Heavy metals in sediment and their accumulation in commonly consumed fish species in Bangladesh

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ABSTRACT
Six heavy metals (chromium [Cr], nickel [Ni], copper [Cu], arsenic [As], cadmium [Cd], and lead [Pb]) were measured in sediments and soft tissues of eleven commonly consumed fish species collected from an urban river in the northern part of Bangladesh. The abundance of heavy metals in sediments varied in the decreasing order of Cr > Ni > Cu > Pb > As > Cd. The ranges of mean metal concentrations in fish species, in mg/kg wet weight (ww), were as follows: Cr, 0.11–0.46; Ni, 0.77–2.6; Cu, 0.57–2.1; As, 0.43–1.7; Cd, 0.020–0.23; and Pb, 0.15–1.1. Target hazard quotients (THQs) and target carcinogenic risk (TR) showed the intake of As and Pb through fish consumption were higher than the recommended values, indicating the consumption of these fish species is associated with noncarcinogenic and carcinogenic health risks.

During the last few decades, rapid urbanization and industrial development have provoked some serious concerns for the aquatic environment. About 80% of the world population is facing an increasing threat regarding water security1,2 because sediments in most of the urban rivers have been contaminated by heavy metals.3–5 It has been well documented that surface sediment acts as a sink of various contaminants and poses a risk to water quality through biogeochemical exchanges with the overlying water body.6 Chromium (Cr), nickel (Ni), copper (Cu), arsenic (As), cadmium (Cd), and lead (Pb) are some of the most common heavy metal pollutants in the environment.7–9 However, these metals from natural and anthropogenic sources10,11 may enter into the aquatic environment and pose serious threats due to their toxicity,12 persistence, and bioaccumulation.13,14 Usually, in unaffected environments, the concentration of most of the heavy metals is low and mostly derived from the mineralogy and weathering.15 Sources such as industrial effluents, agricultural runoffs, transport, burning of fossil fuels, animal and human excretions, geologic weathering, and domestic waste contribute to the accumulation of heavy metals in the water bodies.4,5,16–18

Heavy metal pollution in the environment has become a wide concern owing to the ever-increasing contamination of water, soil, and food in many regions of the world, particularly in some developing countries such as Bangladesh.9,17,19–21 Heavy metals are not only a threat to the public water supply; they also pose risks to human health through consumption of aquatic products, especially fish.17,22,23 Fish that generally accumulate contaminants from aquatic environments have been largely used in food safety studies.24,25 Therefore, studies on bioaccumulation of heavy metals in fish species are important in determining the tolerance limits in fish species, the effects on fish, and biomagnification through the food chain.26 Fish are an important part of the human diet and a good indicator of environmental contamination by a number of substances, including heavy metals. However, fish have been considered as the top of the aquatic food chain;25,27,28 therefore, they normally can accumulate heavy metals from food, water, and sediments.27,28 The accumulated toxic metals in fish can counteract their beneficial effects; several adverse effects of heavy metals to human health have been known for a long time.24,29 These may include serious threats such as renal failure, liver damage, cardiovascular diseases, and even death.5,30 Little is known about the bioavailability to aquatic organisms of sediment-associated contaminants.31 Therefore, many international monitoring
programs have been established to assess the quality of fish (in terms of metal concentration) for human consumption and to monitor the health of the aquatic ecosystem.32

Bangladesh is one of the largest delta regions in the world, formed by the Ganges, Brahmaputra, and Meghna rivers and randomly spreading over 5 countries: Bhutan, Nepal, China, India, and Bangladesh.16 In Bangladesh, huge amounts of untreated industrial waste are discharged daily into the open water bodies and adjacent lands. In addition, a considerable amount of heavy metal–enriched suspended solids come down from neighboring countries such as India through the Teesta and the Brahmaputra rivers.16 Bogra District, known as the northern capital of Bangladesh, is situated on the bank of the river Korotoa, which is connected to the rivers Teesta and Brahmaputra. The area of the Korotoa River that is being studied is the only active section with intensive district traffic and supplies water for the people living adjacent to this river. Industrial activities and irrigation also depend on the Korotoa River. The river sediments are traditionally dredged and used as an amending material for agricultural soil. In addition, the river aquatic products such as fish serve as a key source of food for the local inhabitants. Overexploitation, mismanagement, and discharge of improperly treated industrial effluents into the Korotoa River create a great challenge for the ecosystem balance.16 Thus, the study river recently has become a public concern due to its extreme pollution. To date, no scientific research regarding heavy metal issues on the study river has been conducted. Therefore, the objectives of this study are to evaluate the levels of heavy metals in surface sediment, to observe the metal accumulation in eleven fish species collected from the Korotoa River, and to assess the health risk due to fish consumption in Bangladesh.

Methods

Description of the study area

This study focused on the Korotoa River located at the northern part of Bangladesh. The study river originated from the Himalayas, the mother of numerous rivers. Originating from the northern frontier of Bhutan, the Korotoa enters Bangladesh territory through the Darjeeling and Jalpaiguri districts of West Bengal, India, and forms the boundary between Dinajpur and Rangpur districts in Bangladesh. For the present study, we selected sites of the Korotoa River that flow through the Bogra District urbanized area with an area of about 71.56 km². The total population of this district is about 350,397, and it is situated between 24° 84’ 91.82” N and 89° 37’ 29.57” E.16,33 Thousands of villages, towns, and commercial places such as Shibganj, Mohasthangarh, Bogra, and Sherpur have been built on both sides of the Korotoa River. Mohasthangarh, the capital of ancient Pundranagar, is still there beside the Korotoa as a witness of history in Bangladesh.

