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## Full Length Article

## Dietary intake of heavy metals from eight highly consumed species of cultured fish and possible human health risk implications in Bangladesh

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## ABSTRACT

Concentrations of five heavy metals (Pb, Cd, Cr, As and Hg) in eight highly consumed cultured fish species (*Labeo rohita*, *Clarias gariepinus*, *Hypophthalmichthys molitrix*, *Cyprinus capio*, *Puntius sarana*, *Oreochromis mossambicus*, *Pangasius pangasius* and *Anabas testudineus*) collected from four wholesale markets of Dhaka city, Bangladesh (Karwan Bazar, Mohammadpur Town Hall, Newmarket and Mirpur-1) were measured using atomic absorption spectrometry (AAS) in order to evaluate the potential human health risks from the consumption of fish. The estimated daily intake (EDI) of all the studied heavy metals calculated on the basis of mean fish consumption of 49.5 g person<sup>-1</sup> d<sup>-1</sup> by Bangladeshi households indicated that no risk to people's health with respect to the EDI of investigated heavy metals through the consumption of the fish samples. From the human health point of view, the estimation of non-carcinogenic risk indicated that intake of individual heavy metal through the consumption of fish was safe for human health, whereas, consumption of combined heavy metals suggested potential health risk to highly exposed consumers. However, the estimation of carcinogenic risk of arsenic due to the consumption of fish indicated that consumers remain at risk of cancer.

## 1. Introduction

Global aquatic environments are being heavily polluted with heavy metals as they are highly exposed to the environment with the growing urbanisation and globalisation for the last few decades [1,2]. The presence of heavy metals in the aquatic environments leads to severe adverse effects on fish and has been a subject of concern for many decades [3]. The heavy metals are non-biodegradable and therefore can easily be accumulated in the living organisms including fish [4]. Consequently, human beings are potentially exposed to these contaminants through the food chain with the consumption of fish. The heavy metals result in various adverse health effects. For example, lead may hinder the cognitive development and intellectual performance in children; and increases blood pressure and cardiovascular disease in adults [5]. Cadmium results impaired kidney function, poor reproductive capacity, hypertension, tumorous and hepatic dysfunction [6,7,8]. Moreover, cadmium toxicity may result genotoxicity, endocrine disruption, oxidative damage, disruption of ion regulation [9]. Exposure to chromium may result severe respiratory, cardiovascular, gastrointestinal, hematological, hepatic, renal, and neurological effects as part of the sequelae leading to death [10]. Arsenic exposure can also affect almost all organ systems including the dermatologic, cardiovascular, nervous, renal,

hepatobiliary, gastro-intestinal and respiratory systems [11,12]. The toxicity of mercury includes gastrointestinal toxicity, neurotoxicity, and nephrotoxicity [11,12].

In recent years, world consumption of fish has significantly increased due to their potential nutritional and therapeutic benefits [13]. Eventually, fish is the most important single source of high quality protein contributing about 17 percent of animal protein and 6.7 percent of all protein consumed by the world population [14]. Moreover, in Bangladesh fish accounted for 55 percent of animal protein intake [15] and contributes 49.5 g/capita/day [16]. In addition to being a rich source of protein, fish provides high contents of essential fats, vitamins and minerals [17]. As fish is an important constituent of human diet, it is often deemed as the most suitable object among the bioindicators of aquatic ecosystem [18]. Moreover, metals contents in fishes indicate their accumulation in food chain [19]. But, there is limited information on the heavy metal concentrations in selected few cultured fishes (*Labeo rohita*, *Oreochromis mossambicus*, and *Pangasius pangasius*) [20]. Moreover, due to increasing anthropogenic and industrial stresses, continuous monitoring of the heavy metals in highly consumed cultured fishes with more species is required.

