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Metal Speciation in Sediment and Their Bioaccumulation in Fish Species of Three Urban Rivers in Bangladesh

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Abstract Six trace metals (chromium [Cr], nickel [Ni], copper [Cu], arsenic [As], cadmium [Cd] and lead [Pb]) were measured in sediments and soft tissues of three commonly consumed fish species (*Channa punctatus*, *Heteropneustes fossilis,* and *Trichogaster fasciata*) collected from three urban rivers around Dhaka City, Bangladesh. The abundance of total metals in sediments varied in the decreasing order of Cr > Ni > Pb > Cu > As > Cd. Sequential extraction tests showed that the studied metals were predominantly associated with the residual fraction followed by the organically bound phase. The range of metal concentration in fish species were as follows: Cr (0.75–4.8), Ni (0.14–3.1), Cu (1.1–7.2), As (0.091–0.53), Cd (0.008–0.13), and Pb (0.052–2.7 mg/kg wet weight [ww]). The rank of biota-sediment accumulation factor for fish species were in the descending order of Cu > As > Pb > Ni > Cr > Cd. Metal concentrations in fish exceeded the international permissible standards suggesting that these species are not safe for human consumption.

Trace-metal pollution in the environment has become a concern due to the ever-increasing contamination of water, soil, and food in many regions of the world (Tang *et al*. 2013; Sow *et al*. 2013; Rahman *et al*. 2014). This pollution not only poses a threat to public water supplies, it also poses an ecological and human health risk through the consumption of aquatic products (Waqar 2006; Terra *et al*. 2008). Therefore, studies on trace-metal pollution in fish are important for characterizing health risks (Asuquo *et al*. 2004). Studies have shown that urban and industrial development contributes to metal contamination in freshwater environments (Xia *et al*. 2011; Tao *et al*. 2012). Bangladesh is one of the largest delta regions in the world, formed by the Ganges, Brahmaputra, and Meghna rivers and randomly spreading across five countries, namely, Bhutan, Nepal, China, India, and Bangladesh (Sharif *et al*. 1993). Dhaka is the capital of Bangladesh and has many industries, built with limited planning, that discharge large amounts of untreated effluent into the adjacent rivers

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(Turag, Buriganga, and Shitalakha). This presents a concern in terms of the health of local aquatic ecosystems and the people inhabiting the area. Few studies have evaluated the accumulation and contamination of trace metals in these rivers (Ahmad *et al*. 2010; Alam *et al*. 2003). In recent decades, the accumulation and contamination of trace metals in these rivers have been paid less attention. These rivers are increasingly being polluted with industrial and domestic effluents (Islam *et al*. 2006; Ahmad *et al*. 2010). Trace metals have accumulated in the surface sediments of these rivers (Alam *et al*. 2003; Islam *et al*. 2014). In addition, a considerable amount of trace metals can be accumulated in the surface sediments of these rivers. In sediment, trace metals are present in a number of chemical forms, and they generally exhibit different physical and chemical behaviors in terms of chemical interaction, mobility, biological availability, and potential toxicity (Akcay *et al*. 2003; Singh *et al*. 2005). During sediment transportation, trace metals undergo various changes in their speciation due to dissolution, precipitation, sorption, and complexation phenomena (Dassenakis *et al*. 1998; Akcay *et al*. 2003; Abdel-Ghani & Elchaghaby 2007), all which affect their behavior and bioavailability (Nouri *et al*. 2011). The overall behavior of trace metals in the aquatic environment is strongly influenced by the association of metals with various geochemical phases (Morillo *et al*. 2004).

It is now widely accepted that the role of sediments as a sink for metal pollutants cannot be fully assessed by measuring the total metal concentration (Chandra Sekhar *et al*. 2003). Nevertheless, information on total concentration is not sufficient to assess the environmental impact of contaminated sediments (Chandra Sekhar *et al*. 2003). Therefore, a particular interest is focused on geochemical speciation in assessing the potential environmental impacts and ecotoxicity of trace metals (Sin *et al*. 2001). Chemical speciation can be defined as the process of identification and quantification of different species, forms, or phases of chemicals present in the environment (Abdallah 2007). Metal speciation in sediment is expected to influence metal bioavailability and thereby metal content in biota, in particular in the soft tissues of fish (Pempkowiase *et al*. 1999; Yap *et al*. 2002). To assess the various sediment-bound trace metals into operationally defined fractions, sequential extraction schemes have been developed by several investigators (Tessier *et al*. 1979; Rauret *et al*. 1999; Pustisek *et al*. 2001; Svete *et al*. 2001; Sutherland & Tack 2003; Oyeyiola *et al*. 2011). Among them, the five-stage sequential extraction schemes developed by Tessier *et al*. (1979) is extensively used across the world. Sequential extraction techniques would provide the history of metal input, digenetic transformation within the sediments, and reactivity of trace-metal species of both natural and anthropogenic origin (Sundaray *et al*. 2011).

Previous studies on the Buriganga River have focused on the river water chemistry and the physicochemical properties in the river water (Ali *et al*. 2008; Moniruzzaman *et al*. 2009), and only a few studies have assessed the seasonal and spatial distribution of trace metals in the sediment and biota of peripheral rivers in Dhaka City (Ahmad *et al*. 2010; Alam *et al*. 2003; Mohiuddin *et al*. 2011). No detailed studies have been completed on the trace-metal geochemical speciation in sediments and bioaccumulation in fish species in these rivers. The present study was performed to determine the seasonal variation and geochemical fractionation of metals in the sediment and to assess bioaccumulation of metals in three fish species that are commonly eaten in this area. This research work was performed to determine the seasonal variation and the geochemical fractionation of metals in sediment as well as to assess the bioaccumulation of metals in three edible fish species.

Materials and Methods

Study Area

Dhaka, with a 815.8-km^2 metropolitan areas, is surrounded by three major river—the Turag, the Buriganga, and the Shitalakha (Fig. 1),—which are using as a convenient means for industrial wastes disposal. Dhaka is one of the most densely populated cities in the world with 12 million people of which <25 % are served by a sewage treatment facility (Ahmad *et al*. 2010; Mohiuddin *et al*. 2011). The indiscriminate dumping of domestic and industrial wastes, combined with the failure of authorities to enforce existing regulations to protect the ecological health of these rivers, has aggravated the situation to the point where these rivers are dying biologically and hydrologically. Therefore, these rivers were selected for this study, and the basic information of the study areas is listed in Supplementary Table S1.

Sample Collection and Preparation

Sampling was performed in February through March 2012 (winter) and again in August through September 2012 (summer). During winter, there is no rainfall, and river water levels decrease; during summer, river water levels increase due to heavy rainfall. At each sampling point, composite sediment samples were collected using standard protocol (United States Environmental Protection Agency [USEPA] 2001). River bed sediment samples (approximately 200 g) were taken at a depth of 0 to 5 cm using a portable Ekman grab sampler. Each sediment sample was obtained by mixing sediments randomly collected (three times) at each sampling point, and 18 pairs of composite

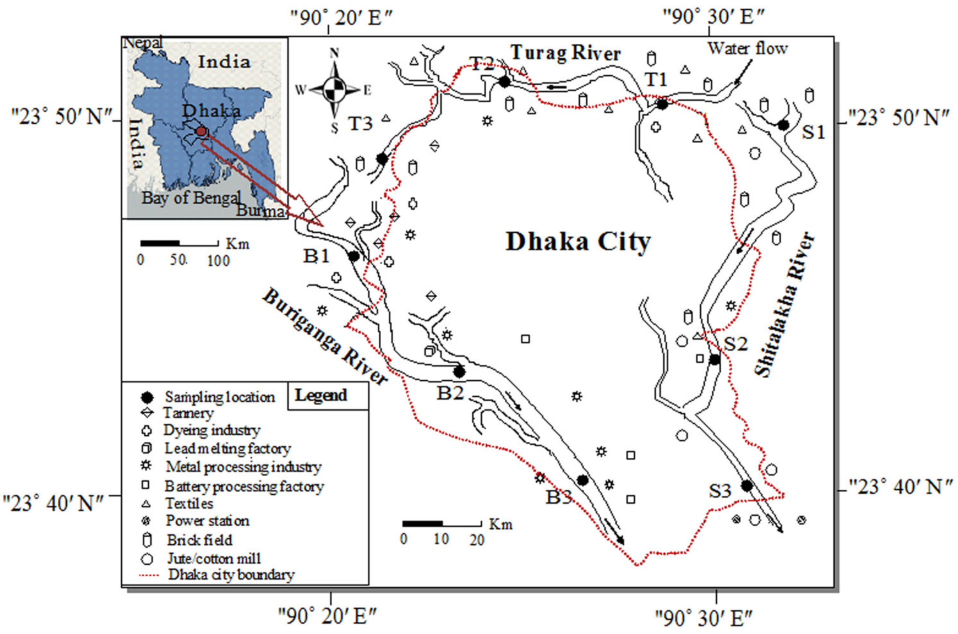

Fig. 1 Map showing sampling locations in Bangladesh

sediment samples were collected. Sediment samples were then freeze dried to obtain constant weight. The samples were homogenized by grinding in an agate mortar, sieved through 106-µm Aperture nylon sieve, and stored in labeled glass bottles until chemical analyses.