Sample collection and preparation

The sampling was conducted in August and September of 2013. A total of 30 composite sediment samples were collected from 10 different locations of the Korotoa River situated at the northern part of Bangladesh (Figure 1). At each point, 3 composite sediment samples were collected using standard protocol.34 The riverbed sediment samples were taken at a depth of 0–5 cm using a portable Ekman grab sampler. Three composite samples of mass approximately 200 g were collected at each station. The upper 2 cm of each sample was taken from the center of the catcher with an acid-washed plastic spatula to avoid any contamination from the metallic parts of the sampler. We collected about 110 samples of eleven different fish species: Channa punctata, Awaous grammepomus, Anabas testudineus, Heteropneus tesfossili, Neotropius atherinoides, Colisa fasciata, Channa striata, Notopterus notopterus, Batasios batasio, Coriscos borna, and Puntius chola. Fish species were collected using nylon net with the help of fishermen at almost the same locations where the sediments were collected. After collection, fish samples were carefully washed immediately with distilled water, and the edible parts of the fish (muscle tissue) were cut into small pieces and oven dried at 70°–80°C to attain constant weight. The moisture content of the fish was calculated by recording the difference between fresh and dry weights. The dried fish samples were crumbled with a porcelain mortar and pestle and sieved through a 2-mm nylon sieve and stored in airtight, clean zip-lock bags in freezer condition until chemical analysis was performed.

Analytical procedure for heavy metals

All reagents used were Merck analytical grade (AR). Deionized water was used for solution preparation. For metal analysis, about 0.3 g of sediment and 0.5 g of the dried fish samples were digested with 15 mL of concentrated HNO₃, H₂SO₄, and HClO₄ in 5:1:1 ratio at 80°C until a transparent solution was obtained.35 The digested samples of sediment and fish species were filtered through Whatman no. 42 filter paper, and the filtrates were diluted to 50 mL for sediment and 25 mL for fish with deionized water. All samples were stored at ambient temperature before analysis. For heavy metals, samples were analyzed using an atomic absorption spectrometer (Perkin Elmer Analyst 300). Blank samples were analyzed after 8 samples. Concentrations were calculated on
a dry weight (dw) basis for sediment and wet weight basis for fish samples. All analyses were replicated 3 times. The precision and analytical accuracy were checked by analysis of standard reference material NMIJ CRM 7303 (lake sediment) and DORM-2 (dogfish muscle) from the National Research Council, Canada. The measured mean and standard deviation of elemental values for reference materials are reported in Table S1. Comparison is made with the certified values, which in both cases confirmed that the sample preparation and operating condition of the instrument provided good levels of accuracy and precision.

**Analysis of physicochemical properties of sediment**

The pH of sediments was measured in a 1:2.5 sediment-to-water ratio. The suspension was allowed to stand overnight prior to pH determination. The pH was measured using a pH meter with the calibration of pH 4.0, pH 7.0, and pH 9.0 standards. For electrical conductivity (EC) determination, 5.0 g of sediment was taken in 50 mL polypropylene tubes. Then, 30 mL of distilled water was added to the tube and was shaken for 5 minutes. After that, EC was measured using a portable EC meter (Horiba D-52). Percent nitrogen (%N) and organic carbon (%C) of sediment were measured using an elemental analyzer (vario EL III, Elementar, Germany). For total nitrogen (TN) and total organic carbon (TOC) determination, sediment samples were weighed in tin or silver vessels and loaded in the integrated carousel. In a fully automatic process, the transfer of the sample through the ball valve into the combustion tube was performed. Each sample was individually flushed with carrier gas to remove atmospheric nitrogen, resulting in a zero blank sampling process. The catalytic combustion was carried out at a permanent temperature of up to 1200°C. The element concentration from the detector signal and the sample weight on the basis of stored calibration curves were measured.

**Metal bioaccumulation in fish species**

Metal concentrations in fish species and sediments from the studied river were used for calculating biota-sediment accumulation factor (BSAF). The BSAF is an index of the ability of fish species to accumulate a particular
metal with respect to its concentration in sediment. It was calculated by the following equation.  

\[ BSAF = \frac{C_{\text{fish}}}{C_{\text{sediment}}} \]  

(1)

where \( C_{\text{fish}} \) is the metal concentration in fish (mg/kg dw) and \( C_{\text{sediment}} \) is the metal concentration in sediment (mg/kg dw).

