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Trace element contamination and health risk assessment from consumption of six commonly consumed fish in a subtropical estuary

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ABSTRACT

Contamination of estuarine fish with trace elements (TE) is a growing concern globally. This study assessed six commonly consumed fish species (*Hilsa ilisha, Acanthopagrus latus, Mugil curema, Glossogobius giuris, Lates calcarifer, and Harpodon nehereus*) for trace elements (TE) pollution levels, and health risks related to consumption from a subtropical estuary along the Bay of Bengal Caost. Fish samples collected in dry (March 2020) and wet (August 2020) season from five sites were analyzed for As, Cd, Cr, Ni, and Pb using Flame Atomic Absorption Spectrometer (FAAS). The average concentration of As, Cd, Cr, Ni and Pb was 0.45 ± 0.23, 0.05 ± 0.03, 0.65 ± 0.36, 0.29 ± 0.16 and 1.35 ± 0.72 mg/kg which were within Food and Agricultural Organization (FAO)/ World Health Organization (WHO) reference values. The concentration of TE was ranked in a decreasing order of Pb > Cr > As > Ni > Cd. As and Pb content was the maximum in *L. calcarifer* and Cd, Cr and Ni in *H. nehereus*. Concentration of all the TE was varied significantly among the fish species except for Cd. Principal component analysis (PCA) and clustering analysis (CA) indicated a general distribution of the elements and they possibly originated from anthropogenic sources. The Average Pollution Load Index (APLI) ranged from 0.84 to 1.95, with *M. curema, G. giuris, L. calcarifer,* and *H. nehereus* exceeding the critical threshold of '1', indicating severe pollution. The estimated daily intake of 0.0002–0.0003 mg/kg/day for children consuming Pb contaminated fish was above the recommended limit. The non-carcinogenic risk (THQ) for As and Pb also exceeded acceptable thresholds, suggesting significant health risks for children. Although carcinogenic risk (CR) values were within acceptable limits for most metals, the potential risk remains, particularly for children. These findings are highlighting the varying contamination risks among fish species and the higher risk for children.

1. Introduction

Trace elements (TE) are ubiquitous in aquatic environments, where their pollution levels are rising at an alarming rate due to escalating anthropogenic pressures (Jewel et al., 2024; Aziz et al., 2023). Among these, arsenic (As), cadmium (Cd), chromium (Cr), nickel (Ni), and lead (Pb) are particularly hazardous, posing severe threats to ecosystems and public health (Senila, 2023; Fan et al., 2021). These contaminants originate from natural processes, such as mineral weathering, and human activities, including urbanization, industrial emissions, agricultural runoff, and land-use changes. Climate change exacerbates this issue by increasing the solubility and bioavailability of trace elements through elevated temperatures and accelerated chemical reactions, further amplifying their environmental and biological impacts (Lazăr

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et al., 2024; Rahman et al., 2012).

While TE are essential micronutrients at trace levels, excessive concentrations result in toxicity, persistence, and bioaccumulation, leading to significant ecological and health risks (Tao et al., 2012; Briffa et al., 2020). Their tendency to form complexes with organic and inorganic substances enables their entry into food webs, where they magnify through trophic levels, a phenomenon known as biomagnification (Angon et al., 2024). This process severely impacts aquatic organisms, particularly fish, which are highly susceptible to heavy metal exposure. Effects include altered blood parameters, erythrocyte abnormalities, genetic damage, and impaired reproductive functions (Mishra et al., 2019; Gárriz & Miranda, 2020). In early developmental stages, TE exposure has been linked to higher mortality rates, cardiac impairments, and skeletal deformities (Taslima et al., 2022). Fish, therefore, serve as critical bioindicators of TE pollution, reflecting environmental contamination levels (Ali et al., 2020). However, chronic human consumption of contaminated fish can lead to severe health issues, such as cardiovascular, neurological, and reproductive disorders (Kalipci et al., 2023; Töre et al., 2021; Aydın et al., 2023).

Bangladesh, one of the world's largest fish-consuming nations, meets an annual demand of 42.38 lakh metric tons of fish (Hossen et al., 2018; DoF, 2019). Coastal fisheries play a pivotal role in food security and economic growth, providing 60 % of the country's daily animal protein intake (FRSS, 2020). Given the high dependence on fish as a dietary staple, monitoring TE concentrations in commercially available fish is essential, as fish consumption is a primary exposure pathway for humans (Köse et al., 2020; Visnjic-Jeftic et al., 2010). Previous studies have documented TE contamination in Bangladeshi coastal fish species (Ahmed et al., 2019b; Rahman et al., 2019; Ali et al., 2019; Biswas et al., 2023; Ali et al., 2023). However, comprehensive assessments of human health risks associated with consuming contaminated fish remain scarce.

Despite significant progress in understanding TE dynamics in aquatic systems, gaps persist in methodological approaches. Early studies primarily focused on TE concentrations in water and sediment, often neglecting their bioaccumulation in aquatic organisms and subsequent health implications. Advanced indices such as the bioaccumulation factor (BAF) and health risk indices (HRI) have improved risk assessment but require greater standardization to ensure cross-study comparability. Various advanced protocols, including Inductively Coupled Plasma Mass Spectrometry (ICP-MS), Graphite Furnace Atomic Absorption (GFAA), and Flame Atomic Absorption Spectrometry (FAAS), have been employed in trace element studies, resulting in variability in reported findings. In regions where advanced technologies like ICP-MS are unavailable, FAAS serves as an effective alternative due to its simplicity, faster operation for single-element analysis, and minimal sample preparation requirements. Its cost-effectiveness, coupled with low operational and maintenance demands, makes FAAS a practical and reliable option for routine testing and environmental monitoring, particularly in laboratories with limited resources. It is established that variations in sampling protocols and analytical thresholds hinder the establishment of universal benchmarks. Using FAAS in this study advances the field by employing an integrative framework that links TE contamination with ecological and human health risks, contributing to a more holistic understanding of aquatic pollution in the study regions.

The Bakkhali River Estuary (BRE), situated on the southeastern coast of the Bay of Bengal, is a vital ecological and economic resource supporting local fishery production and livelihoods (Hena et al., 2007). The estuary's nutrient-rich waters drive high biological productivity, sustaining diverse aquatic life (Kamal & Khan, 2009). However, the BRE faces severe environmental degradation due to unregulated household, municipal, and industrial waste disposal, coupled with sedimentation and urban runoff (Hena et al., 2012). Industrial activities, such as boat repair, further contribute to the contamination of estuarine waters and sediments. While previous studies have documented heavy metal concentrations in the sediments (Siddique et al., 2012) and waters (Jahan et al., 2024) of the Bakkhali River Estuary (BRE), the human health risks associated with fish consumption from this region remain largely unexplored. This study addresses this critical knowledge gap by quantifying trace element (As, Cd, Cr, Ni, Pb) concentrations in six commercially important fish species from the BRE. The objectives of this study were: to quantify trace element concentrations (As, Cd, Cr, Ni, Pb) in six commercially important fish species from the Bakkhali River Estuary (BRE), to evaluate the pollution load in these fish species to determine the extent of contamination, to assess the associated health risks for adults and children using advanced analytical methodologies, to provide a novel and comprehensive assessment of fish safety with implications for public health and environmental management. This research not only contributes to the existing body of knowledge on TE contamination but also informs public health policies and sustainable fisheries management.

