#### **RESEARCH ARTICLE**



# **Spatiotemporal variation and toxicity of trace metals in commercially important fsh of the tidal Pasur River in Bangladesh**

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## **Abstract**

The release of toxic metals in the water creates an adverse condition for the living organisms (e.g., fsh). The aim of this research was to learn more about the spatiotemporal variations and toxicity of heavy metals (As, Cr, Cd, and Pb) among fsh species that are economically important (*Tenualosa ilisha*, *Gudusia chapra*, *Otolithoides pama*, *Setipinna phasa*, *Mystus vittatus*, *Glossogobius giuris*, *Harpadon nehereus*, *Pseudapocryptes elongatus*, *Polynemus paradiseus*, and *Sillaginopsis panijus*) collected from Pasur River. Heavy metal (HMs) concentrations were evaluated using the atomic absorption spectrometry (AAS) technique. Most of the metals showed no signifcant variation spatiotemporally (*p* ˃ 0.05) except As and Cr showed substantial variation in terms of seasons ( $p \text{~} 0.05$ ). All fish species' Cr and Pb concentrations, as well as As and Cd values, were estimated to be greater than FAO/WHO tolerable concentrations, implying that these metals pose danger to humans. HM has a total hazard quotient (THQ) value in individual fsh species reported to be greater than 1, whereas an individual metal, arsenic, exceeds the standard value  $(THQ > 1)$ , causing a significant noncarcinogenic issue in the study region. The target hazard (TR) value for As and Pb exceeds the USEPA norm ( $10^{-4}$ ) suggesting that long-term consumption of fsh poses a chronic cancer risk to the people in the study feld. According to the fndings, the fsh in the Pasur River are unfit for human consumption. The correlation matrix  $(CM)$  indicates that sources of metals are similar (e.g., industries, ships, agricultural inputs, etc.).

**Keywords** Fish · Toxic substances · Carcinogenic hazard · Pasur River · Bangladesh



#### **Introduction**

HMs' high poisonousness, endurance, and bioavailability in the river water ecosystem are regarded as the most harmful chemical pollutants (Han et al. [2021;](#page-13-0) Lao et al. [2019](#page-13-1); Ali et al. [2018](#page-12-0); Bhuyan and Bakar [2017a;](#page-12-1) Islam et al. [2012](#page-13-2)). Since their increased trend in sediments, water, and fsh, metal contamination has become a worldwide problem (Ahsan et al. [2018](#page-12-2); Bhuyan et al. [2017,](#page-12-3) [2019;](#page-12-4) Burger and Gochfeld [2005](#page-12-5); Santos et al. [2004](#page-13-3)). River water pollution has been exacerbated by rapid industrialization, haphazard urbanization, and indiscriminate population development (Bhuyan and Bakar [2017b](#page-12-6); Hajeb et al. [2009](#page-13-4); Islam et al. [2017\)](#page-13-5). The examined river receives immense amounts of untreated effluents from crop fields, agrochemicals, industrial waste, sewage treatment, and industries such as power plants, cotton, fertilizers, oil refneries, and others. Moreover, some brickfelds, feeding sites, and entertainment areas directly discharge chemical wastes to the river. The rising population increased water withdrawal and agriculture and other industrial practices all signifcantly afect the river. The river environment faces signifcant problems from overexploitation and the dumping of unprocessed textile wastewaters into the river (Bebbington et al. [1977](#page-12-7); Singh and Kumar, [2017\)](#page-13-6). Toxic metals discharged by these sources damage marine ecosystems (Bhuyan and Bakar [2017b;](#page-12-6) FAO/WHO [2002](#page-12-8); Habibullah-Al-Mamun et al. [2017\)](#page-13-7). Toxic metal pollution of fsh due to intake of contaminated water and food (zooplankton and phytoplankton) are considered bioindicators of toxic metal contamination (Burger et al. [2002](#page-12-9); Karunanidhi et al. [2017](#page-13-8); Kuklina et al. [2014;](#page-13-9) Saha and Zaman [2013](#page-13-10); Svobodova et al. [2004\)](#page-14-0). The fish's membrane and branchiae could be a good substrate of HM buildup. Because of their bioaccumulation origin in marine environments, these HMs are harmful to aquatic animals and humans (Bhuyan et al. [2016a;](#page-12-10) Islam et al. [2018\)](#page-13-11). Metal poisoning poses a concern to human health since it enters the human food chain through the consumption of a range of aquatic species (e.g., fsh) (Alhashemi et al. [2012;](#page-12-11) Habibullah-Al-Mamun et al. [2017;](#page-13-7) Islam et al. [2016\)](#page-13-12). Adulteration of fsh is a big growing issue because of the health risks involved with eating fsh. Moreover, the livelihood of communities (use river water for their daily activities) living near or adjacent to the river are more prone to threat (Ali et al. [2016;](#page-12-12) Bhuyan and Islam, [2016](#page-12-13); Bhuyan et al. [2016a](#page-12-10); Osman et al. [2016](#page-13-13)).

Fish consumption has increased over the world at the same time as concerns about its nutritious and medicinal benefts have developed. Fish are considered the source house of high protein, minerals, vitamins, and unsaturated lipid (Bhuyan et al. [2016a;](#page-12-10) DoF [2019](#page-12-14); Hossen et al. [2018;](#page-13-14) Medeiros et al. [2012\)](#page-13-15). Ingestion of poisonous metal-contaminated fsh, on the other hand, has been linked to a number of serious disorders. Chromium (Cr) causes anuria, nephritis, and severe lesions in infected fsh, including kidney lesions (Proshad et al. [2018\)](#page-13-16). Cadmium toxicity results in impaired reproductive ability, kidney illness, malignancies, hypertension, and hepatic dysfunction, among other things (Al-Busaidi et al. [2011;](#page-12-15) Ali et al. [2018](#page-12-0); Bhuyan et al. [2016a](#page-12-10)). Lead poisoning damages the liver and produces renal failure (Bhuyan et al. [2019](#page-12-4); Lee et al. [2011](#page-13-1)).

