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Seasonal pollution of heavy metals in water, sediment and tissues of catfish (*Heteropneustes fossilis*) from Gogabil lake of north Bihar, India

Arbind Kumar, Anil Kumar and SK Jha

Abstract
In the present study, seasonal pollution of heavy metals (Cu, Cr, Zn, Cd, Pb) were determined in water, sediment and fish tissues (liver, gills, muscles) of *Heteropneustes fossilis* at three sites of Gogabil lake during 2018-2019. The results showed that total metal concentration in water were higher in summer except Pb, whereas in sediment they were higher in winter except for Zn. In the fish tissues the levels of Zn throughout the season were in the order of liver > gills > muscles, while Cd contents were gills > liver > muscles. Cu contents were highest (41.7±0.862µg g^{-1}) in gills while Cr contents (1.95±0.03µg g^{-1}) and Pb contents (1.703± 0.025µg g^{-1}) were highest in liver during summer. However total metal accumulations were highest in liver and gills and lowest in muscles. The higher accumulation of metals during summer and winter season was attributed to higher influx of disposal of municipal and household wastes and also agricultural runoff of catchment area. In addition, the pollution load index (PLI) values > 1 suggested that deterioration of sediment quality and I-geo values indicated moderate pollution by Cr and high degree pollution by Cd (class 5). Sediment quality guidelines (SQGs) indicating 27.7% sediment samples were moderately and 13.3% were highly polluted, whereas mean probable effect concentration quotients (m-PEC-Q) confirmed that moderate toxicological risks for sediment dwelling organism, with a toxicity incidence of 15-29%. This study suggests that lake water and selected fish are serious concern of human health and potential danger may occur in the future.

Keywords: Bioaccumulation**,** heavy metals, water, sediment, fish tissues, gogabil lake

1. Introduction
Pollution of heavy metals is a threat to the environment due to their toxicity, persistence in the environment, and bioaccumulation in nature **(**Kumar *et al.,* 2015; Kumar and Seema, 2016; Kumar *et al.,* 2017; Xu *et al.,* 2018; Ali *et al.,* 2019) [39, 40, 41, 108, 4]. Heavy metals enter in aquatic ecosystem from natural and anthropogenic sources. Anthropogenic activities continuously increase the amount of heavy metal in the aquatic ecosystem (Xu *et al.,* 2018) [108]. As heavy metals cannot be degraded, they are deposited, assimilated or incorporated in water, sediment and aquatic organisms and thus, causing heavy metal pollution in aquatic ecosystem (Kumar and Kumar, 2018; Farsani, *et al.,* 2019) [43, 30].

Sediment is the lose sand, clay, slit and other soil particles that settle at the bottom of rivers, lakes dams reservoirs and oceans. It is an important edaphic factor which to have effect not only the water quality standing upon it but also influences quantitative and qualitative distribution of aquatic organism along their productivity. Sediments are ecologically important components which play significant role in maintaining the tropic states of water body. Their quality can indicate the states of water pollution (Zahra *et al.,* 2014) [112]. The distribution of heavy metals in the sediments is affected by chemical composition of the sediments, grain size and content of total carbon matter (Zhao *et al.,* 2014; Adhishwar and Choudhary, 2015; Ali-Azidi *et al.,* 2018) [113, 1, 5]. The release of heavy metals from sediment to water bodies is affected by the overlying water conditions, pH, alkalinity, salinity, dissolved oxygen concentration and suspended solid (Li *et al.,* 2013) [53] and found that physical disturbance of sediments released metals more rapidly than biological disturbance (Atkinson *et al.,* 2007) [14].

In recent years, the consumption of fish has increased rapidly with awareness of its nutritional and therapeutic benefits (Sioen *et al.,* 2007; Bawuro *et al.,* 2018) [77, 15]. Fish contains high level of unsaturated acids and low levels of cholesterol and also has high levels of many important nutrients, including high quality of proteins, iodine and various vitamins and minerals (El-Mosclhy 2000; Bawuro *et al.*, 2018; Rajeskumar and Li, 2018) [24, 15, 75]. According to the literature, it contains omega-3 fatty acids, which are beneficial for humans who obtain from their diet, which have potential health benefits, such as helping prevent cardiovascular disease, prevent and treat depression, reduced risk of type -1 diabetics, prevent asthma in children and protect vision in old age and also improve sleep quality (Kris-Etherton *et al.*, 2002; Afshan *et al.*, 2014) [44, 2].Various researches have shown the adverse effect of heavy metals to human health, such renal failure, cardiovascular diseases, liver damage and even death (Castro-Gonzalez and Armenta, 2008; Al-Busaidi *et al.*, 2011; Rahman *et al.*, 2012) [18, 7, 73]. Bioaccumulation of heavy metal in freshwater fish depends upon on the various factors like age, size, sex, reproductive cycle, feeding behaviour, swimming pattern and geographical location (Zhao *et al.*, 2012; Egbeja *et al.*, 2019) [113, 27]. The retention of heavy metal in the body of an organism depends on many factors such as the speciation of the metal concerned and the physical mechanism developed by the organism for the regulation, homeostasis, and detoxification of the heavy metal. The degree of bioaccumulation in different tissues of fish is generally different depending in the active tissue as liver, gills, and kidney have higher accumulation of the heavy metal than other tissues such as skin and muscles (Maurya *et al.*, 2019; Ezekiel *et al.*, 2019) [62, 25].

Gogabil Lake is one of the largest wetlands of Bihar, and is connected with two major Rivers Mahananda and Ganga. It receives large amount of wastes through multifarious human activities. The urban wastes management and garbage disposal organized body around the lake are unproductive, so that some area is used as general dumpsite of the urban. The lake provides level of the local economic activities such as fish production and irrigation system. There are potential irrigable lands around the lake for the cultivation of rice, maize and vegetables for food security. Excessive use of fertilizers, pesticides in the cultivation resulted in elevated metals concentration in the lake. There are no published papers are available to study about trace metals concentration in the water, sediments and fish species of the Gogabil lake. Therefore this study was conducted (1) to assess the seasonal variation of metal load of Cu, Cr, Zn, Cd and Pb in water, sediment and shingi fish tissues (liver, gills and muscles) (2) to estimate bioaccumulation factor and relationship between water/sediment and fish tissues.

Materials and methods

Study area

Gogabil Lake is the first largest freshwater lake in Bihar, and lies in between $25^0$22.5233 North latitude and $87^0$4121.63 East longitudes. It covers an area of about 60 km^2, out of which about 20 km^2 is purely lake throughout the year. The lake is directly linked to Ganga and Mahananda River and during the peak of rainy season and flood; the lake communicates to these rivers through channels and tributaries. Fig. 1 showing location and route map and Fig. 2 showing satellite map of Gogabil Lake.

