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Distribution of heavy metals in water, sediments and fish tissue (*Heteropneustis fossilis*) in Kali River of western U.P. India

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Abstract
The present study the selected heavy metals such as cadmium (Cd), chromium (Cr), lead (Pb), Manganese (Mn) and cooper (Cu) were determined in water, sediment and gill, liver, muscle of fish *H. fossilis*, the concentration of Cd, Cr, Pb, Cu Mn and Zn was observed 0.009, 0.056, 0.056, 0.058, 0.049, 0.883, 0.36 mg/l in river water and 3.38, 20.11, 81.53, 258.48, 258.48, 3.40 µg/g in sediment respectively. The heavy metals concentrations in the tissue of fish *H. fossilis* (µg/g dry weight), Cd 30.89, Cr 20.59, Pb 15.28, Cu 30.66 and Zn 26.67 µg/g in muscle, and the Cd 30.89, Pb 15.28, Cr 20.59, Cu 30.66 and Zn 32.42 in gills and Cd 34.44, Cr 18.00, Pb 17.34, Cu31.18 and Zn 58.44 µg/g in liver. The results showed that the liver appeared to the main heavy metals storage tissues, while the muscle of fish was lover accumulator compare than gill and liver. All the metals concentration exceeded the detection limits except than Mn. The results indicated that the selected fish does not feed directly without proper treatment of riverin ecosystem.

Keywords: Heavy metals, Water, Sediments, Kali River, Fish tissue

Introduction
The heavy metals are considered as critical toxic contaminants of aquatic ecosystems, due to their high potential to enter and accumulate in food chain (Olojo *et al*. 2005) [56]. The main sources of heavy metal pollution of the agriculture, industry and metropolitan cities, the bioaccumulation of toxic heavy metals in fish species from different aquatic systems is dependent on their foreign polluted substances. The distribution of heavy metals in water, sediments and fish play a key role in detecting sources of heavy metal pollution in aquatic ecosystem (Forstner and Wittman, 1981) [31]. The aquatic systems deposition of contaminants, including heavy metals, can lead to elevated sediment concentrations that cause potential toxicity of the aquatic biota (Yang and Rose 2003; Heyvart *et al*. 2000) [89, 34]. Because of the importance of sediments to the overall quality of aquatic systems, sediment analysis is often included in environmental assessment studies (Adekola and Eletta 2007; Li *et al*. 2006; Jain *et al*. 2005; Horsfall and Spiff 2002) [1, 44, 39, 37]. The evaluate of the heavy metal pollution load in the environment, it is usually not sufficient to measure only total concentrations, but also to establish the proportions of heavy metals present in various soluble fractions, which are commonly quantified by a sequential extraction procedure (Chester and Hughes 1967; Tessier *et al*. 1979, Forstner and Wittmann 1981; Horowitz *et al*. 1999; Stamatis *et al*. 2006) [17, 82, 31, 35, 74]. The levels of certain trace elements in river ecosystem have been found to be moderately, to very high polluted as a result of industrial discharges (Al-Masri *et al*. 2002; Coker *et al*. 1995) [6,18]. Sediments conserve important environmental information (Gutierrez *et al*. 2004) [32] and increasingly are recognized as both carriers and possible sources of contaminants in aquatic systems (Tessier *et al*. 1979) [82]. Evaluated heavy metals concentration in river systems are often considered indicators of anthropogenic influence and they are potential risk to the natural environment. Therefore, it is important to assess and track the abundance of these heavy metals. It is well known that the metals toxicity and bioavailability depends on other speciation, either in water or sediment. Heavy metals are distributed in sediments in four fractions, as exchangeable bound, iron– manganese oxide, organic matter and residual species (Dean, 2002) [21]. Sediments are important sinks for various pollutants like heavy metals and also play a significant role in the remobilization of contaminants in aquatic systems under