With a yearly demand of 42.38 lakh metric tons, fish accounts for a major component of the Bangladeshi population's daily diet (DoF [2019](#page-12-14); Hossen et al. [2018](#page-13-14)). Bangladeshis eat fsh on a regular basis (annual ingesting 21.90 kg/person), and it is one of their key sources of protein (DoE, [2019\)](#page-12-16). However, toxic metal concentrations in fish bodies enter into the human body (FAO/WHO, [2002](#page-12-8)), either directly or indirectly, and have an efect on human health (Islam et al. [2016](#page-13-12)). Bangladeshis tend to eat river fish as part of their regular diet. Industrial effluents pollute the Pasur River, resulting in large concentrations of HMs (e.g., As, Cr, Cd, and Pd) being dumped into the river. However, there is little scientifc evidence of HM pollution in the fsh of the study river. As a result, the current research looked into HMs in various commercial species from the Pasur River.

## **Materials and methods**

#### **Sampling sites**

The study took place along the Pasur River, which is surrounded by various industries. It is in Khulna City, right next to the Sundarbans. The river fows into Bangladesh's Bay of Bengal (Fig. [1\)](#page-2-0). The Pasur River is a river in southwestern Bangladesh and a distributary of the Ganges. It continues the Rupsa River. All its distributaries are tidal. It meets the Shibsa River within the Sundarbans, and near to the sea, the river becomes the Kunga River (Dara et al. [2004\)](#page-12-17). It is the deepest river in Bangladesh. The maximum and minimum widths are 650 m and 322 m, respectively, with an average width of 486 m. It is a meandering, perennial river and a considerable number of fsheries, dockyards, shipyards, and industries that are located along this river's bank. Various types of industrial wastes, solid waste, and hazardous pollutants are produced as a result of unrest production activities, and most of them are promptly discharged into the river without adequate treatment. Fe, Cu, Zn, Cd, Pb, Mn, and As are found in the telecommunication, oil, limestone, metallurgical, plating, and battery industries' by-products, respectively (Bhuyan et al. [2016b](#page-12-18); Dara and Mishra [2004;](#page-12-17) Hilal and Ismail [2008\)](#page-13-17).

<span id="page-2-0"></span>



## **Fish sample collection**

During the winter and summer seasons, fish were collected from several water sites along the Pasur River. The detail of the collected fsh is documented in Table [1](#page-3-0). The studied fish species are mostly consumed by the Bangladeshi population on a regular basis. The government of Bangladesh earns foreign currency by exporting the examined fsh species. Therefore, we have selected these fsh species for the present study. A total of 10 individuals were collected for the analysis.

### **Geological information**

The Pasur River is a distributary of the Ganges and one of Khulna's most infuential rivers. It follows the Rupsa River and all of its tributaries, all of which are tidal. Inside the Sundarbans, it reaches the Shibsa River, and near the sea, it merges with the Kunga River to form the Kunga River. It is the country's river with the greatest depth. Its source is the Madhumati River northeast of Khulna, and it runs south for 110 mi (177 km) to the Bay of Bengal, passing through the swampy Sundarbans area and the port of Mongla. Summers have more rainfall and water than winters, which can lead to changes in water and sediment metal concentrations (Ali et al. [2016;](#page-12-12) Islam et al. [2015](#page-13-18)). Changes in HM concentrations in fsh species are possible as a result of this (Ali et al. [2016](#page-12-12); Bhuyan et al. [2016b](#page-12-18)).

#### **Fish sample preservation**

Before being transferred to the lab, the fish were placed on ice in a thermos to preserve roughly at−4 °C. The fsh were washed, scales removed, viscera removed, bone removed, skull removed, and gills removed) before being left to air dry. Samples were pasted and homogenized with an ultrasonic homogenizer after air drying before being kept in a plastic bag at−25 °C (Bhuyan et al. [2016a](#page-12-10)).

#### **Fish species identifcation**

The site has been used to identify fsh species. Species that were difficult to identify at the moment were taken to a lab