2. Materials and methods

2.1. Description of the study sites and fish species

The study was carried out at the Bakkhali River estuary, located in Cox's bazar district, Bangladesh. Five sampling locations were chosen to collect fish samples (Fig. 1). S1 ($21^0$28 30.91 N, $91^0$58 21.32 E) and S2 ($21^0$27 05.84 N, $91^0$58 09.24 E) were near the estuary mouth and had most industrial zones. Thus, industrial effluents damage these two areas considerably. Sites S3 ($21^0$26 47.39 N, $91^0$59 23.46 E) and S4 ($21^0$27 01.12 N, $91^0$59 59.47 E) were located upstream of the estuary and significantly impacted by septic systems, household waste, and market refuse. Site S5 ($21^0$26 29.97 N, $92^0$00 43.31 E) was distant from the estuary mouth and impacted by agriculture. Six highly consumed fish species, namely *Hilsa ilisha*, *Acanthopagrus latus*, *Mugil curema*, *Glossogobius giuris*, *Lates calcarifer* and *Harpodon nehereus* were examined for trace element analysis. Fish species were selected on the basis of its consumer preferences. These species are highly priced and demanded in the market (Sunny et al., 2023). Furthermore, these fish species are reported to contain essential macro- and micronutrients which are beneficial to promote human health (Rifat et al., 2023). *Hilsa ilisha* is the national fish of Bangladesh that contributes around 1 % to the GDP of Bangladesh (Dutta et al., 2021). Furthermore, *A. latus, M. curema, G. giuris, L. calcarifer* and *H. nehereus* form an important part of the coastal fisheries production. Sampled fish species were also processed through drying which have demand in domestic and international markets due to their unique taste and flavor, and their demands are increasing daily in the international markets.

2.2. Fish sampling and preparation for analysis

Fish sample was collected in two different seasons, one in dry (March 2020) and another in wet (August 2020). On each sampling date, desired number of fish species was collected from the local fisherman. A total of 60 fish samples of six species from each individual were collected from the five sampling sites in two seasons (2 season × 5 sites × 6 individuals). Collected fish specimens were wrapped in labeled polyethylene zipper bag (Ziploc, USA) and delivered to the laboratory in an icebox for further analysis (Mohiuddin et al., 2022). In the laboratory, edible muscle from the dorsal side of fish was removed and homogenized using a food processor. The homogenized tissue was subsampled and 1-g aliquots of samples were stored in polyethylene bags lined with aluminum foil at −20°C for the digestion.

2.3. Digestion of the samples

The homogenized tissue sample was placed in a microwave digestion tube along with 5 mL of HNO_3 (65 % pure, Merck, Germany) and 1 mL of H_2O_2 (30 % pure, Merck, Germany). A reagent blank was also prepared which contained only the acids. The digestion was completed in a microwave digestion system (Berghof, Germany). In microwave

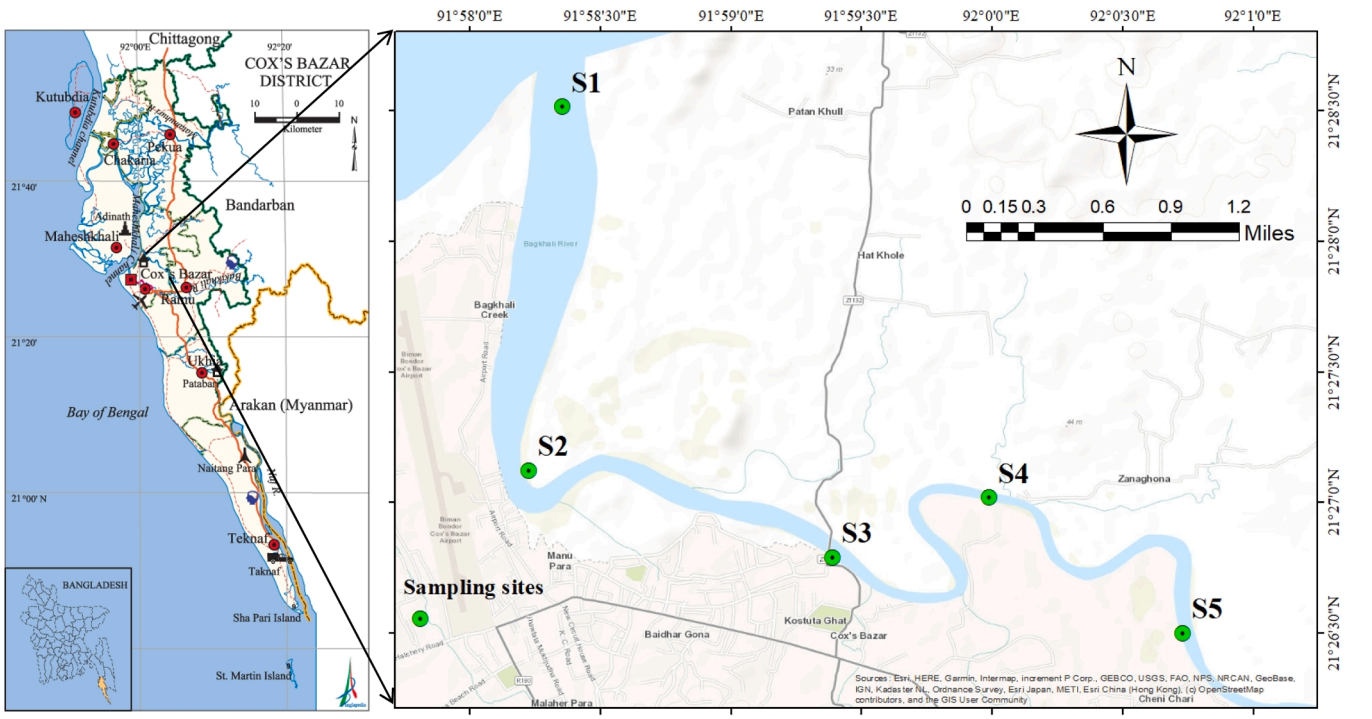

Fig. 1. Location of the sampling sites in the Bakkhali River estuary, Cox's bazar, Bangladesh.

digestion system, the samples were heated in two rounds. In first round, the samples were digested once in low power (200 W) for 1 min and the containers were cooled in an ice-bath. In second round, the samples were digested at high power (1000 W) for six times for a period of 30 sec. After digestion, the samples were cooled to room temperature and diluted upto 10 mL by adding ultrapure deionized water. The diluted samples were transferred to labeled polypropylene falcon tubes and stored at 4°C (AOAC, 2005). A blank sample was prepared using the same digestion protocol as the fish samples, but without the addition of any tissue. This ensured that any contamination from the reagents or equipment during the digestion process could be identified and accounted for. The triplicates were also performed for each sample analysis.

2.4. Analysis of TE

Analysis of TE was conducted following the procedure described by Jahan et al. (2024). The preserved samples were passed through a 0.45 μm Whatmann-42 filter paper using a filtration unit (Rocker-300, Taiwan). The concentrations of As, Cd, Cr, Ni, and Pb were measured with the Flame Atomic Absorption Spectrometer (FAAS) (Shimadzu, AA-6800). Each sample was analyzed at least three times for TE using FAAS, and the results were reported as mg/kg of wet weight (ww) of tissue. The analytical parameters, including detection limits (LOD) and quantification limits (LOQ) for TE in fish samples using FAAS are detailed in Supplementary Table 1.