favourable conditions and in interactions between water and sediment river sediments, therefore, are important sources for the assessment of man-made contamination in rivers. Several studies on the distribution of heavy metals in sediments of the river Ganges and its tributaries have been carried out different types of pollution (Ajmal *et al.*, 1983; Subramanian *et al.*, 1987; Saikia *et al.*, 1988; Jha *et al.*, 1990; Ansari *et al.*, 2000; Dutta and Subramanian, 1998; Ramesh *et al.*, 2000; Singh, 2001, Singh *et al.* 2002) [3, 77, 65, 41, 7, 22, 63, 66, 68] have also reported that highly polluted sediments are adversely affecting the ecological functioning of rivers due to heavy metal mobilization from urban areas into biosphere. Therefore, in the present study, the distribution of heavy metals in both sediment and water of the river Gomti in different seasons at different sites in the Lucknow region have been investigated to initiate remedial measures to assess the pollution load.

The objective of the present study was to determine the levels of selected heavy metals Cd, Cr, Pb, Mn, Co in riverine water, sediment and three different organs like, muscles, gills and liver of *H. fossilis*. The fish *H. fossilis* live and search for food in the mud and sand at the bottom of the river, however occasionally feeds on water surface also. The above component of Kali river have been selected for this study because this river emerges from a very pure water source along different important industrial places situated at bank of river.

Materials and Methods

Description of site

Kali River that flow in western Uttar Pradesh (India) is a small perennial river having a basin area of about 150 km^2, and lies between latitude 29^{o}23'-29^{o}21' N and longitude 77^{o}43'-77^{o}39' E in the Muzaffarnagar district. Samples were taken from four zones, i.e. zone A, Nagal to Rohana; zone B, Muzaffarnagar to Sujru zone C, Mansoorpur to Mandawali, and zone D, Kutubpur village in River Kali shown in Fig 1. The River Kali receives several types of untreated municipal, industrial, and agriculture wastes from different point and non-point sources.

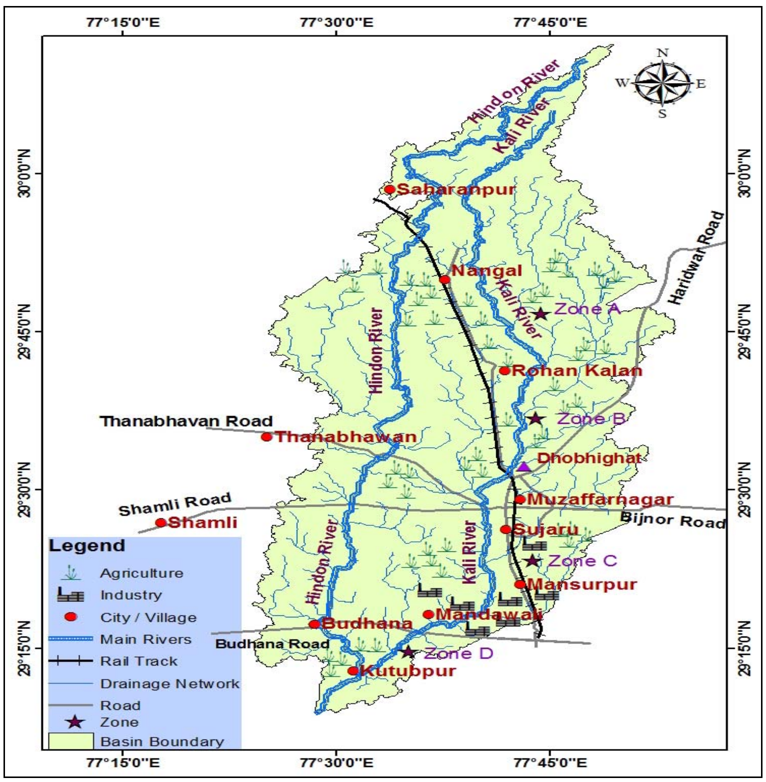

Fig 1: Map showing the study zone of Kali river district Muzaffarnagar.