**Noncarcinogenic risk**

The noncarcinogenic risk was estimated in accordance with that provided in the USEPA Region III Risk-based Concentration Table.\(^{37}\) The noncarcinogenic risk for each metal through the consumption of fish was assessed by the target hazard quotient (THQ)\(^{38}\): “the ratio of a single substance exposure level over a specified time period (eg, subchronic) to a reference dose (RfD) for that contaminant derived from a similar exposure period.” THQ assumes a level of exposure (ie, RfD) below which it is unlikely for the populations to experience adverse health effects. If the exposure level exceeds this threshold (ie, if THQ = E/RfD exceeds unity), there may be concern for potential noncarcinogenic risks.

\[ THQ = \frac{EFr \times ED \times FIR \times C}{RfD \times BW \times AT} \times 10^{-3} \]  

(2)

\[ \text{TotalTHQ(TTHQ)} = \text{THQ}_{\text{metal1}} + \text{THQ}_{\text{metal2}} + \ldots + \text{THQ}_{\text{metaln}} \]  

(3)

where \( EFr \) is the exposure frequency (365 days/year); \( ED \) is the exposure duration (70 years), equivalent to the average human lifespan\(^{39}\); \( FIR \) is the fish ingestion rate (g/person/day); \( C \) is the metal concentration in samples (mg/kg, fresh weight [fw]); \( RfD \) is the oral reference dose (mg/kg/day); \( BW \) is the average body weight (adult, 60 kg); \( AT \) is the averaging time for noncarcinogens (365 days/year \times number of exposure years, assuming 70 years). The daily consumption rate of fish for adult residents was 45.67 g on a fresh weight basis.\(^{40}\) The oral reference doses were based on 1.5, 0.02, 0.04, 0.0003, 0.003, and 0.004 mg/kg/day for Cr, Ni, Cu, As, Cd, and Pb, respectively.\(^{37,41}\) If the THQ is less than 1, the exposed population is unlikely to experience obvious adverse effects. If the THQ is equal to or greater than 1, there is a potential health risk,\(^{42}\) and interventions and protective measures should be taken.

**Carcinogenic risk**

Carcinogenic risks of As and Pb were estimated as the incremental probability that an individual will develop cancer over a lifetime as a result of exposure to that potential carcinogen (ie, incremental or excess individual lifetime cancer risk).\(^{38}\) The equation used for estimating the target carcinogenic risk\(^{38}\) is as follows:

\[ TR = \frac{ED \times FIR \times C \times CSFo}{BW \times AT} \times 10^{-3} \]  

(4)

where CSFo is the oral carcinogenic slope factor from the Integrated Risk Information System\(^{37}\) database. The slope factors were 1.5 for As and 8.5 \( \times 10^{-3} \) (mg/kg/day)\(^{-1} \) for Pb.

**Statistical analysis**

The data were statistically analyzed using the statistical package SPSS 16.0 (SPSS, USA). The means and standard deviations of the metal concentrations in sediment and fish species were calculated. Multivariate post hoc Tukey test was used to examine the statistical significance of the differences among mean concentrations of heavy metals among fish species. A multivariate method in terms of principal component analysis (PCA) was used to obtain the detailed information of the data set and gain insight into the distribution of heavy metals by detecting similarities or differences in samples.

**Results**

**Physicochemical properties and metals in sediment**

Physicochemical properties of sediments are presented in Table 1. The pH of the sediments did not vary much and was slightly acidic for all the sites except S10, which was slightly alkaline. The lower pH at most of the stations of the studied river might be due to discharge of the acidic effluent from nearby industries. The S10 site showed considerably higher pH (8.31), which might be due to the deposition of huge amounts of sediment at this site. This deposited sediment may contain much calcium carbonate and magnesium carbonate, which are calcareous. Hydrolysis of these calcium carbonate and magnesium carbonate releases OH\(^-\) ion, which contributes to alkalinity in sediment.\(^{43}\) Due to the variations in topography, hydrology, and geology within catchment areas, as well as differences in precipitation, local climate, and anthropogenic influence, the water chemistry such as pH, alkalinity, and concentration of heavy metals may differ considerably between streams even within small distances.\(^{44}\) The variation trends of pH at different sampling sites may be due
to the higher concentrations of colloidal and/or particulate matter during high river discharges.45 The composition of the organic carbon in the riverine sediments is varying due to its origin in the aquatic environment. Phytoplankton and zooplankton are the most abundant sources of the organic material in the sediments.46 Total nitrogen content was in the range of 0.112%–0.252%; organic carbon was in the range of 0.74%–2.45%; and metal retention was found to be high in the locations with high organic carbon (sites S6, S9, and S10). The highest percentage of organic carbon might be attributed to the high amount of drainage water. The high rate of organic growth together with the organic detritus introduced by the drainage system can be considered as the main source of organic carbon.47 The lower percentage of TOC are due to the structure of the sediments in the investigated area, which was mainly sand that has a low affinity to absorb contaminants. According to the US soil texture classification, the textural analysis revealed that the sediments in the study region were dominated by sand and sandy loam (Table 1).

The concentrations of heavy metals in sediment are presented in Table 2. The distributions of heavy metals in the sediments were not uniform among the sampling sites of the river. The variations in the concentration might be due to differences in the source of the heavy metals and prevailing physicochemical conditions and complex reactions such as adsorption, flocculation, and redox condition taking place in the sediments.47,48 The concentrations of heavy metals at sites S6–S10 were much higher than at other sites because these sites located at the downstream of the river were influenced by the extensive discharging of urban waste.5,16 Elevated concentrations of heavy metals in surface sediments found at the downstream sites of the Korotoa River close to the urban area of Bogra District indicated that urbanization drove metal contamination in surface sediment.49,50 The urban activities (industrial discharges, municipal waste water, household garbage, and urban runoff) of Bogra District urban area are the main reasons for higher metal input at sites S6–S10. Higher contaminations of heavy metal found in Yuandang Lagoon due to the municipal sewage discharge or other unknown pollution sources from Xiamen City, China,51 are in line with our findings. The average concentrations of heavy metals in sediments were in the following decreasing order: Cr > Ni > Cu > Pb > As > Cd.