2.5. Quality assurance and quality control

Ensuring precision in the tests involved conducting all procedures on three replicates, with the average result being exclusively used. Equipment was thoroughly pre-cleaned with 10 % HNO_3 nitric acid for a minimum of 24 hours. Ultrapure deionized water (resistivity 18.2 MΩ·cm), with a conductivity of 0.05 μS/cm, produced by the Direct-Q3 Ultrapure Water Systems from Millipore (Molsheim, France), was used for sample preparation and standard dilution. Throughout the study, only analytical-reagent-grade chemicals were employed. Blank reagents and Certified DORM-4 from the National Research Council (NRC) Canada were utilized with each batch of samples, including one blank and one standard sample for every ten samples. Blanks were prepared following the same protocol, using reagents only. The results demonstrated a strong agreement between certified and observed values, with recovery rates ranging from 90.51 % to 98.67 % (Supplementary Table 2).

2.6. Assessment of pollution load index

The average pollution load index (APLI) of the analyzed TE was used to assess the harmful impact of these metals on the fish. The APLI was calculated based on the formula provided by Liu *et al.* (2021) as follows

$$APLI = \frac{1}{n}\sum_{n=1}^{n} Ci/Si$$

Here, *Ci* represents the concentration of TE, *Si* is the maximum permissible concentration for these metals, and n denotes the number of TE. According to Chakraborty et al. (2022), the APLI is classified as follows: < 0.1 (unpolluted), 0.1–0.2 (micro polluted), 0.2–0.5 (lightly polluted), 0.5–0.7 (moderately polluted), 0.7–1.0 (heavily polluted), and > 1.0 (seriously polluted).

2.7. Health risk assessment

2.7.1. Estimated daily intake

The estimated daily intake (EDI) of TE in the fish species studied was calculated by considering the maximum concentration of TE in the fish, along with the average daily consumption and body weight of both adults and children residing in Bangladesh. The EDI for marine fish was computed using the formula provided by Chakraborty et al. (2022) as follows

$$EDI = \frac{(E_f \times E_D \times F_{IR} \times C_M)}{(B_W \times T_A)} \times 10^{-3}$$

where*Ef* represents the exposure frequency (365 days/year), E_D denotes

the exposure duration (72.3 and 15 years for adult and children, respectively (BBS, 2022), F_{IR} is the average fish intake (67.8 and 52.5 g/person/day for adult and children, respectively (HIES, 2022), C_M indicates the TE content (mg/kg fresh weight), B_W is the average body weight for an adult and children (60 kg and 15 kg), and T_A is the total exposure time (365 days multiplied by the number of exposure years, which is 72.3 and 15 years for adult and children, respectively in this scenario). To assess the safety of long-term exposure to metals in food, water, and other sources, the Tolerable Daily Intake (TDI) was employed. TDI represents a Health-Based Guidance Value (HBGV) established by leading organizations, including the World Health Organization (WHO), the European Food Safety Authority (EFSA), and the U.S. Environmental Protection Agency (USEPA). In this study, the toxicological reference values (HBGVs) or reference points (BMDLs) used for each element, along with the corresponding establishing bodies, are provided in Supplementary Table S3.

2.7.2. Non-carcinogenic risk assessment

The non-carcinogenic risk of TE was evaluated using the target hazard quotient (THQ). The THQ was calculated by comparing the oral reference dose to the estimated daily intake (EDI) for both adults and children, following the formula provided by USEPA (2016)

$$THQ = \frac{EDI}{RfD}$$

where RfD, the oral reference dose (mg/kg/day). RfDs were determined to be 0.0003, 0.001, 1.5, 0.02 and 0.0035 mg/kg body weight/day for As, Cd, Cr, Ni and Pb, respectively. To retain the accuracy of the RfD in determining THQ, it was assumed that 50 % As ions are in the inorganic form (USEPA, 2023). If the THQ is < 1, fish is considered safe for non-carcinogenic risk, whereas THQ > 1 implies a significant risk of harmful health effects.

2.7.3. The hazard Index

The hazard index (HI) represents the combined non-carcinogenic health risk associated with consuming contaminated fish. The benefit of HI is that, even if the individual THQs for the TE are less than 1, the overall impact of consumption might still lead to negative health consequences. Adverse non-cancer health impacts may occur when the Hazard Index (HI) exceeds 1. The HI was calculated using the following formula:

$$HI(HMs) = THQ(M1) + THQ(M2) + THQ(M3) + \cdots\cdots + THQ(Mn)$$

where M1, M2, M3 and Mn are the number of TE.

2.7.4. Carcinogenic risk assessment

The cancer risk estimate involves calculating the likelihood of developing cancer over a lifetime due to exposure to TE (USEPA, 2011). The target carcinogenic risk (TCR) for both adults and children was determined by multiplying the cancer slope factor (CSF) with the estimated daily intake (EDI), using the following formula:

$$TCR = EDI \times CSF_0$$

where slope factors of oral cancer is expressed in mg/kg/day (*CSFo*). The *CSFo* values for As, Cd, Cr, Ni and Pb were 1.5, 15, 0.5, 1.7 and 8.5×10^{-3} (mg/kg/day), respectively (USEPA, 2016).

2.8. Statistical analysis

All relevant data were coded and reported as means with standard deviations. Parametric assumptions were checked, and data normality was evaluated using the Shapiro-Wilk and Kolmogorov-Smirnov tests. One-way ANOVA was used to analyze significant differences in TE content among fish species, with post-hoc comparisons conducted using the Tukey multiple comparison test ($P < 0.05$). As the preliminary normality test showed the normal distribution of the data, multivariate analyses were performed with the untransformed data. The Pearson correlation coefficient examined relationships between TE in fish species. Principal component analysis (PCA) identified sources of TE in the fish. Before conducting the PCA analysis, the data was checked for the assumptions of PCA. Correlation matrix of the variables showed high correlation among them and most of the values were greater than 0.3 which signifies the use of PCA for examining the relationship among the variables for their source identification (Supplementary Table 3). Number of principal component (PC) to be described was selected based on the eigenvalue. PC with an eigenvalue greater than 1 was considered. A scree plot visually aided for determining the appropriate number of PCs (Supplementary Figure 1). A two-way hierarchical cluster analysis (heat map) with Ward's linkage method and Euclidean distance was also used to assess the distribution of TE. All statistical analyses were performed using Origin (Pro), Version 2024b, OriginLab Corporation, Northampton, MA, USA.

3. Results

3.1. Concentration of TE

Five TE were evaluated in six different fish species that were caught in the estuary of the Bakkahli River (Supplementary Table 4). The average concentration of As, Cd, Cr, Ni and Pb was 0.45 ± 0.23, 0.05 ± 0.03, 0.65 ± 0.36, 0.29 ± 0.16 and 1.35 ± 0.72 mg/kg and arranged in a descending order of Pb > Cr > As > Ni > Cd (Fig. 2). The level of the analyzed TE in six fish species are presented in Fig. 3. Concentrations of As ranged from 0.290 to 0.686 mg/kg, with significantly ($P < 0.05$) highest amount obtained in *L. calcarifer*. Concentrations of Cd ranged from 0.046 to 0.060 mg/kg. Cr concentration was the highest in *H. nehereus* and lowest in *H. ilisha*. Concentrations of Ni ranged from 0.162 to 0.439 mg/kg, with significantly ($P < 0.05$) highest amount recorded in *H. nehereus*. Pb was the most prevalent TE found in the fishes, whereas the maximum concentration of Pb (1.970 mg/kg) was detected in *H. nehereus* and the lowest concentration of 0.789 mg/kg was found in *H. ilisha*.