In the present study, levels of heavy metals lead (Pb), cadmium (Cd), chromium (Cr), arsenic (As) and mercury (Hg) in eight different

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highly consumed cultured fish species (*Labeo rohita*, *Clarias gariepinus*, *Hypophthalmichthys molitrix*, *Cyprinus capio*, *Puntius sarana*, *Oreochromis mossambicus*, *Pangasius pangasius* and *Anabas testudineus*) collected from four wholesale markets of capital city Dhaka, Bangladesh from where fishes are supplied to the local markets were determined. Several analytical techniques such as atomic absorption spectrometry (AAS), inductively coupled plasma-atomic emission spectrometry (ICP-AES), inductively coupled plasma-mass spectrometry (ICP-MS), neutron activation analysis (NAA), differential pulse anodic stripping voltammetry (DPASV), energy dispersive X-ray fluorescence (EDXRF) etc. have been employed for the determination of trace metals in different environmental samples. AAS was used for quantifying the heavy metal contents in fish samples in the present case as it is highly selective, accurate, sensitive and cost effective [21]. Moreover, to the best of our knowledge, this is the first report regarding the heavy metal concentrations in cultured fishes with a good number of varieties highly consumed by Bangladeshi people were measured using validated and accredited test methods [22]. Fishes are well-known for their ability to concentrate heavy metals in muscles; therefore, we selected muscles as a primary site of metal uptake in the present study. Moreover, long term consumption of fish contaminated with heavy metals may result to the accumulation of toxic metals in different essential organs could carry serious health risks [23]. Therefore, an assessment on non-carcinogenic and carcinogenic human health risks due to consumption of those fishes has been conducted.

## 2. Materials and methods

### 2.1. Sample collection and preservation

Eight fish species of *Labeo rohita* (Rui), *Clarias gariepinus* (Magur), *Hypophthalmichthys molitrix* (Silver carp), *Cyprinus capio* (Karfu), *Puntius sarana* (Sharputi), *Oreochromis mossambicus* (Tilapia), *Pangasius pangasius* (Pangas) and *Anabas testudineus* (Koi) those are highly consumed by Bangladeshi population were collected from four different wholesale markets of Dhaka city namely: Mirpur-1, Mohammadpur Town Hall, Newmarket and Karwan Bazar. A total of 32 individuals representing 8 (eight) fish species were collected and wrapped in polyethylene bags to transport to the Analytical Chemistry Laboratory, Atomic Energy Centre Dhaka, Bangladesh Atomic Energy Commission. Immediately after transportation to the laboratory, samples were washed with fresh water to remove the mud or other fouling substances. Then the muscle tissue of each sample was removed and chopped into pieces with the aid of a steam cleaned stainless steel knife. The muscle tissues were then washed with deionized water and air dried to remove the extra water and subsequently, homogenized in a food processor and 200 g of test portions were stored at  $-20^{\circ}\text{C}$ . Metal contents were expressed as  $\text{mg kg}^{-1}$  wet wt. basis of fresh fish.

### 2.2. Analytical methods

#### 2.2.1. Instruments and reagents

A Varian AA280Z atomic absorption spectrometer (AAS) with Zeeman background correction system equipped with graphite furnace (GTA 120) and an auto sampler (PSD 120) was used for the determination of Pb, Cd and Cr. Moreover, Hg and As were determined using cold vapour AAS (CV-AAS) and hydride generation AAS (HG-AAS) techniques using a Varian AA240FS equipped with hydride vapour generator (VGA 77). The purity of argon and acetylene gases were 99.999% and 99.99% respectively. Hollow cathode lamps were used for Pb (283.3 nm and slit 0.5 nm), Cd (228.8 nm and slit 0.5 nm), Cr (357.9 nm and slit 0.2 nm), As (193.7 nm slit 0.5 nm) and Hg (253.7 nm and slit 0.5 nm) and they were operated according to the conditions recommended by the manufacturer. Atomic signals were measured for Pb, Cd and Cr in peak area mode and for As and Hg in integration mode. The digestions were performed using a microwave

