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Original Article

Seasonal heavy metal monitoring of water, sediment and common carp (*Cyprinus carpio*) in Aras Dam Lake of Iran

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Abstract: Heavy metal in aquatic ecosystems are monitored by measuring their concentrations in water, sediments and biota. In the presented study, cadmium, copper, lead, mercury, nickel, and zinc concentrations determined in water, sediment and tissues (liver and muscle) of *Cyprinus carpio* in Aras dam Lake of Iran, during spring, summer, autumn, and winter from 2016 to 2017. The concentration of Pb, Cd, Zn, Cu and Ni of samples were measured using inductively coupled plasma-optical emission spectroscopy. The concentration of Hg was analyzed using Atomic Absorption equipped with MHS 15 CVAAS. The trend in the metal mean concentrations of liver and muscle was Zn>Cu>Pb≈Ni>Cd>Hg, and water and sediment were Cu>Zn>Pb>Cd≈Ni>Hg. Heavy metals concentration was more in sediment than fish tissues and water. This results revealed that heavy metals accumulation of muscle was the highest in summer showing the most contaminated season. Whereas, winter had the lowest contamination in water (Cd،Hg, Cu, Pb, Ni, Zn), sediments (Hg, Cu, Zn, Pb, Ni), and the liver and muscle of fish (Cd, Zn, Ni, Cu). The amount of heavy metals was less or slightly higher than global standards (EPA, WHO).

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Introduction

Nowadays, global concern on metals contamination of aquatic ecosystems is growing (Yi et al., 2017; Jiang et al., 2018). Technically, heavy metals concentration in various natural environments and ecosystems depends on local geology (Fiorino et al., 2018; Plhalova et al., 2018). However, anthropogenic release has been considered as the main source of heavy metals accumulation in water, sediment and aquatic organisms (Savorelli et al., 2017; Capillo et al., 2018; Aliko et al., 2018). According to the guidelines of water quality of the European Water Framework Directive (Directive 2008/105/EC 2008), the list of metals in priority include Hg, Pb, Cd and Ni, while Cu, Cr, As and Zn are on the list of other specific chemical dangerous substances. Heavy metals are distributed in the forms of particulate and dissolved in water bodies and accumulated in the bottom (Vukosav et al., 2014). Therefore, potential source of heavy metals could be sediment which is releasing metals into the surrounding water through natural and anthropogenic processes (Aliko et al., 2018). In addition, toxic metals can transferred via food chain to higher levels by fish grazing from benthos fauna (Wang et al., 2010; Zhang et al., 2015).

Aras dam-reservoir is important for many activities such as fisheries, agriculture, industry, domestic and drinking water supply (Mohebbi et al., 2012). Sewage and industrial effluents discharges daily into Aras River through multifarious man-made activities. Nasehi et al. (2013) stated that enhanced erosion of soil, dissolution of bed load, runoffs and excessive use of pesticides resulted in elevated metals' concentration in the Aras River, which led to heavy metals excessive concentration compare to the permissible values of WHO published.

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Due to the heavy metals persistence in the environment, heavy metals monitoring in the environment would be a key role of ecosystem management (Vukosav et al., 2014). No published papers dealing with the detailed study of trace metals concentrations in the water body, sediment and indicator fish species of the Aras Dam is available. Therefore, this study was conducted on (1) estimation seasonal concentration of trace metals (Cd, Hg, Cu, Pb, Ni and Zn) in water, sediment and Common carp liver and muscle in the Aras Dam and (2) evaluating the transfer factor and relationship between water/ sediment and fish body. The results could help us to understand status and seasonal heavy metals variation in Aras Dam and toxic metals accumulation rate in ecosystems with shallow lake.