A total of 54 samples of 3 mostly consumable fish species—spotted snakehead (*Channa punctatus*), stinging catfish (*Heteropneustes fossilis*), and banded gourami (*Trichogaster fasciata*) —were collected from 3 rivers around Dhaka City, Bangladesh (Fig. 1). These three fish species can change their feeding habits and survive in polluted river conditions. *C. punctatus* is omnivorous—carnivorous and feeds on zooplankton, zoobenthos, and plants from the surface water body (Saikia *et al.* 2012). *H. fossilis* and *T. fasciata* are omnivorous and often feed on insects, larvae, crustaceans, algae, and detritus in sediment. Moreover, they live close to sediments (moreso than do *C. punctatus*). Thus, it is expected that these 3 fish species are differently exposed to metals and other contaminants. Each species of fish, i.e., 18 samples, were collected with the help of fishermen. Immediately after collection, fish samples were washed thoroughly with freshwater to remove mud or other fouling substances and put into plastic bag/containers; samples were then transported to the Department of Fisheries, Dhaka University, Bangladesh. After transportation to the laboratory, the fish samples were allowed to reach room temperature, and nonedible parts were removed with the help of a steam-cleaned stainless steel knife. In the present study, only fish muscles were evaluated for the elemental concentration because Bangladeshi people do not habitually consume the other parts, such as liver, kidneys, gills, and gonads. Muscle tissues of the fish samples were then washed with distilled water and cut into small pieces (2–3 cm) using the cleaned knife over a clean polyethylene sheet. Fish samples were then freeze dried to obtain constant weight and homogenized by grinding in an agate. The preprocessed samples were brought to Yokohama National University, Japan, for chemical analysis.

Analytical Methods for Physicochemical Parameters

Physicochemical parameters, such as pH, EC, % nitrogen (%N), and % carbon (%C), were measured. The pH of sediments was measured in a sediment-to-water ratio of 1:2.5. The sediment/deionized water mixture was stirred with a clean glass rod, and the slurry was allowed to equilibrate for 30 min (Ikem & Adisa 2011; Ahmed *et al.* 2012). The pH values were recorded using a Horiba U-23 instrument with the calibration of pH 4 and pH 7 standards. For electrical conductivity (EC) determination, 5.0 g of sediment was taken in 50-mL polypropylene tubes. Then 30 mL of distilled water was added to the tube. The lid was closed properly and was shaken for 5 min. After that, EC was measured using an EC meter (Horiba D-52) (modification after Niwa *et al.* 2011). Percent N and C of sediment was measured using an elemental analyzer (vario EL III; Elenemtar, Germany) at Yokohama National University, Japan. For N and C determination, sediment samples were weighed in tin or silver vessels and loaded into the integrated carousel. In a fully automatic process, transfer of the sample through the ball valve into the combustion tube was performed. Each sample was individually flushed with carrier gas to remove atmospheric N resulting in a zero blank sampling process. The catalytic combustion was performed at a permanent temperature of $\leq$1,200°C. The element concentration from the detector signal and the sample weight were measured on the basis of stored calibration curves. Sediment texture was determined by the hydrometer method (Ikem & Adisa 2011).

Sample Digestion and Metal Extraction

All chemicals were analytical-grade reagents, and Milli-Q (Elix UV5 and MilliQ, Millipore, USA) water was used for solution preparation. The Teflon vessel and polypropylene containers were cleaned, soaked in 5 % HNO_3 for >24 h, then rinsed with Milli-Q water and dried. Samples were digested in a microwave digestion system (Berghof-MWS2; Berghof Speedwave, Germany). The microwave digestion system was designed to performed chemical digestion procedures under extreme pressure and temperature conditions. Digestion reagents used were 5 mL of 69 % HNO_3 acid (Kanto Chemical Co, Japan) and 2 mL of 30 % H_2O_2 (Wako Chemical Co, Japan). The weighed samples (0.3 g of powdered fish) were then placed into the digestion reagent in a Teflon vessel. DAP-60 K type pressure vessels (Berghof, Germany), which are made entirely of tetrafluoromethoxylene, were used in this study. Three step-digestion procedures were followed: (1) temperature and power were maintained at 180°C and 85 %, respectively, for 15 min; (2) temperature was kept steady at 190°C for 15 min together with 90 % power; and (3) decreased temperature (100°C) and power (40 %) were used for 10 min to cool down the Teflon vessels (maximum microwave power is 1000 W when power is 100 %). After that, all vessels were kept in cold water to decrease the residual pressure inside the Teflon vessel. After digestion, the solution was then filtered using DISMIC-25HP polytetrafluoroethylene syringe filter (pore size 0.45 µm) (Toyo Roshi Kaisha, Ltd., Japan) and stored in 50-mL polypropylene centrifuge tubes (Nalgene, New York). Afterward, the vessels were cleaned with Milli-Q water and air-dried. Finally, blank digestion with 5 mL 69 % HNO_3 after the described digestion procedures was performed to clean up the digestion vessels (Berghof's Product User Manual 2008).

For chemical partitioning of metals, sediment samples were analyzed using the Tessier sequential chemical extraction procedure (Tessier *et al*. 1979). The sequential extraction procedure was divided into five operationally defined chemical fractions: (F1) the exchangeable fraction (readily soluble and exchangeable); (F2) the carbonate bound and specifically adsorbed fraction (carbonate-bound, specifically adsorbed and weak organic and inorganic complexes); (F3) the iron (Fe)–manganese (Mn) oxides fraction (bound to Fe and Mn oxides (Fe–Mn oxides); (F4) the organic/sulphide fraction (bound to stable organic and/or sulphide [organic] complexes); and (F5) the residual fraction (held in primary and secondary minerals within their crystal structure). The detailed geochemical fractionation procedure of sediment is presented in Fig. 2.

Instrumental Analysis and Quality Assurance

For trace metals, samples were analyzed using inductively coupled plasma mass spectrometer (Agilent 7700 series). Multielement standard XSTC-13 (Spex CertiPrep, USA) solutions were used to prepare the calibration curve. The calibration curves with $R^2 > 0.999$ were accepted for concentration calculation. Before starting the sequence, relative SD ([RSD] <5 %) was checked by using tuning solution purchased from Agilent. Internal calibration standard solutions containing 1.0 mg/L of indium, yttrium, beryllium, tellurium, cobalt, and thallium were purchased from Spex CertiPrep, USA. Multielement solution (Agilent), 1.0 µg/L, was used as a tuning solution covering a wide range of masses of elements. All test batches were evaluated using an internal quality approach and validated if they satisfied the defined internal quality controls. For each experiment, a run included blank, certified reference materials (CRMs), and samples, all of which were analyzed in duplicate to eliminate any batch-specific errors. The CRMs (NMIJ CRM 7303–lake sediment and DORM-2–dogfish muscle from the National Research Council, Canada) were analyzed to confirm analytical performance and good precision (RSD <20 %) of the applied method (Table S2).

Metal Bioaccumulation in Fish Species

Average metal concentrations in fish species and sediments from the studied rivers were used for calculating the (BSAF. The BSAF is an index of the ability of a fish species to accumulate a particular metal with respect to its concentration in sediment. It was calculated by the following equation (Abdallah and Abdallah 2008):

$$BSAF = \frac{C_{\text{fish}}}{C_{\text{sediment}}} \tag{1}$$

where C_{fish} is the metal concentration in fish (mg/kg dw), and C_{sediment} is the metal concentration in sediment (mg/kg dry weight [dw]).

Statistical Analysis

Data were statistically analyzed using the statistical package SPSS 16.0 (SPSS, USA). Means, SDs, and percentiles of metal concentrations in sediments and fish species were calculated. Multivariate *post hoc* Tukey tests were employed to examine the statistical significance of the differences among mean concentrations and BSAF of trace metals among the fish species.