oven (MARS'5, CEM Corporation, USA). The working standard solutions of Pb, Cd, Cr, As and Hg were daily prepared by appropriate dilution of the respective  $1000\ \mu\text{g L}^{-1}$  stock standard solutions using 1% (w/w) *suprapur* grade nitric acid (Merck, Darmstadt, Germany). Phosphoric acid (Merck, Darmstadt, Germany) and ammonium phosphate monobasic (Merck, Darmstadt, Germany) were used as chemical modifiers for the determination of Pb and Cd respectively. Sodium borohydride (Acros Organics, USA), sodium hydroxide (BDH), hydrochloric acid (Merck, Germany) were used as reductant for the determination of Hg and As. Moreover, potassium iodide (Merck, Germany) and ascorbic acid (Merck, Germany) were used as reductant during the sample preparation for As analysis. The reagents were of analytical grade and all solutions were prepared using deionized water ( $18\ \text{M}\Omega/\text{cm}$ ) produced using an E-pure system (Thermo Scientific, USA). All containers and glassware were cleaned by soaking into 20 percent nitric acid for at least 24 h and rinsed three times with deionized water prior to use.

#### 2.2.2. Sample digestion

1.00 g of the homogenized fish samples were accurately weighted into the polytetrafluoroethylene digestion vessel and 6 mL of *suprapur* nitric acid were added to it. Subsequently, using a two-step temperature program the samples were digested with the MARS'5 XP-1500 plus (CEM Corporation, USA) microwave oven. The maximum power of the rotating magnetron was 1600 W. During the first step, the temperature was linearly increased to  $180^{\circ}\text{C}$  for 10 min and then it was maintained at that temperature for 15 min during the second step. After digestion and cooling each solution was diluted to a final volume of 10 mL with deionized water.

#### 2.2.3. Quality control programmes

**2.2.3.1. Internal quality control.** Internal quality control charts (IQCs) were constructed in order to monitor whether results were reliable enough to be released. The objective of IQCs was the elongation of method validation: continuously checking the accuracy of analytical data obtained from day to day in the laboratory. The analytical system was under control if no more than 5% of the measured values exceed the warning limits and none of them the action or control limits [24].

**2.2.3.2. External quality control.** The external quality assurance program was maintained through the participation in proficiency programmes organized by Asia Pacific Metrology Program-Asia Pacific Laboratory Accreditation Cooperation (APMP- APLAC) and LGC, UK. The results of proficiency tests were within  $\pm 2$  Z-scores.

**2.2.3.3. Accuracy of the method.** The accuracy of the method was evaluated by analysing a certified reference material NIST CRM 1566a (Oyster Tissue) by the same procedure used for fish samples. Mean recoveries of the analysed metals were between 95.7% to 98.6%, indicating a good agreement between certified and measured values (Table 1).

**Table 1**

Heavy metal concentration (mean  $\pm$  standard deviation) in certified reference material (NIST CRM 1566a, Oyster Tissue),  $n = 3$ .

Heavy metal	Certified value (mg/kg)	Found value (mg/kg)	Mean recovery (%)
Pb	$0.371 \pm 0.014$	$0.357 \pm 0.011$	96.2
Cd	$4.15 \pm 0.38$	$4.01 \pm 0.05$	96.6
Cr	$1.43 \pm 0.46$	$1.41 \pm 0.04$	98.6
As	$14.0 \pm 1.2$	$13.40 \pm 0.58$	95.7
Hg	$0.0642 \pm 0.0067$	$0.0620 \pm 0.0027$	96.6

**Table 2**According to the area, trace elements concentrations (mean  $\pm$  SD) in fish species, number of measurements, n=3.