Sampling procedure

Water

Surface water samples were taken from each zone by using 1-L polyethylene bottles and transported to lab in cooler boxes. The bottles were kept in 1% nitric acid (Analytical Grade) at 4 ^{0}C before their use.

Sediment

Sediment samples were taken from each zone, using the Ekman grab sampler. The sediment, samples were air dried, sieved with 230 mesh (600 μ) to separate only big materials and pebbles from the sediment and digested with 4:1 mixture of nitric acid and perchloric acid. The volume was again made up to 10 ml with 0.1N HNO_3. The sample of water and sediment were ready for analyzed by AAS.

Fish

Freshwater fish species, *H. fossilis* were collected seasonally in the Kali River. The collected fish were transported to the lab for separation of fish organs and muscles for the determination of concentrations of heavy metals.

Results and discussion

The present study indicated the concentration of heavy metals in the Kali river water ranged between, Cd, 0.001-0.0024 mg/l, Cr 0.002 0.087 mg/l, Pb 0.001-0.34 mg/l, and Zn, 0.004-0.367 mg/l were recorded. The mean values of the heavy metals were observed the out of detection limits except than Cd according to (WHO, 1993) [87]. Shown in Fig. 2 and Table 1. The lowest concentration of metals in Shur river water column were identified, there was little special variation in cadmium concentrations indicated this is a conservative metals in this aquatic system, whereas the Mn and Cu were observed under permissible limits. The Orontes river water indicated that highest concentration of all selected heavy metals study according (Ayse Baher 2010)[12] and second highest polluted river Tigaris study on (Hulya *et al* 2007)[38] shown in Table 2. The Zn, Mn and Cu are generally good indicators of anthropogenic inputs and zinc is used in galvanizing steel and iron product (Ayse Baher 2010) [12]. Zinc oxide is used in rubber manufacturing industries as an additive to activate vulcanization process. Zinc carbamates were used as pesticides (Aboud and Nandini 2009) [11]. The concentration of heavy metals varied seasonally and showed highest concentration in pre-monsoon season followed by monsoon and post-monsoon season. A similar trend was observed (Lohani *et al*. 2008) [46] in Gomti River (pre-monsoon > monsoon > postmonsoon).

Table 1: Concentration of heavy metals in water, sediments, *H. fossilis* fish in Kali river.

Heavy metals concentration		**Concentration**					
		Cd	**Cr**	**Pb**	**Zn**	**Mn**	**Cu**
Water (mg/l)	Minimum	0.001	0.002	0.001	0.004	0.002	0.032
	Maximum	0.024	0.087	0.34	0.367	0.287	0.047
	Mean	0.0052	0.013	0.059	0.079	0.044	0.041
	Detection limit	**1**	**0.05**	**0.005-0.008**	**0.05-0.01**	**5.0-3.0**	
Sediment (mg/kg)	Minimum	0.11	0.35	14.22	8.07	91.22	2.59
	Maximum	3.38	20.11	81.53	258.45	258.48	3.40
	Mean	1.23	7.43	32.16	63.84	163.53	3.09
	Detection limit	**8**	**ND**	**2**	**22**	**40**	
***H. fossilis* (mg/kg)**	Muscles	28.15	20.30	14.10	21.43	50.43	29.20
	Gills	31.65	20.92	17.00	24.08	52.44	33.12
	Liver	35.24	17.42	14.39	37.43	62.31	31.35
	Detection limit	**ND**	**0.15**	**0.2**	**1.5**	**150**	

Fresh water WHO mg/l (1993) [87], sediment mg/g Canadian EPA (1976) [27]. Fresh water fish WHO mg/g (1993)[87] ND Not Detected.

Table 2: Concentration of heavy metals in Kali river water and other selected river from literature.