Among the sites in the current study, the average concentration of Cr was 118 mg/kg; the highest Cr was observed in sediment collected from site S10 (179 mg/kg; Table 2). The concentration of Cr in sediment was in line with the other studies and slightly higher than the average shale value (ASV), toxicity reference value

### Table 1. Physicochemical properties of sediment collected from korotoa river of Bogra District, Bangladesh.

| Site | pH  | EC (mS/cm) | % N  | % OC | C/N ratio | % Sand | % Silt | % Clay | Textural class |
|------|-----|------------|------|------|-----------|--------|--------|--------|---------------|
| S1   | 5.12| 25.5       | 0.181| 1.18 | 6.52      | 88     | 7.0    | 5.0    | Sand          |
| S2   | 5.91| 29.8       | 0.177| 1.79 | 10.11     | 77     | 15     | 8.0    | Sand          |
| S3   | 6.62| 23.8       | 0.155| 1.13 | 7.29      | 69     | 18     | 13     | Sandy loam    |
| S4   | 6.93| 42.8       | 0.172| 1.21 | 7.03      | 85     | 9.0    | 6.0    | Sandy loam    |
| S5   | 6.13| 27.5       | 0.114| 0.74 | 6.49      | 66     | 21     | 13     | Sandy loam    |
| S6   | 6.17| 42.5       | 0.216| 2.01 | 9.31      | 72     | 19     | 9.0    | Sandy loam    |
| S7   | 5.43| 19.7       | 0.113| 1.84 | 16.28     | 58     | 15     | 27     | Sandy clayey loam |
| S8   | 6.88| 26.7       | 0.112| 0.89 | 7.95      | 81     | 15     | 4.0    | Loamy sand    |
| S9   | 7.72| 33.2       | 0.155| 1.69 | 10.90     | 66     | 22     | 12     | Sandy loam    |
| S10  | 8.31| 22.9       | 0.252| 2.45 | 9.72      | 72     | 17     | 57     | Clay loam     |

**Note.** EC = Electrical conductivity; % N = Percent nitrogen; % OC = Percent organic carbon.

### Table 2. Heavy metal concentrations (mg/kg dw) in sediment of Korotoa River, Bangladesh (n = 3).

| Site | Cr Concentration | Cr SD | Ni Concentration | Ni SD | Cu Concentration | Cu SD | As Concentration | As SD | Cd Concentration | Cd SD | Pb Concentration | Pb SD |
|------|------------------|-------|------------------|-------|------------------|-------|------------------|-------|------------------|-------|------------------|-------|
| S1   | 86               | 9.4   | 57               | 6.5   | 49               | 14    | 5.4              | 1.7   | 0.79             | 0.20  | 50               | 11    |
| S2   | 58               | 9.7   | 45               | 11    | 54               | 11    | 18               | 6.6   | 0.53             | 0.13  | 46               | 10    |
| S3   | 57               | 5.8   | 46               | 18    | 52               | 13    | 6.9              | 1.4   | 1.1              | 0.32  | 44               | 8.6   |
| S4   | 85               | 8.2   | 135              | 23    | 75               | 17    | 15               | 3.7   | 0.77             | 0.15  | 62               | 11    |
| S5   | 93               | 7.8   | 99               | 9.4   | 89               | 17    | 17               | 4.0   | 1.1              | 0.21  | 49               | 9.4   |
| S6   | 177              | 10    | 126              | 22    | 105              | 14    | 52               | 8.7   | 2.6              | 0.59  | 79               | 9.4   |
| S7   | 183              | 27    | 163              | 29    | 117              | 23    | 51               | 9.1   | 1.9              | 0.58  | 58               | 13    |
| S8   | 149              | 12    | 94               | 16    | 67               | 17    | 39               | 7.4   | 1.4              | 0.48  | 75               | 16    |
| S9   | 115              | 12    | 112              | 14    | 92               | 14    | 27               | 4.9   | 1.6              | 0.24  | 81               | 9.0   |
| S10  | 179              | 20    | 155              | 13    | 118              | 21    | 38               | 10    | 2.8              | 0.95  | 83               | 13    |
| Average | 118            | 50    | 103              | 43    | 82               | 26    | 27               | 17    | 1.5              | 0.77  | 63               | 16    |