Scientific name	English name	Feeding habits	Habitat	Length $(cm)$ Weight $(g)$		<b>IUCN</b> status
Tenualosa ilisha	Hilsa shad	Phytoplankton, zooplank- ton, plants, mollusks, and crustaceans	Pelagic		$33.12 \pm 5.40$ $570.50 \pm 130.20$ LC	
Gudusia chapra	Indian river shad	Phytoplankton, zooplank- ton, and crustaceans	Pelagic	$14.70 + 3.55$	$45.80 \pm 18.35$	LC
Otolithoides pama	Pama croaker	Crustaceans and mall teleost			Benthopelagic $22.11 \pm 3.30$ $185.90 \pm 15.50$	NE
Setipinna phasa	Gangetic hairfin anchovy	Zooplankton, zoobenthos, and crustaceans	Pelagic	$17.50 \pm 4.10$	$48.50 \pm 9.55$	LC
Mystus vittatus	Striped dwarf catfish	Zoobenthos, insects, crus- taceans, and mollusks	Demersal	$13.70 \pm 2.38$	$22.47 \pm 11.15$	LC
Glossogobius giuris	Tank goby	Nekton, detritus, zooben- thos, and insects	Benthopelagic $20.50 \pm 3.75$		$105.50 \pm 30.77$	LC
Harpadon nehereus	Bombay duck	Nekton and small finfish	Benthopelagic	$23.20 \pm 3.50$	$87.40 \pm 32.50$	NE
Pseudapocryptes elon- gatus	Lanceolate goby	Phytoplankton and inver- tebrates	Demersal	$16.52 \pm 2.20$	$19.45 \pm 6.35$	LC
Polynemus paradiseus	Paradise threadfin	Zoobenthos, nekton, invertebrates, and crus- taceans	Demersal	$16.35 \pm 4.41$	$30.21 \pm 11.40$	LC
Sillaginopsis panijus	Flathead sillago	Zoobenthos, plants, nek- ton, and crustaceans	Demersal	$20.57 \pm 3.66$	$50.65 + 27.30$	<b>NE</b>
Lates calcarifer	Barramundi	Zooplankton, zoobenthos, mollusks, crustaceans, and invertebrates	Demersal	$31.25 \pm 4.79$	$730.16 \pm 57.53$	<b>NE</b>
Pampus argenteus	Silver pomfret	Zoobenthos, zooplankton, jellyfish, benthos, inver- tebrates, and mollusks			Benthopelagic $21.42 \pm 3.52$ $388.50 \pm 21.35$	<b>NE</b>

<span id="page-3-0"></span>**Table 1** The ecological parameters and morphometric measurements of ten fsh species gathered from Bangladesh's Pasur River

Note: sample number=*n*

IUCN conservation status: *LC*, least concern; *NE*, not evaluated

for further examination. The photographs of studied fsh species are presented in Fig. [2.](#page-4-0)

#### **Heavy metal determination**

AAS used a common analytical technique to assess the HM content. For metal analysis, sample selection is important. To prevent contamination, samples were regularly treated with care. The reagents were of analytical grade, and the glassware had been thoroughly washed. Throughout the report, distilled water was used. To correct the instrument's readability, blank reagent determinations were used.

#### **Fish sample digestion**

Merck Germany provided all target element standard solutions with the maximum level of purity (99.98%). For sample digestion, The  $HNO<sub>3</sub>$  used was ultrapure. All of the other acids and substances were super pure and were sourced from Germany and Spain (JECFA, [2005](#page-13-19)). Fish that had been homogenized were taken out of the freezer and allowed to come to room temperature for 1 h. For As determination,

2 g of material was poured in a 100 ml beaker and placed 15-m strenuous nitric acid on the hotplate (Ali et al. [2020](#page-12-19); Bhuyan et al. [2016a;](#page-12-10) Rahman et al. [2020\)](#page-13-20). Heat the sample at 130 °C for 5 h or until just 1–2 ml solution remains after digestion. After cooling the sample, 1 ml of hydrogen peroxide was added and heated at 120 °C for another 30 min. The sample was cleaned with Whatman 41 filter paper, rinsed with distilled water, and converted to a weight of 100 ml for AAS investigation. In a 50 ml beaker, 1.5 ml HCl was added to 12.5-ml processed test aliquots. After that, 1 ml of potassium iodide suspension was transferred to the last volume of 50 ml, and the As reduction reaction was allowed to fnish for 2 h (FAO, [2006;](#page-12-20) Ali et al. [2020;](#page-12-19) Bhuyan et al. [2016a;](#page-12-10) Rahman et al. [2020;](#page-13-20) Shaheen et al. [2015](#page-13-21)). The reading was taken after a 5 ml aliquot of the ready sample was poured into the reaction container. Cr, Cd, and Pb analysis: in a 100-ml Pyrex beaker, 2 g of standardized sample was inserted and burned in a muffle furnace for a minimum of 10 h at 150 °C for 1 h. After it has cooled, we added 5 ml of 6 M HCl and heated it until totally dry on the hotplate. Finally, 10 ml of 0.1 M HNO<sub>3</sub> was added and then heated for another 30 min before being fltered with Whatman No. 41

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<span id="page-4-1"></span>**Table 2** Analytical conditions for employing AAS to quantify heavy metals in a sample solution



and leveled with  $0.1$  M HNO<sub>3</sub> in a 50-ml volumetric flask. Finally, using GF-AAS, the samples were tested for Cr, Cd, and Pb (Ali et al. [2020](#page-12-19); Rahman et al. [2020](#page-13-20)).

#### **Analytical technique and accuracy check**

Using a graphite furnace atomic absorption spectrometry (GFAAS) and hydride generator method, all of the media were tested for Pb, Cd, Cr, and As using AAS. All of the procedures were tested in-house in accordance with EC567/2002. Table [2](#page-4-1) summarizes the analytical conditions for measuring HMs in samples using AAS.