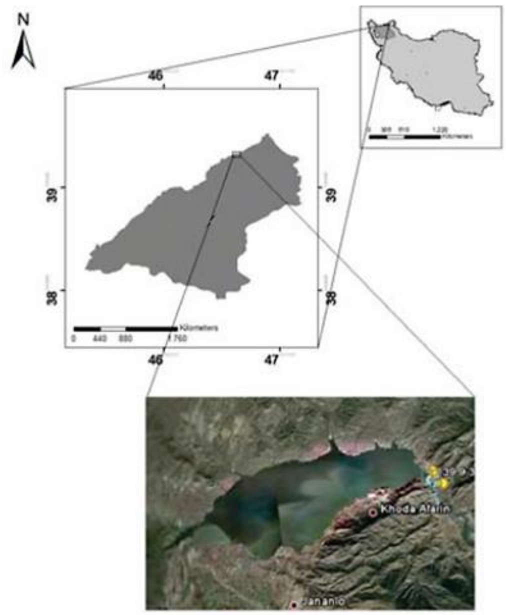

Figure 1. Sampling area of Aras Dam Lake (39°33"82"N, 46°52'99"E).

Materials and Methods

Sampling of fish, water and sediment: For each season, a total of 30 samples of fish, water and sediment were collected from regional stations in Aras Dam Lake (Fig. 1). Before sampling from water and sediments, all sampling bottles were washed with 1:1 nitric acid and distilled water and dried with 60° C for 4 hours. Three to five-liter water samples were taken at the depth of 20 cm using a plastic bottle. Sediment samples were collected using a Van Veen Grab device with 0.0256 m^2 cross section and decanted to plastic bags. Fish samples were collected using gillnet and after weight and length measurement, samples were placed into zipped plastic. All samples put in an iceberg and transferred to the laboratory for further analysis.

Sample preparation: For tissue preparing, at first stainless steel knife disinfected and cleaned with acetone and distilled water, then liver and muscle tissues were dissected. Tissue and sediment samples were oven dried for 24 hours at 105°C. After cleaning teflon vessel and soaking in %5 HNO_3 for 1 day, it was rinsed with ultrapure water and dried. In order to analyze metal concentration in tissue and sediments, 0.5 g of samples were weighted and decanted to 50 ml close teflon vessels. Tissue and sediment samples were treated with 5 ml 60% $HClO_3$ and 8 ml 68% HNO_3 and 4 ml 30% H_2O_2. The solution was shaken well to complete tissues digestion. For determining the concentration of Pb, Cd, Zn, Cu and Ni, the samples put within an inductively coupled plasma optical emission spectrometer (ICP-OES, model 735-ES, Varian Company, Italy). The concentration of Hg was analyzed with Perkin Elmer 4100ZL Atomic Absorption equipped with MHS 15 CVAAS.

For determination of the heavy metals in water samples, 1 L of 60% HNO_3 were added to 250 ml of water samples. Liquid-liquid extraction technique was performed according to Mentasti et al. (1989). Sample evaporation done on a low temperature reaching up to 20 ml. Metal concentration in the final solution was determined using Atomic Absorption Spectrometry (Varian 710-Es).

Transfer factor (TF): The transfer factor of samples, including muscle, liver, sediment and water were calculated according to Kalfakakour and Akrida-Demertzi (2000) and Rashed (2001) as follows:

$$TF = M\ tissue / M\ sediment\ or\ water$$

Where M tissue is the metal concentration in liver and muscle and M sediment or water the metal concentration in sediment and water.

Geochemical index (Igeo) and Pollution load index (PLI): The heavy metal contamination degrees in

Table 1. Seasonal transfer factor of metals (mg/kg) in different tissues of *Cyprinus carpio.*

	`	Cd	Hg	Cu	Pb	Ni	Zn
Muscle	TF from water Sp	16.83	5.04	30.38	1.90	18.52	278.17
	TF from water Su	8.18	5.30	41.89	3.92	13.27	323.58
	TF from water Au	5.85	6.22	26.74	7.08	5.26	227.56
	TF from water Wi	7.67	11.83	28.61	9.23	6.03	273.50
	TF from sediment Sp	0.11	0.07	0.09	0.03	0.07	0.20
	TF from sediment Su	0.06	0.11	0.15	0.05	0.08	0.22
	TF from sediment Au	0.07	0.10	0.10	0.09	0.06	0.15
	TF from sediment Wi	0.04	0.09	0.09	0.13	0.04	0.17
Liver	TF from water Sp	37.58	19.38	93.31	12.28	56.96	801.67
	TF from water Su	20.24	20.25	103.69	10.00	39.40	877.00
	TF from water Au	15.77	25.50	70.50	19.35	20.15	682.05
	TF from water Wi	28.33	31.33	82.04	33.85	20.29	623.10
	TF from sediment Sp	0.25	0.27	0.29	0.19	0.21	0.58
	TF from sediment Su	0.16	0.44	0.37	0.14	0.25	0.59
	TF from sediment Au	0.18	0.43	0.27	0.25	0.21	0.44
	TF from sediment Wi	0.14	0.25	0.25	0.48	0.14	0.39

