

A REVIEW ON THE TOXICITY LEVELS IN DOMINANT FINFISHES FROM SELECTED RIVERS OF INDIA

KAKOLI BANERJEE*, TRISNEHI PRADHAN, ASMITA BASU
AND PRAMOD KUMAR BINDHANI

*The present work is an intensive research review encompassing the data of last two decades to provide a wholesome idea about the health of freshwater ecosystem. The study has been carried out in 7 major rivers of India namely Ganga, Yamuna, Kali, Subarnarekha, Godavari, Gadilam and Thamirabarani. Selected heavy metals (Cr, Cu, Zn, Cd and Pb) in water, sediment and dominant finfish species (*Labeo rohita*, *Heteropneustes fossilis* and *Puntius* sp.) were studied. Results suggest that Yamuna waters have Zn, Cu, Pb levels considerably higher than the WHO (2011) and BIS (2012) standards. Sediment data has shown Cr in Ganga and Subarnarekha, Cu in Yamuna and Subarnarekha and Pb in Subarnarekha and Kali rivers respectively are considerably higher than the USEPA and TEL standards. The records for Cr, Zn and Pb in *Labeo rohita*; Cr, Cd and Pb in *Heteropneustes fossilis* and *Puntius* sp. have shown considerably higher concentration in comparison to FAO/WHO (2019). These records have been found much higher than the FAO/WHO (1989) standards. The target hazard quotient (THQ) and hazard index (HI) were calculated and found consumption of *L. rohita* from river Ganga and *H. fossilis* and *Puntius* sp. from river Kali are not safe for human health.*

Introduction

Heavy metals are the metallic elements having high atomic mass and high mass density (greater than 4.5 g/cm³), which includes Cr, Mn, Fe, Co, Ni, Cu, Zn, Cd, Hg, Pb etc.¹ from geological as well as anthropogenic sources. They are also called trace metals because of the fact that they are present in low concentrations in the environment and their bioavailability is affected by various physical, chemical and biological factors. Some heavy metals like Mn, Fe, Co, Cu, Zn are considered as essential metals due to their role as micronutrients in many microorganisms, plants and animals while others like Cd, Hg, and Pb are called non-essential metals². Based on their capability to induce carcinogenesis in humans, the International Agency for Research on

Cancer³ has classified the heavy metals into two groups. Carcinogenic group includes the prominent metals like Cr, Cd, Pb and As, while Fe, Cu, Ni, Zn, Mn and Co are classified as non-carcinogenic.

The freshwater bodies, especially rivers are the most important sources of drinking water, water for irrigating the agricultural fields, culturing fishes and other domestic activities⁴ which necessitate rigorous water quality monitoring practices. The heavy metal pollution in rivers can be attributed to natural origin (volcanic eruption, weathering of rocks and other geological processes) as well as anthropogenic sources. Sediments are known to act as both source and sink of heavy metals and the heavy metal distribution in the sediments are mainly governed by the grain size, chemical composition and amount of organic matter present in it. Once these trace metals enter an aquatic system, they either get deposited or get associated with the sediments, then get bio accumulated and bio-amplified in various living tissues of the aquatic

* Department of Biodiversity and Conservation of Natural Resources, Central University of Odisha, Koraput 763004, India.

Corresponding author: banerjee.kakoli@yahoo.com

biota⁵. The quantification of the heavy metals accumulated in biota is done in terms of Bio concentration factor (BCF), Bioaccumulation factor (BAF), Bioaccumulation coefficient (BAC) etc.

According to FAO, fish provides almost 20% of the average per capita intake of animal protein to approximately 3.3 billion people all over the globe. The recent trends in the increasing levels of the metals in the fishes has raised concerns about their consumption and potential health risks associated with it. World agencies like Food and Agriculture Organisation (FAO), World Health Organisation (WHO), Environment Protection Agencies (EPA), along with many national agencies like Bureau of Indian Standards (BIS), Central Pollution Control Board (CPCB) have set up safety limits for the heavy metals⁶.

Fishes serve as excellent biological indicators of metal in an aquatic ecosystem due to which they have been used to study the pollution status over the years⁷. The metabolically active tissues like liver, kidneys and gills are easy target sites for the trace metals to get accumulated due to Metallothionein's (metal-binding proteins) induction upon exposure to metals⁸. There are different pathways through which heavy metals enter the fish body and accumulate in the tissues. These includes metal ion exchange through the dermal layer, gills, ingestion of food and water⁹. Then the metals are transported to various tissues via the circulatory system. Long-term exposure of fishes to heavy metals can lead to physical deformities, behavioural anomalies, disrupted growth, swimming imbalance, immune system damage, gills damage, renal and nervous system damage, high embryonic mortality and also induces mutagenesis and genotoxic damage. The health risk assessment (HRA), target carcinogenic risk (TCR) and hazard quotient (HQ) are some of the tools used to estimate the risk of heavy metal contamination to human health. This intensive research review attempts to integrate the data on heavy metal levels in different riverine ecosystems and understand the pattern of heavy metal accumulation in finfishes from different rivers of India.

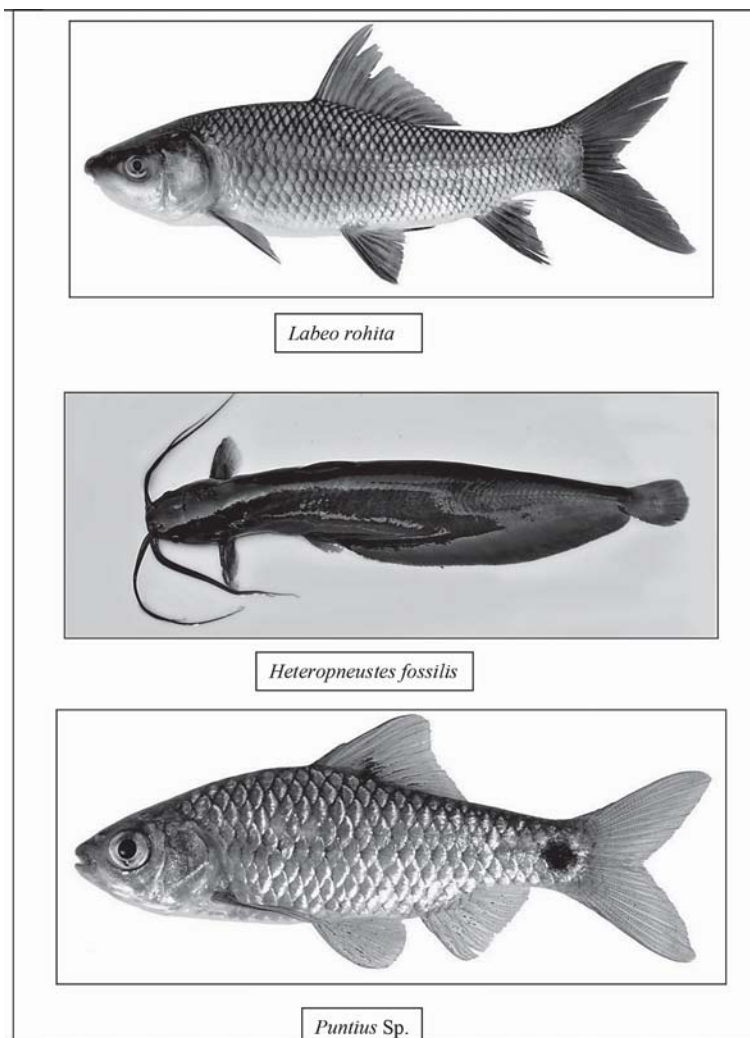
Methods

Study Area: For the study, total of seven rivers namely Ganga, Yamuna, Kali, Subarnarekha,