Collection of water, sediment and fish samples

The water, sediment and freshwater Shingi fish (*Heteropneustes fossilis)* were collected from Gogabli Lake at 3 sites (Fig. 2) during summer, rainy and winter seasons from April 2018 - March 2019. About 500 mL water samples were collected at 0.5 meter below the water surface, filtered in pre cleaned bottle and preserved by adding 5 mL of 20% HNO_3 to it and then packed in ice bath (4 ^{0}C) and brought to laboratory for further digestion (APHA, 2005) [9]. About 500g sediments samples were collected at a depth 0-10 cm by applying method of (US EPA, 2001) [88] and immediately transferred into polyethylene bags, which were already washed with 10% HNO_3 solution. All sediment samples were transferred using ice box to the laboratory for further processing. A total 30 fish sample was collected at the three same sampling locations from the lake basin. After identifying all fishes, total length 20±0.1 to 25±0.1 cm and total weight 250±0.1 to 455±0.1g were recorded. About 3g of the epaxial muscle on the dorsal surface, the entire liver and four gill racers from each sample were dissected by applying the method of (Ali-Busaidi *et al.*,2011; Voegborlo *et al.*, 2012) [7, 91] and put in to Petri plate to dry at 120 ^{0}C until reaching a constant weight.

Digestion of water, sediment and fish samples

100 mL water sample was taken in conical flask and 5 mL of concentrated HCl acid was added to it and then heated on the hot plate for two hours at 105^0C to 25 mL. The concentrated water sample was then transfer into 100 mL volumetric flask and distilled water was added to fill up to the mark and then analysed for Cu, Zn, Cr, Cd and Pb using Atomic Absorption Spectrophotometer. To estimate the heavy metal content, 2g of sediment sample collected from each site was digested separately by applying method of (Allen *et al.*, 1986) [8], modified by (Singh *et al.*, 2017) [80]. After this digested solution was filtered in pre-cleaned 100 mL measuring flask and volumes were made up to mark and then subjected to atomic absorption spectrophotometer for analysis of heavy metals. The dried fish tissues were digested by the method described by (Voegborlo *et al.*, 2012) [95] modified by (Bawuro *et al.*, 2018) [15]. After complete digestion each digested mixture was transferred in to 50 mL volumetric flask and distilled water was added to it to fill up to the mark and analysed heavy metals using Atomic Absorption Spectrophotometer.

Bioaccumulation factor (BAF)

BAF was calculated by following equation described by (Kalvins *et al.*, 1998) [45] and modified by (Ali *et al.*, 2019) [4]:

$$BAF = \frac{C fish\ tissue}{C\ water\ or\ sediment}$$

Where $C_{fish\ tissue}$ is the metal concentration in fish tissue and $C_{water\ or\ sediment}$ is the metal concentration in water or sediment.

Pollution load index (PLI)

The PLI of a single site is obtained as the n^{th} root of *n* number of multiplied together contamination factor (C_f) values. The index is computed as follows (Thomilson *et al.*, 1980) [88]:

$$PLI = n\sqrt{}\,(C_{f1} \times C_{f2} \times C_{f3} \times \text{-------} \times C_{fn})$$

Where, n is the number of metals studied (5 in the study) and the C_f is the contamination factor. The PLI < 1 refers no pollution; PLI =1 present that only baseline, whereas PLI>1 would indicate deterioration of site quality (Thomilson *et al.*, 1980; Mohiuddin *et al.*, 2012) [88, 64].

Geo-accumulation index (I-geo)

I-geo values for heavy metals in sediment were determined by using following equation as introduced by (Muller *et al.*, 1969) [58] and described by (Boszke *et al.*, 2004) [17].

$$I\text{-}geo = \log_2 \frac{Ci}{1.5 \times Bn}$$

Where C_i is measured concentration of metal in sediment and B_n is geochemical background value in average shale of element n.

