post-monsoon seasons, the Igeo values showed that the sediments of Haor were unpolluted to moderately polluted for all metals during the pre-monsoon, monsoon, and post-monsoon seasons (Figure 4). The Igeo values for the studied heavy metals in the sediment of the Shitalakhya river revealed that Igeo was higher in the post-monsoon season followed by pre-monsoon and monsoon season (Kabir et al., 2020b), which is similar to the present study.

Bio-concentration is the accumulation of the contaminant by aquatic organisms through non-dietary uptake routes from the soluble phase (Chaudhuri et al., 2007). In the present study, $BCF_{f/w}$ refers to the ratio of heavy metals concentration in fish and water, while the $BCF_{f/s}$ refers to the ratio of heavy metals concentration in fish and sediments. The results of BCF of heavy metals in fish and water, and fish and sediment are given in Table 13. Fish intake of heavy metals is directly tied to biological requirements, metabolisms, and other elements such as salinity, amount of water pollution, diet, and sediment (Ashraf et al., 2011). The rate of heavy metal buildup in fish is determined by chemical compound characteristics, concentration, and fish species (Sarong et al., 2015). If the BCF value was greater than 1.0, it meant that the metals had been bio-accumulated and bio-magnified to a high degree (Ba and Olanipekun, 2007). From this investigation, fish and water obtained low to very high category bio-concentration factor whereas, fish and sediment obtained low category bio-concentration

factor (Table 13). The heavy metal Zn observed highest loading followed by Cu, Pb and Cd in fish and water. The heavy metal Zn and Cu indicated the very high concentration factor while Pb and Cd indicated high concentration factor. In addition, the heavy metal such as Mn indicated the low bio-concentration factor in fish and water. In case of fish and sediments, Cu showed the highest BCF followed by Cd, Zn, Pb and Mn. Cu has a greater BCF value than Pb and Cd, despite the fact that Cu is a necessary component of several enzymes in organisms (Yap et al., 2016) and serves as a micronutrient for cellular metabolism (Ismarti et al., 2017). Similar studies were reported by Olusola and Festus (2015); Asaolu et al. (1997); Adefemi et al. (2004) and Valipour et al. (2012, 2013). According to Yap et al. (2016), high Cu intake from seafood can cause health concerns such as liver and kidney damage, although it is not carcinogenic to people or animals.

CONCLUSION

The highest concentrations of heavy metals were found in pre-monsoon season followed by post-monsoon and monsoon season might be due to the variation of volume and flow of water over that seasons. Concentrations of heavy metals in water and sediments were lower than the recommended levels but fishes were contaminated by Pb, Zn, Cd and Mn. The haor of Kishoreganj is contaminated by heavy metals and might create an adverse effect on aquatic ecosystem and human health problem in forthcoming days. In order to conserve the aquatic food system, the haor environments are needed to monitor and manage regularly by concerned authority. The result of this study will be provided baseline information to the researchers and decision makers about the heavy metal concentrations in water, fish and sediments in the haor waters of Kishoreganj region.

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