#### **RESEARCH ARTICLE**



# Spatiotemporal variation and toxicity of trace metals in commercially important fish 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., fish). The aim of this research was to learn more about the spatiotemporal variations and toxicity of heavy metals (As, Cr, Cd, and Pb) among fish 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 significant variation spatiotemporally  $(p^{>}0.05)$  except As and Cr showed substantial variation in terms of seasons (p < 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 (THO) value in individual fish 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 fish poses a chronic cancer risk to the people in the study field. According to the findings, the fish 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

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#### Introduction

HMs' high poisonousness, endurance, and bioavailability in the river water ecosystem are regarded as the most harmful chemical pollutants (Han et al. 2021; Lao et al. 2019; Ali et al. 2018; Bhuyan and Bakar 2017a; Islam et al. 2012). Since their increased trend in sediments, water, and fish, metal contamination has become a worldwide problem (Ahsan et al. 2018; Bhuyan et al. 2017, 2019; Burger and Gochfeld 2005; Santos et al. 2004). River water pollution has been exacerbated by rapid industrialization, haphazard urbanization, and indiscriminate population development (Bhuyan and Bakar 2017b; Hajeb et al. 2009; Islam et al. 2017). 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 refineries, and others. Moreover, some brickfields, 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 significantly affect the river. The river environment faces significant problems from overexploitation and the dumping of unprocessed textile wastewaters into the river (Bebbington et al. 1977; Singh and Kumar, 2017). Toxic metals discharged by these sources damage marine ecosystems (Bhuyan and Bakar 2017b; FAO/WHO 2002; Habibullah-Al-Mamun et al. 2017). Toxic metal pollution of fish due to intake of contaminated water and food (zooplankton and phytoplankton) are considered bioindicators of toxic metal contamination (Burger et al. 2002; Karunanidhi et al. 2017; Kuklina et al. 2014; Saha and Zaman 2013; Svobodova et al. 2004). 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; Islam et al. 2018). 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., fish) (Alhashemi et al. 2012; Habibullah-Al-Mamun et al. 2017; Islam et al. 2016). Adulteration of fish is a big growing issue because of the health risks involved with eating fish. 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; Bhuyan and Islam, 2016; Bhuyan et al. 2016a; Osman et al. 2016).

Fish consumption has increased over the world at the same time as concerns about its nutritious and medicinal benefits have developed. Fish are considered the source house of high protein, minerals, vitamins, and unsaturated lipid (Bhuyan et al. 2016a; DoF 2019; Hossen et al. 2018; Medeiros et al. 2012). Ingestion of poisonous

metal-contaminated fish, on the other hand, has been linked to a number of serious disorders. Chromium (Cr) causes anuria, nephritis, and severe lesions in infected fish, including kidney lesions (Proshad et al. 2018). Cadmium toxicity results in impaired reproductive ability, kidney illness, malignancies, hypertension, and hepatic dysfunction, among other things (Al-Busaidi et al. 2011; Ali et al. 2018; Bhuyan et al. 2016a). Lead poisoning damages the liver and produces renal failure (Bhuyan et al. 2019; Lee et al. 2011).

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; Hossen et al. 2018). Bangladeshis eat fish on a regular basis (annual ingesting 21.90 kg/person), and it is one of their key sources of protein (DoE, 2019). However, toxic metal concentrations in fish bodies enter into the human body (FAO/WHO, 2002), either directly or indirectly, and have an effect on human health (Islam et al. 2016). 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 scientific evidence of HM pollution in the fish 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 flows into Bangladesh's Bay of Bengal (Fig. 1). 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). 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 fisheries, 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; Dara and Mishra 2004; Hilal and Ismail 2008).





#### **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 fish is documented in Table 1. 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 fish species. Therefore, we have selected these fish 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 influential 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; Islam et al. 2015). Changes in HM concentrations in fish species are possible as a result of this (Ali et al. 2016; Bhuyan et al. 2016b).