Sample location	Fish species (Local name)	Trace element concentrations (mg/kg)				
		Pb	Cd	Cr	As	Hg
Karwan Bazar	<i>Labeo rohita</i> (Rui)	0.412 $\pm$ 0.082	< 0.01	0.05	0.357 $\pm$ 0.017	< 0.02
	<i>Clarias gariepinus</i> (Magur)	0.820 $\pm$ 0.164	< 0.01	0.363 $\pm$ 0.054	< 0.08	0.473 $\pm$ 0.081
	<i>Hypophthalmichthys molitrix</i> (Silver carp)	0.559 $\pm$ 0.112	< 0.01	0.058 $\pm$ 0.009	< 0.08	< 0.02
	<i>Cyprinus capio</i> (Karfui)	0.362 $\pm$ 0.072	< 0.01	0.119 $\pm$ 0.018	< 0.08	< 0.02
	<i>Puntius sarana</i> (Sharputi)	0.518 $\pm$ 0.104	< 0.01	0.139 $\pm$ 0.021	< 0.08	0.0415 $\pm$ 0.008
	<i>Oreochromis mossambicus</i> (Tilapia)	0.313 $\pm$ 0.063	< 0.01	0.086 $\pm$ 0.013	< 0.08	< 0.02
	<i>Pangasius pangasius</i> (Pangas)	0.947 $\pm$ 0.189	< 0.01	0.121 $\pm$ 0.018	< 0.08	< 0.02
Mohammadpur	<i>Anabas testudineus</i> (Koi)	0.882 $\pm$ 0.176	< 0.01	0.187 $\pm$ 0.028	< 0.08	< 0.02
	<i>Labeo rohita</i> (Rui)	0.443 $\pm$ 0.089	< 0.01	< 0.05	0.429 $\pm$ 0.021	< 0.02
	<i>Clarias gariepinus</i> (Magur)	0.783 $\pm$ 0.157	0.015 $\pm$ 0.001	0.067 $\pm$ 0.010	0.358 $\pm$ 0.018	< 0.02
	<i>Hypophthalmichthys molitrix</i> (Silver carp)	0.116 $\pm$ 0.023	< 0.01	< 0.05	< 0.08	< 0.02
	<i>Cyprinus capio</i> (Karfui)	0.096 $\pm$ 0.019	0.020 $\pm$ 0.002	< 0.05	< 0.08	< 0.02
	<i>Puntius sarana</i> (Sharputi)	0.351 $\pm$ 0.070	< 0.01	< 0.05	< 0.08	0.038 $\pm$ 0.007
	<i>Oreochromis mossambicus</i> (Tilapia)	0.148 $\pm$ 0.030	< 0.01	< 0.05	< 0.08	< 0.02
Newmarket	<i>Pangasius pangasius</i> (Pangas)	0.468 $\pm$ 0.094	< 0.01	0.142 $\pm$ 0.021	< 0.08	< 0.02
	<i>Anabas testudineus</i> (Koi)	0.607 $\pm$ 0.121	0.018 $\pm$ 0.002	0.104 $\pm$ 0.016	< 0.08	< 0.02
	<i>Labeo rohita</i> (Rui)	0.495 $\pm$ 0.099	< 0.01	0.060 $\pm$ 0.009	0.501 $\pm$ 0.025	0.0205 $\pm$ 0.003
	<i>Clarias gariepinus</i> (Magur)	1.019 $\pm$ 0.204	< 0.01	0.018 $\pm$ 0.003	0.272 $\pm$ 0.014	0.0287 $\pm$ 0.004
	<i>Hypophthalmichthys molitrix</i> (Silver carp)	0.977 $\pm$ 0.195	0.017 $\pm$ 0.002	< 0.05	< 0.08	< 0.02
	<i>Cyprinus capio</i> (Karfui)	0.456 $\pm$ 0.091	0.016 $\pm$ 0.002	< 0.05	< 0.08	< 0.02
	<i>Puntius sarana</i> (Sharputi)	1.382 $\pm$ 0.276	0.020 $\pm$ 0.002	0.102 $\pm$ 0.015	< 0.08	0.045 $\pm$ 0.005
Mirpur-1	<i>Oreochromis mossambicus</i> (Tilapia)	0.904 $\pm$ 0.181	< 0.01	< 0.05	< 0.08	< 0.02
	<i>Pangasius pangasius</i> (Pangas)	2.354 $\pm$ 0.470	< 0.01	< 0.05	< 0.08	< 0.02
	<i>Anabas testudineus</i> (Koi)	0.878 $\pm$ 0.176	< 0.01	0.077 $\pm$ 0.012	< 0.08	< 0.02
	<i>Labeo rohita</i> (Rui)	0.228 $\pm$ 0.046	< 0.01	0.854 $\pm$ 0.128	0.143 $\pm$ 0.007	0.121 $\pm$ 0.015
	<i>Clarias gariepinus</i> (Magur)	0.606 $\pm$ 0.121	< 0.01	0.901 $\pm$ 0.135	0.358 $\pm$ 0.018	< 0.02
	<i>Hypophthalmichthys molitrix</i> (Silver carp)	0.626 $\pm$ 0.125	< 0.01	< 0.05	0.358 $\pm$ 0.018	< 0.02
	<i>Cyprinus capio</i> (Karfui)	0.519 $\pm$ 0.104	< 0.01	0.168 $\pm$ 0.025	< 0.08	< 0.02
Mirpur-1	<i>Puntius sarana</i> (Sharputi)	0.271 $\pm$ 0.054	< 0.01	< 0.05	< 0.08	< 0.02
	<i>Oreochromis mossambicus</i> (Tilapia)	0.223 $\pm$ 0.045	< 0.01	0.356 $\pm$ 0.053	< 0.08	< 0.02
	<i>Pangasius pangasius</i> (Pangas)	0.139 $\pm$ 0.028	< 0.01	< 0.05	< 0.08	< 0.02
	<i>Anabas testudineus</i> (Koi)	0.083 $\pm$ 0.017	< 0.01	< 0.05	< 0.08	< 0.02