**Note.** Cr = chromium; Ni = nickel; Cu = copper; As = arsenic; Cd = cadmium; Pb = lead; SD = standard deviation.
| River (location)                  | Cr     | Range       | Ni       | Range       | Cu     | Range       | As       | Range       | Cd     | Range       | Pb      | Range       | Reference |
|----------------------------------|--------|-------------|----------|-------------|--------|-------------|----------|-------------|--------|-------------|--------|-------------|-----------|
| Korotoa (Bangladesh)             | 118    | 57–183      | 103      | 45–163      | 82     | 49–118      | 27       | 5.4–52      | 1.5    | 0.53–2.8    | 63     | 44–83       | This study |
| Paara River (Bangladesh)         | 45     | 17–93       | 34       | 13–63       | 30     | 10–65       | 12       | 2.6–29      | 0.72   | 0.15–1.6    | 25     | 9.1–38      | Islam et al. (2014b) |
| Buriganga River (Bangladesh)     | 177.53 | 200.45      | 27.85    | NA          | 3.33   | 69.75       | 19       | 19          | 0.61   | 59.99       | 14      | 4.3–84      | Ahmad et al. (2010) |
| River around Dhaka (Bangladesh)  | 695    | 112–2,471   | 355      | 139–606     | 191    | 65–405      | 35       | 12–58       | 17     | 8.5–29      | 356    | 45–1,846    | Islam et al. (2014a) |
| Jamuna River (Bangladesh)        | 110    | 33          | 28       | NA          | NA     | NA          | NA       | NA          | 19     | NA          | 4       | 19          | Datta and Subramanian (1998) |
| Bangshi River (Bangladesh)       | 98.10  | 25.67       | 31.01    | 1.93        | 0.61   | 59.99       | 14       | 4.3–84      | 4       | 19          | 4       | 19          | Rahman et al. (2014) |
| River Ganges (India)             | 1.8–6.4| NA          | 0.98–4.4 | NA          | 0.14–1.4 | 4.3–84      | 4       | 19          | 14     | 4.3–84      | 4       | 19          | Gupta et al. (2009) |
| Gomti River (India)              | 8.15   | 15.7        | 5        | NA          | 2.42   | 40.33       | 4       | 19          | 14     | 4.3–84      | 4       | 19          | Singh et al. (2005) |
| Yellow River (China)             | 41–128 | NA          | 30–102   | 14–48       | NA     | 26–78       | 4       | 19          | 14     | 4.3–84      | 4       | 19          | Liu et al. (2009) |
| Gediz River (Turkey)             | 170–220| 101–129     | 108–152  | NA          | NA     | 105–140     | 4       | 19          | 14     | 4.3–84      | 4       | 19          | Akcay et al. (2003) |
| Okumeshi River (Nigeria)         | 0.07   | 1.21        | NA       | NA          | 1.32   | 0.45        | 4       | 19          | 1        | 0.45        | 4       | 19          | Raphael et al. (2011) |
| Shur River (Iran)                | NA     | NA          | 9,174    | NA          | 6.85   | 162         | 4       | 19          | 1        | 0.45        | 4       | 19          | Karbassi et al. (2008) |
| ASV                             | 90     | 68          | 45       | 13          | 0.3    | 20          | 4       | 19          | 1        | 0.45        | 4       | 19          | USEPA (1999) |
| TRV                             | 26     | 16          | 16       | 6           | 0.6    | 31          | 4       | 19          | 1        | 0.45        | 4       | 19          | USEPA (1999) |
| LEL                             | 26     | 16          | 16       | 6           | 0.6    | 31          | 4       | 19          | 4       | 19          | 4       | 19          | USEPA (1999) |
| SEL                             | 110    | 75          | 110      | 33          | 10     | 250         | 4       | 19          | 4       | 19          | 4       | 19          | USEPA (1999) |

Note. Cr = chromium; Ni = nickel; Cu = copper; As = arsenic; Cd = cadmium; Pb = lead; ASV = average shale value; TRV = toxicity reference value; LEL = lowest effect level; SEL = severe effect level; NA = not available. Values of metals in sediment are presented mean concentration (mg/kg dw).
(TRV), lowest effect level (LEL), and severe effect level (SEL; Table 3). There are 2 possible causes for Cr enrichment in sediment: (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.]). Consequently, the waste discharged from such industries is responsible for the elevated Cr level in the exposed sediment.\textsuperscript{48,52}

The mean concentration of Ni was 103 mg/kg; the highest was observed at site S7 (163 mg/kg). Slightly higher levels of Ni were observed at the sites near the district urban area and downstream, indicating that the higher input of Ni in sediment might originate from urban and industrial wastes.\textsuperscript{16,52} Nickel and its salts are used in several industrial applications, such as electroplating and fabric printing, as well as in storage batteries, automobiles, electrodes, cooking utensils, pigments, lacquer cosmetics, and waste water.\textsuperscript{21,30} The effluents from the Bogra District urban area might be the source of Ni for some sites of the Korotoa River.

The average concentration of Cu was 82 mg/kg; elevated levels of Cu were found at sites S6, S7, and S10 (Table 2). The higher level of Cu indicates its higher input in the sites (S6, S7 and S10); Cu input originates from anthropogenic activities such as vehicle and coal combustion emissions\textsuperscript{49} and car lubricants\textsuperscript{62} and from natural phenomena such as the metal content of rocks and parent materials and processes of soil formation.\textsuperscript{50,63}

The highest As was observed at site S6 (52 mg/kg), followed by site S7 (51 mg/kg). The concentration of As in sediment of this study was in line with that in the other studies and higher than the ASV, TRV, LEL, and SEL (Table 3). Recent anthropogenic activities, such as treatment of agricultural land with arsenical pesticides,\textsuperscript{62} treatment of wood with chromated copper arsenate, burning of coal in thermal plant power stations, and sediment excavation that alters the hydraulic regime and/or arsenic source material, increased the rate of discharge into the freshwater habitat.\textsuperscript{20,64}

The mean concentration of Cd in sediment was 1.5 mg/kg with the range of 0.53–2.8 mg/kg (Table 2). This was in agreement with other studies in Bangladesh and other countries and far higher than the ASV, TRV, and SEL (Table 3), indicating that Cd might pose a risk to the surrounding ecosystems. Elevated concentration of Cd in sediments of the Korotoa River might be related to industrial activity, atmospheric emission, and leachates from defused Ni-Cd batteries and Cd-plated items.\textsuperscript{4,16}

The average concentration of Pb was 63 mg/kg, which was 3 times the ASV and 2 times the TRV and LEL values (Table 3). The elevated level of Pb in sediments can be due to the effect from point and nonpoint sources, such as leaded gasoline, municipal runoff and atmospheric deposition,\textsuperscript{20,52} and chemical manufacturing and steel works in the urban area of Bogra District.\textsuperscript{16} The concentration of Pb in sediment was in agreement with some previous studies (Table 3).