#### 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 fish 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).

#### Fish species identification

The site has been used to identify fish 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)	IUCN status
Tenualosa ilisha	Hilsa shad	Phytoplankton, zooplank- ton, plants, mollusks, and crustaceans	Pelagic	33.12±5.40	$570.50 \pm 130.20$	LC
Gudusia chapra	Indian river shad	Phytoplankton, zooplank- ton, and crustaceans	Pelagic	$14.70 \pm 3.55$	$45.80 \pm 18.35$	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	$20.57 \pm 3.66$	$50.65 \pm 27.30$	NE
Lates calcarifer	Barramundi	Zooplankton, zoobenthos, mollusks, crustaceans, and invertebrates	Demersal	31.25±4.79	730.16±57.53	NE
Pampus argenteus	Silver pomfret	Zoobenthos, zooplankton, jellyfish, benthos, inver- tebrates, and mollusks	Benthopelagic	$21.42 \pm 3.52$	$388.50 \pm 21.35$	NE

Table 1 The ecological parameters and morphometric measurements of ten fish 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 fish species are presented in Fig. 2.

#### **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). 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; Bhuyan et al. 2016a; Rahman et al. 2020). 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 finish for 2 h (FAO, 2006; Ali et al. 2020; Bhuyan et al. 2016a; Rahman et al. 2020; Shaheen et al. 2015). 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 filtered with Whatman No. 41





Table 2Analytical conditionsfor employing AAS to quantifyheavy metals in a samplesolution

Elements	Wavelength (nm)	Slit (nm)	Lamp cur- rent (mA)	Mode	Calibration range (mg/L)	Detection limit (mg/L)
As	193.7	0.8	6.0	HG-AAS	0.0–20	0.0007
Cr	357.9	0.8	4.0	GF-AAS	0.0-32	0.0009
Cd	228.8	1.2	3.0	GF-AAS	0.0-1.2	0.00003
Pb	283.3	0.8	4.0	GF-AAS	0.0–40	0.0009

and leveled with 0.1 M  $HNO_3$  in a 50-ml volumetric flask. Finally, using GF-AAS, the samples were tested for Cr, Cd, and Pb (Ali et al. 2020; Rahman et al. 2020).

#### 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 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 fish 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; Lao et al. 2019). To ensure that the analytical process was

**Table 3** Concentrations of metals detected in the National Research Council Canada's Certified 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	Measured	Deviation	Recovery (%)
DORM-4		value	(70)	
As	$6.80 \pm 0.09$	$6.07 \pm 0.13$	8.07	89.19
Cr	$1.87 \pm 0.10$	$1.85 \pm 0.02$	0.65	99.09
Cd	$0.306 \pm 0.005$	$0.298 \pm 0.01$	1.66	97.06
Pb	$0.416\pm0.07$	$0.380 \pm 0.18$	6.28	91.35

correct, fish protein-approved reference substances for trace elements were applied. NRC (Canada) processed and supplied these fish samples (NRC 1989). The certified and observed values were found to be very close to each other. Table 3 demonstrates that the recorded certified 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):

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

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). Fish is consumed at a rate of 59.91 g per day on a fresh wt. basis (HIES 2011; Oguri et al. 2012; Kuklina et al. 2013).

# **Risk of noncarcinogenicity**

The risk-based concentration table was provided by the USEPA (2010). Region III was applied to estimate the non-carcinogenicity of fish. The THQ was used to calculate the noncarcinogenic danger of each metal from fish ingestion (USEPA 1989):

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

Total THQ (THQ) = THQ<sub>toxicant 1</sub> + THQ<sub>toxicant 2</sub> + ... ... ... + THQ<sub>toxicant n</sub>

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). 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; Reddy et al. 2008). There may be a health hazard if the THQ is  $\geq$  1, and relevant measures and safeguards should be undertaken (Islam et al. 2014).

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

HI =  $\Sigma$ TTHQ = 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). 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):

 $TR = \{(EFr \times ED \times FIR \times C \times CSF0)/(BW \times AT)\} \times 10^{-3}$ 

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; USEPA 2010).

# **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 fish

The levels of hazardous metals in twelve fish from the Pasur River are tabulated in Table 4. 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).