Godavari, Gadilam and Thamirabarani were selected across India from the existing literature on the topic of heavy metal concentration pattern in the riverine ecosystem. The river's dimensions, geospatial location, proximity to urban settlements and possible sources of heavy metals were thoroughly understood.

Selection of Fish Species: For the study, 3 major finfish species (*Labeo rohita*, *Heteropneustes fossilis* and *Puntius* sp.) were selected (Plate 1).

Data Collection and Analysis: The secondary data for heavy metal concentration in water, sediments and selected dominant finfish species from different rivers was collected from different literature reviews^{5,6,10-23} where the authors have determined the accumulation level in muscles, gills, liver, kidney and intestine.



(Source: https://www.researchgate.net/publication/341868681_The_non-native_freshwater_fishes_of_Singapore_an_annotated_compilation/figures?lo=1; <https://www.flickr.com/photos/144485618@N05/46839331182>)

Plate 1. Pics of the studied fish species

Statistical Analysis: The heavy metal data in water, sediment and finfishes are expressed as mg/l, mg/kg dry weight and mg/kg dry weight respectively. Two-way analysis of variance (ANOVA) was carried out with $P \leq 0.05$ to compute the variation of individual metals among different rivers and selected finfish species. Microsoft Office Excel 2007 was used to plot the column charts and to carry out two-way ANOVA.

Health Risk Assessment for Fish Consumption: The data of heavy metal concentration in fish published as dry weight were converted to fresh weight using conversion factor 5. Estimated dietary intake (EDI), target hazard quotient (THQ) and hazard index (HI) were calculated for the estimation of health risks due to consumption of contaminated fish. EDI value for metal was computed using the formula as per ²⁴,

$$EDI = \frac{C_{\text{metal}} \times W_{\text{fish}}}{B_w}$$

Where, C_{metal} = the concentration of heavy metals detected in fish ($\mu\text{g/g}$ per day)

W_{fish} = the weight of fish consumed per day (grams per day)

B_w = the body weight of the consumer (kilogram)

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

Where, RfD = reference dose (mg/kg per day)

$$HI = \sum THQ$$

Results and Discussion

Concentration of heavy metals surpassing the maximum permissible limits in fish body can induce serious health concerns in humans when consumed. The health hazards associated with these carcinogenic and mutagenic toxic metals includes damage of nervous system, impairment and dysfunctioning of vital organs like kidney and liver, deterioration in the cognitive function, damaged cardiovascular system, growth abnormalities, skeletal mis-formation and cancers. Progressive diseases like Alzheimer's disease, decreased muscular strength, multiple sclerosis is some of the resultant risks of long-term exposure to the toxic metals²⁵.

The results obtained from the analysis of the compiled data on heavy metal concentration in water, sediment and three dominant finfish species are presented in Figure2, 3, 4, 5, 6. Risk assessment for contaminated fish consumption was done by calculating the hazard indices. The results of the assessment by calculating Estimated Dietary Intake (EDI), Target Hazard Quotient (THQ) and Hazard Index (HI) are presented in Tables 1,2 and 3.

The thoroughly compiled data of heavy metal concentrations in water, sediments, *Labeo rohita*, *Heteropneustes fossilis* and *Puntius* sp. from different rivers of India used for the study has been analysed and discussed below separately.

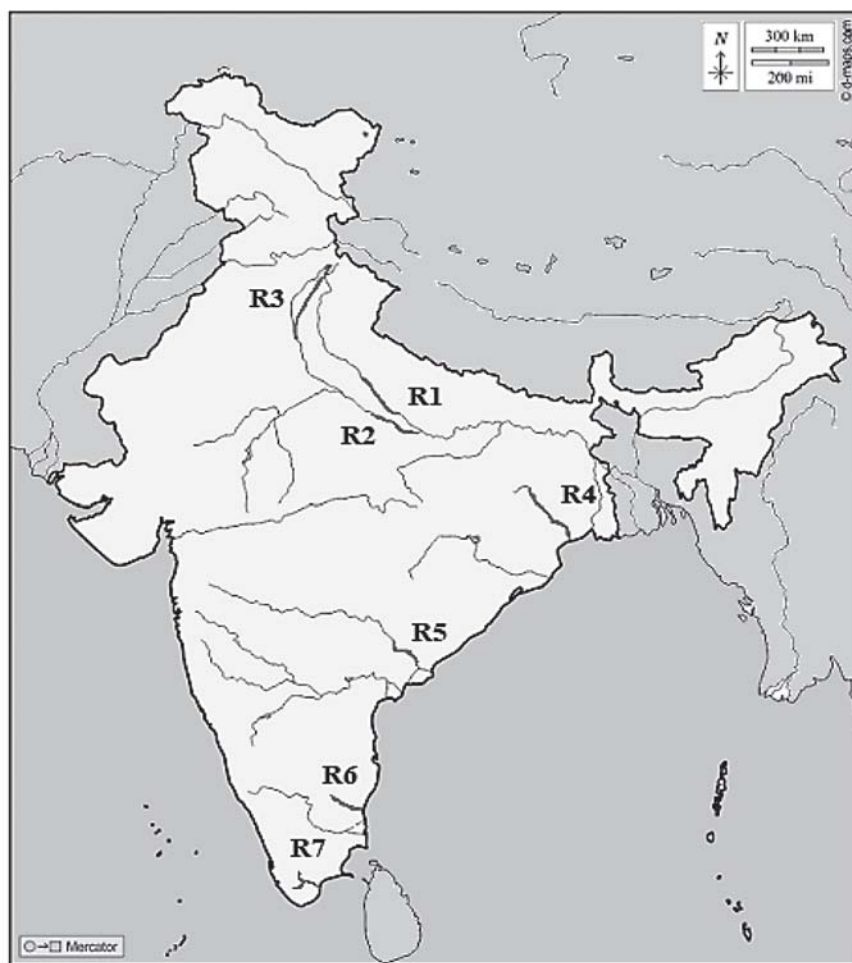


Fig. 1 Map of the rivers showing the stretch studied

Table 1. Estimated dietary intake (EDI), target hazard quotient (THQ) and hazard index (HI) of heavy metals by consuming *Labeo rohita*.