The calibration criteria for instruments were created using Sigma-Aldrich's (Switzerland) diluting standard (1000 ppm); mg/kg was used to denote the fsh weight. Deionized ultrapure (0.05 s) water was utilized during the experiment. All equipment and bottles were washed with 20% nitric acid before being treated with deionized water and placed in an oven to dry (Lakshmanan et al. [2009](#page-13-22); Lao et al. [2019\)](#page-13-1). To ensure that the analytical process was

<span id="page-4-2"></span>**Table 3** Concentrations of metals detected in the National Research Council Canada's Certifed Reference Materials DORM-4 (mean standard errors, in mg/kg as wet wt.) by AAS (mean standard errors, in mg/kg as wet wt.)  $(n=3)$ 

	Element Certified value	Measured value	Deviation $(\%)$	Recovery $(\%)$	
DORM-4					
As	$6.80 + 0.09$	$6.07 + 0.13$	8.07	89.19	
Cr	$1.87 + 0.10$	$1.85 + 0.02$	0.65	99.09	
Cd	$0.306 + 0.005$	$0.298 + 0.01$	1.66	97.06	
Ph	$0.416 + 0.07$	$0.380 + 0.18$	6.28	91.35	

correct, fsh protein-approved reference substances for trace elements were applied. NRC (Canada) processed and supplied these fish samples (NRC [1989\)](#page-13-23). The certified and observed values were found to be very close to each other. Table [3](#page-4-2) demonstrates that the recorded certifed materials' standard deviations of the means ranged from 0.65 to 8%, with a % of a return of 89 to 99%.

## **Data analysis**

## **Estimated daily intakes (EDIs)**

Using the formula below, the EDI for HMs was derived by multiplying the mean content in samples by the wt. of food item intake by a person (60 kg bw/adult in Bangladesh), as determined by the family income and expenditure survey (Shaheen et al. [2015](#page-13-21)):

 $EDI = (FIR \times C)/BW$ .

FIR stands for food intake rate (g/person/day), *C* stands for the metal amount in food (mg/kg), and BW stands for the adult resident's BW (considering 60 kg) (FAO 2004; Pintaeva et al [2011](#page-13-24)). Fish is consumed at a rate of 59.91 g per day on a fresh wt. basis (HIES [2011](#page-13-17); Oguri et al. [2012](#page-13-25); Kuklina et al. [2013](#page-13-22)).

## **Risk of noncarcinogenicity**

The risk-based concentration table was provided by the USEPA ([2010](#page-14-1)). Region III was applied to estimate the noncarcinogenicity of fsh. The THQ was used to calculate the noncarcinogenic danger of each metal from fsh ingestion (USEPA [1989](#page-14-2)):

THQ = {(EFr  $\times$  ED  $\times$  FIR  $\times$  *C*)/(RfD  $\times$  BW  $\times$  AT)}  $\times$  10<sup>-3</sup>

Total THQ (THQ) =  $THQ_{\text{toxicant 1}} + THQ_{\text{toxicant 2}} + \ldots + THQ_{\text{toxicant } n}$ 

The letters THQ stand for target danger quotient, EFr for contact times (365 days/year), ED for exposure period (70 years), FIR for food intake rate (g/day), *C* for the amount of metal in foods (mg/kg dw), RfD for oral reference dosage (mg/kg/day), and AT for average time for noncarcinogens (365 days/year amount of exposure years) (USEPA [2008\)](#page-14-3). For Cr, As, Cd, and Pb, object reference levels of 1.5, 0.0003, 0.0005, and 0.0035 mg/kg/day were adopted accordingly (Nadal et al. [2008;](#page-13-26) Reddy et al. [2008\)](#page-13-27). There may be a health hazard if the THQ is  $\geq$  1, and relevant measures and safeguards should be undertaken (Islam et al. [2014\)](#page-13-28).

To quantify the total possible for noncarcinogenic impacts from several HMs, a hazard index (HI) was developed following the (USEPA [1999USEPA \(2006\).](#page-14-4)) standards for health risk evaluation of chemical combinations. HI is calculated from THQs using the number of (USEPA [2010](#page-14-1)). The following is the equation for calculating the HI:

HI = ΣTTHQ = TTHQ food 1 + TTHQ food 2 +………. + TTHQ food *n*

## **Carcinogenic risks**

To evaluate carcinogen risk, the incremental risk of acquiring cancer throughout a lifetime of exposure to a possible carcinogen was used (USEPA [1989](#page-14-2)). The predicted carcinogenic hazards originating from As and Pb intake were estimated following the USEPA standard. The following equation was applied to estimate target hazard (TR):

TR =  $\{ (EFr \times ED \times FIR \times C \times CSP0) / (BW \times AT) \} \times 10^{-3}$ 

where EFr is for contact time (365 days per year), ED stands for the contact period (70 years) [65], and AT stands for carcinogenic average time (365 days per year, 70 years). The oral carcinogen slope factor for As and Pb was 1.5 and  $8.5 \times 10^{-3}$  (mg/kg/day)<sup>-1</sup>, respectively, as per the IRIS database (USEPA, [2006](#page-14-2); USEPA [2010\)](#page-14-1).

## **Statistical analysis**

The data was analyzed with the SPSS V. 20 statistics software. The metal values in fish were measured, and the means, standard deviations, and correlation coefficients were calculated. Microsoft Excel 2013 was used for the rest of the calculations.

## **Results and discussion**

## **Amount of toxic substances in species of fsh**

The levels of hazardous metals in twelve fsh from the Pasur River are tabulated in Table [4](#page-6-0). In the current study, the mean value of As in fish fluctuated from 0.79 to 3.817 mg/kg dry wt. in the summer and winter seasons, respectively (Table [4\)](#page-6-0).

HMs were estimated from fsh muscle since Bangladeshi people are more likely to eat fsh muscles (edible component) than branchiae, liver, kidneys, sex gland, and other portions of the fsh. Present HMs concentration in fsh fesh compared with different national and international rivers (Table [5](#page-8-0)). Figures [3](#page-9-0) and [4](#page-9-1) show the spatial–temporal variation of HMs.