*Sp= spring, Su= summer, Au= autumn, Wi= winter, Cd: cadmium, Hg: mercury, Cu; Copper, Pb: Lead, Ni; Nickel, Zn; Zinc

sediment were determined using measuring Geochemical index (Igeo) (Muller, 1969; Saleem et al., 2015) and Pollution load index (PLI). Igeo is calculated as follows:

$$Igeo = \log [Cn / 1.5\ Bn]$$

Where Cn is the measured concentration of heavy metal n in the sediment and Bn is the crustal shale background content of the metal. The PLIs defined as the nth root of the multiplications of the metal concentration (Rajeshkumar et al., 2018):

$$PLI = (Cf1 \times Cf2 \times \ldots \times Cfn)1/n$$

Where Cf1 is concentration of the first metal, Cf2 is concentration of the second metal and n is the total number of studied metal in the survey. Classification of Igoe contamination assessment model is as defined as follows: Igoe<0 - uncontaminated; 0≤Igeo ≤1 - moderately contaminated; 1≤Igeo≤3 - considerable contaminated; 3≤Igeo≤5 high contaminated; 5≤Igeo extremely contaminated (Muller, 1969). Zero value of PLI shows excellence condition while a value of one or higher indicates baseline level of pollutants and progressive deterioration of the site and estuarine quality, respectively (Tomlinson et al., 1980).

Results

Transfer factor: The results of transfer factor (TF) in water and sediments are given in Table 1. For all the studied elements within muscle and liver, TFs from water were higher than sediments and exceeds 1 in all seasons. This confirmed intensive accumulation of studied metals from surrounding water within the evaluated species. The highest organ/water ratio was for Zn in fish liver in summer while the lowest was for Pb in muscle in spring. The TF from sediment for all measured heavy metals, fixed less than 1 within muscle and liver tissues throughout the seasons. It confirmed limited accumulation from sediment within fish liver and muscle tissues.

Water metals concentration: Concentration of all heavy metal in surface water samples were higher in both summer and autumn than winter and spring (Table 2). The mean concentration of metals in water followed the order of: Cu> Pb> Zn> Hg> Ni> Cd. The mean concentration of Cu was 0.27, 0.37, 0.54 and 0.22 (mg/L) in spring, summer, autumn and winter, respectively, which were less than the WHO standard level for drinking water (Table 2). However, the concentration of Pb was 0.1, 0.17, 0.13 and 0.04 in spring, summer, autumn and winter, respectively, which were higher than the WHO for drinking water (Table 2).

Metal concentration in sediment: Seasonal concentrations of heavy metal in sediments are presented in Table 3. The average concentration of

Table 2. Comparison of heavy metals in water (mg/L) with different international guideline and other studies in the world. Different letters (a–d) in the same columns indicate significant differences (P<0.05).