HMs were estimated from fish muscle since Bangladeshi people are more likely to eat fish muscles (edible component) than branchiae, liver, kidneys, sex gland, and other portions of the fish. Present HMs concentration in fish flesh compared with different national and international rivers (Table 5). Figures 3 and 4 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 seafood and fish species (USFDA 1993). For human health protection, in freshwater fish samples, the USEPA establishes a reference limit of 1.3 mg/kg (Burger et al. 2004). The

 $TTHQ(individualfood) = THQtoxicant1 + THQtoxicant2 + \dots + THQtoxicantn$ 

Table 4Concentrations of traceelements (mg/kg dw) in 12fish samples obtained from thePasur River in Bangladesh

Sites	Name of fish	Metals	(summe	r season)		Metals	(winter s	season)	
		As	Cr	Cd	Pd	As	Cr	Cd	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	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	0.290	0.775	3 1 1 7	3 717	0.991	0.132	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	11 ystus viitaitus	2.950	0.655	0.721	3 213	3 673	1.23	0.870	3 883
Site 5		2.842	0.73	0.765	3 347	3 71	0.92	0.801	3.84
Site 1		1.63	0.73	0.315	1.025	1 742	0.92	0.428	1 357
Site 2		1.635	0.7734	0.336	1.025	1.742	0.812	0.391	1.324
Site 3	Glassagahius giuris	1.556	0.754	0.305	1.011	1.674	0.012	0.371	1.05
Site 4	0103308001113 511113	1.550	0.725	0.303	1.1	1.883	0.871	0.532	1.05
Site 5		1 591	0.813	0.36	0.96	1 795	0.97	0.332	1.56
Site 1		1 218	0.759	0.305	1.024	1 230	0.92	0.320	1.50
Site 2		1.115	0.753	0.228	1.024	1.237	0.845	0.432	1.521
Site 3	Harpadon nehereus	1.115	0.732	0.220	1.023	1.127	0.045	0.42)	1.012
Site J	nurpudon nenereus	0.086	0.752	0.432	1.014	1.015	1.015	0.302	1.55
Site 5		1 359	0.762	0.452	0.98	1.015	0.76	0.375	1.72
Site 1		0.983	0.702	0.425	0.90	1.23	0.70	0.578	0.985
Site 7		1.026	0.634	0.425	0.000	1.250	0.098	0.578	1.12
Site 3	Psaudanocroptas alongatus	0.036	0.054	0.393	0.921	1.2	0.705	0.552	0.965
Site J	1 seudupocrypies eionguius	1.054	0.550	0.445	0.850	1.305	0.30	0.332	0.905
Site 5		0.05	0.612	0.525	0.012	1.25	0.734	0.415	0.075
Site 1		1 602	0.662	0.555	0.085	1.1.51	0.72	0.564	1.027
Site 7		1.095	0.002	0.075	0.965	2.015	0.008	0.304	1.027
Site 2	Polynamus paradisaus	1.505	0.309	0.303	0.034	2.013	0.723	0.712	1.104
Site J	Totynemus paraaiseus	1.00	0.723	0.71	1.042	1.975	0.048	0.573	1.057
Site 4		1 320	0.710	0.455	0.056	1.973	0.71	0.303	0.074
Site J		1.559	0.039	0.07	1 502	1.7/	0.202	0.131	0.7/4 2.014
Site 2		1.//1	0.733	0.334	1.323	1.000	0.031	0.003	2.014
Site 2	Sillaginonsis navijus	1.000	0.70	0.575	1.305	1.923	0.734	0.071	2.019
Site J	smagmopsis panijus	1.12	0.734	0.012	1.04	1.005	0.912	0.554	1.900
Site 4		1.004	0./1/	0.0	1.592	1.//5	0.804	0.034	1.890