3.2. Correlation Analysis and Source apportionment of TE

The correlation matrix of the analyzed TE in the studied fish species illustrates the interactions and potential common sources of the TE (Fig. 4). Statistically significant correlations between TE ($P < 0.05$)

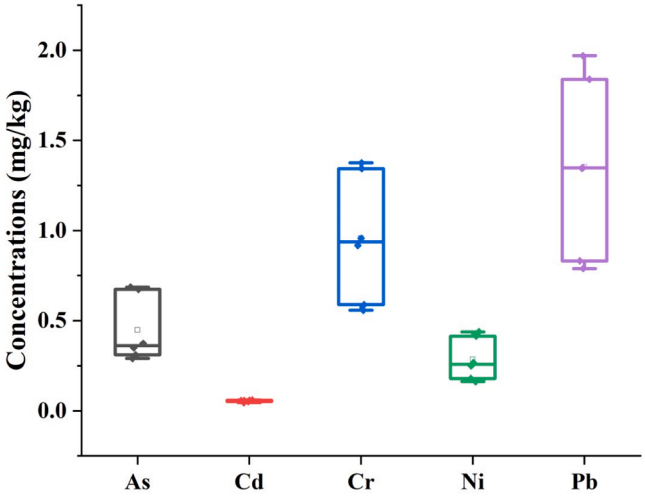

Fig. 2. The average concentration of TE in the studied fishes of Bakkhali River estuary.

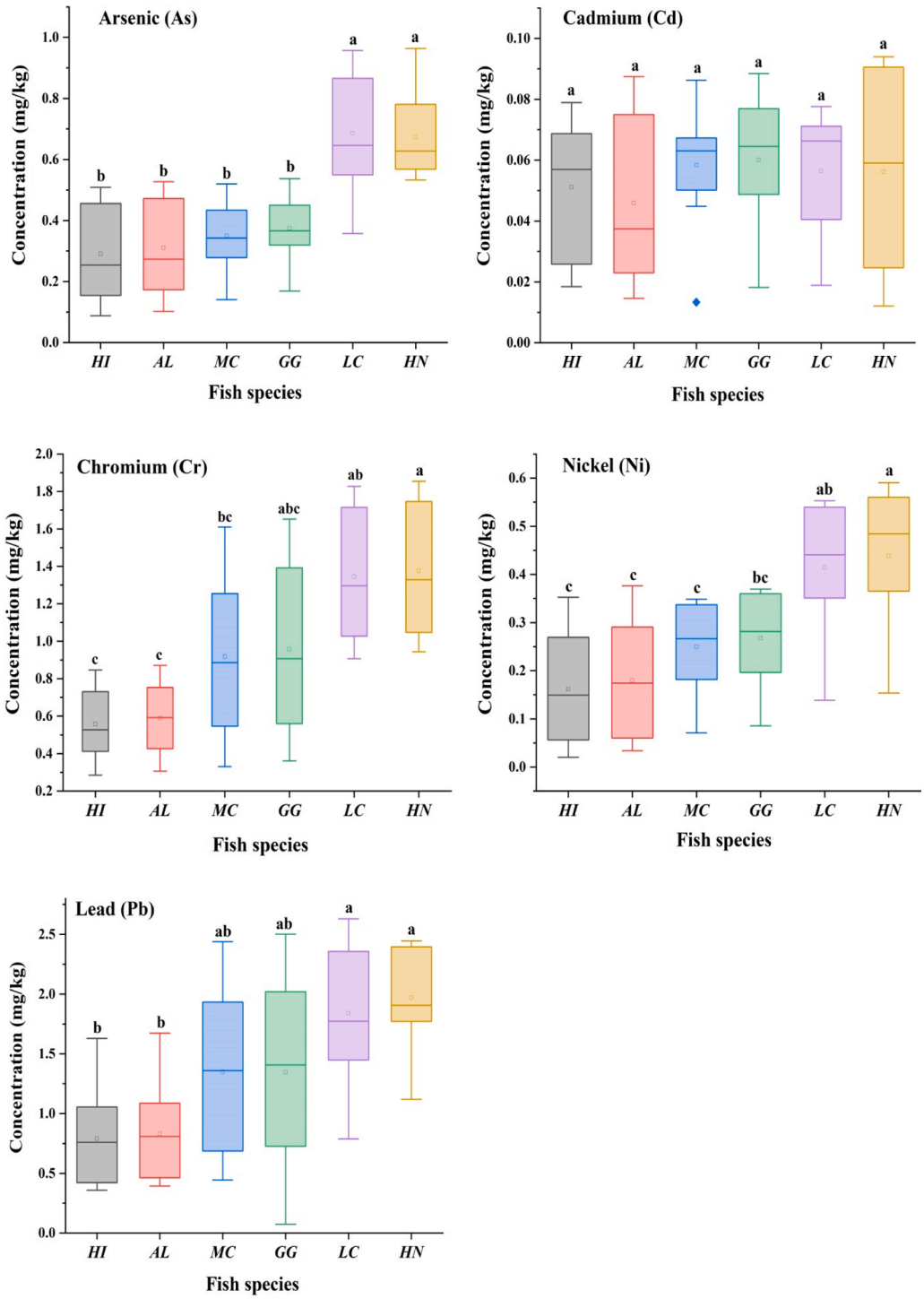

Fig. 3. Comparison of TE content in different fish species.

suggest a shared source of contamination. Positive correlations were notably observed among As-Cr, As-Ni, As-Pb, Cr-Ni, Cr-Pb, Ni-Cr, and Ni-Pb. A scree plot generated by PCA defined two PC with the eigenvalue greater than 1. This two PC cumulatively accounted for 86.71 % (PC 1 = 66.06 % and PC 2 = 20.65 %). In the PCA biplot, PC 1 is consisted of As and Ni and accounted for 66.06 % of the variance. While PC 2 consisted of Cd, Cr and Pb and the variance are 20.65 % (Fig. 5). The two-way hierarchical cluster heat map illustrated the relationships between TE and fish species (Fig. 6). The vertical dendrogram formed two clusters: Cluster 1 included Cr and Pb, while Cluster 2 included As, Ni, and Cd, aligning with PCA results. TE within the same cluster may share similar properties or be closely linked. In the horizontal dendrogram, two clusters were identified, with *L. calcarifer* and *H. nehereus* in Cluster 1, and other fish species in Cluster 2. Fish in Cluster 1 generally had higher levels of TE compared to those in Cluster 2.