River		Heavy Metals				
		Cr	Cu	Zn	Cd	Pb
Ganga	Metal conc. (µg/g fresh wt.)	1.47	7.79	34.91	1.6	1.01
	EDI (mg/kg/day)	0.707	3.745	16.783	0.769	0.486
	RfD (mg/kg/day)	0.003	0.037	0.3	0.001	0.004
	THQ	0.236	0.101	0.056	0.769	0.121
	HI	1.283				
Yamuna	Metal conc. (µg/g fresh wt.)	ND	0.947	9.459	ND	0.53
	EDI (mg/kg/day)	0.0	0.455	4.548	0.0	0.255
	RfD (mg/kg/day)	0.003	0.037	0.3	0.001	0.004
	THQ	0.0	0.012	0.0152	0.0	0.064
	HI	0.0912				
Subarnarekha	Metal conc. (µg/g fresh wt.)	0.674	14.56	59.398	0.082	0.094
	EDI (mg/kg/day)	0.324	7	28.557	0.039	0.045
	RfD (mg/kg/day)	0.003	0.037	0.3	0.001	0.004
	THQ	0.108	0.189	0.095	0.039	0.011
	HI	0.442				
Godavari	Metal conc. (µg/g fresh wt.)	0.242	0.579	3.804	0.098	0.654
	EDI (mg/kg/day)	0.116	0.278	1.829	0.047	0.314
	RfD (mg/kg/day)	0.003	0.037	0.3	0.001	0.004
	THQ	0.039	0.008	0.006	0.047	0.079
	HI	0.179				
Thamirabarani	Metal conc. (µg/g fresh wt.)	0.032	0.12	0.65	0.014	0.106
	EDI (mg/kg/day)	0.015	0.058	0.313	0.007	0.051
	RfD (mg/kg/day)	0.003	0.037	0.3	0.001	0.004
	THQ	0.005	0.002	0.001	0.007	0.013
	HI	0.028				

Threshold of THQ = 1 Threshold of HI = 1 (USEPA, 2011)

Chromium (Cr): Among all the heavy metals, Cr ranks seventh in terms of abundance in nature. It is mainly found in two oxidation states that are Cr ⁺³ (trivalent) and Cr ⁺⁶ (hexavalent). The hexavalent form is known to be more soluble as well as toxic to living beings in comparison to the trivalent one which is predominantly a particle reactive form and serves as a micronutrient in the mammalian bodies²⁶. Chromium mainly targets the pulmonary system and is most commonly known to cause ulcers, perforated nasal septum and lung cancer²⁷.

Figure 2 suggested that the sequence of different rivers in accordance to the mean concentration of chromium in their water was Subarnarekha (0.00075 mg/l) < Kali (0.0131 mg/l) < Thamirabarani (0.213 mg/l) < Ganga

(0.426 mg/l) < Gadilam (0.7 mg/l) < Godavari (0.857 mg/l). The mean concentration of Cr in the Subarnarekha River and Muzaffarnagar stretch of Kali River were within the standards²⁸. Its concentration in the riverine water of Thamirabarani, Ganga, Gadilam and Godavari were much higher than the drinking water safety limit prescribed by BIS²⁹ and notably lower than the permissible standards²⁸. In the water of Ganga River, the level of Cr was about ten times more than the standards²⁸ which can be attributed to the functioning of manufacturing industries along the river bank was reported³⁰. Coal mining company, ITC paper mill se untreated effluents were discharged into the river could be possible reason for the hike of Cr concentration in Godavari water¹⁹.

Table 2. Estimated dietary intake (EDI), target hazard quotient (THQ) and hazard index (HI) of heavy metals by consuming *H. fossilis*.

River		Heavy Metals				
		Cr	Cu	Zn	Cd	Pb
Ganga	Metal conc. (µg/g fresh wt.)	0.88	1.35	6.1	0.5	0.54
	EDI (mg/kg/day)	0.423	0.649	2.933	0.24	0.26
	RfD (mg/kg/day)	0.003	0.037	0.3	0.001	0.004
	THQ	0.141	0.018	0.01	0.24	0.065
	HI	0.474				
Kali	Metal conc. (µg/g fresh wt.)	16.422	ND	12.912	8.472	4.822
	EDI (mg/kg/day)	7.895	0.0	6.208	4.073	2.318
	RfD (mg/kg/day)	0.003	0.037	0.3	0.001	0.004
	THQ	2.632	0.0	0.021	4.073	0.58
	HI	7.306				
Gadilam	Metal conc. (µg/g fresh wt.)	0.158	0.116	0.068	0.154	0.084
	EDI (mg/kg/day)	0.076	0.056	0.033	0.074	0.04
	RfD (mg/kg/day)	0.003	0.037	0.3	0.001	0.004
	THQ	0.026	0.002	0.0001	0.074	0.01
	HI	0.1121				

Threshold of THQ = 1 Threshold of HI = 1 (USEPA, 2011)

The results revealed that in all the rivers the sediments accumulated significantly higher concentrations of Cr than the riverine water as summarised in Figure3. The rivers were arranged in increasing order of Cr accumulation in sediments as - Gadilam (0.94 mg/kg dry wt.) < Kali (7.43 mg/kg dry wt.) < Thamirabarani (8.89 mg/kg dry wt.) < Subarnarekha (77.8 mg/kg dry wt.) < Ganga

(135.9 mg/kg dry wt.), the metal was not determined in Yamuna River. The findings clearly depicted that concentration of Cr detected in the Ganga River (135.9 mg/kg dry wt.) was much higher than in any other river. In Kali River zone wise variation of Cr levels was noted (Zone B > Zone A) which may have occurred due to the differences in volumes of effluents from industries and

sewage being added at different sampling points⁹. The mean concentration exceeded the Canadian EPA limits for sediments. Around 20-25 times higher concentration of Cr was recorded in the downstream sediments as compared to that of the sediments from upstream region of the Thamirabarani river due to large volume discharge of effluents and sewages at the lower stretch of river²². The results of this review indicated that Cr concentration in riverine sediments of Kali, Gadilam and Thamirabarani stayed within the quality standards set by USEPA and TEL while Ganga and Subarnarekha showed higher