Sites	Name of fish	Metals	(summe	r season)		Metals	(winter	season)	
		As	Cr	Cd	Pd	As	Cr	Cd	Pd
Site 5		1.763	0.834	0.471	1.455	1.85	0.883	0.723	2.245
Site 1		0.872	0.487	0.395	0.856	0.943	0.563	0.442	1.023
Site 2		0.79	0.534	0.432	0.872	1.135	0.562	0.465	1.132
Site 3	Lates calcarifer	0.932	0.513	0.365	0.791	0.856	0.534	0.398	0.975
Site 4		0.876	0.45	0.35	0.934	0.934	0.634	0.567	1.1
Site 5		0.822	0.442	0.41	0.812	0.85	0.527	0.375	0.954
Site 1		0.873	0.619	0.438	1.04	0.951	0.72	0.537	1.023
Site 2		0.921	0.623	0.423	0.982	0.856	0.715	0.645	1.13
Site 3	Pampus chinensis	0.845	0.61	0.335	0.975	0.912	0.8	0.486	0.865
Site 4		0.853	0.557	0.356	1.012	1.113	0.663	0.489	1.105
Site 5		0.9	0.642	0.534	1.075	0.953	0.723	0.52	1.015
Site 1		1.372	0.617	0.075	0.963	1.79	0.775	0.287	1.325
Site 2		1.44	0.761	0.056	0.978	1.61	0.723	0.241	1.112
Site 3	Cynoglossus arel	1.354	0.552	0.078	0.885	1.856	0.671	0.189	1.2
Site 4		1.317	0.71	0.113	1.015	1.882	0.853	0.35	1.55
Site 5		1.41	0.5	0.057	0.954	1.8	0.843	0.342	1.373

Table 4 (continued)

highest allowed As concentration in tissue residual was set at 2 mg/kg by the ANZFA (2011). According to the findings of this analysis, approximately 70% of fish 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 pollution due to higher As concentrations than MTC (Table 4).

The mean Cr concentration in the examined fish were fluctuated from 0.012 and 1.015 mg/kg dw between the summer and winter seasons (Table 4). Cr concentrations fluctuated 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 fish is lower in the developed world. According to the reference, Plaskett and Potter (1979) 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 (Amin et al. 2011; Raphael et al. 2011).

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). 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). *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). 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). Cd amounts are capped at 1 mg/kg in Spanish law (DoF 2019; 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 fish 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 0.49–3.89 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 fish 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), and the Spanish regulation sets a limit of 2 mg/kg for Pb (Bristi et all 2019; Ekeanyanwu et al. 2010). 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; Reddy et al. 2007), it was found that the current study had higher Pb concentrations, indicating Pb