Location		Cd	Hg	Cu	Pb	Ni	Zn
This study	Sp	0.02±0.01[a]	0.04±0.01[a]	0.27±0.11[ab]	0.10±0.01[a]	0.02±0.01[a]	0.04±0.02[a]
	Su	0.06±0.02[b]	0.07±0.03[a]	0.37±0.25[abc]	0.17±0.03[b]	0.05±0.02[b]	0.06±0.04[ab]
	Au	0.07±0.01[c]	0.05±0.02[a]	0.54±0.10[c]	0.13±0.02[b]	0.08±0.0[c]	0.07±0.01[b]
	Wi	0.02±0.02[a]	0.02±0.01[b]	0.22±0.12[b]	0.04±0.03[c]	0.02±0.01[a]	0.03±0.02[a]
WHO		0.003	0.006	2	0.01	0.07	3
EPA*		0.001	0.002	1.3	0.05	0.1	0.5
Ataturk Dam Lake[1]		N.D.	N.D.	0.02	N.D.	0.01	0.06
Saline lake in karstic area[2]	Sp	0.1×10^{-4}	0.2×10^{-4}	0.3×10^{-3}	0.4×10^{-4}		0.4×10^{-3}
	Wi	0.2×10^{-4}	0.1×10^{-5}	0.3×10^{-3}	0.5×10^{-4}		0.3×10^{-3}
Enne and Porsuk Dam Lakes[3]		0.0297	BDL	0.008	0.02	0.004	BDL
Buriganga Reservior[4]		0.009		0.163	0.065	0.008	
Taihu Lake[5]	Sp	0.1×10^{-3}		0.1×10^{-3}	0.2×10^{-2}		
	Su	0.2×10^{-3}		0.1×10^{-2}	0.6×10^{-2}		
	Au	0.1×10^{-3}		0.4×10^{-3}	0.2×10^{-3}		
	Wi	0.7×10^{-3}		0.3×10^{-3}	0.5×10^{-3}		

[1]Karadede and Ünlü (2000), [2]Cuculic et al. (2009), [3]Cicek et al. (2009), [4]Ahmad et al. (2010), [5]Rajeshkumar et al. (2018), *Guidelines (2002) for water, BDL= Bellow Detection Limit. N.D.= Not Detected

Table 3. Comparison of heavy metals in sediment (mg/Kg) with different international guideline and other studies in the world. Different letters (a–d) in the same columns indicate significant differences (P<0.05).

Location		Cd	Hg	Cu	Pb	Ni	Zn
This study	Sp	2.98±0.06[a]	2.92±0.25[a]	89.5±5.77[a]	6.38±0.98[a]	5.23±0.45[a]	55.67±0.76[a]
	Su	7.20±0.82[b]	3.10±0.90[a]	103.6±7.09[b]	12.2±1.12[b]	7.97±1[b]	94.00±4.58[b]
	Au	5.55±1.13[c]	3.18±0.33[a]	142±8.19[c]	10.1±1.68[c]	7.55±1.63[b]	111±3.12[c]
	Wi	3.93±1.14[a]	2.53±0.55[a]	73.33±7.64[d]	3.03±0.25[d]	3.23±1.17[c]	53.33±4.73[a]
Etueffont (France)[1]	Su	1.67		177.21	37.29	45.61	371.17
	Au	0.51		63.13	42.12	38.73	132.12
Saline lake in karstic area[2]		14.8	1.76	300	62.8		377
Plitvice Lakes[3]		10.9	1.22	115	11.7		258
EPA Guidelines (2001) for sediments[4]							
Not polluted		/		<25	<40	<20	<90
Moderately polluted		/		25-50	40-60	20-50	90-200
Heavily polluted		>60		>50	>60	>50	>200
Sediment quality guideline-quotient (SQG-Q)[5]		4.21	0.7	108	112		271

[1]Ben Salem (2014), [2]Cuculic et al. (2009), [3]Vukosav et al (2014), [4]nvironmental Protection agency, [5]MacDonald et al. (1996).

studied metals in sediment is followed the order of: Cu> Zn>Pb>Ni>Cd>Hg. In the sediment, metals have higher concentrations in summer and autumn than winter and spring. According to EPA (2001) in case of Cu, sediment was heavily polluted and in case of Zn, it was moderately polluted in summer and autumn. For other heavy metals, sediment were not polluted in this study (Table 3).

Heavy metal content in fish liver and muscle tissue: Seasonal variation of metal (Cd, Hg, Cu, Pb, Ni and Zn) accumulation in fish liver and muscle is shown in Table 4. The decreased tendency of the mean heavy metal concentrations in fish tissues according to the elements was as Zn>Cu>Pb>Ni>Cd. The values in summer and autumn were significantly more than winter and spring. Accumulation of metals in the liver was obviously more than muscle. Among the six studied metals, Zn and Cu concentration were high whereas Hg and Ni were low in fish organs.