Arsenic is widely distributed due to both man-made and natural origins. At site 3, the maximum value of As (3.817 mg/kg) was discovered in *Mystus vittatus*. *Lates calcarifer* at site 2 had the lowermost As amount (0.79 mg/kg). As levels varied between 0.79 and 2.94 mg/kg in the summer to 0.85 and 3.82 mg/kg in the winter. As is a possibly poisonous metal that is found in approximately 90% of sea-food and fish species (USFDA [1993\)](#page-14-5). For human health protection, in freshwater fsh samples, the USEPA establishes a reference limit of 1.3 mg/kg (Burger et al. [2004\)](#page-12-21). The

TTHQ(individualfood) = THQtoxicant1 + THQtoxicant2 +………⋯ + THQtoxicant*n*

<span id="page-6-0"></span>**Table 4** Concentrations of trace elements (mg/kg dw) in 12 fish samples obtained from the Pasur River in Bangladesh

Sites	Name of fish	Metals (summer season)				Metals (winter season)			
		As	$\mathbf{C}$ r	Cd	Pd	As	$\mathbf{C}\mathbf{r}$	C <sub>d</sub>	Pd
Site 1		1.078	0.028	0.078	0.352	1.107	0.057	0.092	0.51
Site 2		1.054	0.012	0.057	0.322	1.123	0.062	0.077	0.441
Site 3	Tenualosa ilisha	1.113	0.023	0.086	0.296	1.174	0.044	0.095	0.562
Site 4		0.965	0.031	0.082	0.367	1.137	0.053	0.115	0.49
Site 5		1.046	0.037	0.079	0.363	0.97	0.062	0.081	0.531
Site 1		1.083	0.205	0.097	0.512	1.428	0.295	0.109	0.612
Site 2		1.055	0.186	0.091	0.44	1.221	0.251	0.125	0.6
Site 3	Gudusia chapra	1.07	0.217	0.085	0.538	1.45	0.31	0.095	0.578
Site 4		1.117	0.225	0.127	0.5	$1.5\,$	0.278	0.145	0.634
Site 5		1.081	0.188	0.077	0.583	1.459	0.308	0.075	0.627
Site 1		1.721	0.519	0.095	0.985	1.802	0.622	0.109	1.052
Site 2		1.522	0.542	0.069	0.786	1.75	0.7	0.11	1.156
Site 3	Otolithoides pama	1.83	0.47	0.097	1.056	1.823	0.654	0.098	0.98
Site 4		1.754	0.573	0.134	0.878	1.8	0.588	0.078	1.033
Site 5		1.7	0.455	0.085	1.134	1.832	0.571	0.2	1.042
Site 1		1.412	0.295	0.099	0.632	1.501	0.395	0.129	0.712
Site 2		1.357	0.285	0.085	0.734	1.531	0.356	0.1	0.721
Site 3	Setipinna phasa	$1.5\,$	0.312	0.124	0.612	1.4	0.397	0.136	0.635
Site 4		1.276	0.235	0.105	0.593	1.584	0.412	0.115	0.732
Site 5		1.523	0.298	0.084	0.534	1.492	0.375	0.132	0.71
Site 1		2.83	0.705	0.775	3.117	3.717	0.991	0.81	3.852
Site 2		2.771	0.774	0.823	3.034	3.625	0.882	0.712	3.776
Site 3	Mystus vittatus	2.936	0.653	0.761	3.012	3.817	0.93	0.876	3.902
Site 4		2.8	0.687	0.721	3.213	3.673	1.23	0.864	3.883
Site 5		2.842	0.73	0.765	3.347	3.71	0.92	$\rm 0.8$	3.84
Site 1		1.63	0.773	0.315	1.025	1.742	0.854	0.428	1.357
Site 2		1.635	0.734	0.336	1.011	1.68	0.812	0.391	1.324
Site 3	Glossogobius giuris	1.556	0.756	0.305	1.1	1.674	0.774	0.44	1.05
Site 4		1.7	0.725	0.27	1.32	1.883	0.871	0.532	1.42
Site 5		1.591	0.813	0.36	0.96	1.795	0.92	0.326	1.56
Site 1		1.218	0.759	0.395	1.024	1.239	0.852	0.432	1.521
Site 2		1.115	0.773	0.228	1.023	1.127	0.845	0.429	1.612
Site 3	Harpadon nehereus	1.089	0.732	0.45	1.014	1.534	0.779	0.502	1.33
Site 4		0.986	0.71	0.432	1.126	1.015	1.015	0.419	1.72
Site 5		1.359	0.762	0.4	0.98	1.25	0.76	0.375	1.361
Site 1		0.983	0.612	0.425	0.885	1.236	0.698	0.578	0.985
Site 2		1.026	0.634	0.395	0.921	1.2	0.705	0.649	1.12
Site 3	Pseudapocryptes elongatus	0.936	0.556	0.443	0.856	1.365	0.56	0.552	0.965
Site 4		1.054	0.645	0.325	0.874	1.23	0.734	0.413	0.87
Site 5		0.95	0.612	0.535	0.912	1.151	0.72	0.62	0.975
Site 1		1.693	0.662	0.675	0.985	1.98	0.668	0.564	1.027
Site 2		1.565	0.569	0.563	0.884	2.015	0.723	0.712	1.104
Site 3	Polynemus paradiseus	1.88	0.723	0.71	0.934	1.973	0.648	0.573	1.037
Site 4		1.621	0.716	0.455	1.042	1.975	0.71	0.583	1.051
Site 5		1.339	0.659	0.67	0.956	1.97	0.583	0.757	0.974
Site 1		1.771	0.753	0.554	1.523	1.853	0.851	0.665	2.014
Site 2		1.853	0.76	0.573	1.385	1.923	0.734	0.671	2.019
Site 3	Sillaginopsis panijus	1.72	0.734	0.612	1.64	1.865	0.912	0.554	1.956
Site 4		1.664	0.717	0.6	1.592	1.775	0.864	0.654	1.896