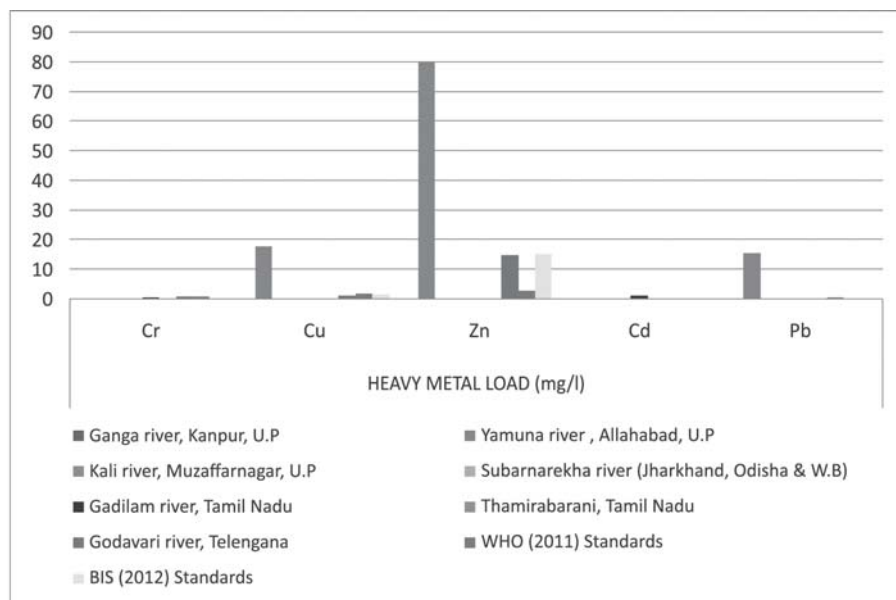


Fig. 2. Mean heavy metal concentrations (mg/l) in the water of different rivers of India.

Table 3. Estimated dietary intake (EDI), target hazard quotient (THQ) and hazard index (HI) of heavy metals by consuming *Puntius* sp.

River		Heavy Metals				
		Cr	Cu	Zn	Cd	Pb
Ganga	Metal conc. (µg/g fresh wt.)	0.67	2.37	12.2	0.45	1.72
	EDI (mg/kg/day)	0.322	1.139	5.865	0.216	0.827
	RfD (mg/kg/day)	0.003	0.037	0.3	0.001	0.004
	THQ	0.107	0.031	0.02	0.216	0.207
	HI	0.581				
Kali	Metal conc. (µg/g fresh wt.)	5.426	ND	7.364	4.166	1.724
	EDI (mg/kg/day)	2.609	0.0	3.54	2.003	0.829
	RfD (mg/kg/day)	0.003	0.037	0.3	0.001	0.004
	THQ	0.87	0	0.012	2.003	0.207
	HI	3.092				
Subarnarekha	Metal conc. (µg/g fresh wt.)	0.526	8.1	40.566	0.08	0.44
	EDI (mg/kg/day)	0.253	3.894	19.503	0.038	0.212
	RfD (mg/kg/day)	0.003	0.037	0.3	0.001	0.004
	THQ	0.084	0.105	0.065	0.038	0.053
	HI	0.345				

Threshold limit for THQ = 1

Threshold limit for HI = 1 (USEPA, 2011)

values than the permissible limits (Figure3). In the study³⁰ found that the detected values in the Ganga River sediments exhibited considerable variations from the maximum allowance limits possibly because of the influence of industries around the river which was similar to the results obtained from the present study.

Concentration of Chromium in *Labeo rohita* of different rivers followed the trend - Thamirabarani (0.16 mg/kg dry wt.) < Godavari (0.242 mg/kg dry wt.) < Subarnarekha (3.37 mg/kg dry wt.) < Ganga (7.35 mg/kg dry wt.). The studied tissues of *Labeo rohita* from the Ganga River bio concentrated high amount (7.35 mg/kg dry wt.) of Cr which might be the resultant of their large body size and voracious column feeding habit in addition to high influence of industrial discharge on the riverine environment³¹. Concentrations in *L. rohita* obtained from the Ganga and Subarnarekha were found to be higher than standards³² while it remained within the standard limit set by the regulatory body for Cr in the species from Godavari and

Thamirabarani (Figure4). From Figure5 the order of Chromium concentration in the species *Heteropneustes fossilis* of different rivers was found as - Kali (82.11 mg/kg dry wt.) > Ganga (4.4 mg/kg) > Gadilam (0.792 mg/kg dry wt.). *Heteropneustes fossilis* from the Kali River concentrated several folds higher level of chromium than those caught from other rivers. Also, Cr accumulated maximally in the muscle tissue of this fish species as compared to other metals⁹. The findings of the study by¹⁴ stated that the mean concentration of Cr in *H. fossilis* of

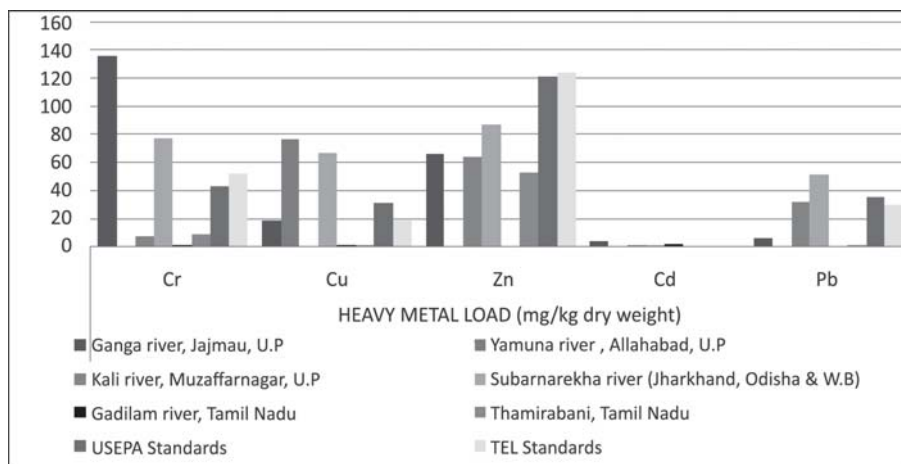


Fig. 3. Mean heavy metal concentrations (mg/kg dry weight) in the sediments of different rivers of India.

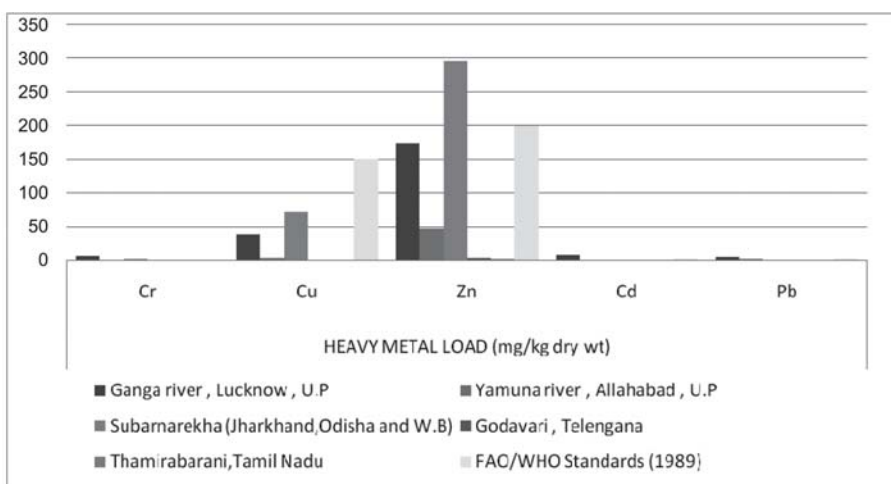


Fig. 4. Mean heavy metal concentrations (mg/kg dry weight) in *Labeo rohita* from different rivers of India.