Pollution load index (PLI) and Geoaccumulation index (Igeo): Figures 2 and 3 show the indexes of PLI and Igeo of heavy metals in sediment. Based on the

Table 4. Comparison of heavy metals in liver and muscle of common carp (µg/g) with different international guideline and other studies in the world. Different letters (a–d) in the same columns indicate significant differences (P<0.05).

Location			Cd	Hg	Cu	Pb	Ni	Zn
This study	Muscle	Sp	0.33±0.12^{a}	0.20±0.01^{a}	8.36±0.40^{a}	0.18±0.03^{a}	0.36±0.04^{a}	11.12±0.71^{a}
		Su	0.46±0.09^{a}	0.35±0.04^{b}	15.36±0.52^{b}	0.65±0.08^{b}	0.66±0.10^{b}	20.49±0.86^{b}
		Au	0.38±0.06^{a}	0.33±0.06^{b}	14.48±1.29^{b}	0.93±0.08^{c}	0.42±0.08^{a}	16.30±0.36^{c}
		Wi	0.15±0.03^{b}	0.24±0.04^{a}	6.39±0.78^{c}	0.40±0.08^{d}	0.14±0.05^{c}	9.11±0.46^{d}
	Liver	Sp	0.75±0.12^{a}	0.78±0.04^{a}	25.66±0.77^{a}	1.19±0.16^{a}	1.09±0.18^{a}	32.06±0.08^{a}
		Su	1.15±0.14^{b}	1.35±0.17^{b}	38.02±0.92^{b}	1.67±0.24^{a}	1.97±0.12^{b}	55.54±1.44^{b}
		Au	1.03±0.27^{b}	1.36±0.26^{b}	38.19±1.30^{b}	2.55±0.27^{b}	1.60±0.21^{b}	48.88±1.22^{c}
		Wi	0.57±0.06^{c}	0.63±0.10^{c}	18.32±1.21^{c}	1.47±0.25^{a}	0.46±0.12^{c}	20.77±0.61^{d}
FAO (1983)			0.05		30	0.5		30
FAO/WHO limit			0.5		30	0.5		40
WHO 1989			1		30	2		100
Nasser Lake *Tilapia Nilotica*[1]	Muscle				0.260		0.62	0.63
	Liver				7.5		0.13	2.28
Lake Kasumigaura *Cyprinus Caripio*[2]	Muscle		0.009		0.24	0.03	0.04	5.43
	Liver		0.01		0.74	0.08	0.05	201
Karoon River *Barbus* sp[3]	Muscle		0.84	0.73		1.75	1.15	
	Liver		1.07	0.79		2.10	1.30	

[1]Rashed (2001), [2]Alam et al. (1980), [3]Mohammadi et al. (2011).