### 2.3. Calculations

#### 2.3.1. Estimated daily intakes (EDI)

The estimated daily intakes (EDI) for the analysed metals were calculated by multiplying the respective mean concentration of the metal determined in the targeted fish samples by the weight of fish consumed by an average individual in Bangladesh which was obtained from the “Report of the household income and expenditure survey 2015” [16], and calculated by using the formula [20].where,

$$EDI = DFCXMC$$

where, DFC = daily food (fish) consumption, MC = mean metal concentration of metal in fish sample. The daily fish consumption rate for an adult (60 kg) was an average of 49.5 g on fresh weight basis “Report of the household income and expenditure survey 2015” [16].

#### 2.3.2. Non-carcinogenic risk

The non-carcinogenic risk assessments are typically conducted to estimate the potential health risks of pollutants using the target hazard quotient (THQ). The THQ values through the consumption of fish species by local inhabitants can therefore be assessed for each heavy metal and calculations were made using the standard assumption for an integrate USEPA risk analysis as follows [25],where,

$$THQ = \frac{EFr \times ED \times FIR \times C}{RfD \times BW \times TA} \times 10^{-3}$$

where, EFr is the exposure frequency (365 d year<sup>-1</sup>); ED is the exposure duration (70 years) equivalent to the average human life time; FIR is the food ingestion rate (g person<sup>-1</sup> d<sup>-1</sup>); C is the metal concentration in samples (mg kg<sup>-1</sup>, wet weight); BW average body weight (adult: 60 kg); TA is the averaging time for non-carcinogens 365 d year<sup>-1</sup> X number of exposure years, assuming 70 years); RfD is the oral reference

dose (mg kg<sup>-1</sup> d<sup>-1</sup>); RfDs are based on 0.004, 0.001, 1.5, 0.003, and 0.0005 mg kg<sup>-1</sup> bw d<sup>-1</sup> for Pb, Cd, Cr, As and Hg respectively [26]. If the THQ value is less than 1, the exposed population is unlikely to experience any adverse health hazard. Conversely, if the THQ is equal to or higher than 1, there is a potential health risk [27], and related interventions and protective measurements should be taken.

It has been reported that exposure to two or more pollutants may result in additive and/or interactive effects [28]. Thus, in this study, cumulative health risk was evaluated by summing THQ value of individual metal and expressed as total THQ (TTHQ) as follows

$$TTHQ = THQ(\text{toxicant1}) + THQ(\text{toxicant2}) + \dots THQ(\text{toxicantn})$$

The greater the value of TTHQ, the greater the level of concern.