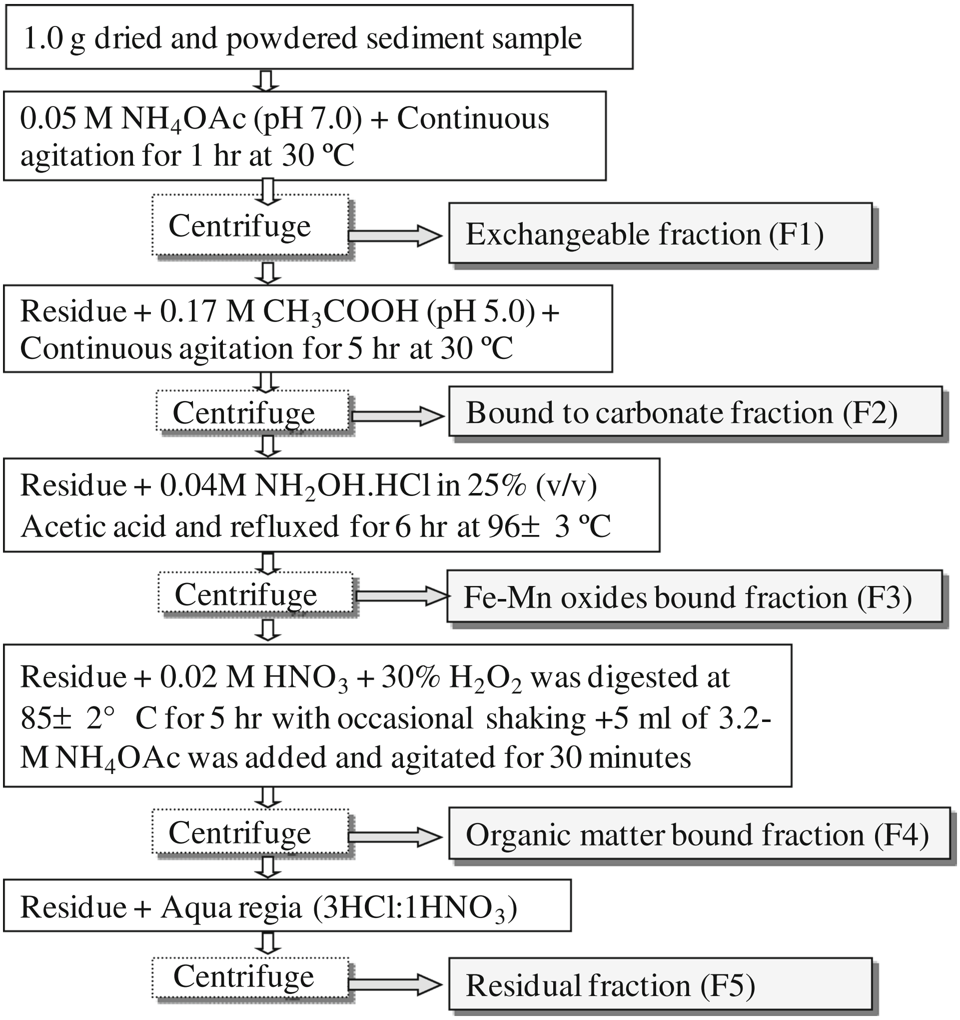

Fig. 2 Flow chart of the sequential extraction scheme used in the present study

Results and Discussion

Physicochemical Properties and Speciation of Metals in Sediment

The sites generally have pH ranging from 6.3 to 7.4 during winter and from 6.5 to 8.7 during summer, which is slightly acidic, except at the T1 site, which was alkaline (Table 1) due to the decomposition of organic matter and subsequent formation of carbonic acid (Ahmad *et al*. 1996). The composition of organic carbon in sediment samples was varied among the sites due to its origin in the aquatic environment. Organic carbon in sediments ranged from 0.18 % to 3.3 % (Table 1). The highest percentage of organic carbon might be attributed to the high amount of drainage water at site B3. According to the United States soil texture classification, textural analysis showed that the sediment belonged to the following classes: sandy loam, silt loam, silty clay loam, silty clay, and clay loam (Table 2). Fine-sediment textures are important for geochemical carriers, which can control metal bioavailability for the aquatic organisms exposed through dissolved metals.

The mobility and toxicity of metals are mainly dependent on metal speciation in the aquatic environment. The relative distribution of trace metals in different geochemical fractions are listed in Table 3 and Fig. 3. The metals associated with different fractions in sediments followed the descending order of Cr = residual > organic > Fe–Mn oxides > carbonate > exchangeable; Ni = residual > Fe–Mn oxides > organic > carbonate > exchangeable; Cu = organic > residual > carbonate > Fe–Mn oxides > exchangeable; As = residual > Fe–Mn oxides > organic > carbonate > exchangeable; Cd: residual > exchangeable > organic > carbonate > Fe–Mn oxides; and Pb = residual > organic > carbonate > Fe-Mn oxides > exchangeable. Considering the five defined chemical fractions, metals in sediments were greater in winter compared with summer (Table 3). Seasonal variation was due to (1 the lack of rainfall in winter, which could allow metal precipitation and resuspension and deposition (Kenniburgh *et al*. 1996; Islam *et al*. 2014) or (2) to changes in pH, redox conditions, and inorganic or

Table 1 Physicochemical properties as well as bioavailable and nonbioavailable metals in sediment during two different seasons

Sites	%Bioavailable		%Nonbioavailable		pH		Ec		%N		%C	
	Winter	Summer	Winter	Summer	Winter	Summer	Winter	Summer	Winter	Summer	Winter	Summer
T1	17	22	83	78	6.9	8.7	28	40	0.09	0.16	0.18	1.8
T2	12	14	88	86	6.9	7.2	40	5.4	0.17	0.15	2.8	1.6
T3	16	19	84	81	6.9	6.7	30	6.5	0.11	0.13	0.93	1.2
B1	19	18	81	82	6.3	6.5	41	26	0.17	0.23	1.4	1.6
B2	18	17	82	83	6.3	6.9	27	3.3	0.09	0.14	0.74	1.0
B3	18	16	82	84	7.2	6.7	13	24	0.31	0.05	3.3	0.51
S1	21	21	79	79	6.4	6.9	9.7	6.2	0.09	0.07	0.84	0.59
S2	24	24	76	76	6.5	6.9	17	10	0.11	0.13	0.89	1.1
S3	28	26	72	74	7.4	7.2	39	14	0.09	0.18	0.66	1.6

Table 2 Textural class of sediments collected from three rivers around Dhaka City, Bangladesh

Sites	Sediment texture (%)						Type[a]
	Sand		Silt		Clay		
	Winter	Summer	Winter	Summer	Winter	Summer	
T1	17	20	54	43	29	37	Silt loam
T2	16	10	60	64	24	26	Silty clay loam
T3	56	49	28	26	16	25	Sandy loam
B1	71	58	17	25	12	17	Sandy loam
B2	76	67	18	23	6.0	10	Sandy loam
B3	68	66	19	7.0	13	27	Sandy loam
S1	8.0	13	50	54	42	33	Silty clay
S2	16	18	24	27	60	55	Clay loam
S3	6.0	9.0	61	55	33	36	Silty clay loam

[a] According to the United States Department of Agriculture soil classification system

Table 3 Total and fraction concentration of metals in sediments (mg/kg dw [minimum–maximum])

Metals	Exchangeable		Carbonates		Fe–Mn oxides		Organic		Residual		Total	
	Winter	Summer	Winter	Summer	Winter	Summer	Winter	Summer	Winter	Summer	Winter	Summer
Cr	10–50	4.4–27	5.0–99	15–40	20–122	10–274	45–997	28–595	256–1195	70–1107	395–2471	112–2039
Ni	1.4–4.1	0.82–16	2.8–99	15–63	64–121	27–114	42–117	34–130	110–335	74–236	242–606	139–522
Cu	2.6–12	1.7–9.9	7.5–25	4.4–27	3.4–12	2.8–23	80–280	16–111	42–135	25–65	127–405	65–205
As	0.65–1.1	0.32–1.6	2.1–5.5	1.8–3.8	6.1–13	2.8–15	4.7–10	0.74–6.8	20–34	10–30	33–58	16–46
Cd	3.1–9.3	1.9–6.3	1.9–5.1	1.1–3.7	0.74–2.3	0.50–3.9	1.5–5.2	1.5–4.8	4.9–9.9	2.2–7.6	14–29	8.5–26
Pb	5.1–49	3.7–59	20–400	6.0–312	13–120	5.2–346	17–462	11–483	22–815	19–598	70–1846	45–1756

organic complexation (Christl *et al.* 2001; Liang & Wong 2003; El Nemr *et al.* 2006; Bastami *et al.* 2012). Slightly greater level of metals in winter might be attributed to the variation in water capacity of the river when water input to the river is generally limited in winter, thus resulting in the precipitation of metals in sediment.

Chemical fractionation differentiates metals those derived from natural or anthropogenic sources. In general, results of sequential extraction indicated that the residual fraction dominated Cr (48–74 %), Ni (29–65 %), As (40–66 %), and Pb (21–44 %) contents during winter and Cr (34–63 %), Ni (38–49 %), As (43–62 %), and Pb (25–42 %) content during summer. This result suggested that Cr, Ni, As, and Pb had the strongest associations to the crystalline sedimentary components and were likely to be reflected by the geological characteristics (Hamilton *et al.* 1984). Cr, Ni, As, and Pb were the least mobilisable because considerable proportions of these metals were in the nonmobile fraction. This further indicates their less availability to the aquatic fauna and that they have less

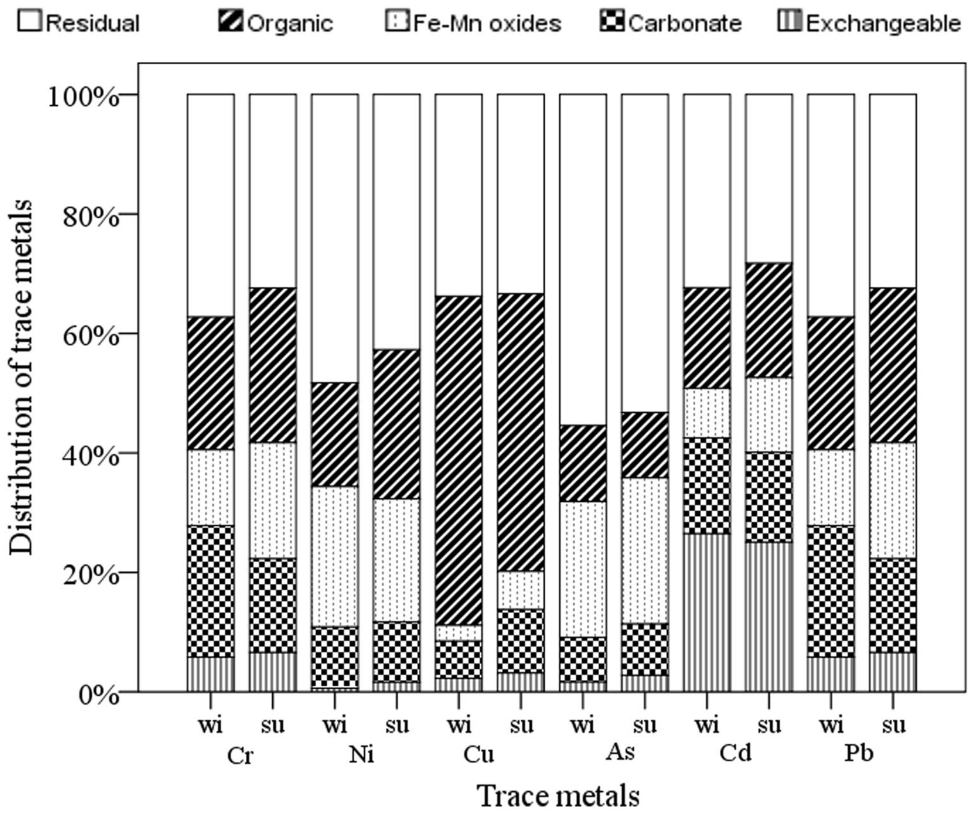

Fig. 3 Relative distribution of metals in sediment of different chemical fractions

chance of entering into the food chain (Chandra Sekhar *et al*. 2003).