**Comment**

**Heavy metals in fish species**

Heavy metals may act as a source of contamination when significant changes of pH, redox potential, salinity, particulate matter, or microbial activity occur in the environment. These changes can increase the mobility and transport of the metals in the aquatic media and make them bioavailable to the biota.\textsuperscript{48} It is well known that metals can be bioaccumulated in fish tissues.\textsuperscript{55} The magnitude of bioaccumulation is a function of age, species, and trophic transfer. Within the same species, the concentrations of metals may vary with age and body weight.\textsuperscript{66} The concentrations on fresh wet basis of 6 metals (Cr, Ni, Cu, As, Cd, Pb) in 11 different fish species are listed in Table 4. Overall, the mean concentrations of heavy metals in fish species were in the following descending order: Ni (1.4 mg/kg) > Cu (1.0 mg/kg) > As (0.69 mg/kg) > Pb (0.43 mg/kg) > Cr (0.23 mg/kg) > Cd (0.10 mg/kg). The concentration of metals varied considerably among the fish species. However, the overall concentrations of studied metals among the fish species were in the following descending order: \textit{H. fossilis} > \textit{A. testudineus} > \textit{B. batasio} > \textit{C. fasciata} > \textit{C. soborna} > \textit{P. chola} > \textit{A. grammepomous} > \textit{C. punctuate} > \textit{N. atherinoides} > \textit{C. striata} > \textit{N. notopterus}. Bottom-dwelling fish were found to exhibit higher concentrations of heavy metals than were pelagic fishes.\textsuperscript{67} Chromium does not normally accumulate in fish, and hence, low concentration was reported even from the industrialized part of the world. A study has shown a higher rate of uptake in young fish, but the body burden of Cr declines with age due to the rapid elimination.\textsuperscript{30} The mean concentration of Cr in fish species was found to be 0.23 mg/kg with a range of 0.11–0.46 mg/kg (Table 4). No significant difference was observed for Cr concentration among the investigated fish species. The mean concentration of Ni was 1.4 mg/kg; the highest concentration was observed in \textit{A. testudineus} and \textit{H. fossilis} (2.6 mg/kg). The Ni concentration in fish species was higher than the maximum allowable concentration (MAC) in fish (0.8 mg/kg), indicating that Ni might pose a risk to humans through consumption of these contaminated fish species.

Copper was detected in all examined fish species, and a significant difference \((p < .05)\) was observed for Cu content in \textit{A. testudineus} and \textit{H. fossilis} compared with
other species. The mean concentration of Cu was 1.0 mg/kg with a range of 0.57–2.1 mg/kg (Table 4). Arsenic is widespread in the environment from both anthropogenic and natural sources. The highest concentration of As was observed in H. fossilis (1.7 mg/kg), followed by A. testudineus (1.2 mg/kg). Arsenic concentration in these 2 species (A. testudineus and H. fossilis) showed significant differences (p < 0.05) compared with the other species (Table 4). The USEPA has set a value of 1.3 mg/kg fw in tissues of freshwater fish as the criterion for human health protection. Therefore, 2 species of fish (A. testudineus and H. fossilis) are a concern for health risk due to As exposure; the other species showed concentrations of As lower than the MAC. In fish samples, the mean concentrations of Cd ranged from 0.020 mg/kg (C. striata) to 0.23 mg/kg (H. fossilis; Table 4). The average concentrations of Cd in the fish species A. testudineus, H. fossilis, N. notopterus, B. batasi, and C. soborna were higher than the MAC (0.10 mg/kg; Table 4), indicating potential health hazards due to the consumption of these fish species from the studied river. In the investigated fish species, the mean concentration of Pb ranged from 0.15 mg/kg (C. soborna) to 1.1 mg/kg (A. testudineus). Lead concentrations in fish species C. punctate, A. testudineus, H. fossilis, and N. atherinoides were higher than the safe limit of 0.5 mg/kg, indicating these 4 fish species were contaminated by Pb and might pose risks to humans.

A PCA was conducted to infer the hypothetical sources of heavy metals (natural or anthropogenic) following the standard procedure reported in the literature, which showed clustering of the variables into different groups, where variables belonging to one group are highly correlated with each other. The PCA was performed on the dimensionless standardized form of the data set and is presented in Figure 2. The Varimax rotation was used to maximize the sum of the variance of the factor coefficients. Multivariate PCA of heavy metals in the samples explained about 96% (sediment) and 69% (fish) cumulative variance of the data. In the PCA analysis, first 3 PCs were computed, and the variances explained by them were 41.4%, 31.6%, and 22.6% for sediment and 27.1%, 22.2%, and 19.7% for fish, respectively (Figure 2). Overall, the PCA revealed 3 major groups of the metals for both sediment and fish. One group consisted of Pb for sediment and Cu and Pb for fish, which were predominantly contributed by lithogenic sources. The second group showed mutual associations of Cd and Ni for sediment and Cd and As for fish, which were mostly contributed by the industrial emissions in the vicinity of the sampling sites. The third group revealed similar loadings of Cr, Cu, and As in sediment and Cr and Ni in fish, indicating anthropogenic activities.