Gadilam river exceeded the standards prescribed by different regulatory agencies like WHO and FEPA which was similar to our results obtained in Figure5. The possible sources of Cr contamination in fish tissues were large volume of waste water being discharged from the agricultural fields into the river. The results of this review revealed that when comparison was done between the recorded levels of Cr in fish body and standards³², *H. fossilis* from all the three-river showed higher levels of metals than the suggested standards. *Puntius ticto* bioconcentrated relatively lower concentration of Cr than the *H. fossilis* (carnivorous) from Lucknow stretch of the Ganga River due to its herbivore nature³¹. Data from Figure6 indicated the order of Cr accumulation in tissues of *Puntius* sp. of different rivers to be - Subarnarekha (2.63 mg/kg dry wt.) < Ganga (3.35 mg/kg dry wt.) < Kali (27.13 mg/kg dry wt.). Concentration of the metal in the species from all three rivers crossed the safety limits³². Highest Cr level in Kali River may be because of its close proximity to tannery industries.

Two-way analysis of variance (ANOVA) that was computed for studying the significant differences of Cr among the rivers and fish species clearly revealed that there was no significant difference neither between the rivers nor the fish species for this particular metal ($F_{cal} = 2.068$; $df=5$; $p \geq 0.05$ and $F_{cal} = 0.853$; $df=2$; $p \geq 0.05$).

Copper (Cu): Cu is an essential heavy metal for the living

bodies owing to its important functions in several biological processes like haemoglobin synthesis but excess of this metal causes toxic effects in living organisms^{14,26}. Short term exposure to Cu can lead to gastro-intestinal distress, jaundice and destruction of RBCs while chronic exposure may result in liver damage, Wilson's disease (inheritable), Cirrhosis syndrome, heart failure and kidney related issues²⁷.

Low concentration of copper was found in the water of four rivers (Ganga, Gadilam, Godavari

and Subarnarekha) except for that in the Yamuna water. Cu was not determined in Kali and Thamirabarani. The order of rivers according to Cu concentration in their water was noted as - Subarnarekha (0.0069 mg/l) < Ganga (0.023 mg/l) < Gadilam (0.5 mg/l) < Godavari (1.401 mg/l) < Yamuna (17.64 mg/l) as presented in Figure2. Its mean concentration in water samples of Ganga, Godavari, Subarnarekha and Gadilam were lower than the acceptable limits²⁸ and drinking water standard²⁹. The results obtained in this review were in accordance with the study³⁰ which stated that Cu concentration in Gadilam river water was about 45 times lower than the safety limits. The results of the review also exhibited that concentration of Cu detected in Yamuna River water exceeded the permissible limits by several folds which may be the result of effluents from small scale industries set along the river bank, automobile discharges and untreated sewage from urban areas of Allahabad.

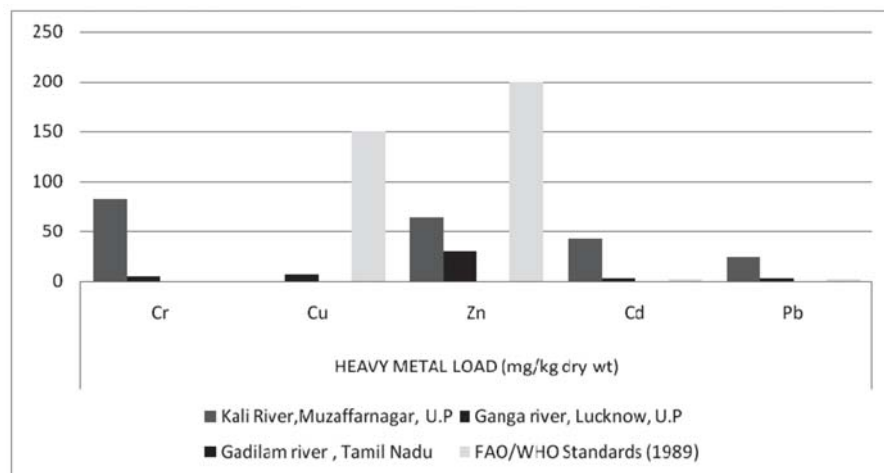


Fig. 5. Mean heavy metal concentrations (mg/kg dry weight) in *Heteropneustes fossilis* from different rivers of India.

Copper concentration in sediments were much higher than that detected in water from all the studied rivers. Rivers were found to be following a decreasing trend according to the Cu concentration detected in their sediments as - Yamuna (76.64 mg/kg) > Subarnarekha (67.23 mg/kg) > Ganga (19.17 mg/kg) > Thamirabarani (1.4 mg/kg) > Gadilam (0.6 mg/kg), Cu went undetermined in Kali River (Figure3). The elevated levels in Yamuna and Subarnarekha may be due to the functioning of industries nearby and presence of copper mining regions respectively^{17,33}. When compared with the maximum allowance limit of USEPA and TEL for sediment quality, concentrations detected in all the rivers remained within the standards.

The concentration of Cu reported in *Labeo rohita* from different rivers were comparatively higher than that in riverine water but lower than the sediments except in Subarnarekha and the Ganga. The sequence of rivers on the basis of magnitude of Cu in tissues of *L. rohita* was observed as - Subarnarekha (72.8 mg/kg dry wt.) > Ganga (38.95 mg/kg dry wt.) > Yamuna (4.735 mg/kg dry wt.) > Thamirabarani (0.6 mg/kg dry wt.) > Godavari (0.579 mg/kg dry wt.). All the mean Cu concentrations in fish were lower than the prescribed food standards³² (Figure4). Comparison between Cu concentration in *L. rohita*, *H. fossilis*, *Puntius* sp. inferred that *L. rohita* showed higher concentration than other two species due to its feeding habit (predominantly herbivorous)⁷. Cu traces in *H. fossilis* were found in the concentration of 6.75 mg/kg dry wt. in Ganga river³¹ which was higher than the level detected in fish from Gadilam river (0.58 mg/kg dry wt.)¹⁴. Concentration in the body of the fish species did not cross the food standards by WHO/FAO in any of the rivers. As *H. fossilis* is a carnivorous bottom dwelling fish, the low concentrations detected in its tissues can possibly be related to the low levels of Cu in the sediments. Similar results were derived in case of *Puntius* sp. where concentration was lower in all rivers than food safety limits by international agency. Higher concentration was recorded in Subarnarekha (40.5 mg/kg dry wt.) and lower in Ganga (11.85 mg/kg dry wt.). Cu concentration in fishes from Subarnarekha River showed elevated levels which can be attributed to higher influence of copper mining industries,