According to the partitioning pattern, a considerable proportion of Cd (26 % during winter and 25 % during summer) was associated with the exchangeable fraction (Fig. 3) and can be more mobile in sediments (Zhang *et al*. 2012). The highest proportion of Cd was bounded for the sum of exchangeable and carbonate fractions (approximately 40 %), thus placing the availability of Cd into the high-risk category, meaning that it could easily enter the food chain (Sundaray *et al*. 2011). Metals such as Cd and Pb represent an appreciable portion in the carbonate phase because these metals have a special affinity toward carbonate and may coprecipitate with its minerals. Substantial proportions of Cd (16 % in winter and 15 % in summer) and Pb (24 % during winter and 16 % during summer) were found in the carbonate-bound fraction (Fig. 3). This result suggests that Cd and Pb can be easily released back into the water column, which may cause secondary pollution and at the same time potential effects on the fish themselves or the organisms that consume them, including upper-trophic level organisms and humans (Burger & Gochfeld 2005; Dhanakumar *et al*. 2013). Numerous earlier reports have also considered the exchangeable fraction to be the most mobile and bioavailable phase present in sediments followed by the carbonate-bound fraction (Tessier *et al*. 1979; Ahumada *et al*. 1999; Howari & Banat 2001). Slight variations were observed in labile (exchangeable and carbonate-bound) and lithogenic fractions, which could be attributed to numerous factors, such as weathering, mineral transport, anthropogenic inputs, and physicochemical components of sediment (Sundaray *et al*. 2011). The lower association of Cr (1.2 to 19 % during winter and 1.9 to 13 % during summer) in the carbonate fraction of sediments might arise due to the inability of Cr^{3+} to form a precipitate or complex with carbonates (Sundaray *et al*. 2011).

Fe–Mn oxides and organic matter plays excellent scavenging role for the removal of trace metals from the water column in aquatic environments (Dhanakumar *et al*. 2013). Metals associated with Fe–Mn oxides fraction can be mobilized by changes in the redox potential of the sediment and become bioavailable in special fish that feed on sediments and/or sediment-associated benthic biota. Among the nonlithogenic fractions, Fe–Mn oxyhydroxide is the scavenger for Ni, As, and Pb (Fig. 3). This attributes to the adsorption, flocculation, and coprecipitation of trace metals with the colloids of Fe and Mn oxyhydroxide (Rath *et al*. 2009). Furthermore, organic-bound Cr, Cu, and Pb seem to be the second dominant fraction among the nonlithogeneous fractions. Cu was identified as the highest proportion (45 to 61 % during winter and 24 to 58 %

Table 4 Metal concentration in fish species (mg/kg ww) of three rivers in Bangladesh ($n = 3$)

Sites	Fish species		Cr		Ni		Cu	
			Winter	Summer	Winter	Summer	Winter	Summer
Turag River	*C. punctatus*	Mean ± SD	2.5 ± 0.55^{a}	1.3 ± 0.29^{a}	0.44 ± 0.08^{a}	0.18 ± 0.04^{a}	1.9 ± 0.35^{a}	1.2 ± 0.08^{a}
		Range	1.9–2.9	1.0 –1.6	0.37–0.53	0.14–0.22	1.5–2.2	1.1–1.3
	H. fossilis	Mean ± SD	2.4 ± 0.25^{a}	1.5 ± 0.05^{a}	1.1 ± 0.16^{b}	0.91 ± 0.10^{b}	5.1 ± 0.67^{b}	3.4 ± 0.41^{b}
		Range	2.2–2.7	1.5–1.6	0.99–1.3	0.80–1.0	4.4–5.7	3.1–3.9
	T. fasciata	Mean ± SD	2.9 ± 0.67^{a}	2.3 ± 0.08^{b}	2.5 ± 0.20^{c}	2.3 ± 0.23^{c}	3.7 ± 0.38^{c}	2.2 ± 0.27^{c}
		Range	2.3–3.6	2.2–2.4	2.3–2.7	2.1–2.6	3.3–4.0	1.9–2.5
Buriganga River	*C. punctatus*	Mean ± SD	$3.5 \pm 0.47^{a,b}$	1.4 ± 0.39^{a}	2.9 ± 0.02^{a}	1.5 ± 0.26^{a}	$5.5 \pm 0.35^{a,b}$	2.7 ± 0.69^{a}
		Range	3.1–4.0	1.0–1.8	2.8–2.9	1.3–1.8	5.2–5.9	2.3–3.5
	H. fossilis	Mean ± SD	2.8 ± 0.93^{a}	$2.1 \pm 0.44^{a,b}$	$3.0 \pm 0.018^{b,c}$	1.6 ± 0.29^{a}	6.0 ± 1.1^{a}	4.4 ± 0.41^{b}
		Range	1.8–3.6	1.6–2.5	2.9–3.0	1.3–1.9	5.2–7.2	4.2–4.9
	T. fasciata	Mean ± SD	4.5 ± 0.24^{b}	2.5 ± 0.38^{b}	3.0 ± 0.04^{c}	2.4 ± 0.12^{b}	4.1 ± 0.56^{b}	$4.1 \pm 0.50^{a,b}$
		Range	4.3–4.8	2.3–3.0	3.0–3.1	2.3–2.5	3.5–4.5	3.7–4.7
Shitalakha River	*C. punctatus*	Mean ± SD	2.3 ± 0.26^{a}	1.1 ± 0.11^{a}	0.69 ± 0.28^{a}	0.55 ± 0.29^{a}	4.2 ± 0.5^{a}	2.5 ± 0.5^{a}
		Range	2.1–2.6	1.0–1.2	0.40–0.95	0.29–0.86	3.8–4.7	2.3–3.1
	H. fossilis	Mean ± SD	$2.9 \pm 0.64^{a,b}$	1.1 ± 0.38^{a}	$1.5 \pm 0.12^{a,b}$	0.94 ± 0.06^{a}	5.8 ± 0.20^{b}	3.6 ± 0.44^{a}
		Range	2.4–3.6	0.75–1.5	1.4–1.6	0.89–1.0	5.7–6.0	3.1–3.9
	T. fasciata	Mean ± SD	3.6 ± 0.42^{b}	1.4 ± 0.42^{a}	1.8 ± 0.67^{b}	1.4 ± 0.53^{a}	3.0 ± 0.48^{c}	3.8 ± 1.4^{a}
		Range	3.2–4.0	1.1–1.9	1.1–2.4	0.85–1.9	2.5–3.4	2.3–5.0
Permissible limits (FAO/WHO 2004)			1.0		0.9		4.5	

Sites	Fish species		As		Cd		Pb	
			Winter	Summer	Winter	Summer	Winter	Summer
Turag River	*C. punctatus*	Mean ± SD	0.25 ± 0.02^{a}	0.093 ± 0.002^{a}	0.011 ± 0.003^{a}	0.008 ± 0.000^{a}	0.63 ± 0.12^{a}	0.13 ± 0.11^{a}
		Range	0.24–0.28	0.091–0.094	0.008–0.013	0.007–0.008	0.49–0.72	0.052–0.26
	H. fossilis	Mean ± SD	0.28 ± 0.02^{a}	$0.14 \pm 0.008^{a,b}$	0.026 ± 0.004^{b}	$0.024 \pm 0.001^{b,c}$	1.3 ± 0.08^{b}	$0.65 \pm 0.14^{a,b}$
		Range	0.27–0.30	0.13–0.15	0.023–0.03	0.023–0.025	1.2–1.4	0.53–0.80
	T. fasciata	Mean ± SD	0.29 ± 0.11^{a}	0.28 ± 0.12^{b}	0.015 ± 0.002^{a}	0.025 ± 0.006^{c}	1.5 ± 0.05^{c}	0.85 ± 0.41^{b}
		Range	0.22–0.41	0.20–0.42	0.014–0.018	0.019–0.030	1.5–1.6	0.41–1.2
Buriganga River	*C. punctatus*	Mean ± SD	$0.30 \pm 0.03^{a,b}$	0.092 ± 0.001^{a}	0.025 ± 0.002^{a}	0.044 ± 0.014^{a}	1.4 ± 0.16^{a}	0.81 ± 0.02^{a}
		Range	0.26–0.32	0.091–0.092	0.022–0.026	0.028–0.053	1.2–1.5	0.78–0.83
	H. fossilis	Mean ± SD	0.34 ± 0.04^{a}	$0.24 \pm 0.019^{a,b}$	0.034 ± 0.01^{a}	0.053 ± 0.000^{a}	$1.4 \pm 0.10^{a,b}$	$1.0 \pm 0.07^{a,b}$
		Range	0.32–0.39	0.23–0.26	0.026–0.045	0.052–0.53	1.3–1.5	0.91–1.0
	T. fasciata	Mean ± SD	0.28 ± 0.01^{b}	0.36 ± 0.14^{b}	0.049 ± 0.018^{a}	0.095 ± 0.034^{a}	2.5 ± 0.26^{c}	1.2 ± 0.18^{b}
		Range	0.27–0.28	0.28–0.53	0.033–0.068	0.060–0.13	2.2–2.7	1.0–1.3