#### 2.3.3. Carcinogenic risk

Carcinogenic risk (CR) indicates an incremental probability of an individual of developing cancer over a lifetime due to exposure to a potential carcinogen. Cancer risk over a lifetime exposure to Pb and As were obtained using cancer slope factor (CSF), provided by USEPA [29]. The equation used for estimation of the cancer risk is as follows USPEA, 2000:

$$CR = CSF \times EDI$$

where, CSF is the carcinogenic slope factor of 0.0085 (mg/kg/day)<sup>-1</sup> for Pb and 1.5 (mg/kg/day)<sup>-1</sup> for As set by USPEA (USPEA, 2010). EDI is the estimated daily intake of heavy metals. Acceptable risk levels for carcinogens range from 10<sup>-4</sup> (risk of developing cancer over a human lifetime is 1 in 10000) to 10<sup>-6</sup> (risk of developing cancer over a human lifetime is 1 in 1000000).

### 3. Results and discussion

#### 3.1. Concentration of heavy metals in fish muscles

Concentration of heavy metals, Pb, Cd, Cr, As and Hg in muscle tissues of eight cultivated fish species highly consumed by the Bangladeshi people were listed in Table 2. All metal concentrations were determined on a wet weight basis. Heavy metal contents in fish samples were found 0.083–2.354 mg kg<sup>-1</sup> for Pb, 0.015–0.020 mg kg<sup>-1</sup> for Cd, 0.058–0.901 mg kg<sup>-1</sup> for Cr, 0.143–0.501 mg kg<sup>-1</sup> for As, and 0.021–0.121 mg kg<sup>-1</sup> for Hg. According to these data, the ranking order of mean concentration of the heavy metals in fish muscles were Pb (0.593 mg kg<sup>-1</sup>) > As (0.332 mg kg<sup>-1</sup>) > Cr (0.193 mg kg<sup>-1</sup>) > Hg (0.050 mg kg<sup>-1</sup>), and Cd (0.017 mg kg<sup>-1</sup>).

Lead is a non-essential heavy metal and endures many adverse health effects including neurotoxicity and nephrotoxicity [30]. The minimum and maximum lead level observed were 0.358 mg kg<sup>-1</sup> in *Cyprinus capio* and 0.977 mg kg<sup>-1</sup> in *Pangasius pangasius* respectively. Lead contents in the literature have been reported in the range of 4.25–8.17 mg/kg dry weight in fish species from Dhaleshwari river, Bangladesh [31], 1.76–10.27 mg/kg dry weight in some edible fishes from Bangshi River, Savar, Dhaka, Bangladesh [8], 8.03–13.52 mg/kg dry weight in some freshwater fishes of Buriganga River, Bangladesh [32], 0.052–2.7 mg/kg wet weight in some fish species from urban rivers around Dhaka city, Bangladesh [2], 0.017–0.090 mg/kg wet weight in some cultured fishes highly consumed by Bangladeshi people [20]. The maximum legislative value of lead as described by the Commission Regulation (EC) NO. 1881/2006 and Bangladesh Gazette S. R. O. No. 233-Act 2014 [33] is 0.30 mg kg<sup>-1</sup> as wet weight basis. The present observation showed that level of lead in every fish species was above the proposed acceptable limit for human consumption.

Cadmium is a highly toxic element capable of causing severe toxicity even when it is present at a very low concentration of ~1 mg/kg [34]. The accumulation of Cd in the human body may give rise to hepatic, pulmonary, renal, skeletal, reproductive effects and even cancer [20]. The lowest and highest Cd content in fish species were found as 0.015 mg kg<sup>-1</sup> in *Clarias gariepinus* and 0.019 mg kg<sup>-1</sup> in *Puntius sarana* respectively. Cd contents in the literature have been reported in the range of 0.51–0.73 mg/kg dry weight in fish species from Dhaleshwari river, Bangladesh [31], 0.09–0.87 mg/kg dry weight in some edible fishes from Bangshi River, Savar, Dhaka, Bangladesh [8], 0.008–0.13 mg/kg wet weight in some fish species from urban rivers around Dhaka city, Bangladesh [2], 0.001–0.003 mg/kg wet weight in some cultured fishes highly consumed by Bangladeshi people [20]. The amount of cadmium measured in all the fish species was below the standard of 0.05 mg kg<sup>-1</sup> set by [35] European Union and Government of the People's Republic of Bangladesh, but long period of cadmium accumulation in fish may pose health hazards.