Rivers	**Cd**	**Cr**	**Pb**	**Cu**	**Mn**	**Zn**	**Unit**	**Reference**
Ganga, India	0.012	0.018	0.086	0.03	NA	0.122	µg/l	Aradhana *et al* 2009 [9]
Hindon, India	NA	NA	37.0	3.45	NA	272.3	µg/l	Reza & Singh 2010 [64]
Gomti, India	0.0005	0.004	0.027	0.003	0.008	0.0287	µg/l	Vinod *et al* 2005 [85]
Shur, Iran	0.026	NA	0.116	0.771	NA	0.688	µg/l	Karbassi *et al* 2008 [43]
Tigris Turkey	ND	NA	ND	0.075	ND	0.1	µg/l	Hulya *et al* 2007 [38]
Sakarya Turkey	0.236	0.027	1.786	0.851	NA	0.173	µg/l	Mustafa *et al* 2007 [55]
Orontes Turkey	11.0	15.3	27.0	40.3	NA	39.0	µg/l	Ayse Baher 2010 [12]
Anujang Korea	BDL	2.72	BDL	1.43	47.6	8.11	Mg/l	Sanghoon Lee 2003 [79]
Khoshk Iran	NA	0.19	0.07	0.03	0.25	1.7	Mg/l	Salati 2010 [80]
Kali India	0.009	0.056	0.058	0.049	0.883	0.36	Mg/l	Present Study

Sediments act as the most important component of river ecosystem received the reservoir or sink of metals and other pollutants in the aquatic environment. The heavy metals concentration increases with decrease in the particle size and increase of organic matter content (Halcrow *et al*. 1973)[33]. Contaminated sediment can cause decrease in the ecosystem biodiversity and affect the aquatic systems and food chain. The organisms' food chains have been used as biomarkers in assessing level of contaminants in sediments. The special distribution of Kali river sediments observed that maximally Cr 20.11 mg/kg, Pb 81.53 mg/kg, Cu 258.45 mg/kg respectively in Fig 3 & Table 3. (U.S. EPA 2001)[83], the elevated level of trace metals especially in the sediments can be a good indication of pollution and high level of heavy metals and often can be attributed to anthropogenic influences, rather than natural enrichment of the sediment by geological weathering (Davies *et al.* 1991) [23]. Table.1 and Table. 3 and other selected river like Sakarya river (Mustafa *et al* 2007) [55], Shur river (Karbassi *et al* 2008) [43]. And Ganga river (Aradhana *et al* 2009) [9], represented same values of the heavy metals in sediment. During the rainy season (July–August) rivers are heavily flooded and the drainage system is drastically affected which results in mixing of polluted and unpolluted waters shown in Table 2. This leads to decrease in heavy metal concentration (Collvin 1985) [19]. whereas increase in the concentration of metals during summer seasons could be due to drought and decrease in water level. These metals exert sub lethal effect on aquatic organisms and predators adversely influence their reproduction and behaviour (Beyer *et al*. 2000) [15]. Metals are generally precipitated at alkaline pH in the form of insoluble oxides and carbonates. It has been proved that lethality increases of living animals and decrease oxygen concentration in river ecosystem. Increase in temperature also increases toxicity due to depletion in dissolved oxygen, increase in energy demand causing rise in respiration rate in the organisms, which leads to rapid assimilation of wastes (Bonga and Lock 2003) [16].The study reveals that there is a considerable variation in the concentration of heavy metals in water and sediment samples at various sites. These variations may be due to the change in the volume in the industrial sewage being added. In general, among different metals, Pb and Zn were higher than detection limits. The same trend was observed in case of sediments except for Cd, Cr, Pb and Cu in Table 3. Muzaffarnagar district, most of the people lives in catchment basin of Kali River and consumes river water for different uses such as bathing and cleaning of clothes (without any treatment and filtration). As these toxic substances like pesticides and heavy metals do not degraded, they are persistent in the water, sediment and fish tissues and other component of river ecosystem.

Table 3: Concentration of heavy metals in sediment in Kali River and other selected riverin sediments.