Table 4 continued

Sites	Fish species		As		Cd		Pb	
			Winter	Summer	Winter	Summer	Winter	Summer
Shitalakha River	*C. punctatus*	Mean ± SD	0.28 ± 0.05^{a}	0.12 ± 0.005^{a}	0.036 ± 0.009^{a}	0.031 ± 0.009^{a}	0.64 ± 0.26^{a}	0.16 ± 0.04^{a}
		Range	0.23–0.33	0.12–0.13	0.026–0.043	0.020–0.038	0.35–0.79	0.13–0.20
	H. fossilis	Mean ± SD	0.37 ± 0.07^{a}	$0.22 \pm 0.01^{b,c}$	$0.024 \pm 0.01^{a,b}$	$0.040 \pm 0.01^{a,b}$	1.2 ± 0.29^{a}	$1.0 \pm 0.14^{b,c}$
		Range	0.29–0.43	0.21–0.22	0.011–0.034	0.029–0.051	0.85–1.4	0.88–1.1
	T. fasciata	Mean ± SD	0.37 ± 0.04^{a}	0.22 ± 0.03^{c}	0.014 ± 0.002^{b}	0.071 ± 0.019^{b}	1.3 ± 0.36^{a}	0.69 ± 0.29^{c}
		Range	0.34–0.41	0.20–0.25	0.012–0.016	0.049–0.086	1.1–1.7	0.40–1.0
Permissible limits (FAO/WHO 2004)			1.0		0.1		0.5	

Vertically, letters a, b, and c indicate statistically significant differences (*$P < 0.05$*) among fish species from each river

during summer) in the organic fraction. This phenomenon can be explained by the affinity of metals to organic matter, especially humic substances, which are the component of natural organic matter and chemically active in complexing metals (Fytianos & Lourantou 2004; Jain *et al.* 2008). In addition, organic substances are known to exhibit a high degree of preferential selection for divalent ions (Stone & Marsalek 1996). Cu can be retained by sediment through exchange and specific adsorption, but precipitation may also be an important mechanism of retention in polluted sediments. Cu is generally adsorbed to a greater extent than other metals; the high affinity of Cu^{2+} ions for soluble organic ligands may greatly increase their mobility in sediments (McLean & Bledsoe 1992).

It is recognized that sequential extraction technique enables the prediction of possible metal impact on biota in aquatic ecosystems. The exchangeable and carbonate fractions are considered to be weakly bounded. These may equilibrate with the aqueous phase, thus becoming more rapidly bioavailable and causing environmental toxicity (Morrison *et al.* 1996; Kim *et al.* 1998; Karbassi & Shankar 2005). The metal in sediment of Fe–Mn oxides, as well as the organic matter bound, can be mobilized when environmental conditions become increasingly reduced or oxidized (Karbassi & Shankar 2005). Metal present in the inert fraction, being of detrital and lattice origin or primary mineral phases, can be regarded as a measure of contribution by natural sources (Salmonas & Forstner 1980) such as metal content in rocks and parent materials (Yang *et al.* 2009; Yi *et al.* 2011), anthropogenic industrial activities such as tanneries and textile factories (Mohiuddin *et al.* 2011), vehicle and coal combustion emissions (Li *et al.* 2012), treatment of agricultural land with arsenical pesticides (Fu *et al.* 2014), and electroplating, production of Ni–Cd batteries, waste incineration, *etc.* (El Nemr *et al.* 2006; Islam *et al.* 2014). During our sampling, we observed unplanned tanning activities from 270 tanneries, leachates from defused Ni–Cd batteries, Cd-plated items, and lead smelting and lead products manufacturing at the sampling sites, which can be coherent with the presence of metals in sediment.

Metal Concentrations in Fish Species

Concentrations of Cr, Ni, Cu, As, Cd, and Pb in the muscles of three fish species are listed in Table 4. The ranking order of mean concentrations of trace metals in *C. punctatus* Cu (3.0) > Cr (2.0) > Ni (1.0) > Pb (0.63) > As (0.19) > Cd (0.026), in *H. fossilis* were Cu (4.7) > Cr (2.1) > Ni (1.5) > Pb (1.1) > As (0.27) > Cd (0.033), and in *T. fasciata* were Cu (3.5) > Cr (2.9) > Ni (2.2) > Pb (1.3) > As (0.30) > Cd (0.045) (mg/kg ww). However, as a whole, the concentration of studied metals among the fish

Table 5 Correlation of metal concentrations between fish and sediments

Metals	Sediment fish species Sediment *C. punctatus*	 Sediment *H. fossilis*	 Sediment *T. fasciata*
Cr	0.487*	0.339	0.42
Ni	0.560*	0.526*	0.447
Cu	0.433	0.623**	0.634**
As	0.297	0.329	−0.02
Cd	0.074	0.103	0.364
Pb	0.538*	0.289	0.344

* Correlation is significant at the $P = 0.05$ level (two-tailed)

** Correlation is significant at the $P = 0.01$ level (two-tailed)

species were in the descending order of *T. fasciata* > *H. fossilis* > *C. punctatus*. Concentrations of metals varied considerably among the fish species by season, which is influenced by the age (size), growth cycle, and feeding habits of the species (Yilmaz 2005; Al Sayegh *et al*. 2012). In addition, the differences noted in metal concentrations in tissues of fish species between seasons could have been the result of local pollution (Dural *et al*. 2007). In the present study, relatively low levels of As, Cd, and Pb were found in fish, which can be due to lower levels of Cd in the environment as well as a greater proportion in the nonmobile forms of As and Pb (Campbell *et al*. 2005; Mazej *et al*. 2010).

In the present study, the greatest mean concentration of Cr was observed in *T. fasciata* of Buriganga River (4.5 and 2.5 mg/kg ww in winter and summer, respectively) (Table 4), which can be attributed to the wastewater coming from various industries such as dyeing and tanneries (Mohiuddin *et al.* 2011). Considering both seasons, the greatest mean concentrations of Ni were found in *T. fasciata* and *H. fossilis* (3.0 mg/kg ww) of Buriganga River (Table 4), which is due to the fact that Ni and its salts are used in several industrial applications, such as electroplating, storage batteries, automobiles, aircraft parts, sparks, electrodes, cooking utensils, pigments, lacquer cosmetics, water, and fabric printing (Rahman *et al.* 2012). Although the present study depicted that the examined fish species were not highly contaminated by Cr and/or Ni, long-term discharge of untreated industrial wastes could pollute riverine organisms. Although considerable proportions of these metals were found in the residual form, these are cumulative body poisons so their concentrations should remain as low as possible. Cu was detected in all examined fish samples, and its concentration ranged from 1.1 to 7.2 mg/kg ww with the highest mean content found in *H. fossilis* (6.0 mg/kg ww) during winter and the lowest mean content in *C. punctatus* (1.2 mg/kg ww) during summer (Table 4). Mean concentrations of As in fish species were observed 0.22–0.43 and 0.091–0.53 mg/kg ww during winter and summer, respectively. Differences in As concentration in the fish species were examined and showed that *T. fasciata* had higher concentrations than other two species ($P < 0.05$) (Table 4).

The fish species from three different rivers varied significantly for metal concentrations, wherein Buriganga River showed greater concentrations than did the other two rivers. This study has provided the evidence that effluents discharged from tanneries, dyeing, metal processing, batteries and auxiliary industries, and urban sewage system were the main sources of Cr in the river systems of the Hazaribagh area of southwestern Dhaka City, Bangladesh (Ahmad *et al*. 2010; Mohiuddin *et al*. 2011; Rahman *et al*. 2012; Islam *et al*. 2014). Concentrations of trace metals in fish samples were greater than the permissible levels in fish per Food and Agriculture Organization/World Heath Organization (FAO/WHO) guidelines (Table 4), thus indicating that consumption of these fish species might pose toxic effects on human health.