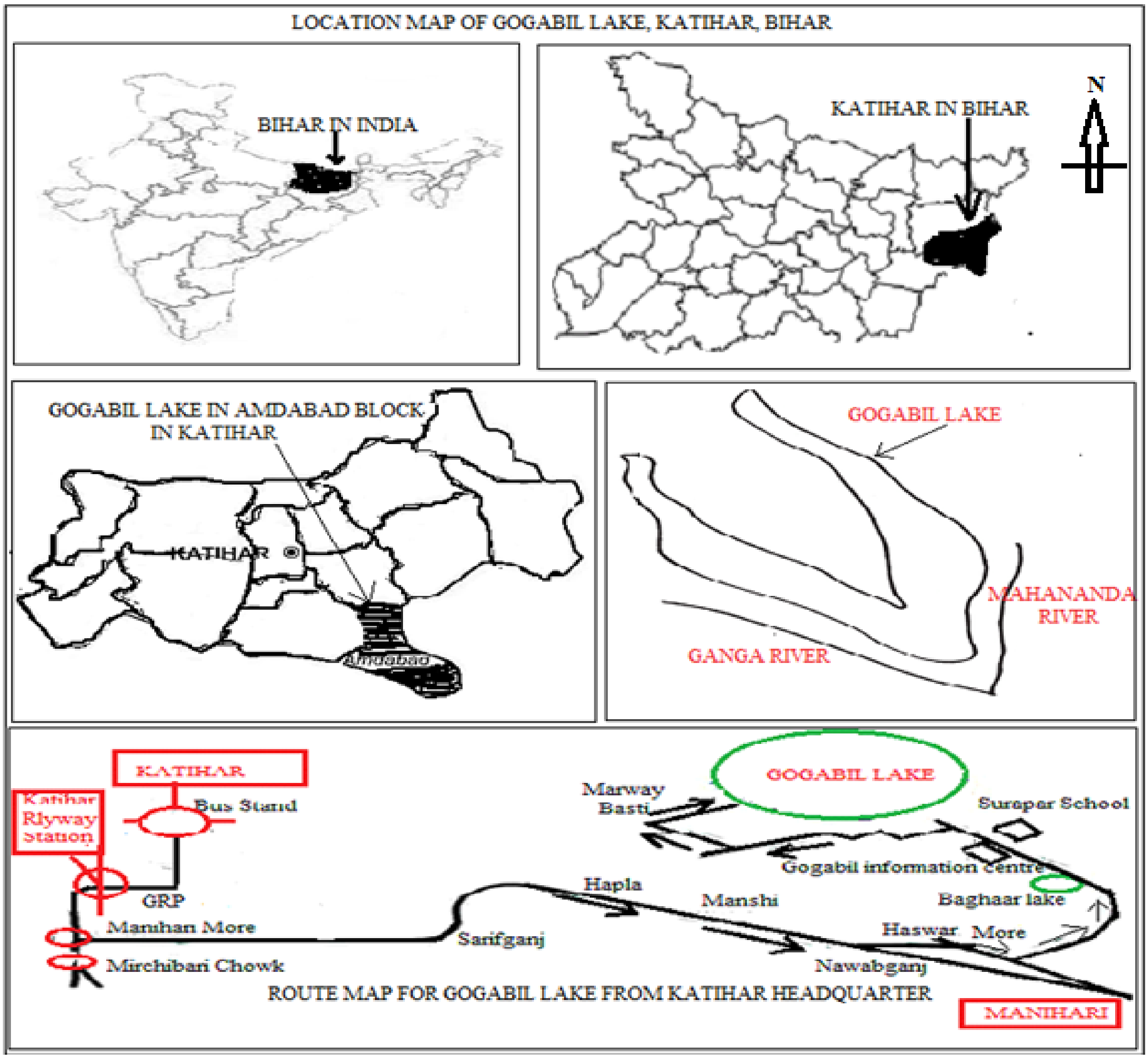

Fig 1: Map showing location and route of Gogabil Lake

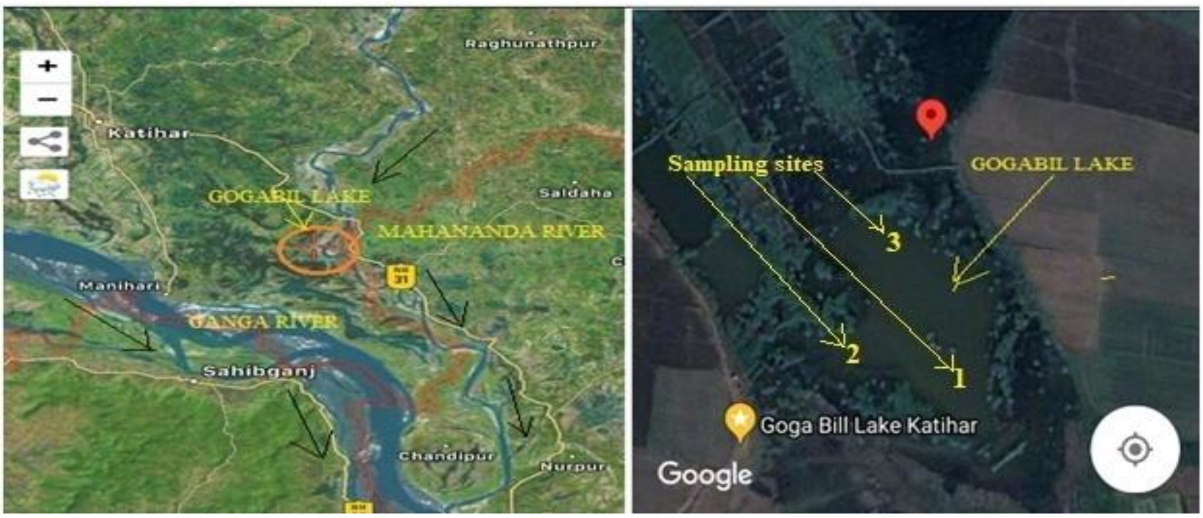

Fig 2: Satellite map of Gogabil Lake, Katihar (Taken from maps.google.com)

Sediment quality guidelines

The consensus-based sediment-quality guidelines (SQGs) were proposed by (MacDonald *et al.*, 2000) [61], which included a threshold effect concentration (TEC) and a probable effect concentration (PEC) (MacDonald *et al.*, 1996; Swartz, 1999; MacDonald *et al.*, 2000; Varol *et al.*, 2012; Lin *et al.*, 2013; Wang *et al.*, 2016) [60, 79, 61, 96, 54, 104]. The toxicity of heavy metals present in bed sediment on aquatic-organism was evaluated by determining the mean probable concentration quotients (m-PEC-Q) in sediment samples. The m-PEC-Q values for heavy metal in sediment were calculated by applying following equation as described (MacDonald *et al.*, 2000) [61]:

$$\text{m} - \text{PEC-Q} = \sum_{i=1}^{n} \left(\frac{Ci}{PECi}\right)/n$$

Where C_i = content of metal in sediment sample 'i', PECi is the PEC for individual metal 'i' and 'n' is the number of study metals.

Statistical Analysis

Statistical analysis were performed by using lenovo ™ computer using the Microsoft EXCEL and Word 2007 format. The mean, standard deviation of heavy metals concentration in water, sediments and fish tissues were calculated by using Casio calculator (made in China) *fx-991 MS*. A probability level of $p < 0.05$ was considered statistically.

Results and Discussion

Metal ions concentration in water

Seasonal variation of levels of heavy metals in the water is present in Table 1. Significant variations of heavy metals were found with higher values in summer followed by winter and rainy. Heavy metal concentrations in water samples are in the following order Cu > Pb > Cr > Zn > Cd in summer and Cu > Pb > Zn > Cd > Cr in winter and rainy seasons. The average concentration of Zn and Cd remained relatively constant in the study period. The average level of Cu (0.478 mg/l), Cr (0.06 mg/l), Zn (0.067 mg/l), Cd (0.0.65 mg/l) and Pb (0.14 mg/l) in water samples were significantly higher the heavy metals levels of various river, Koshi (Singh *et al.*, 2016) [81], Ghaghra (Singh *et al.*, 2016) [82], Kali (Marurya amd Malik, 2016) [66], Gomati (Vinod *et al.*, 2005) [97] and Ganga (Gupta *et al.*, 2009) [35] except Zn (Table 1). The levels of Cu and Zn were lower and concentrations of Cd, Cr and Pb were higher than (WHO joint FAO/WHO, 2011) [102] and EPA (Environmental Protection Agency 2002) [23] during all three seasons. The concentrations of all studied metals did not exceed the EC (Eruption Commission, 1998) [28] water quality direction for human water consumption. Difference between three sampling seasons with higher concentration of metals during summer, may be explained by metrological conditions. The summer season's combined effect of increased vaporisations and decreased rainfall may lead to higher concentration. Similar trends was reported by various authors (Terkin-Ozan, 2007; Terkin-Ozan and Kir, 2008; Duman and Kar, 2012; Salem *et al.*, 2014; Rajeshkumar *et al.*, 2018; Farsani *et al.*, 2019) [90, 91, 22, 87, 74, 30].Moreover reverse result was observed in rainy might be due to rainfall effect which caused increase the lixiviation process and continue to the dilution of heavy metals during wet season (Khattabi *et al.*, 2007) [49].