3.3. Average pollution load index

The average pollution index is an indicator that measures the total contamination of TE in the fishes being researched. The APLI value of the studied fishes was ranged from 0.84 to 1.95. The distribution arrangement of APLI in the fishes follows a descending order of:

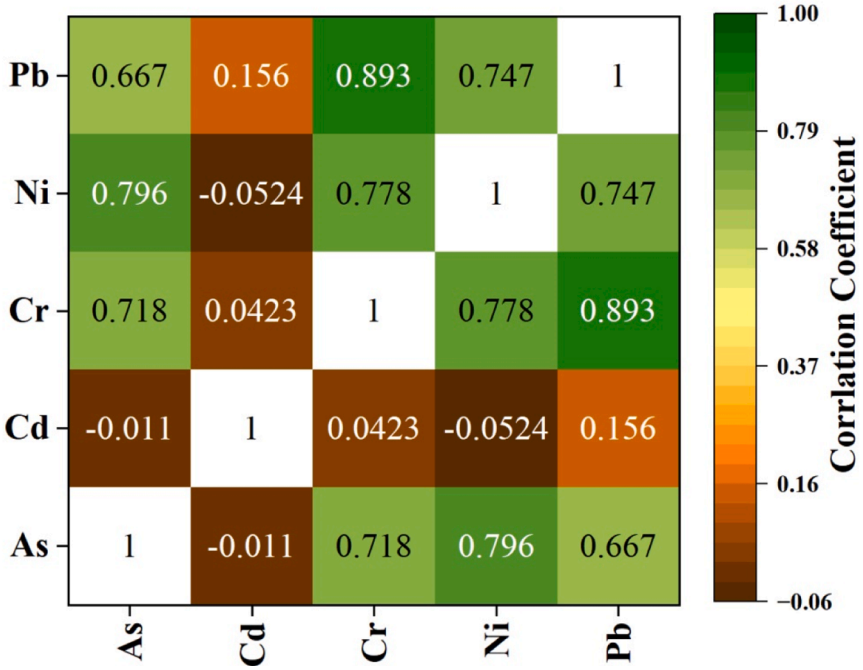

Fig. 4. Correlation-coefficient of TE in the studied fish species.

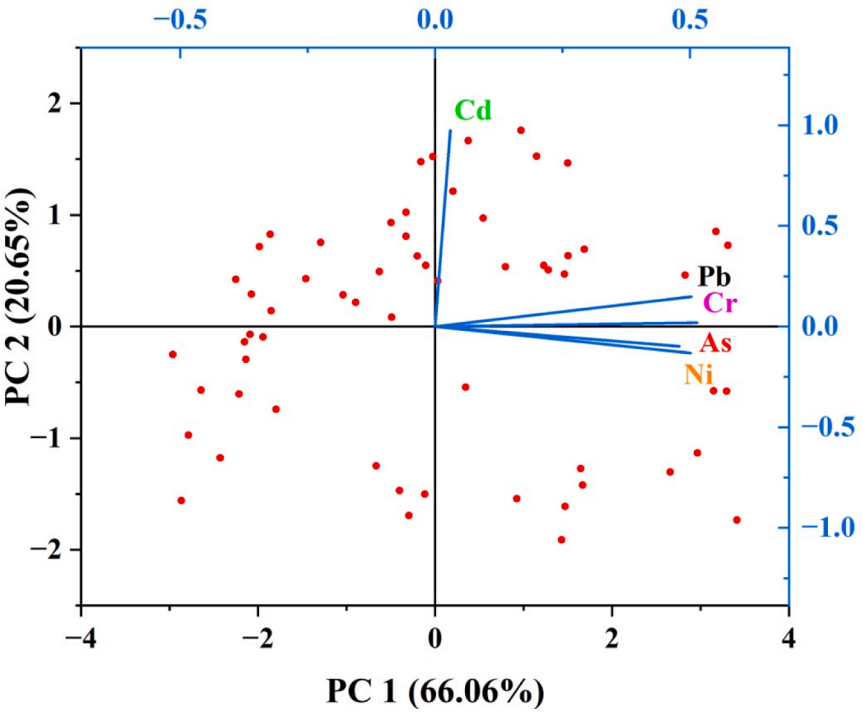

Fig. 5. Principal component analysis of TE in the studied fish species.

H. nehereus > *L. calcarifer* > *G. giuris* > *M. curema* > *A. latus* > *H. ilisha* (Fig. 7). According to the APLI value *H. ilisha* and *A. latus* indicating heavy pollution by the studied metals. However, APLI value of *M. curema*, *G. giuris*, *L. calcarifer* and *H. nehereus* exceeded the critical value of > 1 and indicating serious pollution of these fishes with the TE.

3.4. Health risk assessment

3.4.1. Estimated daily intake

Estimated Daily Intake (EDI) values (Table 1) for TE in six fish species for both adults and children. The average EDI values for Cr and Pb were higher in children, with As (for children) and Pb (both adult and children) contaminated fish leading to EDI values that exceeded recommended limits. In contrast, the EDI values for other TE were below the daily intake reference values. The average EDI of TE was ranked as Pb > Cr > As > Ni > Cd for both adult and children. **Non-carcinogenic risk assessment**

THQ of the studied TE are showed in Table 2. Compared to adults, children's THQ scores were significantly higher in this study. The estimated mean THQs were ranked in a decreasing order of As > Pb > Cd

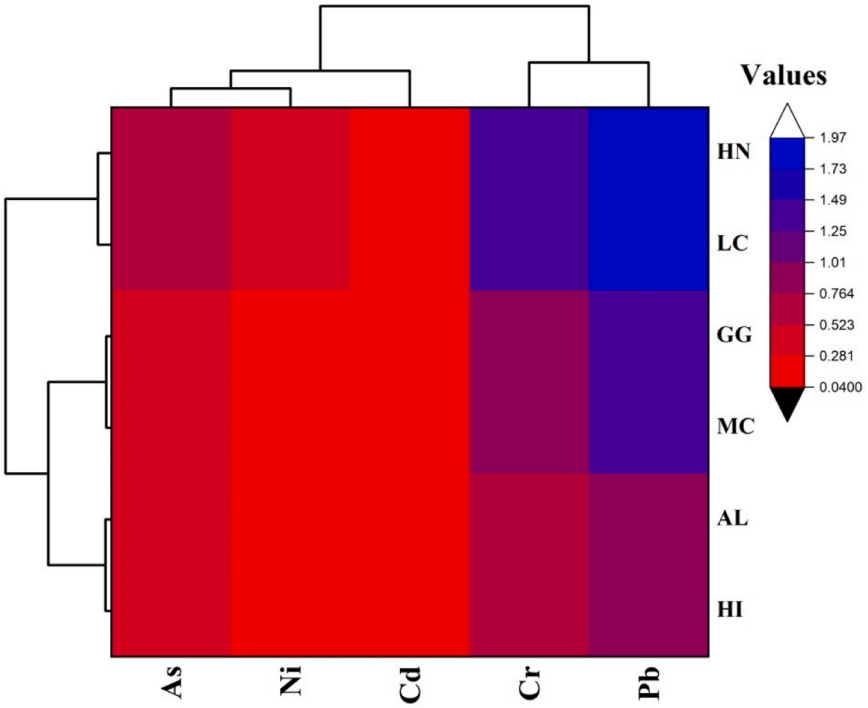

Fig. 6. Cluster analysis of TE in the studied fish species.