### Metal bioaccumulation in fish species

Metals in the aquatic environment, particularly in sediment, can be bioaccumulated in fish tissues. Bioaccumulation of heavy metal in fish species is dependent not only on metal exposure and its environment, but also on the different physiological and biochemical activities through which a specific organism deals with metals. Hence, different organisms accumulate metals from the environment depending on their filtration rate, ingestion rate, and gut fluid quality, as well as on the detoxification

| Local name | Scientific name | Cr Concentration | Cr SD | Ni Concentration | Ni SD | Cu Concentration | Cu SD | As Concentration | As SD | Cd Concentration | Cd SD | Pb Concentration | Pb SD |
|------------|----------------|------------------|-------|------------------|-------|------------------|-------|------------------|-------|------------------|-------|------------------|-------|
| Taki       | Channa punctata| 0.17             | 0.13  | 0.69             | 0.62  | 0.78             | 0.39  | 0.54             | 0.35  | 0.032            | 0.03  | 0.52             | 0.50  |
| Balla      | Awaous grammepomus| 0.32             | 0.29  | 1.1              | 0.90  | 0.98             | 0.72  | 0.38             | 0.39  | 0.031            | 0.02  | 0.27             | 0.18  |
| Koi        | Anabas testudineus| 0.46             | 0.61  | 2.6              | 4.0   | 2.1              | 0.69  | 1.2              | 0.92  | 0.16             | 0.24  | 1.1              | 0.91  |
| Shing      | Heteropeusates fossilis| 0.46             | 0.48  | 2.6              | 4.0   | 1.6              | 1.0   | 1.7              | 1.0   | 0.23             | 0.33  | 0.82             | 1.0   |
| Batashi    | Neotropius atherinoides| 0.17             | 0.20  | 0.77             | 0.66  | 0.71             | 0.30  | 0.44             | 0.26  | 0.031            | 0.03  | 0.60             | 0.32  |
| Khalisa    | Colisa fasciata| 0.24             | 0.32  | 1.5              | 1.3   | 0.81             | 0.86  | 0.59             | 0.37  | 0.040            | 0.04  | 0.27             | 0.33  |
| Shoil      | Channa striata| 0.13             | 0.22  | 1.2              | 1.5   | 0.69             | 0.56  | 0.43             | 0.42  | 0.020            | 0.02  | 0.25             | 0.23  |
| Foli       | Notopterus notopterus| 0.15             | 0.22  | 0.86             | 0.87  | 0.57             | 0.47  | 0.47             | 0.23  | 0.10             | 0.22  | 0.16             | 0.19  |
| Tengra     | Batasio batasi| 0.13             | 0.20  | 1.6              | 1.9   | 0.97             | 0.67  | 0.55             | 0.25  | 0.20             | 0.28  | 0.29             | 0.19  |
| Kachki     | Corica soborna| 0.11             | 0.15  | 1.5              | 1.8   | 0.89             | 0.64  | 0.61             | 0.22  | 0.11             | 0.18  | 0.15             | 0.12  |
| Punti      | Puntius chola| 0.18             | 0.18  | 1.1              | 1.1   | 1.0              | 1.0   | 0.59             | 0.59  | 0.043            | 0.04  | 0.34             | 0.34  |
| Average    |                 | 0.23             | 0.32  | 1.4              | 1.8   | 1.0              | 0.75  | 0.69             | 0.61  | 0.10             | 0.18  | 0.43             | 0.53  |
| MAC        |                 | 1                | 0.8   | 4.5              |       | 1                |       | 0.1              |       | 0.5              |       |

Note. Cr = chromium; Ni = nickel; Cu = copper; As = arsenic; Cd = cadmium; Pb = lead; SD = standard deviation; MAC = maximum allowable concentration of heavy metals in fish. Vertically, different letters (a and b) indicate the significant difference at the 0.05 level.
strategies they adopt (e.g., storage in nontoxic form or elimination). Table 5 clearly shows a large variation in BSAF among different fish species and metals. Among the studied metals, the ranking order of mean BSAF values were Cd > As > Ni > Cu > Pb > Cr (Table 5). Among the selected 6 metals, Cd showed the highest BSAF value, suggesting a higher rate of accumulation in fish species. At some sites, levels of metal might be high but accumulation lower than expected due to metal complexation.

The BSAFs for the studied metals in A. testudineus and H. fossilis were slightly higher than the values obtained for other fish species (Table 5). This can be explained by the ingestion of sediment as well as the omnivorous feeding behavior of A. testudineus and H. fossilis, which may lead to greater BSAFs for these species. Therefore, 2 of the fish species investigated in this study, A. testudineus and H. fossilis, can be potential bioindicators for the assessment of heavy metal contamination in this riverine environment. This study revealed a trivially higher accumulation of metals in 2 fish species (A. testudineus and H. fossilis). These 2 fish species are bottom feeders, and therefore, sediments could be the major source of trace metal accumulation in these fish species. Bottom-dwelling fish are found to exhibit higher concentrations of heavy metals than pelagic fishes. The BSAFs of individual metals among the fish species and sampling sites did not display similar patterns due to the environment-specific phenomenon. The ingested sediments found in the digestive tracts of fish accelerated the accumulation of metal.