Metal ions concentration in sediment

Seasonal pollution of heavy metal concentrations in the sediment of Gogabil lake are statistical different are presented in Table 2 ($p < 0.05$). The level of heavy metals in the sediments was higher in winter and summer than rainy season. The mean concentration of heavy metal in the sediments was in decreasing order of Cu >> Zn >> Pb > Cr ≈ Cd in winter, Cu ≈ Zn >> Pb > Cr > Cd in summer and Cu >> Zn >> Cd ≈ Cr > Pb in rainy season. The average concentration of Cr and Cd were approximately same during study period. High concentration of heavy metal in sediment in winter and summer than rainy may be due to accumulation of heavy metals in sediment was also supported by so many researchers (Wang *et al.*, 2012 a,c; Islam *et al.*, 2015; Mohammad Ali *et al.*, 2016; Salam *et al.*, 2014; Rajeshkumar *et al.*, 2018; Farsani *et al.*, 2019) [103, 105, 38, 63, 87, 74, 30]. The mean concentration of Cu 119 ± 3.3 mg kg^{-1} in winter, 114.46 ± 2.42 mg kg^{-1} in summer and 75.37 ± 2.6 mg kg^{-1} in rainy season was higher than LEL, TEC, and ASV (Table 2). The high concentration of Cu in sediments during winter and summer might be attributed to agricultural activities and may be Cu based fertilizers and pesticides used in agricultural land around the Gogabil lake as winter and summer season are the main season for agricultural activities (Fu *et al.*, 2014; Ahmed *et al.*, 2016; Neschi *et al.*, 2013; Rajeshkumar and Li, 2018; Farsami *et al.*, 2019) [31, 12, 67, 75, 30].

Table 1: Seasonal variation heavy metal in water (mg/l) of Gogabil Lake with International guidelines and World's River

Season	Cu	Cr	Zn	Cd	Pb	Reference
Summer	0.613 ± 0.03	0.076± 0.005	0.073±0.002	0.071±0.003	0.123± 0.005	Present study
Rainy	0.40 ± 0.05	0.051± 0.001	0.062±0.003	0.062±0.002	0.12 ± 0.017	
Winter	0.42 ± 0.04	0.053± 0.001	0.065±0.001	0.063±0.002	0.176± 0.005	
WHO	1.5	0.05	15	0.005	0.01	Joint FAO/WHO, (2011) [102]
EPA	1.3	0. 05	0.5	0.01	0.05	Guidelines, (2002) [23]
Kovada lake, Turkey	0.54 ± 0.28	0.92 ± 0.48	5.18 ± 1.06	0.19 ± 0.14	BDL	Kayrak & Ozan, (2018) [47]
Gadilamn River, India	0.50 ± 0.025	0.70 ± 0.26	0.10 ± 0.002	1.46 ± 0.054	0.46 ± 0.018	Ambedkar &Muniyn, (2012) [10]
Koshi River India	0.014-0.026	0.001± 0.006	0.017-0.024	0.009-0.026	0.004-0.123	Singh &Shukla, (2016) [81]
Ghaghara River, India	0.016-0.032	0.001 ±0.007	0.013-0.031	0.003-0.043	0.005-0.019	Singh *et al.*, (2016) [82]
Kali River India	0.049	0.056	0.36	0.009	0.058	Maurya & Malik, (2016) [66]
Gomti River, India	0.003	0.004	0.287	0.0005	0.027	Vinod *et al.*, (2005) [97]
Ganga, India	0.03	0.018	0.122	0.012	0.086	Gupta *et al.*, (2009) [35]
	Aras Dam Lake, Iran					Farsani *et al.*, (2019) [30]
Summer	0.37 ± 0.25	------	0.06 ± 0.04	0.06 ± 0.02	0.17± 0.03	
Autumn	0.54 ± 0.10	------	0.07 ± 0.01	0.07± 0.01	0.13 ± 0.02	
Winter	0.22 ± 0.12	------	0.03 ± 0.02	0.02 ± 0.02	0.04 ± 0.03	

	Meiliang Bay of Talu lake, China					Rajeshkumar *et al.*, (2018) [74]
Summer	0.0001	------	------	0.0002	0.0006	
Rainy	0.0004	------	------	0.0001	0.0002	
Winter	0.0003	------	------	0.0007	0.0005	
EC	2	50	------	5	10	European Commission, (1998)[28]

The mean concentration of Cd was 7.47 ± 0.371mg kg^{-1} in winter, 5.94 ± 0.085 mg kg^{-1} in summer, 4.33 ± 0.21mg kg^{-1} in rainy. High concentration of Cd was found in water, which might be due to the difference in water capacity of the lake where low water flow in water resulting the precipitation of Cd in the sediment thereby raising its concentration (Mohammad Ali *et al.*, 2016; Rajeskumar *et al.*, 2018) [63, 74]. The mean level of Pb was observed 13.4 ± 0.82, 11.87 ± 1.65, and 3.65 ± 0.58 mg kg^{-1} in winter, summer and rainy respectively, was lower than LEL, TEC, PEC, SEL and ASV (Table 2), which could be due to the effect from point and non point sources such as municipal runoff and atmospheric deposition (Yu *et al.*, 2012; Mohiuddin *et al.*, 2012) [109, 64]. The mean concentration of Cr 7.92 ± 1.27, 7.45 ± 0.41 and 4.00 ± 0.67 mg kg^{-1} in winter, summer and rainy respectively, which was lower than LEL, TEC, PEC, SEL and ASV(Table 2) which could be due to large amount of waste water had been received from Mahananda and Ganga River.

Table 2: Seasonal variation of heavy metal in sediment (mg kg^{-1}) of Gogabil Lake with International guidelines and World's River