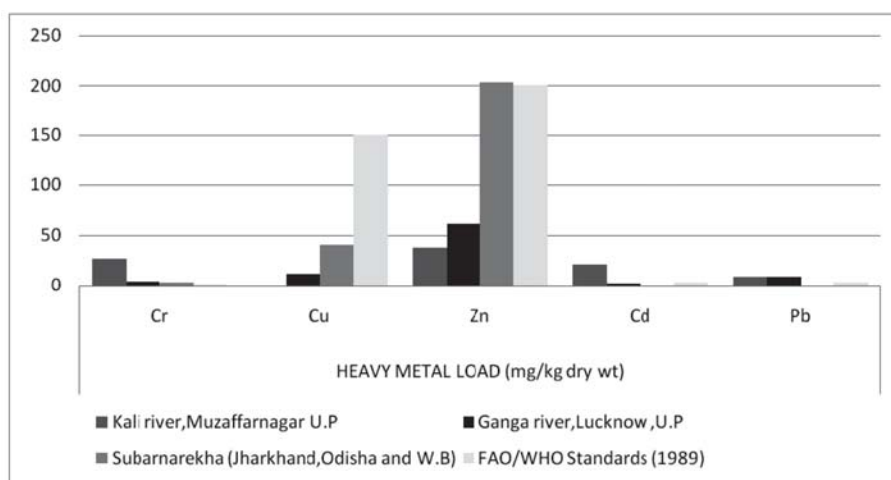


Fig. 6. Mean heavy metal concentrations (mg/kg dry weight) in *Puntius* sp. from different rivers of India.

metallurgical industries and other anthropogenic stresses on the sampling sites⁷.

The output of two-way ANOVA between rivers and fish species clearly suggested that there was no significant difference neither between the rivers nor the fish species for copper ($F_{cal} = 1.481$; $df=5$; $p \geq 0.05$ $F_{cal} = 2.324$; $df=2$; $p \geq 0.05$).

Zinc (Zn): Zinc is an essential element which serves as micronutrient in the living organisms as it has an important biological role to play in the transcription factors. Different types of growth abnormalities, malabsorption, immunological defects, liver related diseases arise due to zinc deficiency¹⁹. However, high dose of Zn ingested through food can pose serious threat of health to humans²⁶. Zinc toxicity in humans can cause zinc induced copper deficiency, lower HDL cholesterol levels, sideroblastic anaemia etc. due their lethal nature.

From Figure2 it is evident that the concentration of zinc detected in rivers were low except for that in Yamuna and Godavari where it was found to be very high. The increasing order of rivers according to the mean concentration of Zn detected in their waters was - Subarnarekha (0.0093 mg/l) < Kali (0.0792 mg/l) < Ganga (0.029 mg/l) < Gadilam (0.1 mg/l) < Godavari (14.657 mg/l) < Yamuna (79.92 mg/l) and in Thamirabarani it was not determined. In all the riverine waters the Zn levels were higher than the water quality guidelines²⁸ and drinking water safety limit²⁹. The results of this review were in accordance with the findings³³ which informed that elevated Zn level in Yamuna River water due to industrial effluent discharged into it makes it inapposite for survival of aquatic life and usage by humans for drinking purpose.

In sediments higher level of Zn was recorded in comparison to that in the riverine waters. Yamuna (0.029 mg/kg dry wt.) < Gadilam (0.2 mg/kg dry wt.) < Thamirabarani (52.92 mg/kg dry wt.) < Kali (63.84 mg/kg dry wt.) < Ganga (66.35 mg/kg dry wt.) < Subarnarekha (87.27 mg/kg dry wt.) was the observed trend of rivers according to mean Zn concentration detected in their sediments. Although high concentrations were seen in some riverine sediments, the levels remained below the USEPA and TEL sediment quality standards (Figure3). High industrial influence on the Subarnarekha River may be reason of highest detected levels of the Zn in sediments among all the rivers.

The values of Zn concentration in *Labeo rohita* was much more than that observed in the water and sediments of the rivers. Order of Zn mean concentration in *L. rohita* of different rivers was noted as - Thamirabarani (3.25 mg/kg dry wt.) < Godavari (3.804 mg/kg dry wt.) < Yamuna (47.295 mg/kg dry wt.) < Ganga (174.55 mg/kg dry wt.) < Subarnarekha (296.99 mg/kg dry wt.). Compared to the safety guidelines by WHO/FAO, fish samples from all other rivers exhibited lower concentration except for the fish from Subarnarekha River (Figure 4). *L. rohita* from mainly two sampling sites (Tatanagar and Mosabani) of the Subarnarekha River showed higher Zn concentration than other sites, the reason could be tremendous influence of mining activity, industrial setups and vehicular inputs⁷. The Zn concentration trend in *Heteropneustes fossilis* from different river was found to be - Kali (64.56 mg/kg dry wt.) > Ganga (30.5 mg/kg dry wt.) > Gadilam (0.34 mg/kg dry wt.). Although Zn in muscles of the fish species of Kali River was the highest, it did not cross the permissible limits²⁸ for fishes as investigated⁹. Results of present review suggested that mean concentration of Zn in *H. fossilis* of all the three rivers were lower than the acceptable safety limits by the international agencies (Figure5). *Puntius* sp. from the studied rivers showed Zn concentration higher than the *H. fossilis* but lower than *L. rohita*. The sequence of rivers owing to the magnitude of Zinc in tissues of *Puntius* sp. was - Kali (36.82 mg/kg dry wt.) < Ganga (61 mg/kg dry wt.) < Subarnarekha (202.83 mg/kg dry wt.). *Puntius ticto* from Kali River showed much lower concentrations of Zn than *H. fossilis* of the same river. Levels of the heavy metal in *Puntius* sp. from Kali and Ganga River were lower than the prescribed standard by WHO/FAO. In contrast Zn concentration in *Puntius conchoni* exceeded the safety limits which was found to be similar with the findings⁷.

No significant difference found either between the rivers or the fish species from the two-way ANOVA conducted between rivers and fish species for zinc ($F_{\text{cal}} = 1.177$; $df=5$; $p \geq 0.05$; $F_{\text{cal}} = 1.87$; $df=2$; $p \geq 0.05$).

Cadmium (Cd): Cadmium is a non-essential heavy metal exhibiting high solubility property in water due to which it shows fast propagation rate in the nature. It is highly toxic to human body even at low exposure levels that is why it has been classified as carcinogenic metal by the International Agency for Research on Cancer³. In human cadmium as a toxicant is known to affect mainly the skeletal system and kidneys adversely causing growth abnormalities, teratogenesis, osteomalacia and renal dysfunction²⁷.