The presence of chromium in the diet is of great importance due to its active involvement in lipid metabolism and insulin function [20]. The minimum and maximum chromium values were found as 0.058 mg kg<sup>-1</sup> in *Hypophthalmichthys molitrix* and 0.206 mg kg<sup>-1</sup> in *Pangasius pangasius* respectively. Chromium contents in the literature have been reported in the range of 6.92–12.23 mg/kg dry weight in fish species from Dhaleshwari river, Bangladesh [31], 0.47–2.07 mg/kg dry weight in some edible fishes from Bangshi River, Savar, Dhaka, Bangladesh [8], 5.27–7.38 mg/kg dry weight in some freshwater fishes of Buriganga River, Bangladesh [32], 0.75–4.8 mg/kg wet weight in some fish species from urban rivers around Dhaka city, Bangladesh [2], 1.054–1.349 mg/kg wet weight in some cultured fishes highly consumed by Bangladeshi people [20]. From our measured value it was observed that chromium in the selected fish species was below the legislative value of 1.00 mg kg<sup>-1</sup> set for the Bangladeshi people.

Arsenic is one of the most potential toxic heavy metals present in the environment and originates from both natural and anthropogenic

processes [36]. According to USFDA [37] about 90 percent of total human exposure of As instigates from fish and other seafood. The lowest and highest arsenic contents were observed as 0.309 mg kg<sup>-1</sup> in *Hypophthalmichthys molitrix* and 0.357 mg kg<sup>-1</sup> in *Labeo rohita* respectively. Arsenic contents in the literature have been reported in the range of 1.01–15.2 mg/kg in fresh water fish species [38], 1.97–6.24 mg/kg dry weight in some edible fishes from Bangshi River, Savar, Dhaka, Bangladesh [8], 0.091–0.53 mg/kg wet weight in some fish species from urban rivers around Dhaka city, Bangladesh [2], 0.077–1.486 mg/kg wet weight in some cultured fishes highly consumed by Bangladeshi people [20]. According to Bangladeshi standard, the maximum permitted concentration for arsenic is 5.00 mg kg<sup>-1</sup>, which was beyond the proposed acceptable limit for human consumption.

Mercury is a non-essential heavy metal and cannot be excreted easily. It could be retained in the tissues for long periods resulting behavioural and cognitive changes, neurological impairment and lesions [39]. Moreover, during pregnancy, mercury can pass through the placenta to the fetus and may affect the development of central nervous system [23]. The minimum and maximum mercury contents were found as 0.038 mg kg<sup>-1</sup> in *Clarias gariepinus* and 0.071 mg kg<sup>-1</sup> in *Labeo rohita* respectively. The European Commission Regulation as well as Bangladesh Government Act stated permitted mercury concentration of 0.50 mg kg<sup>-1</sup> which was higher than our values found for selected fish species. A correlation matrix was also calculated for the analysed metals in fish species in order to identify the common sources of metals. Table 3 showed the correlation between the investigated heavy metals in fish samples, listing the Pearson correlation coefficient. Significant correlations were found between Cr and As ( $r = 0.597$ ), Cr and Hg ( $r = 0.785$ ), and As and Hg ( $r = 0.574$ ) at  $p < 0.05$ , suggesting a common source of these metals (possibly fish feed) [40,41]. On the other hand, no correlation between Pb and Cd with other metals was noted suggesting that Pb and Cd contaminations might be different origin than other metals. The contamination of Pb was possibly due to industrial discharge from battery and Cd possibly for industry and mining activities.