Season	Cu	Cr	Zn	Cd	Pb	Reference
Summer	114.46 ± 2.42	7.45 ± 0.41	113.46 ± 2.1	5.94 ± 0.085	11.87 ± 1.65	Present study
Rainy	75.37 ± 2.6	4.00 ± 0.67	55.16 ± 2.13	4.33 ± 0.21	3.65 ± 0.58	
Winter	119 ± 3.3	7.92 ± 1.27	94.0 ± 2.09	7.47 ± 0.371	13.4 ± 0.82	
	Environment Protection Agency guideline for sediment					
Unpolluted	< 25	< 25	< 90	-----	> 40	EPA,(2002) [23]
Moderately	25-50	25-75	90-200	-----	40-60	
Heavily polluted	> 50	> 75	> 200	> 60	>60	
Mahananda River India	49.51 -82.78	121.34 -198.7	83.64 -141.68	0.53-1.05	17.91 -29.52	Kumar *et al.*, (2019) [42]
Ghaghara River, India	2.76 -11.74	15.29 -25.59	13.26 - 17.59	0.21 - 0.28	10.71 - 14.26	Singh *et al.*, (2017) [80]
Kali River India	258.45	20.11	3.4	3.38	81.53	Maurya & Malik, (2016) [66]
Cauvery River, India	11.2	38.9	93.1	1.3	4.3	Raju *et al.*, (2012) [72]
Ganga, India	39-73	113-230	72-140	0.45 -0.95	15 -27	Singh *et al.*, (2013) [80]
Boriganga, Bangladesh	184.4	101.2	502.3	0.8	79.8	Saha and Hossan (2010) [76]
Gadilm River, India	0.60 ± 0.022	0.94 ± 0.04	0.20 ± 0.009	1.64 ± 0.05	0.48 ± 0.014	Ambedkar & Muniyan (2012) [10]
Plitvice Lakes Park	115	------	258	10.9	62.8	Vukosav *et al., (*2014) [101]
Mljet National Park	300	------	377	14.8	62.8	Cuculic *et al.*, (2009) [20]
	Aras Dam Lake, Iran					Farsani *et al.*, (2019) [30]
Summer	103.6 ±7.09	------	94.0 ± 4.58	7.20 ± 0.82	12.2 ± 1.12	
Autumn	142 ± 8.19	------	111 ± 3.12	5.55 ± 1.13	10.1 ± 1.68	
Winter	73.33 ± 7.64	------	53.33 ± 4.73	3.93 ± 1.14	3.03 ± 0.25	
	NOAA, (2009) [68]					
LEL	16	26	-----	0.6	31	Lowest Effect Level
TEC	31.6	43.4	-----	0.99	35.8	Threshold Effect Concentration
PEC	149	111	-----	4.9	128	Probable Effect Concentration
SEL	110	110	-----	10	250	Severe Effect Concentration
ASV	45	90	95	0.3	20	
European background	2-100	------	10-200	0.1-1	2-80	Samecka-Cymerman and Kempers, (2001) [78]

The mean concentration of Zn in sediment was observed to be 113.46 ± 2.1mg kg^{-1} in summer 94.0 ± 2.09 mg kg^{-1} in winter and 55.16 ± 2.13 mg kg^{-1} in rainy season which was higher in summer, lower in rainy and same in winter as ASV (95 mg kg^{-1}) value (Table 2). The main source of Zn at study area was excess fertilizers and pesticides used in agriculture, soil erosion due to rain fall, and land construction (Higgins *et al.*, 2007; Chen *et al.*, 2004) [36, 19].

Pollution load index, Geo-accumulation index and Sediment Quality Guidelines

The PLI of heavy metals in sediment of Gogabil lake is given in Fig. 3, which was ranged from 0.4028 -1.7363 during studied period. The highest value of PLI was recorded in winter at site- 3 and lowest value was recorded in rainy at site-2. I-geo values based on average shale value recommended by (Turkian and Wedephohi, 1961) [89] are presented in Fig. 4. The results revealed that I-geo values varied from 0.1591 to 0.8181 for Cu, -5.077 to - 4.09 for Cr, -1.369 to – 0.3288 for Zn, 3.267 to 4.054 for Cd and -3.039 to – 1.163 for Pb. PLI value > 1 suggested that deterioration of sediment quality especially in winter and summer at all sites of Gogabil Lake. The results thus obtained are very close to 1.145 -1.209 our previous study (Kumar *et al.*, 2019) [42] on Mahananda River in Seemanchal zone. The PLI values also indicated that trend of heavy metals contamination in the sediments of winter > summer > rainy. The high PLI values indicated that Cd, Cu and Zn are the major contributors to the sediment pollution and can provide some understanding to the inhabitants about the quality of the environment. In addition, it also provides valuable information to the decision maker on the pollution status of the area.

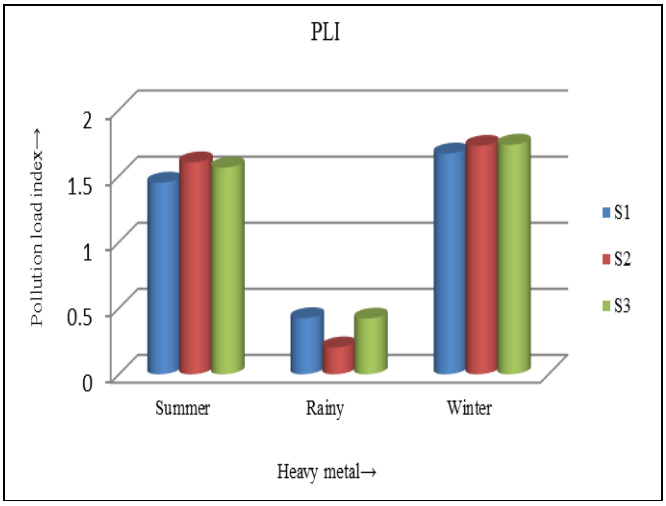

Fig 3: PLI of heavy metal in Gogabil Lake

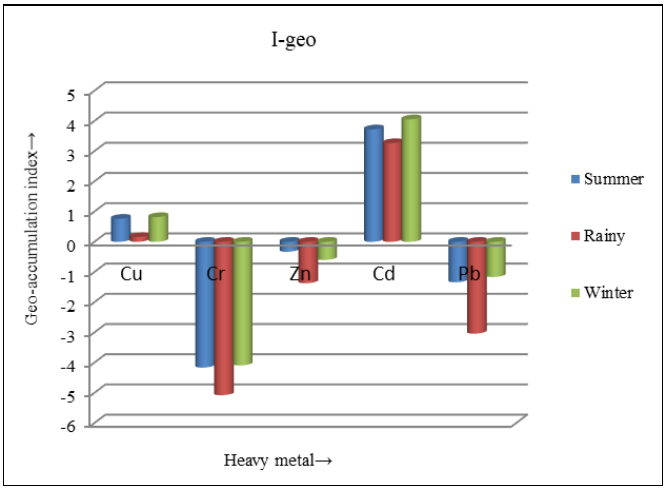

Fig 4: I-geo of heavy metal in Gogabil Lake

According to I-geo classification, sediments of Gogabil Lake may be unpolluted with reference to Cr, Zn and Pb as their I-geo values ranged from -5.078 to -0.6003 along the study area. On the other hand I-geo value of Cu ranged from unpolluted to moderately pollute. In addition to high degree of pollution was found in the sediments by Cd (class 5). Several negative I-geo values were noted for all considering metals at many sites in all three seasons showed that Gogabil lake bed sediment is uncontaminated for the most of the trace metals in the study area. Previous study carried out by (Kumar *et al.*, 2019) [42] on Mahananda River in Seemanchal zone noted that several negative I-geo values for Cu, Zn, Mn and Pb at various sites except Cd and Cr. Similar results were reported by (Singh *et al.*, 2013) [84] on Ganga River at various sites. As Gogabil lake is direct linked to Ganga and Mahananda River and receives effluents from many small industries, textile battery producing unit from these Rivers and also agricultural runoff from its catchment area.