Rivers	Cd	Cr	Pb	Cu	Mn	Zn	Mg/kg	Reference
Luan, Chaina	0.37	152.73	38.29	178.61	NA	25.66	Mg/kg	Jingling *et al* 2009 [42]
Yangtze Chaina	3.40	205.00	98.00	129.00	NA	1,142.00	Mg/kg	Yang *et al* 2008 [90]
Huaihe Chaina	0.33	73.70	113.00	54.60	NA	83.10	Mg/kg	Zhang and Shan 2008 [93]
Pasvik Northern	3.84	NA	62.00	6,495.00	NA	439.00	Mg/kg	Dauvalter and Rognerud 2001 [20]
Gomti, India	8.38	19.13	75.30	35.03	NA	101.70	Mg/kg	Singh *et al* 2005 [73]
Po, Italy	2.10		98.50	90.10		305.00	Mg/kg	Farkas *et al*. 2007 [30]
Almendares, Cuba	4.30	23.40	189.0	420.80	NA	708.80	Mg/kg	Olivares- Rieumont *et al* 2005 [57]
Lahn,Germany	1.13	NA	68.40	48.20		245.20	Mg/kg	Martin 2004 [47]
Buriganga, Bangladesh	0.16	1.43	0.50	0.16	NA	0.26	µg/g	Mohiuddin *et al* 2011 [51]
Ganga,Allahabad, India	1.40	6.40	8.40	4.42	NA	20.40	Mg/g	Aradhana *et al* 2009 [9]
Tigris, Turkey	ND	NA	ND	69.2	337.6	25.73	µg/g	Hulya *et al* 2007 [38]
Kabini India	NA	254.52	11.67	110.55	115.66	87.94	Mg/g	Mohiuddin 2011 [51]
Kali, India	3.38	20.11	81.53	258.45	258.48	3.40	Mg/kg	Present Sudy

Note: All results are given in terms of the dry weight of material analyzed
NA: Not Analysis
ND: Not detected

Table 4: Comparison of heavy metal concentrations (µg/g dry weight, mean±SD) in different tissues of *H. fossilis*

Organs	Location	Pb	Cd	Cr	Zn	Cu	References
Muscle	Egypt	-	-	0.08±0.03	0.63±0.06	0.26±0.08	Rashed 2001 [62]
	Serbia	-	0.09±0.12	-	25.18±5.64	0.98±0.38	Jaric *et al*.2011 [40]
	Iran	1.11±0.40	0.09±0.06	0.57±0.15	20.974.42	21.812.79	Fallah *et al*. 2011 [29]
	Chaina	-	0.02±0.00	-	3.51±0.19	0.17±0.02	Onsanit *et al* 2010 [58]
	Turkey	023±0.07	0.02±0.03	0.56±0.34	25.5±8.64	5.36±3.30	Yilmaz *et al* 2010 [92]
	Bangladesh	1.990±03	0.36±0.01	1.56±0.11	26.67±1.37	8.05±0.00	Aleya *et al* 2013 [3]
	Kali river	15.28±0.99	30.89±0.21	20.59±0.65	52.1±1.83	30.66±0.92	Present study
Gill	Italy	2.43±0.06		0.33±0.13	34.53±18.28	2.33±0.40	Storelli *et al* 2006 [75]
	Egypt	-	-	0.22±0.09	1.37±0.44	0.34±0.12	Rashed 2001 [62]
	Serbia	-	0.15±0.13	-	62.39±15.62	2.05±0.38	Jaric *et al*.2011 [40]
	Bangladesh	5.83±0.41	3.57±0.26	3.62±0.26	17.81±1.93	6.31±2.46	Aleya *et al* 2013 [3]
	Kali, India	17.24±0.26	33.83±2.85	19.86±1.85	54.28±1.68	32.42±0.98	Present study
Liver	Italy	0.29±0.17	-	0.74±0.32	10.93±2.89	34.84±3.33	Storelli *et al* 2006 [75]
	Egypt	-	-	0.14±0.03	2.28±0.38	7.50±2.70	Rashed 2001 [62]
	Serbia	-	2.83±3.39	-	123.99±46.44	104.02±58.50	Jaric *et al* 2011 [40]
	Iran	2.89±0.57	1.87±0.56	1.32±0.32	81.07±8.92	58.49±9.81	Fallah *et al* 2011 [29]
	China	-	0.29±0.02	-	26.83±1.06	3.83±0.31	Onsanit *et al* 2010 [58]
	Turkey	2.42±1.75	0.29±0.15	1.37±0.33	6.73±1.07	34.50±3.27	Yilmaz *et al* 2010 [92]
	Bangladesh	18.16±0.52	3.92±0.40	6.30±0.91	60.81±0.14	45.61±1.29	Aleya *et al* 2013 [3]
	Kali, India	17.33±2.38	34.44±0.79	18.00±3.56	58.44±3.67	31.18±0.95	Present study