### Noncarcinogenic and carcinogenic risks

Target hazard quotients (THQs) of 6 heavy metals from consuming fish species are listed in Table 6. The THQ values for individual metals (except As) in fish species were less than unity, which is considered safe for human consumption. Nevertheless, total values of THQ for As and Pb were greater than 1.0 (Table 6);

![Component Plot in Rotated Space](image_url)

**Figure 2.** Principal component analysis (PCA) of heavy metals in sediment (A) and fish (B) collected from Korotoa River, Bangladesh.

### Table 5. Biota-sediment accumulation factor of heavy metals in fish species collected from Korotoa River, Bangladesh.

| Fish species | Scientific name | Cr Mean BSAF | Cr SD | Ni Mean BSAF | Ni SD | Cu Mean BSAF | Cu SD | As Mean BSAF | As SD | Cd Mean BSAF | Cd SD | Pb Mean BSAF | Pb SD |
|--------------|-----------------|--------------|-------|--------------|-------|--------------|-------|--------------|-------|--------------|-------|--------------|-------|
| Taki         | Channa punctata | 0.007        | 0.008 | 0.038        | 0.033 | 0.037        | 0.025 | 0.106        | 0.106 | 0.073        | 0.054 | 0.029        | 0.029 |
| Baila        | Awaous grammepomus | 0.010 | 0.010 | 0.094        | 0.046 | 0.047        | 0.035 | 0.089        | 0.056 | 0.067        | 0.040 | 0.015        | 0.011 |
| Koi          | Anabas testudineus | 0.014 | 0.019 | 0.142        | 0.094 | 0.092        | 0.043 | 0.254        | 0.248 | 0.459        | 0.392 | 0.061        | 0.060 |
| Shing        | Heteropneus testudineus | 0.013 | 0.013 | 0.087        | 0.096 | 0.082        | 0.074 | 0.297        | 0.210 | 0.488        | 0.592 | 0.039        | 0.041 |
| Batashi      | Neotroplus atherinoles | 0.007 | 0.010 | 0.039        | 0.034 | 0.033        | 0.019 | 0.123        | 0.182 | 0.068        | 0.048 | 0.034        | 0.019 |
| Khalisa      | Colisa fasciata | 0.007        | 0.010 | 0.058        | 0.061 | 0.038        | 0.045 | 0.124        | 0.151 | 0.082        | 0.057 | 0.014        | 0.014 |
| Sholi        | Channa striata | 0.003        | 0.004 | 0.074        | 0.048 | 0.028        | 0.024 | 0.099        | 0.179 | 0.042        | 0.039 | 0.014        | 0.014 |
| Foli         | Notopterus notopterus | 0.005 | 0.007 | 0.040        | 0.036 | 0.028        | 0.033 | 0.095        | 0.144 | 0.171        | 0.291 | 0.011        | 0.014 |
| Tengra       | Batasiaobatusio | 0.005        | 0.009 | 0.094        | 0.074 | 0.044        | 0.039 | 0.120        | 0.134 | 0.294        | 0.390 | 0.017        | 0.013 |
| Kachli       | Coriascoborna | 0.004        | 0.008 | 0.088        | 0.070 | 0.039        | 0.037 | 0.141        | 0.139 | 0.259        | 0.391 | 0.009        | 0.009 |
| Puti         | Puntius chola | 0.006        | 0.008 | 0.045        | 0.048 | 0.049        | 0.032 | 0.119        | 0.123 | 0.094        | 0.102 | 0.019        | 0.009 |

*Note. Cr = chromium; Ni = nickel; Cu = copper; As = arsenic; Cd = cadmium; Pb = lead; BSAF = biota-sediment accumulation factor; SD = standard deviation.*
consequently, the consumption of these fish species was considered to be unsafe, and their consumption was not recommended. Therefore, consumers are at high risk due to the exposure of As and Pb from fish that were associated with noncancerogenic risks. Given all metals in consideration, total THQ (sum of individual metal THQs) for the consumption of fish species was 1.20–4.59 (Table 6); therefore, potential health risks from studied fish species are of some concern.

The target carcinogenic risks (TRs) derived from the intake of As and Pb were calculated and are presented in Table 6. In fish species, TR values for As ranged from 0.243 to 0.951; TR values for Pb ranged from 0.001 to 0.006 (Table 6). TR values for As and Pb were higher than the acceptable risk limit (0.000001), indicating the inhabitants consuming these fish species are exposed to As and Pb with lifetime cancer risk. According to the results of this study, the potential health risk for the inhabitants due to metal exposure through consumption of fish should not be ignored.

### Conclusions

In conclusion, this study revealed that the concentrations of heavy metals in sediment from some sites exceeded the sediment quality standards, indicating their risk to the surrounding ecosystems. Fish species from the study river were also contaminated by the relevant metals, particularly Ni, As, Cd, and Pb, which could be a potential health concern to the local inhabitants. The concentrations and biota-sediment accumulation factors (BSAFs) of heavy metals in *H. fossilis* and *A. testudineus* were slightly higher than those of other species, which might be due to their mode of feeding behavior. *H. fossilis* and *A. testudineus* could be potential bioindicators for metal pollution study. The target hazard quotients (THQs) of individual metals (except As) would not pose any potential risk; however, combined effects of heavy metals can pose significant risks. The carcinogenic and noncancerogenic risks of As and Pb due to fish consumption showed a considerable risk.

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