The ecological risk of single heavy metal and the combined ecological effect of five heavy metals for Gogabil Lake using the SQG method are presented in Table 3. SQG method revelled that the metal concentration were within TEC and PEC ranges for Cu and Cd at 100% and 33.3% of the sites respectively and lowers than the TEC for Cr, Zn and Pb at 100% sites, while concentration of Cd exceeded the PEC value at 66.7% sites of Gogabil Lake. The toxicity, derived from m- PEC-Q quotients, that results from the mixture of the six heavy metals at each sampling site of Gogabil Lake. The mean PEC quotients for all of the samples were well within the range of 0.058 – 1.21 with average value 0.590, indicating moderate toxicological risks for sediment dwelling organism, with a toxicity incidence of between 15 and 29% as also supported by **(**Swartz, 1999**;** Long et al., 2006**;** Varol *et al.*, 2012) [79, 56, 96].

Table 3: Comparison between sediment quality guidelines with heavy metals concentration of all sites in the Gogabil lake

Sediment quality guideline (SQGs)	Cu	Cr	Zn	Cd	Pb	Ecological risk
TEC	31.6	43.4	121	0.99	35.8	--------
PEC	149	111	459	4.98	128	--------
% of samples < TEC	0	100	100	0	100	60% are not effected
% of samples between TEC and PEC	100	0	0	33.3	0	26.7% are moderately effected
% of samples > PEC	0	0	0	66.7	0	13.3% are effected

Bioaccumulation factor (BAF)

Bioaccumulation factor of heavy metal ions from water and sediment to the fish tissues are represented in Table 4 and found that BAFs from water were significantly higher than sediment and exceed 1 in all three seasons. The highest organs/water ratio was found (771.64) in summer in liver while lower value (2.17) was obtained for Pb in muscles in winter season. In this study BAFs showed that concentration of measured metals ions in the fish tissues followed the rate liver > gills > muscles and their magnitude ranking was as follows Zn > Cu > Cr > Pb > Cd. The BACs from sediments for all metals were less than 1 within liver, gills and muscles throughout the season.

Table 4: Bioaccumulation actor of metals in different organs of *H. fossilis*

Organ	Season	BAF	Cu	Cr	Zn	Cd	Pb
Gills	Summer	water to gills	68.023	25.53	749.04	17.46	12.48
	Rainy	water to gills	62.8	24.5	740.6	17.25	10.16
	winter	water to gills	40.26	9.62	315.08	10.68	8.64
	Summer	sediment to gills	0.3643	0.2657	0.4819	0.2087	0.1293
	Rainy	sediment to gills	0.5593	0.3925	0.8324	0.2487	0.7452
	winter	sediment to gills	0.142	0.0644	0.2178	0.0901	0.4164
Liver	Summer	water to liver	66.96	25.66	771.64	16.56	13.84
	Rainy	water to liver	51.15	23.7	768.8	16.19	10.41
	winter	water to liver	42.67	7.74	317.69	9.52	8.18
	Summer	sediment to liver	0.3586	0.2617	0.4964	0.1979	0.1434

	Rainy	sediment to liver	0.5663	0.4125	0.8642	0.2441	0.7315
	winter	sediment to liver	0.1505	0.0063	0.2196	0.0803	0.1074
Muscle	Summer	water to muscles	26.39	8.33	288.08	6.66	5.06
	Rainy	water to muscles	25.32	4.25	247.9	3.09	3.72
	winter	water to muscles	16.64	2.77	150.76	2.17	2.33
	Summer	sediment to muscles	0.1413	0.0849	0.1853	0.796	0.0525
	Rainy	sediment to muscles	0.1874	0.1042	0.2786	0.0907	0.2574
	winter	sediment to muscles	0.0587	0.0185	0.1042	0.0183	0.0305

As sediment is a main source of metal accumulation in aquatic environment, in the present study BAFs indicate that bioaccumulation of heavy metals in fish tissues mainly come from water. Similar results were also reported by (Abdel *et al.*, 2011; Salam *et al.*, 2014 and Farsani *et al.*, 2019) [11, 87, 30]. Metabolic active tissue like liver and gills showed higher accumulation of heavy metal ions than muscles (Ali et al., 2019) [4].

Metal ions concentration in fish tissues

The highest mean concentration of Cu was found in gills (41.7 ± 0.862µg g^{-1}), followed by liver (41.05 ± 1.050.404µg g^{-1}) and in muscle (16.18 ± 0.246µg g^{-1}) and the level of Cu in the different tissue samples of *H. fossilis* were varied between 16.18±0.246 – 41.7 ± 0.862µg g^{-1}in summer, 10.13 ± 0.98 – 25.13 ± 1.97µg g^{-1} in rainy and 6.99 ± 0.021-17.92 ± 0.715µg g^{-1}in winter respectively. The level of Cu ion in summer followed the order of gills ≈ liver >> muscles and in rainy gills > liver >> muscles and in winter is of following order liver ≈ gills >> muscles. The mean concentration of Cu present in this study was exceeded the several folds than the literature (Ambedkar and Muniyan, 2012; Dhanakumar *et al.*, 2015; Rajeshkumar and Li, 2018) [10, 21, 75] and also exceeded the several folds than the permissible limit (3mg/kg) recommended by WHO, 2008. However, lower the maximum permissible limit (MPL) recommended by (FAO, FAO/WHO, WHO) [33, 107] for human consumption in liver and gills in rainy and winter seasons and in muscles throughout the season (Table 5). The high level of Cu in the liver than other fish tissues has been also observed by several researchers (Storelli *et al.* 2006; Frang *et al.* 2007; Yilmaz *et al.*, 2007; Uysal *et al.*, 2009; Karunanidhi *et al.*, 2017) [85, 30, 110, 93, 46]. Pyle *et al.*, (2005) [70] reported that in the liver, Cu concentration are usually regulated by homeostatic control below 50 µg g^{-1}dw and can exceed this threshold only if the control mechanisms are overloaded. The high levels of Cu in the different tissues of *H. fossilis* may be due to domestic waste, agricultural and industrial wastes and also due to increased boating activities, recurrent usage of antifouling paint, oil dropping from boat and commercial fishing in the study area. Cu is an essential element for the formation of haemoglobin and some enzymes in human (Sivaperumal *et al.*, 2007) [86]; however, high intake can result in damage to liver and kidneys (Alipour *et al.*, 2015) [13].