The composition of heavy metals in river water and sediment may be used to identify major pollution sources entering river Kali. The mean concentration of heavy metals in the muscles, gill and liver of commercially valuable freshwater edible *H. fossilis* are presented in Fig. 4 &Table 4. Although our previous work evaluated water, sediments and fish tissue for elemental contents (Maurya and Malik 2015) [48]. The metals of Cadmium, chromium, lead, copper and manganese were selected as the analysed elements from the viewpoint based on the basis of types of industry near the Kali River and the metal pollution anticipated. The elemental concentration in the tissues of *H. fossilis* did not very significantly measure among the all station. This means that the water quality of the river hardly changed among the all station. In order to evaluate the elemental concentrations in freshwater fish tissues in the Kali river, we compared measured values with those in the tissues obtained at order places (Storelli *et al*. 2006; Rashed 2001; Jaric *et al*. 2011; Shah *et al* 2009; Fallah *et al* 2011; Onsanit *et al*. 2010; Yilmaz *et al*. 2010) [75, 62, 40, 67, 29, 58, 91].

Present study has suggested that Cadmium, Chromium, lead act as an endocrine disruptor material at extremely high concentration were observed of Cd in muscle, gill, liver 28.18, 31.65, 35.24 mg/kg, chromium in muscle, gill, liver were observed 20.30, 20.92, 17.42 mg/kg, in lead concentration muscle, gill, liver 14.10. 17.00, 14.39, and zinc in muscle, gill, liver were observed 21.43, 24.08, 37.43 mg/kg respectively in Fig. 4 & Table 4. As the concentration in the muscle, gill and liver observed in this study is different from the result in Pakistan (Shah *et al* 2009) [67].

The highest cadmium concentration was found in the liver of *H. fossilis* (35.24 mg/kg), while the lowest cadmium concentration in muscles (28.15 mg/kg). The same distribution pattern of Cd recorded in the studies (highest levels in the liver, lowest in the muscles) was found in other fish species as well as (Jaric *et al* 2011; Onsanit *et al* 2010). The highest concentration of Pb (17.00 mg/kg) in the gills approached the 0.5 µg/g threshold, which is considered harmful to fish and predators (Walsh *et al* 1977). The maximum admissible value for fish is 0.05 mg/kg wet weight (EU commission 2001) [26]. From our experimental study, it was sharply observed that Cd in the muscle of *H. fossilis* from Kali River the values, the long period of accumulation of Cd in fish may pose health hazards.

The Cr level in the muscles of *H. fossilis* was maximally 20.30 mg/kg in higher than the result observed by other authors (Rashed 2001; Fallah *et al* 2011; Yilmaz *et al* 2010) [62, 29, 92]. According to these results, it could be suggested that there is dense Cr pollution that might have resulted from the effluent coming from the tannery industries near the Kali River. (Mohanta *et al* 2010; Ahmed *et al* 2011)[54, 5], was observed that the high concentration of heavy metals cause adversely affected of fish consumption of the local people.