Correlation analysis of metal concentrations between fish species and sediment indicated the significant relationship between *C. punctatus* and sediment and *H. fossilis* and sediment for Cr, Ni, Cu, and Pb. Meanwhile, no

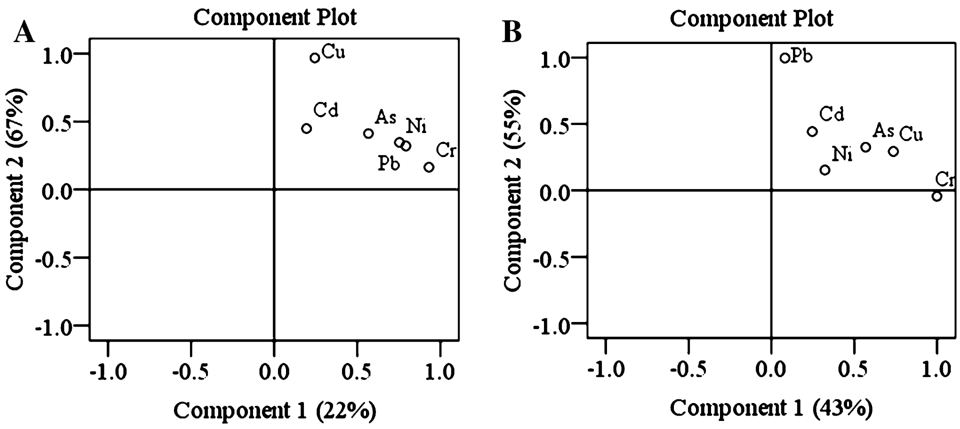

Fig. 4 PCA loading plot of the studied metals in **a** fish and **b** sediments from three rivers (Turag, Burigangam, and Shitalakha) around Dhaka City, Bangladesh

Table 6 Biota-sediment accumulation factors (BSAFs) of metals in fish species (two-season average)

Metal	Geochemical fraction	BSAF of *C. punctatus*			BSAF of *H. fossilis*			BSAF of *T. fasciata*		
		Mean	SD	CV	Mean	SD	CV	Mean	SD	CV
Cr	F1	0.56	0.21	0.37	0.56	0.25	0.44	0.76	0.34	0.44
	F2	0.27	0.16	0.59	0.28	0.203	0.72	0.37	0.24	0.64
	F3	0.24	0.093	0.39	0.24	0.108	0.45	0.33	0.15	0.46
	F4	0.066	0.029	0.44	0.070	0.042	0.61	0.093	0.057	0.61
	F5	0.030	0.009	0.28	0.031	0.015	0.47	0.042	0.018	0.43
	Total Cr	0.017	0.005	0.31	0.018	0.008	0.46	0.024	0.011	0.45
Ni	F1	1.3	1.1	0.80	2.2	1.2	0.54	3.1	1.7	0.53
	F2	0.12	0.079	0.66	0.19	0.093	0.48	0.30	0.23	0.77
	F3	0.051	0.038	0.74	0.074	0.025	0.34	0.11	0.038	0.36
	F4	0.056	0.042	0.74	0.083	0.029	0.35	0.123	0.056	0.45
	F5	0.029	0.027	0.95	0.040	0.023	0.57	0.053	0.021	0.40
	Total Ni	0.012	0.010	0.79	0.018	0.007	0.39	0.025	0.009	0.38
Cu	F1	2.8	1.6	0.58	3.2	0.94	0.30	2.6	1.2	0.45
	F2	0.84	0.26	0.31	1.06	0.38	0.36	0.89	0.51	0.57
	F3	1.9	1.3	0.68	2.2	0.7	0.33	1.8	0.84	0.47
	F4	0.13	0.07	0.52	0.16	0.053	0.33	0.12	0.04	0.35
	F5	0.21	0.11	0.56	0.23	0.070	0.30	0.18	0.06	0.32
	Total Cu	0.073	0.038	0.52	0.086	0.024	0.28	0.068	0.021	0.30
As	F1	1.4	0.54	0.40	1.5	0.62	0.41	1.5	0.54	0.37
	F2	0.36	0.13	0.36	0.41	0.16	0.40	0.40	0.14	0.35
	F3	0.11	0.031	0.27	0.13	0.040	0.31	0.13	0.062	0.47
	F4	0.35	0.34	0.98	0.42	0.42	1.0	0.36	0.29	0.81
	F5	0.049	0.013	0.27	0.054	0.012	0.22	0.057	0.026	0.45
	Total As	0.028	0.006	0.23	0.031	0.007	0.23	0.032	0.013	0.41
Cd	F1	0.016	0.006	0.36	0.019	0.011	0.59	0.018	0.010	0.54
	F2	0.027	0.010	0.37	0.033	0.020	0.62	0.031	0.018	0.57
	F3	0.048	0.036	0.74	0.044	0.023	0.53	0.043	0.018	0.42
	F4	0.024	0.011	0.45	0.027	0.015	0.55	0.026	0.012	0.48
	F5	0.015	0.009	0.58	0.015	0.007	0.46	0.014	0.006	0.45
	Total Cd	0.005	0.002	0.50	0.005	0.002	0.47	0.005	0.002	0.46
Pb	F1	0.21	0.16	0.73	0.35	0.28	0.78	0.46	0.34	0.75
	F2	0.088	0.052	0.59	0.15	0.10	0.68	0.19	0.12	0.65
	F3	0.10	0.072	0.71	0.18	0.12	0.70	0.22	0.15	0.69
	F4	0.095	0.064	0.67	0.16	0.097	0.60	0.21	0.14	0.66
	F5	0.056	0.036	0.63	0.096	0.055	0.58	0.12	0.076	0.62
	Total Pb	0.019	0.012	0.68	0.032	0.020	0.48	0.040	0.027	0.48

relationship was observed (except for Cu) between *T. fasciata* and sediment (Table 5). Multivariate principal component analysis (PCA) was performed to establish the relationship between the sources and levels of metals in sediments and fish. PCA analysis incorporated the six metal concentration data of all three rivers and explored the possible similar distribution pattern of metals. In our study, two PCs extracted approximately 89 % (PC1 22 %; PC2 67 %) and 98 % (PC1 43 %; PC2 55 %) of the metal variance for fish and sediment, respectively (Fig. 4). It was estimated that all metals were strongly associated with PC2 (positive loading), mainly existing in the nonbioavailable fractions (>70 %), thus reflecting their lithogenic origin. Hence, PC2 was supposed to reflect the contribution of natural geological sources of metals into the river. Subsequently, the PCA results showed that PC1 was associated with Cu, Cd, and Pb, which might represent anthropogenic sources of metal pollution.

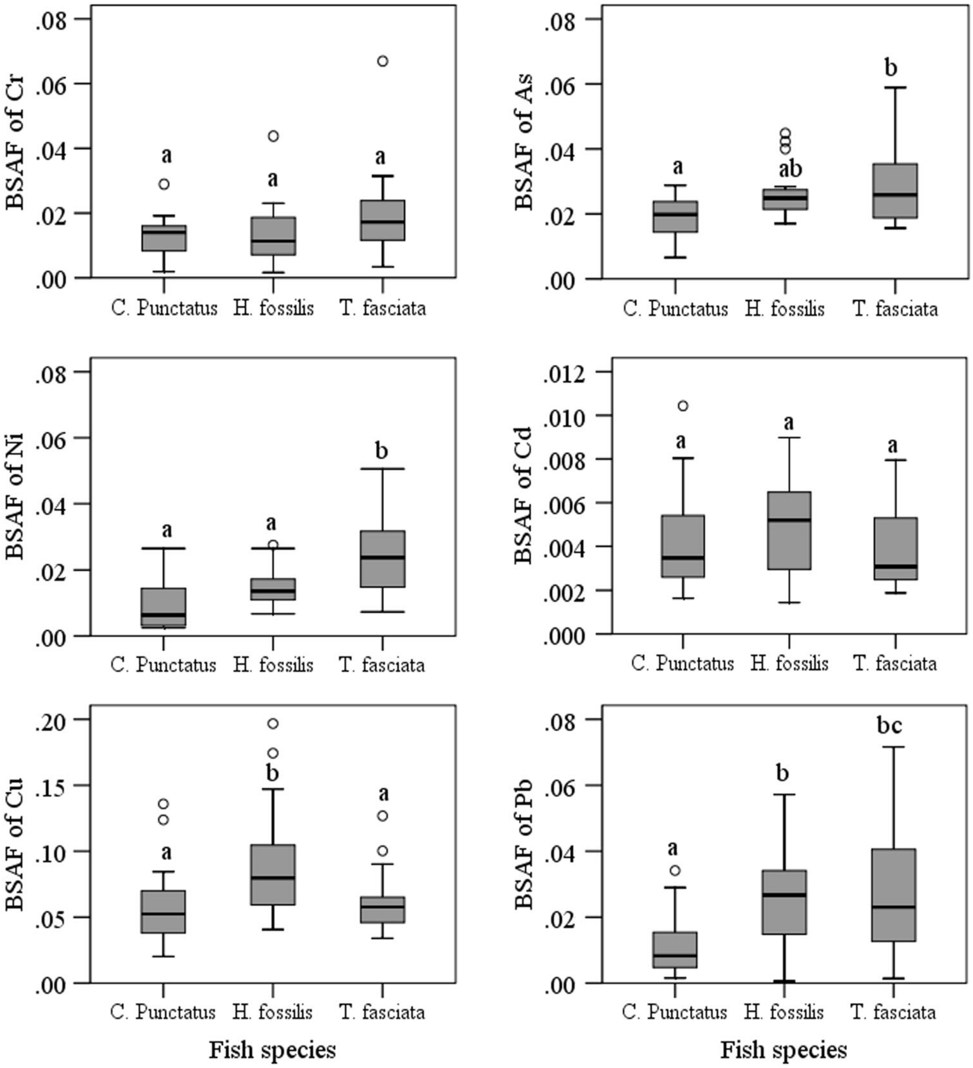

Fig. 5 BSAF of trace metals in fish species. Different letters indicates significant differences ($P < 0.05$) among the fish species, and the *circle* indicates the outlier

Metal Bioaccumulation in Fish Species

Metals contained in sediment can be bioaccumulated in fish tissues (van der Oost *et al*. 2003; Yi *et al*. 2011). Bioaccumulation of a specific metal is not only dependent on metal exposure and its environment but also different physiological and biochemical activities through which a specific organism deals with metals (Luoma & Rainbow 2008). Hence, different organisms accumulate metals from the environment depending on their filtration rate, ingestion rate, gut fluid quality, and detoxification strategies (e.g., storage in nontoxic form or elimination) (Wang & Rainbow 2008). The accumulation of metals in muscle tissues of fish could have a direct impact on health throughout the food chain. Table 6 and Fig. 5 clearly show that large variations in BSAF were observed among different fish species and metals. The ranking order of mean BSAF values of metals for *C. punctatus* was Cu > As > Pb > Cr > Ni > Cd; for *H. fossilis* was Cu > Pb > As > Cr > Ni > Cd; and for *T. fasciata* was Cu > Pb > As > Cr > Ni > Cd (Table 6). Among the selected six metals, Cu showed the greatest BSAF value, thus suggesting a greater rate of accumulation in fish species. At some sites, metal levels might be high but accumulation is lower than expected due to metal complexation (Hare & Tessier 1996).