highest allowed As concentration in tissue residual was set at 2 mg/kg by the ANZFA  $(2011)$  $(2011)$ . According to the findings of this analysis, approximately 70% of fsh species surpass the value of the 1.3 limit set by the USEPA. *G. chapra*, *T. ilisha*, *S. phasa*, *P. paradiseus*, *O. pama*, *H. nehereus*, and *S. panijus* are among the fish species that cause As pollu-

tion due to higher As concentrations than MTC (Table [4\)](#page-6-0).

The mean Cr concentration in the examined fish were fuctuated from 0.012 and 1.015 mg/kg dw between the summer and winter seasons (Table [4\)](#page-6-0). Cr concentrations fuctuated from 0.012 to 0.834 mg/kg dw in the summer and 0.044 to 1.015 mg/kg dw in the winter. *Sillaginopsis panijus* at site 5 had the maximum amount (0.834 mg/kg) in the summer. *Tenualosa ilisha* at site 2 had the lowest, while *Tenualosa ilisha* at site 1 had the highest. In the winter, *Harpadon nehereus* had the highest value (1.015 mg/kg), and *Tenualosa ilisha* had the lowest value (0.044 mg/g) at site 3. In reality, Cr accumulation in the body of fsh is lower in the developed world. According to the reference, Plaskett and Potter ([1979\)](#page-13-29) established a recommended reference value for Cr of 5.5 mg/kg dw in Western Australia, which was greater than the mean Cr concentration detected in the study. This study's Cr amount was greater than earlier studies in the Kichera River, Okumeshi River, Gumti River, and Kichera River, indicating increased Cr pollution in this study region is shown in Table [4](#page-6-0) (Amin et al. [2011](#page-12-23); Raphael et al. [2011](#page-13-30)).

The mean Cd value in fish was recorded between 0.06 and 0.82 mg/kg dw in the summer and 0.08 and 0.88 mg/ kg dw in the winter (Table [4](#page-6-0)). During the winter, the maximum value (0.88 mg/kg) was found in *Mystus vittatus* at site 3. During the summer, the lowest amount (0.06 mg/kg)

was found in *Cynoglossus arel* at site 2 (Table [4\)](#page-6-0). *O. pama*, *G. chapra*, *H. nehereus*, *S. phasa*, *S. panijus*, *P. chinensis*, and *P. paradiseus* surpass the maximum permissible value (0.10 mg/kg dw) for Cr, which is regarded a risk to human health when consumed. Cd in seafood has an allowable value of 2.0 mg/kg set by the ANHMRC (Plaskett and Potter [1979](#page-13-29)). Cd is a lethal HM that can cause extreme toxicity at very low concentrations (less than 1 mg/kg), and its deadly characteristics are greater than other metallic elements (Friberg et al. [1971](#page-12-8)). Cd amounts are capped at 1 mg/kg in Spanish law (DoF [2019](#page-12-14); JECFA 2004). The Cd levels in the fish were found above the MTC value. The amount of Cd was recorded below the limit set by ANHMRC. Long-term Cd buildup in fsh could pose a major threat to public health.

In the summer, the mean Pb amount in the examined fish was 0.5–3.3 mg/kg dw, while in the winter, it was 0.[4](#page-6-0)9–3.89 mg/kg dw (Table 4). The maximum amount (3.89 mg/kg dw) of Pb was reported in *Mystus vittatus* during winter at site 4, and the lowest value (0.49 mg/kg dw) was reported in *Tenualosa ilisha* during winter at site 4. *Tenualosa ilisha* and *Gudusia chapra* are some of the fsh species found in the region. Humans living in the study area may be exposed to chronic toxicity from eating the studied fish species. ANHMRC proposed a maximum permissible value of 9.6 mg/kg dw (Plaskett and Potter [1979](#page-13-29)), and the Spanish regulation sets a limit of 2 mg/kg for Pb (Bristi et all [2019](#page-12-24); Ekeanyanwu et al. [2010\)](#page-12-25). When comparing Pb concentrations in the current study to those in the Gulf of Cambay, Okumeshi River, Wadi Hanifah, and Kichera River (Abdel-Baki et al. [2011](#page-12-26); Reddy et al. [2007\)](#page-13-31), it was found that the current study had higher Pb concentrations, indicating Pb



<span id="page-8-0"></span>J,  $\frac{4}{3}$  $\frac{1}{2}$  $\ddot{\cdot}$  $\frac{1}{2}$  $\cdot$ J. J. √. ÷, ć

ND, not detectable; NA. not analyzed *ND*, not detectable; *NA*. not analyzed

 $\rm ^a\rm{Values}$  present the ranges or mean expressed as mg/kg dry wt <sup>b</sup>Values present the ranges or mean expressed as mg/kg wet wt bValues present the ranges or mean expressed as mg/kg wet wt aValues present the ranges or mean expressed as mg/kg dry wt



<span id="page-9-0"></span>Fig. 3 Variation of heavy metal concentrations of fish in different sites



<span id="page-9-1"></span>**Fig. 4** Variation of heavy metal concentrations of fsh during summer and winter seasons

toxicity is higher in fsh species from the Pasur River. HM led is extremely toxic.