Table 5   Comparison of trace elements in 1	ish muscles in various national and international rivers with th	he current study				
River	Studied fish species	As	Cr	Cd	Pb	References
Pasur River <sup>a</sup>	Tenualosa ilisha, Gudusia chapra, Otolithoides pama, Setipima phasa, Mystus vittatus, Glossogobius giuris, Harpadon nehereus, Pseudapocryptes elongatus, Polyne- mus paradiseus, and Sillaginopsis panijus	0.79–3.817	0.012–1.015	0.06–0.82	0.49–3.89	Present study
Kamaphuli River <sup>a</sup> (Bangladesh)	Mystus vittatus, Polynemus paradiseus, Sillaginopsis paniju, Lates calcarifer, Cynoglossus arel, and Pseudapocryptes elongates	0.85–5.38	0.55–1.54	0.29–1.17	0.62–6.95	Ali et al. (2020)
Dhaleshwari River <sup>a</sup> (Bangladesh)	Channa punctatus, Mastacembelus armatus, Mystus vit- tatus, Puntius puntio, Amblyceps mangois, and Metap- enaeus spinulatus	< 0.005-0.128	0.006-0.159	0.002-0.019	0.086-0.288	Ahsan et al. (2018)
Shitalakkhya River <sup>a</sup> (Bangladesh)	Chanda nama, Colis afasciatus, Mastacembelus Armatus, Channa punctatus, and Lepidocephalus guntea	NA	0.322-0.407	0.0035-0.0085	0.01-0.3159	Brishti et al. (2018)
Okumeshi River <sup>a</sup> (Nigeria)	Chrysichthys nigrodidatatus and Tilapia nilotica	NA	0.06	0.62	< 0.01	Ekeanyanwu et al. (2010)
Bangshi River <sup>a</sup> (Bangladesh)	Mastacembelus armatus, Gudusia chapra, Puntius ticto, Notopterus notopterus, Corica soboma, Setipinna phasa, Amblypharyngodon mola, Mystus vittatus, Heterponeustes fossilis, and Clupisoma pseudeutropius	1.97–6.24	0.47–2.07	0.09-0.87	1.76–10.27	Rahman et al. (2012)
Gumti River <sup>a</sup> (Bangladesh)	Corica soboma, Clupisoma pseudeutropius atherinoides, Gudusia chapra, Trypauchen vagina, and Mystus vitta-tus	NA	NA	NA	0.5-4.05	Amin et al. (2011)
Three urban rivers (Turag, Buriganga, and Shitalakha) <sup>b</sup> (Bangladesh)	Channa punctatus, Heteropneustes fossilis, and Trichogaster fasciata	0.091-0.53	0.75-4.8	0.007-0.13	0.052–2.7	Islam et al. (2015b)
Parana River Delta <sup>b</sup> (Argentina)	Lycengraulis grossidens and Odontesthes bonariensis	0.1 - 0.9	NA	NA	NA	Avigliano et al. (2016)
Red Sea <sup>a</sup> (Jordan)	Abudefduf saxatilis, Chaetodon austricus, C. fasciatus, Epinephelus fasciatus, Fistularia petimba, Kyphosus sp., Mugil sp., Mulloidichthys auriflama, Parupenus cyclosto- mus, Polysteganus coeruleopnctatus, and Thalassoma sp.	NA	1.0–10.3	0.5–2	1.5-8.3	Hilal and Ismail (2008)
Parangipettai <sup>a</sup> (India)	Upeneus vittatus, Anchovilla commersonii, Pomadasys maculates, Lutjanus adetii, and Ambassis commersoni	NA	0.415-1.562	0.004-0.114	0.062–1.569	Lakshmanan et al. (2009)
Aegean and Mediterranean Sea <sup>b</sup> (Turkey)	Pagellus acame, Trigla lyra, Serranus scriba, Scomber japonicus, Scyliorhimis canicula, Pomadasys incisus, Uranoscopus scaber, Liza ramado, Dicentrarchus labrax, Pagrus caeruleostictus, and Sphyraena viridensis	NA	0.07–1.48	<0.01-0.39	0.21–1.28	Turkmen et al. (2009)
Kichera River <sup>b</sup> (Russia) <sup>b</sup>	Rutilus rutilus, Perca fluviatilis, and Esox Lucius	NA	NA	< 0.01-0.10	0.07 - 0.30	Pintaeva et al. (2011)
Wadi Hanifaha (KSA) <sup>a</sup>	Tilapia nilotica	NA	0.23	0.008	0.039	Abdel-Baki et al. (2011)

ND, not detectable; NA. not analyzed

 $^{\mathrm{a}}\mathrm{Values}$  present the ranges or mean expressed as mg/kg dry wt

 $^{\rm b}Values$  present the ranges or mean expressed as mg/kg wet wt



Fig. 3 Variation of heavy metal concentrations of fish in different sites



Fig. 4 Variation of heavy metal concentrations of fish during summer and winter seasons

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

## Spatiotemporal variation of metals in fish (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  $(p \ge 0.05)$  (Fig. 3). While As (F = 4.354; p = 0.04) and Cr (F = 5.422; p = 0.02) showed substantial fluctuation in respect of seasons (p < 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).

#### Heavy metal source identification in fish

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 fish (Ahsan et al. 2018; Avigliano et al. 2015; Bebbington et al. 2012; Bhuyan et al. 2016a). 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). The parameters were correlated and may have derived from the same sources in the study field, as the correlation between the metals was found to be positive and significant (Abbasi et al. 2013; Bhuyan and Bakar 2017a). Strong connections between heavy metals suggest mutual dependence, similar influence activity, and release from the same sources (Bhuyan et al. 2017; Jiang et al. 2014).