#### 3.2. Estimated daily intake (EDI)

The EDI of heavy metals through the consumption of eight fish species by people is given in Table 4. The result shown in Table 4 revealed that Cd contributed the lowest daily intake and Pb contributed the highest daily intake, which agreed well with the earlier result [44]. The EDI was calculated by considering that a 60 kg person consumes 49.5 g fish per day. The results of EDI revealed that the EDI values for the examined fish samples were below the recommended values, indicated no risk to people's health associated with the intake studied heavy metals through the consumption of the selected fish samples.

#### 3.3. Noncarcinogenic risk

THQs of individual heavy metal through fish consumption by average Bangladeshi adults are presented in Table 4. Average heavy metal concentration in fish species was used in order to calculate THQ for the people of Dhaka city, Bangladesh. The THQ values for the

**Table 3**  
Correlation analysis of heavy metals among the fish species.

	Pb	Cd	Cr	As	Hg
Pb	1				
Cd	-0.247	1			
Cr	0.071	-0.339	1		
As	-0.031	0.292	<b>0.597</b>	1	
Hg	-0.106	0.388	<b>0.785</b>	<b>0.574</b>	1

Statistically significant correlations are indicated in bold where  $P < 0.05$ .



**Table 4**

Comparison of the estimated daily intake of heavy metals from fish species studied with the recommended daily dietary allowances.

Metal	Mean concentration (mg/kg -wet wt.)	EDI (mg/day /person)	Recommended daily dietary allowance (mg/day /person)	THQ	TTHQ	Carcinogenic risk (CR)
Pb	0.593	0.0293	0.21 [42]	0.1223	1.1319	4.2E-06
Cd	0.017	0.0008	0.06 [42]	0.0140		
Cr	0.193	0.0096	0.20 [43]	0.0001		
As	0.332	0.0164	0.13 [42]	0.9130		2.7E-04
Hg	0.050	0.0025	0.03 [42]	0.0825		

targeted heavy metal followed the descending order of As > Pb > Hg > Cd > Cr, which agreed well with the earlier report [20]. Table 4 indicated that the THQ value of each metal was less than 1 suggested that people would not experience significant health risks if they only intake individual heavy metal through the consumption of fish. However, the TTHQ was greater than 1, indicated that there was potential health risk through the consumption of fishes involved exposure to a mixture of five examined metals. In this study, the major risk contributor was As with 80.66%, followed by Pb (10.80%), Hg (7.29%), Cd (1.24%) and Cr (0.01%).

### 3.4. Carcinogenic risk

The CR values of Pb and As due to exposure from the consumption of targeted eight fish species were  $4.2 \times 10^{-6}$  and  $2.7 \times 10^{-4}$  respectively, which were slightly higher than the previous study [20] indicating that the fishes were becoming polluted. Generally, the values of CR lower than  $10^{-6}$  are considered as negligible, above  $10^{-4}$  are considered to be unacceptable and lying in between  $10^{-6}$  and  $10^{-4}$  are considered as acceptable range [26]. In the present study, CR for As was higher than the unacceptable range indicating the risk of cancer due to exposure to As through fish consumption was of concern.

## 4. Conclusion

The heavy metal contents were found to be varied in fish species, and the estimated daily intake of Pb, Cd, Cr, As and Hg from the targeted fish samples were below the respective recommended daily dietary allowance for these elements. From the human health point of view, the THQ values for individual element were lower than 1, suggesting that there was no health risk for consumers due to intake of individual heavy metal, however, total THQ for combined heavy metals was higher than 1, indicating potential health risk to highly exposed consumers. The carcinogenic risk of arsenic due to the consumption of fish was also of concern since the carcinogenic rate in fish was above the acceptable risk level of  $10^{-4}$ . It may be mentioned here that, in order to determine the health risk due to intake of trace metals only fish consumption was considered which constitutes only 3% of per capita per day calorie intake by food items. It is thus suggested that other food sources, particularly staple rice, vegetables, fruits, cereals, piscine and non-piscine protein sources need to be considered in order to evaluate the exact health risks due to intake of trace metals from dietary intake. Moreover, constant monitoring of heavy metals is needed on all food commodities in order to evaluate if any potential health risks from heavy metal exposure do exist, to assure food safety, and to protect the end user from food that might injure their health.

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