## **Spatiotemporal variation of metals in fsh (ANOVA analysis)**

There was no substantial variance in As  $(F = 0.032)$ ; *p* = 0.998), Cr (*F* = 0.084; *p* = 0.987), Cd (*F* = 0.023; *p*=0.999), and Pd (*F*=0.042; *p*=0.997) according to sites (*p* ˃ 0.05) (Fig. [3\)](#page-9-0). While As (*F*=4.354; *p*=0.04) and Cr  $(F = 5.422; p = 0.02)$  showed substantial fluctuation in respect of seasons ( $p \text{ }^{\textdegree}$  0.05). Cd ( $F = 2.917$ ;  $p = 0.09$ ) and Pd  $(F = 1.842; p = 0.08)$  exhibited lower variations in metal concentrations during summer and winter seasons (Fig. [4\)](#page-9-1).

#### **Heavy metal source identifcation in fsh**

To better understand the relationships between the HMs tested and to know the sources/origin of metals, the correlation matrix was used. The correlation among the elements in fish is shown in Table  $6$ . Associations between metals can serve as sources and ways for metals found in the fsh (Ahsan et al. [2018;](#page-12-2) Avigliano et al. [2015;](#page-12-29) Bebbington et al. [2012](#page-12-30); Bhuyan et al. [2016a](#page-12-10)). In the summer, there was a very good positive association in As Vs Pb. In the winter, As Vs Pb (*r*=0.852) had a very strong positive correlation, and Cd Vs Cr (*r*=0.725) and Pd Vs Cr (*r*=0.706) had a strong relationship. Pb and Cd had a moderately positive correlation  $(r=0.694)$  (Table [6\)](#page-9-2). The parameters were correlated and may have derived from the same sources in the study feld, as the correlation between the metals was found to be positive and signifcant (Abbasi et al. [2013;](#page-12-31) Bhuyan and Bakar [2017a](#page-12-1)). Strong connections between heavy metals suggest mutual dependence, similar infuence activity, and release from the same sources (Bhuyan et al. [2017;](#page-12-3) Jiang et al. [2014](#page-13-19)).

#### **Estimated daily intake (EDI)**

Adults in the study region who consume fsh species in their regular diet have their dietary exposure to HMs determined by estimated daily consumption. To estimate daily consumption, utilize the mean value of each harmful element and the individual ingestion frequency of that element (Santos et al. [2004\)](#page-13-3). We will fnd out how much HM is consumed on a daily basis based on the average daily consumption. Table [7](#page-10-0) displays the average daily intake of HMs from fsh eating in the current analysis. Fish consumption resulted in a lower total daily intake of HMs than the permissible value. Due to Bangladeshi people's low fsh consumption rate, the EDI is lesser than the allowable limit. As, Cr, Cd, and Pb had mean EDI concentrations of 0.42, 1.09, 0.28, and 0.86 mg/ day, respectively.

Despite the fact that Bangladesh's total EDI is low due to limited fsh intake, long-term ingesting of polluted fsh from the research area could have lethal health consequences



<span id="page-9-2"></span>**Table 6** Heavy metal correlation matrix  $(CM)$  in f during summer and winter

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<span id="page-10-0"></span>



a JECFA (2005)

<sup>b</sup>FAO/WHO [\(2004](#page-12-32))

c *PTDI*, provisional tolerable daily intake (60-kg body weight)

d *ESADDI*, estimated safe and adequate daily dietary intake

for the people of Bangladesh (Islam et al. [2016\)](#page-13-12). For the formulation of numerous regulatory criteria for fsh intake, periodic surveillance is required. In this scenario, the EDI was calculated using a 60 kg person eating 59.91 g fish/day dw basis. Table [7](#page-10-0) shows that the average EDI of HMs from fish is reported below than the reference amount (JECFA [1989,](#page-13-33) [2000](#page-13-34); NRC [1989](#page-13-23)) implying that these fsh species may not create an immediate risk, but continuous eating of these fish species may have a chance to create adverse risks to the consumers.

## **Risks of noncarcinogenic (THQ) and carcinogenic substances**

Table [8](#page-11-0) shows the THQ and carcinogenic risks of four HMs (As, Cr, Cd, and Pb) when consumed with HM contaminated fsh. Due to fsh consumption, the THQ values in As, Cr, Cd, and Pb were 60.66, 0.0046, 1.53, and 4.08, correspondingly. THQ values for As, Cd, and Pb are higher than the permissible value (1) that is considered important for human consumption, and these fsh should not be consumed (Table [8](#page-11-0)). Again, people are exposed to many noncarcinogenic risks from high exposure to toxic metals as a result of eating these fish. In fish species, the total THQ for individual metals ranged from 0.005 to 60.66. In this study, mainly As is a single metal that can pose a noncarcinogenic danger (THQ for As is  $> 1$ ) (Table [8\)](#page-11-0). Assuming a single metal THQ, the highest THQ was found in As in *Mystus vittatus* (10.89), followed by *Sillaginopsis panijus* (6.003). Each fsh's THQ and total individual factor (THQ  $> 1$ ) has the potential to generate noncarcinogenic hazards such as cardiovascular, kidney, nervous, and bone diseases, according to the current investigation. In fsh, As and Cd play the most important roles in HI (Table [8](#page-11-0)). Out of all fish species, *Mystus vittatus* (12.15) had the highest HI, followed by *Sillaginopsis panijus* (6.71) (Table [8](#page-11-0)). The HI for the fish declining in order of *Mystus vittatus* (12.15) > *Sillaginopsis panijus* (6.71)>*Polynemus paradiseus* (6.49)>*Otolithoides pama* (6.16) > *Glossogobius giuris* (6.09) > *Setipinna phasa* (5.08) > *Harpadon nehereus* (4.47) > *Gudusia chapra* (4.34) > *Pseudapocryptes elongatus* (4.14) > *Tenualosa ilisha* (3.73)*.*