BSAF of metals in fish species is regarded to decrease from readily available to unavailable form due to the strength of reagents in the extraction sequence (Tessier & Campbell 1987). Hence, the exchangeable fraction indicated the mostly bioaccumulated form in fish species. BSAF values of metals were found in the following descending order: exchangeable > carbonate > Fe–Mn oxides > organic > residual (Table 6). Metals bound in the residual phase are unlikely to be reactive during sedimentation and thus have little potential bioavailability (Jones & Turki 1997). BSAF values for Ni, As, and Pb in *T.*

fasciata were significantly greater than those obtained for *C. punctatus* and *H. fossilis* (Fig. 5). This can be explained by their ingestion of sediment as well as omnivore-feeding behavior of *T. fasciata*, which may lead to the much greater BSAF value found in this study. Therefore, of the three fish species investigated in this study, *T. fasciata* can be used as a potential bioindicator for the contamination of trace metals in those riverine environments. The present study showed that slightly greater accumulations of metals were observed in two fish species (*T. fasciata* and *H. fossilis*). From the literature survey, it was noticed that the fish species of *T. fasciata* and *H. fossilis* are bottom dwelling, and therefore sediments could be the major sources of trace-metal accumulation in these fish species (Tao *et al.* 2012). Bottom dwelling fishes are found to exhibit greater concentration of trace metals than pelagic fishes (Gupta *et al.* 2009). Bioaccumulations of individual metals among the sampling sites were not similar in pattern due to environment-specific phenomenon. It was considered that ingested sediments found in the digestive tract of fish acted as acid ambients, which accelerated the bioaccumulation of greater metal concentrations than were expected.

Conclusion

Sequential extraction technique was applied for portioning of six trace metals in sediments of three urban rivers showing its association with various geochemical forms. The abundance of metals in chemical fractions was the following decreasing order: residual > organic > Fe–Mn oxides > carbonate > exchangeable. The concentration of trace metals in *T. fasciata* was slightly greater than that it *C. punctatus* and *H. fossilis*, which might be due to their mode of feeding behavior. Similarly, the calculated values of BSAF in *T. fasciata* were greater than in *C. punctatus* and *H. fossilis*, suggested that *T. fasciata* could be used as potential bioindicator for metal pollution. This study has shown that metals (Cr, Ni, Cu, Cd, and Pb) in the Turag, Buriganga, and Shitalakha rivers have accumulated in fish species (*C. punctatus*, *H. fossilis*, and *T. fasciata*) and exceed the permissible limits established by FAO/WHO (FAO/WHO), thus suggesting that these fish are not safe for consumption by local residents.

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References

Abdallah MAM (2007) Speciation of trace metals in coastal sediments of El-Mex bay south mediterranean sea–west of alexandria (Egypt). Environ Monit Assess 132:111–123

Abdallah MAM, Abdallah AMA (2008) Biomonitoring study of heavy metals in biota and sediments in the South Eastern coast of Mediterranean sea. Egypt. Environ Monit Assess 146:139–145

Abdel-Ghani NT, Elchaghaby GA (2007) Influence of operating conditions on the removal of Cu, Zn, Cd and Pb ions from wastewater by adsorption. Int J Environ Sci Technol 4(4):451–456

Ahmad S, Siddiqui EN, Khalid S (1996) Studies on certain physico chemical properties of soil of two freshwater ponds of Darbhanga. Environ Pollut 31:31–39

Ahmad MK, Islam S, Rahman S, Haque MR, Islam MM (2010) Heavy metals in water, sediment and some fishes of Buriganga River, Bangladesh. Int J Environ Res 4(2):321–332

Ahmed F, Bibi MH, Asaeda T, Mitchell CPJ, Ishiga H, Fukushima T (2012) Elemental composition of sediments in Lake Jinzai, Japan: assessment of sources and pollution. Environ Monit Assess 184:4383–4396

Ahumada I, Mendoza J, Navarrete E, Ascar L (1999) Sequential extraction of heavy metals in soils irrigated with wastewater. Commun Soil Sci Plant Anal 30:1507–1519

Akcay H, Oguz A, Karapire C (2003) Study of heavy metal pollution and speciation in Buyak Menderes and Gediz river sediments. Water Res 37:813–822

Al Sayegh PS, Grudnik ZM, Pokorny B (2012) Heavy metals and arsenic concentrations in ten fish species from the Šalek lakes (Slovenia): assessment of potential human health risk due to fish consumption. Environ Monit Assess 184:2647–2662

Alam AMS, Islam MA, Rahman MA, Siddique MN, Matin MA (2003) Comparative study of the toxic metals and nonmetal status in the major river system of Bangladesh. Dhaka Univ J Sci 51(2):201–208

Ali MY, Amin MN, Alam K (2008) Ecological health risk of Buriganga River, Dhaka, Bangladesh. Hydro Nepal 3:25–28

Asuquo FE, Ewa-Oboho I, Asuquo EF, Udo PJ (2004) Fish species used as biomarker for heavy metal and hydrocarbon contamination for Cross River, Nigeria. Environmentalist 24:29–37

Bastami KD, Bagheri H, Haghparast S, Soltani F, Hamzehpoor A, Bastami MD (2012) Geochemical and geo-statistical assessment of selected heavy metals in the surface sediments of the Gorgan Bay Iran. Mar Pollut Bull 64:2877–2884

Burger J, Gochfeld M (2005) Heavy metals in commercial fish in New Jersey. Environ Res 99:403–412

Campbell LM, Norstroom RJ, Hobson KA, Muir DCG, Backus S, Fisk AT (2005) Mercury and other trace elements in a pelagic Arctic marine food web (Northwater Polynya, Baffin Bay). Sci Total Environ 351–352:247–263

Chandra Sekhar K, Chary NS, Kamala CT, Suman Raj DS, Sreenivasa Rao A (2003) Fractionation studies and bioaccumulation of sediment-bound heavy metals in Kolleru lake by edible fish. Environ Int 29:1001–1008

Christl I, Milne CJ, Kinniburgh DG, Kretzschmar R (2001) Relating ion binding by fulvic and humic acids to chemical composition and molecular size. 2. Metal binding. Environ Sci Technol 35:2512–2517

Dassenakis M, Scoullos M, Foufa E, Krasakopoulou E, Pavlidou A, Kloukiniotou M (1998) Effects of multiple source pollution on a small Mediterranean river. Appl Geochem 13(2):197–211

Dhanakumar S, Rutharvel Murthy K, Solaraj G, Mohanraj R (2013) Heavy metal fractionation in surface sediments of the Cauvery River estuarine region, southeastern coast of India. Arch Environ Contam Toxicol 65:14–23

Dural M, Goksu MZL, Ozak AA (2007) Investigation of heavy metal levels in economically important fish species captured from the Tuzla lagoon. Food Chem 102:415–421

El Nemr A, Khaled A, ElSikaily A (2006) Distribution and statistical analysis of leachable and total heavy metals in the sediments of the Suez Gulf. Environ Monit Assess 118:89–112

FAO/WHO (2004) Safety evaluation of certain food additives and contaminants. WHO Food Additives Series No. 52. World Health Organization, Geneva, Switzerland

Fu J, Zhao C, Luo Y, Liu C, Kyzas GZ, Luo Y et al (2014) Heavy metals in surface sediments of the Jialu River, China: Their relations to environmental factors. J Hazard Mater 270:102–109

Fytianos K, Lourantou A (2004) Speciation of elements in sediment samples collected at lakes Volvi and Koronia, N. Greece. Environ Int 30:11–17

Gupta A, Rai DK, Pandey RS, Sharma B (2009) Analysis of some heavy metals in the riverine water, sediments and fish from river Ganges at Allahabad. Environ Monit Assess 157:449–458

Hamilton RS, Revitt DM, Warren RS (1984) Levels and physicochemical associations of Cd, Cu Pb and Zn in road sediments. Sci Total Environ 33:59–74

Hare L, Tessier A (1996) Predicting animal cadmium concentrations in lakes. Nature 380:430–432

Howari FM, Banat KM (2001) Assessment of Fe, Zn, Cd, Hg, and Pb in the Jordan and Yarmouk River sediments in relation to their physicochemical properties and sequential extraction characterization. Water Air Soil Pollut 1321:43–59

Ikem A, Adisa S (2011) Runoff effect on eutrophic lake water quality and heavy metal distribution in recent littoral sediment. Chemosphere 82:259–267

Islam MM, Akhtar MK, Masud MS (2006) Prediction of environmental flow to improve the water quality in the river Buriganga. In: Proceedings of the 17th IASTED International Conference on Modelling and Simulation, Montreal, QC, Canada

Islam MS, Han S, Masunaga S (2014) Assessment of trace metal contamination in water and sediment of some rivers in Bangladesh. J Water Environ Technol 12:109–121

Jain CK, Gupta H, Chakrapani G (2008) Enrichment and fractionation of heavy metals in bed sediments of River Narmada, India. Environ Monit Assess 141:35–47