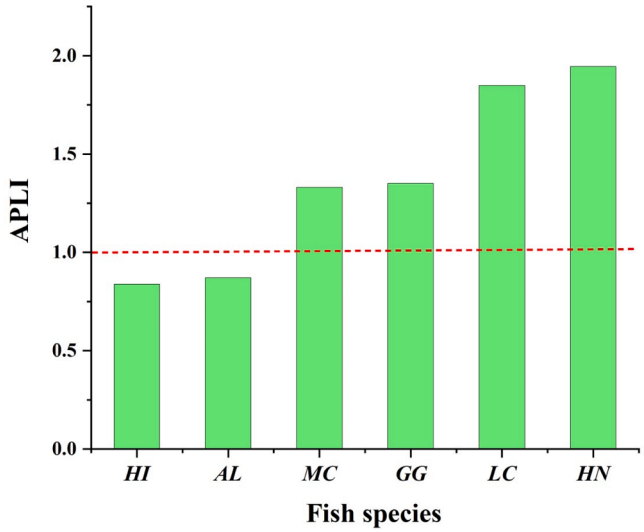

Fig. 7. Average pollution load index (APLI) for TE in the studied fish species.

> Ni > Cr. For adults, THQ values for all TE, except As, were well below the threshold of 1. However, children exhibited THQ values exceeding the acceptable limit (THQ > 1) for As and Pb, indicating significant non-carcinogenic health risks for them. The Hazard Index (HI) for both adults and children, from consuming the studied fish species, was significantly above the threshold limit of 1, suggesting potential non-carcinogenic health hazards. Arsenic was identified as the major contributor to the HI, accounting for 76.57 %, followed by Pb (21.34 %), Cd (2.87 %), Ni (0.73 %), and Cr (0.03 %).

3.4.2. *Carcinogenic risk assessment*

Table 3 represents the CR linked with the ingestion of the studied fishes exposed to TE. For adult, mean CR value for all the TE was within the acceptable limit of 10^{-4}–10^{-6} accept for Cd (1.24E-03) indicating no considerable cancer risk from the contamination of these TE. However, in children CR value of As, Cd, Cr and Ni levels were acceded the safe limit and suggesting that there might be a significant concern for potential carcinogenic risk. In all these cases, CR indicating more vulnerability of child compared to adults due to the consumption of TE exposed fishes.

Table 1
Estimated daily intake (EDI) of trace elements for adults and children with reference daily intake.

Species	EDI									
	As		Cd		Cr		Ni		Pb	
	Adult	Child	Adult	Child	Adult	Child	Adult	Child	Adult	Child
Hilsa ilisha	0.0005	0.0018	0.0001	0.0003	0.0008	0.0030	0.0003	0.0012	0.0016	0.0057
Acanthopagrus latus	0.0005	0.0018	0.0001	0.0003	0.0008	0.0030	0.0004	0.0013	0.0016	0.0059
Mugil curema	0.0005	0.0018	0.0001	0.0003	0.0016	0.0056	0.0003	0.0012	0.0024	0.0085
Glossogobius giuris	0.0005	0.0019	0.0001	0.0003	0.0016	0.0058	0.0004	0.0013	0.0024	0.0088
Lates calcarifer	0.0009	0.0034	0.0001	0.0003	0.0018	0.0064	0.0005	0.0019	0.0025	0.0092
Harpodon nehereus	0.0009	0.0034	0.0001	0.0003	0.0018	0.0065	0.0006	0.0021	0.0024	0.0086
Mean	0.0006	0.0023	0.0001	0.0003	0.0014	0.0051	0.0004	0.0015	0.0021	0.0078
* *TDI (mg/kg/day)	0.002		0.001		0.300		0.012		0.002	

* *Tolerable Daily Intake, Varol and Sünbül (2018)

Table 2
Target hazard quotient (THQ) of trace elements for adults and children.

Species	THQ										HI	
	As		Cd		Cr		Ni		Pb			
	Adult	Child	Adult	Child	Adult	Child	Adult	Child	Adult	Child	Adult	Child
Hilsa ilisha	1.642	5.934	0.076	0.276	0.001	0.002	0.017	0.062	0.451	1.628	2.19	7.90
Acanthopagrus latus	1.703	6.153	0.085	0.306	0.001	0.002	0.018	0.066	0.463	1.674	2.27	8.20
Mugil curema	1.678	6.062	0.084	0.302	0.001	0.004	0.017	0.061	0.675	2.439	2.45	8.87
Glossogobius giuris	1.735	6.270	0.086	0.310	0.001	0.004	0.018	0.065	0.692	2.502	2.53	9.15
Lates calcarifer	3.090	11.167	0.075	0.272	0.001	0.004	0.027	0.097	0.728	2.629	3.92	14.17
Harpodon nehereus	3.112	11.244	0.091	0.329	0.001	0.004	0.029	0.103	0.677	2.445	3.91	14.13
Mean	2.160	7.805	0.083	0.299	0.001	0.003	0.021	0.076	0.614	2.220	2.88	10.40

Table 3
Carcinogenic risk (CR) of trace elements for adults and children.

Species	CR									
	As		Cd		Cr		Ni		Pb	
	Adult	Child	Adult	Child	Adult	Child	Adult	Child	Adult	Child
Hilsa ilisha	7.39E−04	2.67E−03	1.15E−03	4.14E−03	4.10E−04	1.48E−03	5.80E−04	2.10E−03	1.34E−05	4.84E−05
Acanthopagrus latus	7.66E−04	2.77E−03	1.27E−03	4.59E−03	4.22E−04	1.52E−03	6.19E−04	2.24E−03	1.38E−05	4.98E−05
Mugil curema	7.55E−04	2.73E−03	1.25E−03	4.53E−03	7.80E−04	2.82E−03	5.74E−04	2.07E−03	2.01E−05	7.26E−05
Glossogobius giuris	7.81E−04	2.82E−03	1.28E−03	4.64E−03	8.00E−04	2.89E−03	6.08E−04	2.20E−03	2.06E−05	7.44E−05
Lates calcarifer	1.39E−03	5.03E−03	1.13E−03	4.07E−03	8.85E−04	3.20E−03	9.11E−04	3.29E−03	2.16E−05	7.82E−05
Harpodon nehereus	1.40E−03	5.06E−03	1.37E−03	4.94E−03	8.98E−04	3.24E−03	9.72E−04	3.51E−03	2.01E−05	7.27E−05
Mean	9.72E−04	3.51E−03	1.24E−03	4.49E−03	6.99E−04	2.53E−03	7.11E−04	2.57E−03	1.83E−05	6.60E−05

4. Discussion

Trace elements level is considered one of the most important markers which provide essential information about fish health and their surrounding environment (Padrilah et al., 2018). In this study, the average concentrations of TE in fish from the BRE ranked as Pb > Cr > As > Ni > Cd. This ranking contrasts with the findings of Ali et al. (2020), who reported a different order in the Karnaphuli River: As > Pb > Cr > Cd. The highest levels of TE were recorded in *H. nehereus*, followed by *L. calcarifer, G. giuris, M. curema, A. latus,* and *H. ilisha*. Noman et al. (2022) observed that demersal fish typically showed higher TE concentrations compared to benthopelagic and pelagic species, likely due to

Table 4
Comparison of trace elements in fish in the present study to other studies and reference value.