Consumption of the studied fsh species in excess and on a regular basis could pose a number of noncarcinogenic risks. When individual metal THQ was considered, As had the maximum THQ due to its low RfD value relative to its amount, and As in Bangladesh's Pasur River could create major human health problems. The carcinogenic risk was calculated using As and Pb concentrations in several fsh. Depending on the exposure amount, As and Pb have both noncarcinogenic and cancer-posing effects. Obtained from animal experiments, Pb is a likely carcinogen and a possibly dangerous component classed as a carcinogen. Table [8](#page-11-0) illustrates the As and Cr cancer risk for people in the study location who consume HMs from fish species. For all fsh, the cancer hazard value for As was 0.81 to 2.45, and for Pb, it was 0.003 to 0.03. The reference value

	Target hazard quotients (THQs)				Hazard index (total)	<b>Target carcinogenic</b> $risk$ (TR)		
<b>Fish species</b>	As	$C_{r}$	C <sub>d</sub>	Pb		As <sup>a</sup>	Pb	
Tenualosa ilisha	3.583617	$2.72258E - 05$	0.028025	0.12079	3.732458	0.806314	0.003594	
Gudusia chapra	4.148435	0.000163954	0.034149	0.160445	4.343192	0.933398	0.004773	
Otolithoides pama	5.8359	0.000379031	0.03578	0.288196	6.160254	1.313077	0.008574	
Setipinna phasa	4.851379	0.000223664	0.036911	0.188717	5.07723	1.09156	0.005614	
Mystus vittatus	10.89064	0.00056595	0.263171	0.997815	12.15219	2.450394	0.029685	
Glossogobius giuris	5.620224	0.000534663	0.123248	0.345966	6.089972	1.26455	0.010292	
Harpadon nehereus	3.971367	0.000531668	0.135197	0.362627	4.469723	0.893558	0.010788	
Pseudapocryptes elongatus	3.704768	0.000431086	0.164253	0.267113	4.136565	0.833573	0.007947	
Polynemus paradiseus	5.994661	0.000443401	0.20842	0.285115	6.488639	1.348799	0.008482	
Sillaginopsis panijus	6.003315	0.000535329	0.202263	0.505669	6.711782	1.350746	0.015044	
Total	60.65754	0.004629312	1.529702	4.083637	66.27551	13.64795	0.121488	

<span id="page-11-0"></span>**Table 8** Consumption of Pasur River fsh has both noncarcinogenic and carcinogenic trace element risks

<sup>a</sup> Assuming 50% inorganic As in foods Uneyama et al. [\(2007](#page-14-7)), Oguri et al. ([2012\)](#page-13-25)

for cancer risk between  $10^{-6}$  and  $10^{-4}$  is the normal cancer risk number (Turkmen et al. [2009](#page-14-6); Uneyama et al. [2007\)](#page-14-7).

The cancer posing threat is insignifcant if the target hazard (TR) value is less than  $10^{-6}$ , and TR values greater than  $10^{-4}$  are not healthy for humans and may be responsible for causing cancer (USEPA [1989\)](#page-14-2). As was associated with a signifcantly greater risk of cancer when the TR value in this investigation was compared to the typical value  $(10^{-4})$ , and the risk for Pb was likewise higher than the standard value. People exposed to higher amounts of As and Pb from the present research fsh species are at risk for cancer for the rest of their lives. In this analysis, the risk of cancer was assessed based on the ingestion of fsh species. Other food sources are also available, but they are not included in this report.

# **Conclusion**

According to the fndings of this analysis, the majority of the fish species studied were found to be unsuitable for human consumption. As, Cr, Cd, and Pb contents in fsh samples were greater than the allowable limit. Since  $(THQ > 1.0)$ was confrmed as a posing health hazard that is not carcinogenic individually and collectively, the analyzed HMs were documented powerful enough to be assumed chronic. As and Pb risk levels were found above the recommended threshold based on cancer risk  $(10^{-4})$ . People who consume the infected fsh on a daily basis are in danger of developing chronic cancer in the long run, according to the study.

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**Author contribution** Mir Mohammad Ali (MMA) and Mohammad Lokman Ali (MLA) were theinvestigators of this study. They have drafted the preliminary manuscript. Md. Simul Bhuyan (MSB) and Md. Saiful Islam (MSI) were the supporting investigators who collected data. Md. Zillur Rahman (MZR) and Md. Wahidul Alam (MWA) validated the experimental and laboratory analysis. Monika Das (MD) and Sobnom Mustary (SM) supported the analysis of the data and compiled the manuscript with the frst and second authors. Md. Nazrul Islam (MNI) supported the analysis of the data for the manuscript and provided technical support to improve the manuscript. All authors read and approved the fnal manuscript.

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**Availability of data and materials** Though this research is not relevant with big data. On reasonable request, the frst and second authors of this manuscript will provide the datasets created and/or evaluated during this investigation.

## **Declarations**

**Ethics approval and consent to participate** In this investigation, there are no ethical problems, but we have included a statement certifcate on ethics approval and for experimental studies involving local fsh and others.

**Consent for publication** Not applicable.

**Competing interests** The authors declare no competing interests.

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