#### Estimated daily intake (EDI)

Adults in the study region who consume fish 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). We will find out how much HM is consumed on a daily basis based on the average daily consumption. Table 7 displays the average daily intake of HMs from fish 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 fish 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 fish intake, long-term ingesting of polluted fish from the research area could have lethal health consequences

Table 6 Heavy metal   correlation matrix (CM) in fish   during summer and winter	CM in	summer				CM in wi	CM in winter				
		As	Cr	Cd	Pb	As	Cr	Cd	Pb		
	As	1				1					
	Cr	0.387	1			0.486	1				
	Cd	0.480	0.642	1		0.471	0.725	1			
	Pb	0.836	0.556	0.720	1	0.852	0.706	0.694	1		

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Table 7	Estimated daily intakes
(EDIs)	of heavy metals for
consum	ing Pasur River fish

Fish species	As	Cr	Cd	Pb
Tenualosa ilisha	0.064505	0.002450319	0.005044	0.025366
Gudusia chapra	0.074672	0.014755833	0.006147	0.033693
Otolithoides pama	0.105046	0.034112754	0.00644	0.060521
Setipinna phasa	0.087325	0.02012976	0.006644	0.03963
Mystus vittatus	0.196032	0.050935482	0.047371	0.209541
Glossogobius giuris	0.101164	0.048119712	0.022185	0.072653
Harpadon nehereus	0.071485	0.047850117	0.024335	0.076152
Pseudapocryptes elongatus	0.066686	0.038797716	0.029566	0.056094
Polynemus paradiseus	0.107904	0.039906051	0.037516	0.059874
Sillaginopsis panijus	0.10806	0.048179622	0.036407	0.10619
Total	0.416638104	1.091836	0.275346	0.857564
MTDI	$1.00^{a}$	1.00 <sup>b</sup>	2.00 <sup>a</sup>	0.30 <sup>a</sup>
Recommended daily dietary allowance (mg/day/person)	0.13 <sup>c</sup>	$0.05 - 2^{d}$	0.06 <sup>c</sup>	0.21 <sup>c</sup>
References	JECFA (1989)	NRC (1989)	JECFA (1989)	JECFA (2000)

<sup>a</sup>JECFA (2005)

<sup>b</sup>FAO/WHO (2004)

<sup>c</sup>PTDI, provisional tolerable daily intake (60-kg body weight)

<sup>d</sup>ESADDI, estimated safe and adequate daily dietary intake

for the people of Bangladesh (Islam et al. 2016). For the formulation of numerous regulatory criteria for fish 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 shows that the average EDI of HMs from fish is reported below than the reference amount (JECFA 1989, 2000; NRC 1989) implying that these fish 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 shows the THQ and carcinogenic risks of four HMs (As, Cr, Cd, and Pb) when consumed with HM contaminated fish. Due to fish 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 fish should not be consumed (Table 8). 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). Assuming a single metal THQ, the highest THQ was found in As in *Mystus vittatus* (10.89), followed by Sillaginopsis panijus (6.003). Each fish'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 fish, As and Cd play the most important roles in HI (Table 8). Out of all fish species, Mystus vittatus (12.15) had the highest HI, followed by Sillaginopsis panijus (6.71) (Table 8). 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 fish 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 fish. 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 illustrates the As and Cr cancer risk for people in the study location who consume HMs from fish species. For all fish, 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 haza	ard quotients (TH	Qs)		Hazard index (total)	Target carc risk (TR)	inogenic
Fish species	As	Cr	Cd	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

Table 8 Consumption of Pasur River fish has both noncarcinogenic and carcinogenic trace element risks

<sup>a</sup>Assuming 50% inorganic As in foods Uneyama et al. (2007),Oguri et al. (2012)

for cancer risk between  $10^{-6}$  and  $10^{-4}$  is the normal cancer risk number (Turkmen et al. 2009; Uneyama et al. 2007).

The cancer posing threat is insignificant 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). As was associated with a significantly 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 fish 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 fish species. Other food sources are also available, but they are not included in this report.

# Conclusion

According to the findings 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 fish samples were greater than the allowable limit. Since (THQ > 1.0) was confirmed 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 fish 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 first 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 final manuscript.

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Availability of data and materials Though this research is not relevant with big data. On reasonable request, the first 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 certificate on ethics approval and for experimental studies involving local fish and others.

Consent for publication Not applicable.

Competing interests The authors declare no competing interests.

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