Figure 2 revealed that the cadmium mean concentration detected in water of four rivers: Ganga (0.053 mg/l), Kali (0.0053 mg/l), Godavari (0.023 mg/l) and Gadilam (1.46 mg/l) was several times higher than the maximum tolerable limit²⁸ and safe drinking water standard²⁹. The results obtained from the present investigation were found to be similar with the findings³⁰ which states that Cd concentration in water of Ganga River was about ten times higher than the permissible limits. The order according to Cd levels was recorded as - Kali < Godavari < Ganga < Gadilam, the metal was not determined by the previous studies in Yamuna, Subarnarekha and Thamirabarani. The highest level of Cd detected in the waters of Gadilam could be due to agricultural runoffs, sewage from households and other anthropogenic activities¹⁴.

In sediments the order of rivers on the basis of cadmium mean concentration was noted to be: Ganga (3.89 mg/kg dry wt.) > Gadilam (1.64 mg/kg dry wt.) > Kali (1.23 mg/kg dry wt.) > Subarnarekha (1.11 mg/kg dry wt.) > Thamirabarani (0.023 mg/kg dry wt.) and was undetermined in the Yamuna River. Cd in sediments from all the rivers except Thamirabarani exceeded the sediment quality guidelines set by USEPA and TEL (Figure3). Its concentration in Ganga was much higher than the maximum tolerable limits, it could be due to the influence of small-scale industries functioning in Ganga River bank.

From Figure4 it is clearly evident that *Labeo rohita* from the Ganga River presented higher bioconcentration of Cd than the same fish species from other rivers. The sequential order was recorded to be: Ganga (8 mg/kg dry wt.) > Subarnarekha (0.41 mg/kg dry wt.) > Godavari (0.098 mg/kg dry wt.) > Thamirabarani (0.07 mg/kg dry wt.). Concentration detected in fish from Subarnarekha, Godavari

and Thamirabarani was lower than the internationally recommended standard limits. While *L. rohita* from Ganga River concentrated higher amount of Cd than the safety guidelines. This could be due to larger size and extensive feeding habit of *L. rohita*³¹. Figure5 shows that the bio concentration of Cd in *Heteropneustes fossilis* is higher in Kali River (42.36 mg/kg dry wt.) followed by Ganga (2.5 mg/kg dry wt.) and lower in the Gadilam river (0.768 mg/kg dry wt.). Investigative study⁹ suggested that *H. fossilis* is a bottom dwelling fish due to which higher concentration of Cd was found as compared to the surface feeders. It also stated that the prominent sources of the metal were the industrial effluent that was being discharged into the Kali River. Comparison of the detected Cd mean concentration in the fish species with the food standards³² demonstrated that all the levels exceeded the standards which means the consumption of this particular fish species from the above-mentioned rivers may have pronounced effect on the human health (Figure5). Similar findings were obtained for *Puntius* sp., the fish samples from Kali River (20.83 mg/kg dry wt.) bio concentrated higher levels of Cd followed by Ganga River (2.25 mg/kg dry wt.) (Figure 6). These values were above the permissible food standards proposed by the international regulatory agencies. Low levels of Cd were detected in the tissues of *Puntius* sp. from the Subarnarekha River which was well within the limits³² due to which this fish species could be considered safe for consumption.

Two-way ANOVA conducted between rivers and fish species for cadmium revealed that there was no significant variation ($F_{\text{cal}} = 2.68$; $df=5$; $p \geq 0.05$; $F_{\text{cal}} = 0.727$; $df=2$; $p \geq 0.05$) was found either between the rivers or the different fish species.

Lead (Pb): Lead is considered as a potential environmental toxicant and acts as a cumulative metabolic poison in the human body²⁶. Low concentration of Pb is capable enough to induce serious health related issues in the living bodies. Even short-term exposure to high lead levels may cause poisoning, to which children are more prone than the adults²⁵. The target organs are renal system, nervous system and blood vascular system²⁷. The most common effects are anaemia, kidney dysfunction, brain damage, deteriorating intelligence, reduced fertility and can be even fatal. Lead has the capability to pass through the placenta barrier, which means the foetus is also at risk if the mother is exposed to lead.

Results of this review showed that the concentration of lead detected in the water samples of different rivers

was considerably higher than the acceptable limits set for water quality prescribed by international and national regulatory agencies (WHO and BIS) (Figure2). Yamuna river water exhibited highest mean concentration among all the rivers studied due of regular inputs of industrial and domestic wastes because of which it was not suitable for domestic use and drinking purpose according to the investigation³³. The recorded order of rivers in accordance to the Pb levels in their water was: Thamirabarani (0.022 mg/l) < Kali (0.059 mg/l) < Ganga (0.223 mg/l) < Gadilam (0.46 mg/l) < Godavari (0.7 mg/l) < Yamuna (15.26 mg/l).

Sediments of all the rivers concentrated higher concentration of lead than the water samples as suggested by the obtained results. However, in contrast to Yamuna water, the sediments from same rivers exhibited the lowest level of Pb among all other river sediments. Metal was detected in the riverine sediments in the following trend: Yamuna (0.0183 mg/kg dry wt.) < Gadilam (0.48 mg/kg dry wt.) < Thamirabarani (1.68 mg/kg dry wt.) < Ganga (6.55 mg/kg dry wt.) < Kali (32.16 mg/kg dry wt.) < Subarnarekha (51.35 mg/kg dry wt.). When compared with the USEPA and TEL sediment quality tolerable limits, it was noted that except for Kali and Subarnarekha all other rivers concentrated lower amount of lead in their sediments (Figure3). The above results indicated low potential risk from the sediments to the humans. Highest range of Pb detected in Subarnarekha sediments can be attributed to the influence of various industries, mining companies as well as mineral rich status of Jharkhand¹⁷.

The prescribed safety limits³² for Pb in fish was 2.5 mg/kg dry wt. *Labeo rohita* samples studied for Pb in Subarnarekha, Thamirabarani and Godavari did not exceed the above-mentioned permissible limit but the species from Ganga and Yamuna showed considerably higher values of Pb bio concentration in the tissues as compared to the international standards (Figure4). The increasing order in which rivers were arranged according to lead's mean concentration detected in body of *L. rohita* was written as: Subarnarekha (0.47 mg/kg dry wt.) < Thamirabarani (0.53 mg/kg dry wt.) < Godavari (0.654 mg/kg dry wt.) < Yamuna (2.65 mg/kg dry wt.) < Ganga (5.05 mg/kg dry wt.). While that in *H. fossilis* was found to be : Gadilam (0.42 mg/kg dry wt.) < Ganga (2.7 mg/kg dry wt.) < Kali (24.11 mg/kg dry wt.). The fish species bio concentrated Pb below the acceptable limit of food standards while fish from Ganga and Kali River exceeded the threshold³² which indicated that the particular fish species from these rivers might be unsafe for consumption by humans (Figure5). The

outcomes of the investigation for *Puntius* sp. revealed that this fish species from the Subarnarekha River contained least concentration of Pb which was also the case in *Labeo rohita*. It was also seen that the outcomes for *Puntius* sp. matched with that for *H. fossilis* as both the species from Kali River showed highest mean Pb concentration among all the rivers studied. The decreasing trend of Pb level in *Puntius* sp. was recorded as: Kali River (8.62 mg/kg dry wt.) > Ganga River (8.6 mg/kg dry wt.) > Gadilam river (0.22 mg/kg dry wt.). The species from Kali and Ganga River exhibited almost same concentration of lead in their tissues and these levels were several folds higher than the maximum allowance limit³² while the levels in Gadilam river's *Puntius* sp. showed lower magnitude (Figure6).