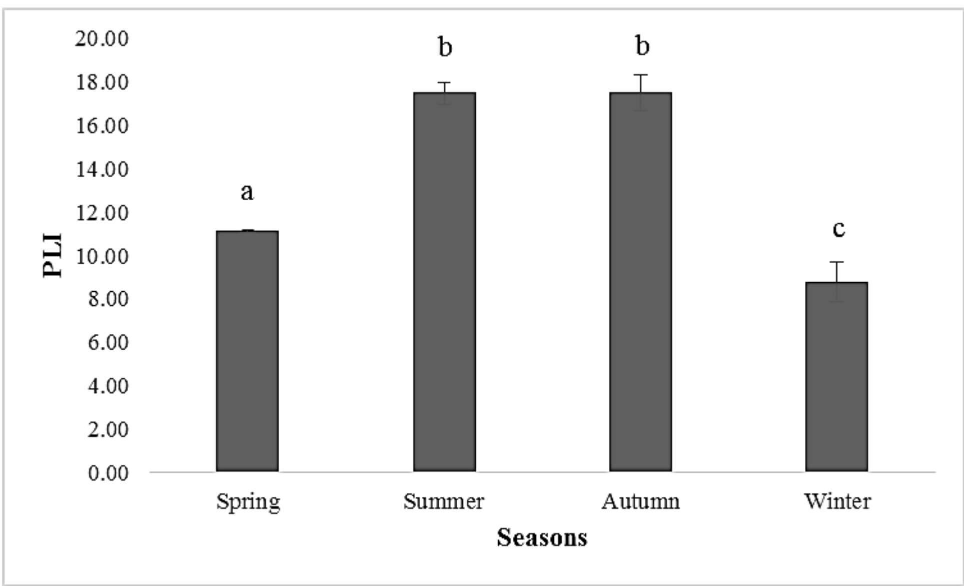

Figure 2. Pollution load index (PLI) value of heavy metals in sediment in Aras dam Lake, Iran. Different letters (a–c) in the same bars indicate significant differences (P<0.05).

results, the value of PLI in autumn and summer significant higer (P<0.05). In the case of Igeo index, Hg and Cu showed the highest (high pollution) and lowest (no pollution) Igeo value in sediment, respectively.

Discussions

Aquatic environment easily destroyed as a consequences of human activities impairing water quality (Zhang et al., 2015; Rajeshkumar et al., 2018). The results revealed seasonal variation of heavy metals pollution within water, sediment and liver as well as muscle tissue of fish in Aras Lake, Iran, throughout a year. Heavy metal concentration results exhibited more density in summer and autumn than winter and spring.

The transfer factor (TF) was measured for all of the heavy elements studied within fish liver and muscle

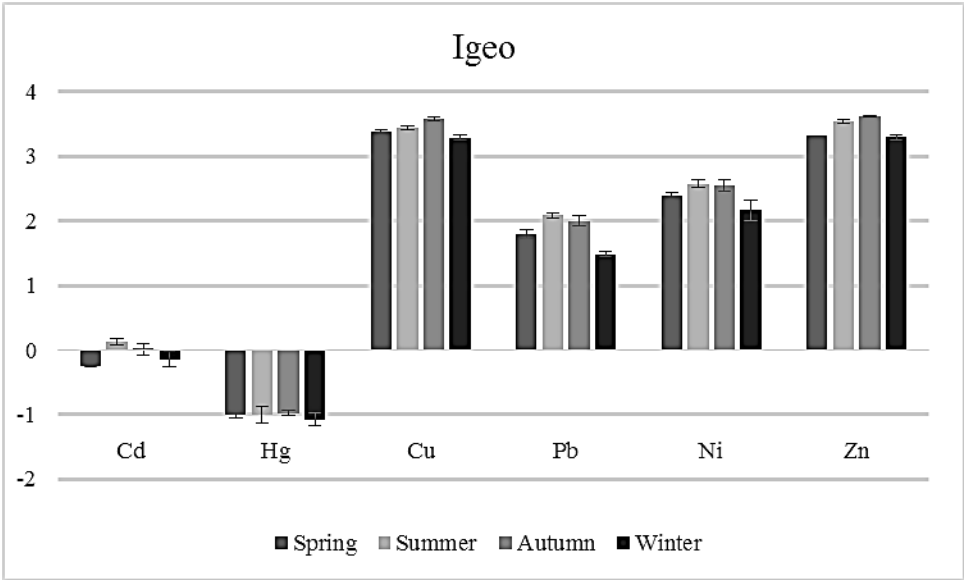

Figure 3. Geoaccumulation index of selected metals in the sediment in in Aras dam Lake, Iran.

tissue for water and sediment. Water TFs of heavy metals, which were higher than 1 in liver and muscle of all fish, were greater than sediments. The results show the fact that although the main source of heavy metals accumulation in aquatic environment is sediment, bioaccumulation of elements in fish mainly come from water. These results obtained from studies by Ali and Fishar (2005), Abdel-Baki et al. (2011) and Salem et al. (2014). However, it must be noted that our findings are not essentially related to high transfer from water.