The highest concentration of Zn (57.22 µg g^{-1}) was observed at site -2 in the liver of *H. fossilis* with mean value 56.33 ± 0.826 µg g^{-1}, while the lowest concentration (9.2 µg g^{-1}) was detected it the same site in the muscle tissue with mean value 9.8 ± 0.458 µg g^{-1}. The distribution of Zn in fish tissues during summer, rainy and winter season is of the following order liver > gills > muscles. Similar results were reported in high concentration in the liver (Zn 26.83 µg g^{-1}) than in muscle (Zn 3.51 µg g^{-1}) (Onsanit *et al.*, 2010) [69]. Seasonal variation of Zn in the fish tissues were observed in the order of summer > rainy > winter. Similar trends were reported by some researches (Yehia and Sebaee, 2012; Maurya and Malik, 2016; Singh and Kumar, 2017) [111, 66, 83]. In these finding Zn level was within the range of permissible limit (10-75mgkg^{-1}) as recommended by (WHO, 2008) but higher than MPL, recommended by (FAO, FAO/WHO) [33] for human consumption in liver and gills in summer and rainy but lower in winter, while in muscle tissue its level was lower in all three seasons. However approximately same as 13.08 ± 0.30 - 78.15 ± 2.04mg kg^{-1}as reported by (Yehia and Sebaee, 2012) [111] and 26.67±1.37 - 58.44± 3.67mg kg^{-1}as reported by (Maurya and Malik, 2016) [66] in liver, gills and muscle of different fish species. The sources of Zn in the study area may be human activities such as industrial, domestic wastes water discharges and agricultural runoff of catchment area. Zn is an essential element as more than one hindered specific enzyme require for their catalytic function (Kayrak and Ozan, 2018) [47]. However at higher levels, Zn produced adverse effect in fish by structural damage, which effects the improvement, growth and survival of fish (Kori *et al.*, 2008) [48]. Zn is potential toxicant to fish (Vosylien *et al.*, 2006) [94] which causes ion regulation, disturbances, disruption of gills tissue and hypoxia (Murugan *et al.*,2008) [65]. In human beings large level of Zn can cause prominent health problem and extensive doze of Zn damage the pancreas and disturb the protein metabolism and cause arteriosclerosis (Afshan *et al.*, 2014) [2].

Among the all studied metal Cd was recorded as minimum amount in shingi fish tissues, it was also detected as lowest in water and sediment throughout the season. The highest mean concentration of Cd (1.24 ± 0.05µg g^{-1}) was found in gills followed by liver (1.176 ± 0.006µg g^{-1}) and muscles (0.473 ± 0.006µg g^{-1}).The distribution of Cd throughout the season was in the following order: gills > liver > muscles and seasonal variation was in sequence of summer > rainy > winter. (Yehia and Sebaee, 2012) [111] reported as same pattern while (Begum *et al.*, 2013; Ambedkar and Muniyan, 2012; Maurya and Malik, 2016) [16, 10, 66] observed high level of Cd in liver followed in gills and muscles in different species of fish. The level of Cd in liver and gills was higher than 0.5µg g^{-1}set by (FAO/WHO) [33] guidelines but lowers in muscles in all three seasons. The high level of Cd in the gills and in the liver tissue than 0.5µg g^{-1} (threshold value), is considered to be harmful to fish and predators (Walsh *et al.*, 1977; Begum *et al.*, 2013) [111, 16].

Table 5: Heavy metal concentration in organs of *H. fossilis* (µg g^{-1}) of Gogabil Lake with International guidelines and World's River

Organ	Season	Cu	Cr	Zn	Cd	Pb	Reference
Gills	Summer	41.7±0.862	1.94 ± 0.026	54.68 ± 0.495	1.24 ± 0.05	1.535 ± 0.021	Present study
	Rainy	25.13±1.97	1.25 ± 0.264	45.92 ± 0.525	1.08 ± 0.035	1.22 ± 0.196	
	winter	16.91±0.26	0.51 ± 0.01	20.48 ± 0.251	0.673 ± 0.02	1.52 ± 0.02	
Liver	Summer	41.05±1.05	1.95 ± 0.03	56.33 ± 0.826	1.176 ± 0.006	1.703± 0.025	

	Rainy	20.4 ± 1.49	1.21 ± 0.221	47.67 ± 0.531	1.06 ± 0.011	1.25 ± 0.225	
	winter	17.92±0.715	0.503 ±0.015	20.65 ± 0.214	0.60 ± 0.01	1.44 ± 0.032	
Muscles	Summer	16.18±0.246	0.633 ±0.035	21.03 ± 0.985	0.473 ± 0.006	0.623± 0.025	
	Rainy	10.13±0.98	0.217 ±0.012	15.37 ± 0.567	0.192 ± 0.009	0.447± 0.012	
	winter	6.99±0.021	0.147 ±0.011	9.8 ± 0.458	0.137 ± 0.011	0.41 ± 0.01	
FAO		30	-----	30	0.05	0.5	FAO, (1983) [32]
FAO /WHO		30	-----	40	0.5	0.5	FAO/WHO (1989) [33]
WHO		30	-----	100	1	2	WHO (1995) [107]
Gills		6.31 ± 2.46	3.52±3.72	17.81 ±1.93	3.57 ±0.26	5.83 ±0.41	Buriganga, Bangladesh, Begum *et al.*, (2013) [16]
Liver		45.61±1.29	6.30±0.91	60.81 ± 0.14	3.92 ± 0.40	18.16± 0.52	
Muscles		8.05 ± 0.00	1.56±0.11	26.67 ± 1.37	0.36 ±0.01	1.99 ± 0.03	
Gills		32.42±0.98	19.86±1.85	54.28 ±1.68	33.83 ± 2.85	17.24 ± 0.26	Kali Rivr, India, Maurya and Malik, (2016) [66]
Liver		31.18±0.95	18.00±3.56	58.44 ± 3.67	34.44 ± 0.79	17.33 ± 2.38	
Muscles		30.66±0.92	2039±0.65	52.1 ± 1.83	30.39 ± 0.21	15.28 ± 0.99	
Gills		0.60±0.029	0.72 ±0.017	0.34 ±0.011	0.78 ± 0.036	0.24 ±0.018	Gadilam River, India Ambedkar & Muniyn, (2012) [10]
Liver		0.76±0.029	1.10 ±0.031	0.48 ± 0.018	0.90 ± 0.038	0.62 ± 0.029	
Muscles		0.40±0.115	0.56 ±0.021	0.20 ±0.009	0.64 ± 0.022	0.34 ±0.011	
Gills		-----	2.25	-----	4.8	7.3	Madivala lakes, India Begum *et al.*, (2009) [114]
Liver		-----	2.6	-----	6.23	7.3	
Muscles		-----	1.54	------	1.1	2.05	

From our findings, it was vividly observed that Cd in the muscle of shingi fish from the Gogabil Lake was below the above-discussed standard values, but long period of accumulation of Cd in fish may pose health hazards. In the study points Cd enter into the fresh water by disposal of industrial, municipal and household waste and also agricultural runoff. Cd is the non-essential and most toxic heavy metal which is widely distributed in aquatic environment and earth's crust. The nutritive need of different tissues of fishes depends on their biochemical configuration of mineral contents, amino acids, protein and vitamins, etc. (Afshan *et al.*, 2014) [2].