The outcomes of two-way ANOVA conducted between rivers and fish species for cadmium was found to be similar with the results obtained for other four metals. It revealed that there was no significant variation ($F_{\text{cal}} = 2.076$; $df=5$; $p \geq 0.05$; $F_{\text{cal}} = 0.46$; $df=2$; $p \geq 0.05$) found either between the rivers or the different fish species.

Human health risk Assessment by Estimated Dietary Intake (EDI), Target Hazard Quotient (THQ) and Hazard Index (HI) of Heavy Metals in Three Finfish Species

Health risk assessment is an important and efficient tool to estimate the health risks in humans preceded by exposure to heavy metals. This tool helps to determine the levels of metals bio available and the extent of risk to human health posed by them²². The EDI values, THQ and HI that were evaluated for three finfish species obtained from different rivers were presented in Tables 1,2 and 3 and the findings are discussed below.

Estimated dietary intake (EDI), developed by USEPA is the preliminary explication of human health risk due to consumption of heavy metal contaminated fish. The comparison of mean concentration detected in fish tissues with the acceptable standards prescribed by USEPA i.e., reference dose (RfD) gives a baseline idea about the extent of exposure. Reference dose is the maximum permissible limit for the daily intake of a particular metal that doesn't induce any adverse health effect. The results of analysis revealed that the EDI values for Cr, Cu, Zn, Cd and Pb in all the three fishes (*L. rohita*, *H. fossilis* and *Puntius* sp. obtained from different rivers was much higher than the RfD proposed by USEPA except for Zn in *H. fossilis* of the Gadilam river.

Target Hazard Quotient (THQ) is the ratio of exposure (EDI) to RfD used to assess the non-carcinogenic hazards

associated with consumption of metal contaminated fish. The threshold limit of THQ as suggested by USEPA is 1, $THQ > 1$ for a metal indicates that the daily intake of metal at this level will have significant adverse health effects. The examined data indicated that in *L. rohita* the highest THQ was exhibited by Cadmium (0.769) in the Ganga River and lowest by Zn (0.006) in Godavari River (Table 1). The index calculated for all the five metals (Cr, Cu, Zn, Cd and Pb) in *L. rohita* did not surpass the threshold limit of 1 in each river. Table 2 suggested that in *H. fossilis*, THQ evaluated for cadmium in all the three rivers (Ganga, Kali and Gadilam) was higher than rest of the four metals studied in those rivers. Its value for all elements studied in the three rivers found to be appreciably < 1 except for Cr (2.632) and Cd (4.073) in Kali River which were approximately 2 to 4 times higher. Similarly, in *Puntius* sp. all the metals showed THQ lower than 1 except cadmium (2.003) in Kali River as it exceeded the optimum standard limit by two folds (Table 3).

Hazard index (HI) is efficiently used to appraise the overall risk to human health because of over-exposure to more than one metal in finfish species from every single river. It is computed by summing up the THQ of all metals in a fish species obtained from a particular river. The acceptable limit of HI set by USEPA is 1 so the rivers with $HI < 1$ means they pose no chronic risk to human health. For *L. rohita* the HI of metals was noted in the following increasing order: Thamirabarani (0.028) < Yamuna (0.0912) < Godavari (0.179) < Subarnarekha (0.442) < Ganga (1.283) as presented in Table 1. In Ganga $HI > 1$ for the fish species means that the consumption of *L. rohita* from this particular river may induce non-carcinogenic detrimental effects in the consumers. From Table 2 it is evident that Gadilam (0.1121) < Ganga (0.474) < Kali (7.306) is in order of HI value in *H. fossilis*. Hazard index of metals in Ganga and Kali were within the acceptable limit, but that in the Kali River was several folds higher. *H. fossilis* consumed from Kali River will cause thought provoking chronic deleterious health issues in the consumers and is unsafe for consumption as the extremely high hazard index suggests. The investigation revealed the results obtained for *Puntius* sp. was similar to that in case of *H. fossilis*. The highest HI was determined in Kali River for *Puntius* sp. and the order was recorded as - Subarnarekha (0.345) < Ganga (0.581) < Kali (3.092). $HI > 1$ in *Puntius* sp. which indicated a potential risk to human health if consumed while other two rivers showed lower HI than the threshold limit which means the fish from these rivers are safe to consume.

A study³⁴ recommended the treatment of heavy metal contaminated fishes with pomegranate peels (natural agent) before cooking.

Conclusion

Results suggest that Yamuna waters have Zn, Cu, Pb levels considerably higher than the WHO (2011) and BIS (2012) standards which is not safe for drinking. Sediment data has shown Cr in Ganga and Subarnarekha, Cu in Yamuna and Subarnarekha and Pb in Subarnarekha and Kali rivers respectively are considerably higher than the USEPA and TEL standards and hence can be hazardous for organisms residing and procuring food from the sediments. Considerable high concentration of Cr (above FAO/WHO (1989) standards) in Ganga and Subarnarekha, Zn in Subarnarekha, Pb in Ganga rivers respectively were documented for *L. rohita*. Very high concentrations of Cr in Ganga and Kali, Cd in Kali and Gadilam, Pb in Kali rivers were seen in *H. fossilis*; Quite high concentrations of Cr in Kali, Ganga, Subarnarekha, Cd in Kali and Pb in Ganga and Kali rivers respectively were documented in *Puntius* sp. Since Hazard Index (HI) values are >1 (optimum value) in case of *L. rohita* from Ganga, *H. fossilis* from Kali and *Puntius* sp. from Kali, they are not safe for human consumption and may lead to adverse health effects in human. The Indian way of cooking fish which includes steaming, stewing, grilling, microwaving and particularly frying is considered as the best way to reduce the heavy metal content in fish tissues. The elevated magnitude of heavy metals in the water, sediment and tissues of fin fishes demands a constant monitoring of these riverine systems, implementation of immediate measure at the sources to reduce the discharge of untreated industrial and anthropogenic effluents into the river.

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