Jones B, Turki A (1997) Distribution and speciation of heavy metals in surficial sediments from the Tees Estuary, north–east England. Mar Pollut Bull 34:768–779

Karbassi AR, Shankar R (2005) Geochemistry of two sediment cores from the west coast of India. Int J Environ Sci Technol 1:307–316

Kenniburgh DG, Milne CJ, Benedetti MF, Pinheiro JP, Filius J, Koopal LK et al (1996) Metal ion binding by humic acid: Application of the NICA-‘Donnan model. Environ Sci Technol 30:1687–1698

Kim KW, Myung JH, Ahn JS, Chon HT (1998) Heavy metal contamination in dusts and stream sediments in the Taejon area, Korea. J Geochem Explor 64:409–419

Li HB, Yu S, Li GL, Liu Y, Yu GB, Deng H et al (2012) Urbanization increased metal levels in lake surface sediment and catchment topsoil of waterscape parks. Sci Total Environ 432:202–209

Liang Y, Wong MH (2003) Spatial and temporal organic and heavy metal pollution at Mai Po Marshes Nature Reserve, Hong Kong. Chemosphere 52:1647–1658

Luoma SN, Rainbow PS (2008) Metal contamination in aquatic environments: Science and lateral management. Cambridge University Press, Cambridge, pp 93–123

Mazej Z, Sayegh-Petkovsek SA, Pokorny BT (2010) Heavy metal concentrations in food chain of Lake Velenjsko Jezero, Slovenia: An artificial lake from mining. Arch Environ Contam Toxicol 58:998–1007

McLean JE, Bledsoe BE (1992) Behavior of metals in soils. EPA Groundwater Issue, EPA/540/s-92/018

Mohiuddin KM, Ogawa Y, Zakir HM, Otomo K, Shikazono N (2011) Heavy metals contamination in water and sediments of an urban river in a developing country. Int J Environ Sci Technol 8(4):723–736

Moniruzzaman M, Elahi SF, Jahangir MAA (2009) Study on temporal variation of physicochemical parameters of Buriganga River Water through GIS (Geographical Information System) technology. Bangladesh J Sci Ind Res 44(3):327–334

Morillo J, Usero J, Gracia I (2004) Heavy metal distribution in marine sediments from the southwest coast of Spain. Chemosphere 55(3):431–442

Morrison HA, Gobas FAP, Lazar R, Haffner GA (1996) Development and verification of bioaccumulation model for organic contaminants in benthic invertebrates. Environ Sci Technol 30:3377–3384

Niwa Y, Sugai T, Saegusa Y, Ogami T, Sasao E (2011) Use of electrical conductivity to analyze depositional environments: Example of a Holocene delta sequence on the Nobi Plain, central Japan. Quaternary Int 230:78–86

Nouri J, Lorestani B, Yousefi N, Khorasani N, Hasani AH, Seif S et al (2011) Phytoremediation potential of native plants grown in the vicinity of Ahangaran lead–zinc mine (Hamedan, Iran). Environ Earth Sci 62(3):639–644

Oyeyiola AO, Olayinka KO, Alo BI (2011) Comparison of three sequential extraction protocols for the fractionation of potentially toxic metals in coastal sediments. Environ Monit Assess 172:319–327

Pempkowiase J, Sikora A, Biernacka E (1999) Speciation of heavy metals in marine sediments vs. their bioaccumulation by mussels. Chemosphere 39(2):313–321

Pustisek N, Milacic R, Veber M (2001) Use of the BCR three-step sequential extraction procedure for the study of the partitioning of Cd, Pb, and Zn in various soil samples. J Soil Sediments 1:25–29

Rahman MS, Molla AH, Saha N, Rahman A (2012) Study on heavy metals levels and its risk assessment in some edible fishes from Bangshi River, Savar, Dhaka, Bangladesh. Food Chem 134:1847–1854

Rahman MS, Saha N, Molla AH (2014) Potential ecological risk assessment of heavy metal contamination in sediment and water body around Dhaka export processing zone, Bangladesh. Environ Earth Sci 71:2293–2308

Rath P, Panda UC, Bhatta D, Sahu KC (2009) Use of sequential leaching, mineralogy, morphology and multivariate statistical technique for quantifying metal pollution in highly polluted aquatic sediments—A case study: Brahamani and Nandira rivers, India. J Hazard Mater 163:632–644

Rauret G, Lopez-Sanchez JF, Sahuquillo A, Rubio R, Davidson C, Ure A, Quevauviller Ph (1999) Improvement of the BCR three step sequential extraction procedure before the certification of new sediment and soil reference materials. J Environ Monit 1:57–61

Saikia AK, Abujam SKS, Biswas SP (2012) Food and feeding habit of Channa punctatus (Bloch) from the paddy field of Sivsagar District, Assam. Bull Environ Pharmacol Life Sci 1:10–15

Salmonas W, Forstner U (1980) Trace metal analysis on polluted sediments, part-II. Evaluation of environmental impact. Environ Technol Lett 1:506–517

Sharif AKM, Alamgir M, Mustafa AI, Hossain MA, Amin MN (1993) Trace element concentrations in ten species of freshwater fish of Bangladesh. Sci Total Environ 138:117–126

Sin SN, Chua H, Lo W, Ng LM (2001) Assessment of heavy metal cations in sediments of Shing Mun River, Hong Kong. Environ Int 26:297–301

Singh KP, Mohan D, Singh VK, Malik A (2005) Studies on distribution and fractionation of heavy metals in Gomati river sediments—A tributary of the Ganges, India. J Hydrol 312:14–27

Sow AY, Ismail A, Zulkifli SZ (2013) An assessment of heavy metal bioaccumulation in Asian swamp eel, Monopterus albus, during plowing stages of a paddy cycle. Bull Environ Contam Toxicol 91:6–12

Stone M, Marsalek J (1996) Trace metal composition and speciation in street sediment: Sault Ste. Marie, Canada. Water Air Soil Pollut 87:149–168

Sundaray SK, Nayak BB, Lin S, Bhatta D (2011) Geochemical speciation and risk assessment of heavy metals in the river estuarine sediments—A case study: mahanadi Basin, India. J Hazard Matter 186:1837–1846

Sutherland RA, Tack FMG (2003) Fractionation of Cu, Pb, and Zn in certified reference soils SRM 2710 and SRM 2711 using the optimized BCR sequential extraction procedure. Adv Environ Res 8:37–50

Svete P, Milacic R, Pihlar B (2001) Partitioning of Zn, Pb, and Cd in river sediments from a lead and zinc mining area using the BCR three-step sequential extraction procedure. J Environ Monit 3:586–590

Tang W, Zhao Y, Wang C, Shan B, Cui J (2013) Heavy metal contamination of overlying waters and bed sediments of Haihe Basin in China. Ecotoxicol Environ Saf 98:317–323

Tao Y, Yuan Z, Xiaona H, Wei M (2012) Distribution and bioaccumulation of heavy metals in aquatic organisms of different trophic levels and potential health risk assessment from Taihu Lake, China. Ecotoxicol Environ Saf 81:55–64

Terra BF, Araujo FG, Calza CF, Lopes RT, Teixeira TP (2008) Heavy metal in tissues of three fish species from different trophic levels in a tropical Brazilian river. Water Air Soil Pollut 187:275–284

Tessier A, Campbell FGC (1987) Partitioning of trace metals in sediments: relationships with bioavailability. Hydrobiology 149:43–52

Tessier A, Campbell PGC, Bisson M (1979) Sequential extraction procedure for the speciation of particulate trace metals. Anal Chem 51:844–851

United States Environmental Protection Agency (2001) Methods for collection, storage and manipulation of sediments for chemical and toxicological analyses. Technical manual EPA-823-B-01-002. Office of Water, USEPA, Washington, DC

van der Oost R, Beyer J, Vermeulen PEN (2003) Fish bioaccumulation and biomarkers in environmental risk assessment: a review. Environ Toxicol Pharmacol 13:57–149

Wang WX, Rainbow PS (2008) Comparative approaches to understand metal bioaccumulation in aquatic animals. Comp Biochem Physiol C 148:315–323

Waqar A (2006) Levels of selected heavy metals in tuna fish. Arab J Sci Eng 31:89–92

Xia XH, Chen X, Liu RM, Liu H (2011) Heavy metals in urban soils with various types of land use in Beijing, China. J Hazard Mater 186:2043–2050

Yang ZF, Wang Y, Shen ZY, Niu JF, Tang ZW (2009) Distribution and speciation of heavy metals in sediments from the mainstream, tributaries, and lakes of the Yangtze River catchment of Wuhan, China. J Hazard Mater 166:1186–1194

Yap CK, Ismail A, Tan SG, Omar H (2002) Correlation between speciation of Cd, Cu, Pb and Zn in sediments and their concentrations in total soft tissue of green-lipped mussel Perna viridis from the west coast of Peninsular Malaysia. Environ Int 28:17–26

Yi Y, Yang Z, Zhang S (2011) Ecological risk assessment of heavy metals in sediment and human health risk assessment of heavy metals in fishes in the middle and lower reaches of the Yangtze River basin. Environ Pollut 159:2575–2585

Yilmaz A (2005) Comparison of heavy metal levels of grey mullet (Mugil cephalus L.) and sea bream (Sparus aurata L.) caught in Uskenderun Bay (Turkey). Turk J Vet Anim Sci 29:257–262

Zhang WF, Liu XP, Cheng HF, Zeng EY, Hu YN (2012) Heavy metal pollution in sediments of a typical mariculture zone in South China. Mar Pollut Bull 64:712–720