The chromium (Cr) concentration among the selected tissues of *H. fossilis* ranged from 0.147 ± 0.011 -1.95 ± 0.03µg g^{-1}. European Union Commission suggested the daily tolerable chromium concentration to be 1µg/g, while the (FEPA, 2003) [34] and (WHO, 1995) [107] suggested 0.15µg g^{-}1. The highest level of Cr was recorded in liver followed by gills and muscles. The same distribution pattern of Cr (liver > gills > muscles) in the *H. fossilis* was reported by (Begum *et al.*, 2013) [16] and (Ambedkar and Muniyan, 2012) [10] but (Mauyrya and Malik, 2016) [66] observed opposite pattern (muscles > gills > liver) as shown in Table 5. High level of Cr at sampling point may be due to agricultural runoff, paints used in boats, and leaching from rocks in the study area (Mauyrya and Malik, 2016; Varsha *et al.*,2017) [66, 98].

The Pb concentration ranged from 0.41± 0.01 – 1.703 ± 0.025µg g^{-1} among the singhi fish tissues from the study area. Highest Pb concentration was detected 1µg $g^{-1}$with mean value 1.703 ± 0.025µg g^{-1}in liver followed by gills (1.55µg g^{-1}) with mean 1.535 ± 0.021µg $g^{-1}$and in muscle (0.65 µg g^{-1}) with mean value 0.623 ± 0.025µg g^{-1}. The distribution pattern of Pb in different tissues of *H. fossilis* was in the order of gills ≈ liver >> muscles and seasonal variation was in the sequence of summer > winter > rainy. In the present study level of Pb was higher than 0.5µg $g^{-1}$and lower than 2.0µg g^{-1}set by (FAO, FAO/ WHO; FEPA, WHO) [33, 110, 111] respectively. (Begum *et al.*, 2013; Mauryn and Malik, 2016) [16, 66] reported higher Pb concentration in liver followed by gills and muscles, while (Ambedkar and Muniyan, 2012) [10] observed lower Pb concentration (compare to the present study) but in opposite distribution pattern (liver > muscles > gills).

Seasonal variation of metal in fish may be due to varying seasonal growth rate, reproductive cycle, water salinity and temperature may be the cause of high metal accumulation of metal mainly during summer compare to rainy and winter season. In addition during summer season water temperatures in the lake have a positive major effect on biological activity and breathing rates of fish, that reduces haemoglobin/oxygen affinity, so that metabolic rate and feeding activity of fish, resulting in an increase in heavy metal take up and accumulation in summer season. Similar pattern was reported by (Singh and Kumar, 2017; Rajeshkumar and Li, 2018; Rajeshkumar *et al.*, 2018) [83,75, 74].

According to our findings, heavy metals were more accumulated in liver and gills than muscles, as active organs metabolite organs accumulate higher amount of metal (Terkin-Ozan, 2007; Terkin-Ozan and Kir, 2007) [86, 87] than muscles that have a weak accumulating potential (Uysal *et al.*, 2009)[89]. The difference in the level of accumulation of metal in different organs of a fish can be attributed to the differences in the physiological role of each organ (Rajeshkumar and Li, 2018; Rajeshkumar *et al.*, 2018) [73, 72], regular ability, behaviour and feeding habits. This finding is an agreement with those of other studies regarding fish tissue (Karaded and Unlu, 2007; Karaded- Akin, 2009; Ebrahimpour *et al.*, 2011; Liu *et al.*, 2012; Rajeskumar and Li, 2018) [51, 50, 26, 109, 73]. Liver is vital organ in vertebrates and has a major role in metabolism (Liu *et al.*, 2012) [109]. The high accumulation of metals in liver is due to the greater tendency of the element to react with the oxygen carboxylate, amino group, nitrogen, sulphur of mercapto group in the matllothionein protein, whose level is highest in liver as supported by (Ali-Yousuf *et al.*, 2000) [6]. As fish gills are in direct contact with water and sediment and an important site for the entry of heavy metals (Vohodhani and Narayanan, 2008; Rajeshkumar and Li, 2018) [95,73]. In the present work higher metal concentration in the gills is due to element complextion with the mucus, which is difficult to be removed completely from the tissue before analysis (Khalil and Faragallah, 2008) [52]. Thus level of metals in the gill reflects the level of the metals in the water system where the fish lives, whereas the level in liver and kidney storage of metals (Vohodhani and Narayanan, 2009) [96].Thus, the gills in fish are more often recommended as environment indicator organs of water pollution than any other fish organ. Level of metals were lower in muscles compared to liver and gills because at being inactive tissue in accumulating heavy metals (Storelli *et al.*, 2006; Ploetz *et al.*, 2007; Agah *et al.*, 2009) [85, 71, 3].

Conclusion

It may be concluded that levels of Cu and Zn in water were lower whereas concentrations of Cr, Cd and Pb were higher than permissible limit set by (WHO, Joint FAO/WHO) [102]. PLI and C_f exposed that sediment pollution was high in summer and winter compared with wet season. In addition, I-geo values indicated high degree of sediment pollution was found by Cd and SQGs method confirmed that 26.7% sediment samples were moderately and 13.3% samples were moderate to high pollute. Significant differences were identified among liver, gills and muscle of *H. fossilis.* The metal accumulation was found to be high in liver and gills and higher in summer and winter season. Though liver and gills are rarely consumed, they may represent good bio-monitor of metals present in the water ecosystem. The seasonal variation of heavy metals in the fish tissues might be due to anthropogenic wastes especially municipal and household discharge and agricultural runoff are released in to lake. The concentrations of Cr, Zn, Cd and Pb in liver and gills of Singhi fish were found to be above than maximum permissible limit (MPL) set by (FAO, FAO/WHO) [33] and (WHO, 1995) [107] for human consumption. This study suggested that water, sediment and fish collected from lake were polluted by heavy metals and is not safe for animals and human. Therefore precaution need to be taken, otherwise they pose threat for animals and human.

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