In the present study, Cu concentrations were maximum 33.12 mg/kg in the gill, with the lower levels in the muscles 29.20 mg/kg found shown in Table 1. The level of Cu in the liver in the other fish tissues has been also observed by other authors (Yilmaz *et al* 2007; Storelli *et al* 2006; Rashed 2001; Uysal *et al* 2009; Wu *et al* 2006; Farag *et al* 2007)[92, 75, 62, 84, 88, 28]. According to Pyle *et al* 2005, the liver Cu concentrations are usually regulated by a homeostatic control below 50 µg/g dry weight and can exceed this threshold only if the control mechanisms are overloaded. High Cu level found in the present study was observed 31.17 mg/kg (dry weight) might imply loss of regulatory control of the lever of Cu (Pyle *et al* 2005) [61]. Overall, the highest concentration of Cd, Zn and Mn were recorded in the liver and Cr and Pb in gill were found. Mormede and Devies (2001) [53]. have suggested that the liver was the target organ, showing the detoxification and accumulation role of the liver. The muscles is generally considered to have a weak accumulating potential (Erdogrul and Erbilir 2007; Uysal *et al*. 2009; Bervoets and Blust 2003) [25, 84, 13]. The liver is the preferred organ for metals accumulation as could be deduced from the present study. Such a pattern has been observed in a number of other studies, covering a wide spectrum of fish species (Storelli *et al* 2006 ; Rashed 2001; Dural *et al* 2006 ; Pyle *et al* 2005 ; Ploetz *et al* 2007; Agah *et al* 2009) [75, 62, 24, 61, 59, 2].

This research showed that all heavy metals concentration in the liver, gill, muscles were out of detection limits except than Mn. Moreover, continuous monitoring of all heavy metals will be needed for the fish consumption of the people of Muzaffarnagar city because heavy metal pollution in the Kali river has been reported (Malik and Maurya 20015) [48].

The gill, liver is the preferred organs for heavy metals accumulation as could be deduced from the present study. This research showed that the gill, liver have almost higher metal concentration than the muscles. Such pattern has been observed in a number of other studies, covering a wide spectrum of fish species (Stprelli *et al* 2006; Rashed 2001; Dural *et al* 2006; Pyle *et al* 2005; Ploelz *et al* 2007; Agah *et al* 2009) [75, 62, 24, 61, 59, 2]. Bottom dwelling fishes are found to exhibit higher concentration of heavy metals than pelagic fishes (Atchison *et al*. 1977) [10]. The increase in concentration of metals in fish could be mainly due to metal contaminated diet which comes from discharge of effluents into rivers from different industries and other sources in the form of particulates and solutions (Mount and Stephan 1969) [52].

The concentration of the heavy metals are higher in the tissues than the recommended value, which suggest that the Kali river is, to a certain extent, a heavy metals polluted river, and the fish consumption are not completely safe for health. Therefore, further continuous monitoring of heavy metals concentration in fish will be needed in India. Increase in concentration of Cd and Cu in fish liver is greater than other organs. Cd and Cu at sublethal concentration in fish decreases survival growth and reproductive health and mainly accumulate in gills and liver (Beyer *et al*. 2000) [15]. Its bioaccumulation can decrease oxygen consumption. In several studies *H. fossilis* species obtained from different selected countries.

Conclusion

The results of this study revealed that the concentrations of metals are high toxic and many pose hazard. The Kali River showed that the pollution load increases when Kali River enters in to the Muzaffarnagar city. This is evident by the increase in Cd, Cr and Pb concentration which exceeded the standard detection limits. The concentrations of heavy metals in fish organs like, Cd, Cr, Pb and Zn in *H. fossilis* from gill, liver from exposed were clearly out of detection limits of (WHO, 1993)[87]. The concentration of the heavy metals are higher in the tissues than the recommended value, which suggest that the Kali river water, sediment and fish *H. fossilis* were highly polluted and fish are not safe for human health therefore, further continuous monitoring of heavy metal concentration in water, sediments and fish in Kali river.

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