River and location	Fish species	As	Cd	Cr	Ni	Pb	Ref.
Bakkhali River estuary, Bangladesh	*T. ilisha*, *A. latus*, *M. curema*, *G. giuris*, *L. calcarifer* and *H. nehereus*	0.29 – 0.69	0.05 –0.06	0.56 – 1.38	0.16 – 0.44	0.79 – 1.97	**Present study**
Rupsha River basin, Bangladesh	*T. ilisha*, *Gudusia chapra*, *Otolithoides pama*, *Setipinna phasa*, *Mystus vittatus*, *G. giuris*, and *Pseudeutropius atherinoides*	1.63	0.44	0.58	–	1.38	Ali et al., (2023)
Coastal area, Bangladesh	*Liza parsia*, *O. pama*, *Escualosa thoracata*, *Hilsa ilisha*, *Nemipterus virgatus*, *Polynemus paradiseus*, *Sillaginopsis panijus*, *Pampus chinensis*, *M. cephalus* and *Thunnus thynnus*	2.64	0.09	2.89	–	9.09	Biswas et al. (2023)
Karnaphuli River Estuary, Bangladesh	*O. pama*, *Awaous grammepomus*, *S. phasa*, *P. paradiseus*, *Cirrhinus cirrhosus*	NA	NA	0.059–1.605	0.023–0.355	0.025–0.385	Mohiuddin et al., (2022)
Meghna River, Bangladesh	*Apocryptes bato*, *P. chinensis*, *L. parsia*, *M. cephalus*, *Hyporhamphus limbatus*, and *T. toil*	1.04	0.76	0.11	–	3.66	Ahmed et al., (2019)
Ennore coast, India	*M. cephalus*, *L. calcarifer*, *Terapon jarbua* and *Oreochromis niloticus*	–					Kumar et al. (2020)
Liaodong Bay, China	*Cynoglossus semilaevis*, *Ablennes hians*, *Enedrias fangi*, *Paralichthys olivaceus*, *Ditrema temmincki*, *Gymnocorymbus ternetzi*, *C. robustus*, *Scomberomorus niphonius*, *Navodon septentrionalis*, *Platycephalus indicus*, *Tridentiger barbatus*, *Acanthogobius hasta*	–	0.007–0.076	0.15–2.65	0.05–0.82	0.33–2.82	Wang et al., (2022)
Coastal areas of Zhejiang, China	*Larimichthys crocea*, *Larimichthys polyactis*, *Trichiurus lepturus*, *Collichthys lucidus*, *Pleuronectiformes*, *Lateolabrax japonicus*, *Leuciscus waleckii*, *Cololabis saira*, *Miichthys miiuy*, *Pneumatophorus japonicus*, *S. niphonius*, *P. argenteus*, *M. cephalus*, *Ilisha elongata*, *H. nehereus*	0.783	0.009	0.114	–	0.043	Han et al., (2021)
Tigris River, Turkey	*Chondrostoma regium*, *Cyprinion macrostomus*, *Barbus rajanorum mystaceus*, *Capoeta trutta*, *Carassius gibelio*, and *M. abu*	0.25	0.05	1.68	2.76	0.33	Töre et al., (2021)
Bangladesh (fish)		5.0	0.25	1.0	0.3	–	MOFL (2014)
MAC*		1.0	0.1	1.0	0.8	0.3	FAO/WHO (2002, 2022)

their bottom-dwelling nature and diet of zooplankton, fish, and benthic macroinvertebrates. This finding supports Ahmed et al. (2019a), who also noted elevated TE levels in demersal fish. Moreover, demersal fish, due to their consumption of HM-contaminated substances, tend to accumulate these toxins at higher trophic levels (Zaghloul *et al.*, 2022). The metal concentrations in fish muscle from our study were compared with those from other estuarine rivers both domestically and internationally. Ikem and Garth (2022) highlighted that fish are exposed to metals through their diet and environment, with bioaccumulation posing serious risks to public health and aquatic ecosystems. Comparison of the present findings with other national and international rivers and guideline standard is given in Table 4.

Concentrations of As ranged from 0.29 to 0.69 mg/kg, whereas the highest and lowest amount was recorded in *H. ilisha* and *H. nehereus*, respectively. The average concentration of As was within the maximum allowable concentration (MAC) WHO/FAO and MOFL (2014). Concentration of As was lower in comparison to national river such Ali et al. (2024) and Biswas et al. (2023) who detected higher As concentration (1.63 and 2.64 mg/kg) in the fishes of Rupsha River and coastal areas of Bangladesh. The present findings was also slightly below the study of Han et al. (2021) who reported the mean concentration of 0.783 mg/kg in marine fish population of Zhejiang, China. It was noted that the study area was heavily polluted by agricultural and household activities, which could be a potential source of As in the examined fish. Cd is a very hazardous contaminant that causes damage to kidney, liver, and respiratory organ (Biswas et al., 2023). Cd content in the fishes ranged from 0.046 to 0.060 mg/kg in which *G. giuris* exhibited the highest mean value 0.060 mg/kg. It is observed that mean Cd was well below the MOFL (2014) and FAO/WHO (2002) guidelines. Moreover, in comparison with other findings, average Cd concentration was also lower than that of national rivers e.g. Rupsha river (0.44 mg/kg), Karnaphuli River (0.41 mg/kg in summer and 0.52 mg/kg in winter), and international rivers e.g. Coastal areas of Zhejiang, China (0.009 mg/kg), Tigris River, Turkey (0.05 mg/kg) and Pearl River Delta (0.02–0.06 mg/kg) (Ali et al., 2024; Han et al., 2021; Tore et al., 2021 and Li et al., 2020; Ali et al., 2019). The calculated mean Cr concentration (0.957 mg/kg) was lower than MAC (1 mg/kg) proposed by FAO/WHO (2002) and MOFL (2014) except *L. calcarifer* and *H. nehereus*. The average Cr concentration was higher than the Rupsha River (0.58 mg/kg) (Ali et al., 2024) and Meghna River (0.11 mg/kg) (Ahmed et al., 2019a) but lower compared to Karnaphuli River Estuary (0.059–1.605 mg/kg) (Mohiuddin et al., 2022), Tigris River, Turkey (1.68 mg/kg) (Tore et al., 2021), and Liaodong Bay, China (0.15–2.65 mg/kg) (Wang et al., 2022). The highest and lowest value of Ni was detected in *H. nehereus* (4.83 mg/kg) and *H. ilisha* (1.18 mg/kg) which surpassed MOFL (2014) and FAO/WHO (2002) safety limit. Ni concentration of the examined fishes ranged from 0.162 to 0.439 mg/kg with the mean value of 0.285 mg/kg which was within the value reported in Karnaphuli River Estuary (0.023–0.355 mg/kg) by Mohiuddin et al. (2022). However, in comparison to the Pearl River this value was much lower (0.44–9.75 mg/kg) reported by Li et al. (2020). The concentration of Pb fluctuated from 0.789 to 1.970 mg/kg with the mean value 1.354 mg/kg which exceed the threshold limit (0.3 mg/kg) proposed by FAO/WHO (2022). The recorded finding was slightly below the concentration (1.38 mg/kg) reported by Ali et al. (2024) in Rupsha River, Bangladesh. However, it was within the findings (0.33–2.82 mg/kg) of Liaodong Bay, China (Wang et al., 2022).