Heavy metals concentration in fish tissues are depends on other factors such as feeding behavior and size (Vinodhini and Narayanan, 2008) which were not considered in this study. According to the results, maximum TF found in summer. Increased summer water temperature has favorable effect on biological activity of freshwater fish which led to higher metabolic activities of fish in this season, increased up taking and accumulation of heavy metal in tissues and finally increasing in TF (Salem et al., 2014). Based on Table 1, overall heavy metal is more accumulated in liver than muscle. It is well-known that liver is the first organ in detoxification and excretion of heavy metals which is the target organ of metal absorption and considered as water pollution indicator in aquatic environments (Kim and Kang, 2004; Farombi et al., 2007). Nevertheless, heavy metals concentration in muscle is substantial because heavy metals accumulation in fish muscles effect on human through transferring high levels of accumulated heavy metals by fish consumption. Whereas, the muscle is not potentially able to accumulate heavy metals, so the amount of heavy metals is less in this organ (Visnjic-Jeftic et al., 2010).

In comparison, the concentration of all studied metals was higher than other published literatures for water contamination (Tables 2 and 3). However, concentration of Cd, Cu, Pb and Zn in the Aras Dam Lake was significantly lower than those reported for Saline Lake in karstic area (Cuculic et al., 2009) and Plitvice Lakes (Vukosav et al., 2014).

Seasonal fluctuation of heavy metals in aquatic ecosystem might be affected through some water physical traits such as temperature and salinity as well as chemical traits of water including pH and dissolved oxygen levels (Wong et al., 2001). In this survey, the highest heavy metals concentration in water was in summer. This could be due to more evaporation and less rainfall which resulted in higher metal concentrations as observed in the previous literatures (Rajeshkumar et al., 2018). Moreover, lower concentration of heavy metals in winter and spring than autumn and summer might be due to increasing in seasonal rainfall which could lead to alleviation of heavy metals throughout the rainy seasons as reported by Duman and Kar (2012). These trends were also observed for sediments.

The results on Cu, showed the highest concentration between heavy metals in both water and sediment in summer. Summer is the main seasons for agricultural activities and many Cu based pesticides and fertilizers uses agricultural lands in Aras reservoirs catchment area (Nasehi et al., 2013). In addition, many industrial shutdowns through winter due to low temperatures and harsh weather, will cause relatively lower metal concentrations in. On the other hand, lower temperatures will reduce the affinity of metals to be dissolved in liquid phase (Drever, 1997).

The pollution load index (PLI) values was significantly higher in autumn and summer. The PLI values ranged from 17 in autumn and summer to 11 in spring and 7 in winter which indicates all seasons, sediment were contaminated. Cu and Zn were the more contributed factors in increasing PLI in the sediments. Researchers reported increasing in PLI values in summer due to the effects of urban activities (Suresh et al., 2012; Ali et al., 2016). The Igeo values showed the decreasing order as Cu> Zn> Ni> Pb> Cd> Hg. Furthermore, small standard deviations indicated that variation between seasons is low and the heavy metal contamination in surface sediments is constant during a year.

Due to the muscle tissue of fish is the most important part of human cosnsumption, therefore, assessment heavy metals in this organ is more vital than other organs. According to Table 4, concentration of all studied heavy metals in fish muscle was lower than guidelines. However, in comparison with *C. carpio* OF Lake Kasumigaua (Alam et al., 1980), heavy metals concentration of this study was much higher. In addition, compare to *Barbus* sp. in Karoon River (Mohammadi et al., 2011), Cd, Hg, Pb and Ni was lower. The results indicated that the concentration of all heavy metals elevated more in summer and autumn than winter and spring.

Furthermore, increasing heavy metals in Aras Lake in summer and autumn could be contributed as high influx of metals as a result of pollution from the surrounding industries thereby increased bioavailability to the fish. Based on the results, Aras Lake sediments are highly polluted. Common carp is a sediment dependent species feed on benthic macro invertebrates. Fish are considered as second trophic level in aquatic ecosystems with capacity of both essential and non-essential heavy metals accumulation (Phillips, 1977). In addition, it is better to know the concentration of heavy metals in fish body in different seasons for fishing ban in high polluted times during a year.

Based on our findings, summer and autumn are the most contaminated seasons. Winter had the lowest contamination in water (Cd.Hg, Cu, Pb, Ni, Zn), sediments (Hg, Cu, Zn, Pb, Ni) and the liver and muscle of fish (Cd, Zn, Ni, Cu), and the amount of heavy metals in them was less or slightly higher than global standards (EPA, WHO).

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