Substantial positive inter-correlations across TE revealed shared sources and routes from which they may be collected in fish muscle. Proshad et al. (2019) found that the sources and pathways of metals in samples may be identified by their inter-metal interactions. Significant correlation between the metals in the ecosystem indicated their parallel level of contamination and identical source of contamination (Li et al., 2009). Conjoint dependence, and common sources and indistinguishable behavior of positively correlated metals were also previously reported by Ali et al. (2019) and Jiang *et al.* (2014). The PCA and clustering analyses revealed that TE contamination stems from both natural and anthropogenic sources. Cr and Pb (PC 1) are linked to industrial discharges near S1 and S2, while As and Ni (PC 2) are associated with agricultural runoff from S5. Higher TE levels in demersal fish like *L. calcarifer* and *H. nehereus* reflect proximity to pollution hotspots. Anthropogenic activities, including industries, agriculture, and fertilizer use, significantly influence As distribution, posing health risks (Nargis et al., 2019).

Consumption of fish with elevated APLI may constitute a health risk. Maximum APLI value was recorded in *H. nehereus* (1.95) whereas minimum was observed in *H. ilisha* (0.84). The average APLI recorded in the present study was 1.36, which is above the threshold value of > 1 and indicating the serious pollution load in the studied fish species (Chakraborty et al., 2022). Different environmental sources, quantities, habitats, dietary practices, and physiology may affect fish species' essential and harmful metal accumulation (Varol et al., 2019).

The toxic nature of TE may lead to health risks when consuming contaminated seafood, making it crucial to conduct an examination (Feng et al., 2020). The average EDI of metals was Pb > Cr > Ni > As > Cd. Demersal species exhibited higher EDIs for both adult and child compared to benthopelagic and pelagic fishes which aligns with the findings of Ali et al. (2024) who also reported higher EDI for demersal fish species. In this research, children have greater EDI than adults for all metals, similar to Hossain et al. (2022). The EDI value of individual metal is lower compared to tolerable daily intake (TDI) (except for As in child for *A. latus*, *M. curema* and *G. giuris*) values depending on the fish species. Therefore, daily intake of contaminated fish containing As may have significant negative impact on child health.

The use of health risk evaluations has been extensive in evaluating the possible hazards linked to the intake of fish species that are polluted, and in presenting compelling evidence of these risks to those responsible for making decisions (Gao et al., 2022). In the present study, the THQs for both adults and children were observed in the decreasing order of As > Pb > Cd > Ni > Cr where THQ of all the metals were far lower in adult than standard threshold limit (THQ <1). Ali et al. (2024) conducted a similar study on Rupsha River and found THQs follows decreasing order of As > Cd > Pb > Cr which support our findings. The present THQ values of individual TE except for As in case of adult and As and Pb in case of child were found below the critical limit of < 1 indicating safe limit of non-carcinogenic risk through the contamination of these metals. However, values of As and Pb above the threshold limit in some cases are considered contamination in fishes and are believed to cause considerable health effects (Shaheen et al., 2024; Biswas et al., 2023). HI indicated the additive impacts of the analyzed TE in the studied fish species and this index was found to increase in the order of *H. ilisha* < *A. latus* < *M. curema* < *G. giuris* < *H. nehereus* < *L. calcarifer*. The calculated HI exceed the threshold limit of 1 and child represent higher HI values than adult which supports Biswas et al. (2023) and Kumar et al. (2021). Compared to other studies, the calculated HI in this study was higher than those reported for fish from the Rupsha River (Ali et al., 2024), Barguzin River (Bazarsadueva et al., 2023), Karnaphuli River estuary (Mohiuddin et al., 2022), and Lower Meghna River (Hossain et al., 2022). These findings indicate that the daily consumption of the studied fish species poses a risk to consumers.

Trace elements concentrations in fish species were assessed to determine CR. According to Baki et al. (2018), CR values greater than 10^{-4} are deemed unacceptable, values between 10^{-6} and 10^{-4} are considered within an acceptable range, and values less than 10^{-6} are considered negligible. The results of the current study indicate that CR values for all metals suggest no cancer risk for adults except for Cd. However, children may face potential cancer risk with prolonged exposure to contaminated fish. This study also highlights that child are more susceptible to cancer risk compared to adults, consistent with findings by Ali et al. (2024), Mohiuddin et al. (2022), and Noman et al. (2022), who also observed higher risks for children due to TE contamination in fish. Supporting these results, Biswas et al. (2023) identified

cancer risks associated with As and Cr in coastal and marine fish species. Although this study focused solely on fish consumption, other aquatic food sources may also pose cancer risks, which were not evaluated here. Thus, carcinogenic risks related to aquatic product consumption, particularly in the study area, warrant further attention. Future research should prioritize investigating the ecosystem-level impacts of heavy metal contamination in key marine environments while identifying and tracing the primary sources and pathways of these pollutants into marine ecosystems. Emphasis should also be placed on developing advanced, eco-friendly remediation strategies and implementing stricter regulatory frameworks to mitigate contamination. Furthermore, establishing comprehensive, long-term monitoring programs for heavy metal levels in marine organisms and promoting public awareness initiatives are critical to safeguarding both ecosystem health and human well-being.

5. Conclusion

This study offered a thorough evaluation of TE contamination in some commonly consumed estuarine fish and associated health risks from consumption. The data indicated that, in most cases, TE concentrations did not exceed the maximum allowable limits. However, demersal fish were found to be more prone to TE contamination due to their scavenging feeding habits. The sources of the studied metals were identified as anthropogenic. The APLI value classified *H. nehereus* as heavily contaminated. Consumption of the studied fish species posed non-carcinogenic health risks to adults, with HI values exceeding the safe threshold of '1'. Carcinogenic risks also surpassed safe limits for adults, with children being more vulnerable. Given the existing amount of pollution in the BRE, law enforcement is critical to protecting the riverine ecology and human health. Additional research is also needed to assess the contamination of other commercial fish species. Technical measures should be considered to improve the quality of waste discharges so that contamination of fish can be reduced. Government policy should be guided towards environmental legislation and holistic planning regarding the production and disposal of potentially toxic elements in the estuaries. Furthermore, a thorough investigation should be done in other estuary rivers to improve our knowledge of the toxicological effects of TE on fish.

Ethical clearance Statement

Sampling and analysis of fishes was approved by the guidelines and regulations of the Department of Fisheries, Faculty of Agriculture, University of Rajshahi, Bangladesh.

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Author statement

There is no conflict of interest

CRediT authorship contribution statement

Md. Ayenuddin Haque: Writing – original draft, Resources, Methodology, Investigation. **Md. Abu S Jewel:** Supervision, Writing – review & editing. **Md. Shahidul Islam Kazal:** Writing – original draft, Methodology, Data curation. **Bithy Khatun:** Writing – original draft, Investigation, Data curation. **Takaomi Aari:** Writing – review & editing, Data curation. **Jesmin Ara:** Validation, Investigation. **Norhayati Ngah:** Writing – review & editing, Resources, Formal analysis. **Bilal Ahamad Paray:** Writing – review & editing, Resources, Funding acquisition. **jimmy Yu:** Writing – review & editing. **Sharmin Jahan:** Writing – original draft, Methodology, Investigation, Formal analysis. **Mohammad Belal Hossain:** Writing – review & editing, Writing – original draft, Resources, Investigation, Formal analysis, Data curation.

CRediT authorship contribution statement

All authors contributed significantly

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jfca.2025.107220.

Data availability

Data will be made available on request.

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