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Concentration of Some Heavy Metals in Water, Sediment and Fish (Common Carp) Organs in Ranya Lakelet in Sulaimani Governorate

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ABSTRACT

This study was conducted in Ranya Lakelet/Dukan, the study started at the end December of 2014 to early April of 2015, heavy metal concentration in water, sediment and fish organs (skin, gills, intestine, liver and muscle.). The experiment was conducted in the laboratory of Chemistry Department, Faculty of Science at University of Raparin, Sulaimani, Kurdistan Region - Iraq. The trials for the sampling species lasted for three months (January, February and March). Sixty eight common carp taken as a sample, then divided into four length groups (20-30(19 fishes); 31-40(29 fishes); 41-50(11 fishes); 51> cm (9 fishes)). Heavy metals contents analysed in water, sediment and fish organs, including Cadmium (Cd), Zinc (Zn), Nickel (Ni), Chromium (Cr), Copper (Cu) and Lead (Pb). The study results showed that the mean concentrations heavy metals (Cd, Zn, Ni, Cr, Cu and Pb) were below detection limit (1µg/L) except Zn, which it was 8.18 µg/L in filtered water. Whereas, the mean concentrations of heavy metals (Cd, Zn, Ni, Cr, Cu and Pb) in sediments were 6.90 µg/L, 146.29 µg/L, 151.46 µg/L, 161.74 µg/L, 38.86 µg/L and 92.49 µg/L, respectively. The higher concentrations of heavy metal (Cd, Zn, Ni, Cr, Cu and Pb) in different organs were 0.69 µg/L, 6352.84 µg/L, 16.52 µg/L, 9.25 µg/L, 42.18 µg/L, and 11.44 µg/L, respectively. While the lower concentration of heavy metal (Cd, Zn, Ni, Cr, Cu and Pb) in different organs were BDL, 208.53 µg/L, 2.93 µg/L, 2.93 µg/L, 20.13 µg/L, BDL, respectively. This study showed that storing of heavy metals occurs by this order; Zn>Cu>Ni>Cr>Cd>Pb. Although the trend of heavy metals concentrations according to length groups generally represented as follows; group4> group3> group2> group1.

تركيز بعض العناصر الثقيلة في الماء والرواسب واعضاء الاسماك (الكارب الشائع) في دربند رانية في محافظة السليمانية

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الخلاصة

الكلمات المفتاحية:
العناصر الثقيلة، الماء ، الرواسب ، اعضاء الاسماك
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أجريت الدراسة الحالية في بحيرة رانية/ التابعة لبحيرة دوكان، للفترة من نهاية شهر كانون الاول من 2014 الى بداية شهر مايس من 2015. شملت الدراسة قياس بعض العناصر الثقيلة (الكادميوم، الزنك، النيكل، الكروم، النحاس، الرصاص) في الماء والرواسب وبعض أعضاء (الجلد، الكبد، الغلاصم، الامعاء، عضلات) جسم الاسماك. أجريت الدراسة في مختبر الاسماك في قسم علوم الكيميا / جامعة رابةرين/ السليمانية/إقليم كردستان، العراق. أجريت هذه الدراسة لبيان تأثيرها على الحالة الصحية العامة لأسماك الكارب الشائع (*Cyprinus carpio*) في البحيرة. صيدت 68 سمكة كارب عادي والتي قسمت الى أربع مجاميع طولية (20–30 (19 سمكة): 31–40(29 سمكة) : 41–50(11 سمكة) : <51(9 سمكة)) سم، والتي أستخدمت لدراسة تركيز محتواها من بعض العناصر الثقيلة (الكادميوم، الزنك، النيكل، الكروم، النحاس، الرصاص)في كل من الماء والرواسب وفي كل من جلد وغلاصم و الامعاء وكبد وعضلات أسماك الكارب العادي. وجد أن متوسط تركيز العناصر الثقيلة (الكادميوم، الزنك، النيكل، الكروم، النحاس، الرصاص) كانت غير محسوسة ماعدا الزنك التي 8.18 µg/L وفي الرواسب كانت

[1] This research is a part of M.Sc.D. thesis for the First Author.

6.90 µg/L, 146.29 µg/L, 151.46 µg/L, 161.74 µg/L, 38.86 µg/L و 92.49 µg/L على التوالي. كانت اعلى التراكيز للعناصر الثقيلة (الكادميوم، الزنك، النيكل، الكروم، النحاس، الرصاص) في بعض أعضاء جسم السمكة 0.69 µg/L, 6352.84 µg/L, 16.52 µg/L, 9.25 µg/L, 42.18 µg/L و 11.44 µg/L على التوالي، بينما أقل التراكيز كانت BDL, 208.53 µg/L, 2.93 µg/L, 2.93 µg/L, 20.13 µg/L, BDL على التوالي. أظهرت نتائج الدراسة الحالية بأن تسلسل العناصر كانت كالآتى: الزنك> النحاس> النيكل> الكروم> الكادميوم> الرصاص. وتمثل ترتيب العناصر الثقيلة بالنسبة للمجاميع الطولية بالآتي: group4> group3> group2> group1.

Introduction:

Heavy metals, those defined as "heavy", arising from industrial and mining activities are discharged into coastal waters and estuaries at many sites. The term heavy metal refers to any metallic chemical element that has a relatively high density and is toxic, highly toxic or poisonous at low concentrations (Bryan and Langston, 1992). Aquatic ecosystems are very vulnerable to water pollution, where contaminants (heavy metals) are either accumulated in aquatic organisms (Ashraf, 2005) or in the sediment (Karadede and Ünlü, 2007). Fish are widely used as a bio-indicator to evaluate the health of aquatic ecosystems (Farkas *et al*., 2003). Metals such as Cu, Zn, Cr, Ni and Manganese (Mn), at very low concentrations are essential metals since they play an important role in biological systems, whereas Pb, Cd and Mercury (Hg) are non-essential metals because they are toxic, even in traces, and are known to cause severe damage in organisms even at very low concentrations (Gadd, 1993), however, the essential metals can also produce toxic effects when the metal intake was excessively elevated (Türkmen *et al*., 2008). Zn and Cu are known as essential elements that activate many enzymatic systems, Zn especially has an important function since it is a component of metallothionein, In contrast to Zn, Cu and Cd, and there is no evidence that Pb produces the metal-binding protein metallothionein (Smirnov *et al*., 2005). The aim of this project is to investigate the Concentration of some heavy metals contents (Zn, Ni, Cr, Cu, Cd and Pb) of water, sediment and in the skin, gills, intestine, liver and muscle of common carp.

Materials and Methods:

The lakelet of Ranya is a part of Little Zap River which is located east of Ranya district (8.49km) and western north of Sulaimani governorate (83.97km) on 36°11'58.2°N 44°56'50.7°E (Fig 1, 2). The study started at the end December of 2014 to early April of 2015. The depth of water can be determined by the height of a hill in the region which was built between 1956-1958 on high region and its 36m high. Recently this hill was sunken and it's called Basmusian. Yearly or seasonally the depth of water fluctuates, most times it covers the hill completely, so it can be said that the depth of water at least is 36m, the water of region such as Qaladza, Zharawa, Sultanade stream, Shahidan, Garfen, and others pour into water Ranya lakelet, as a result the average of water flow is in a year 222.2 m^3/second and yearly income is about 7 milliard m^3, so it's composed about 17% of Tigris River (Ghafoor, 2008).

Water samples were collected between 10:00 am to 2:00 pm (January, February and March). and collected from the depth of 30 cm from the river. One liter polyethylene bottles used to collect the water sample, each was pre-washed by river water. After that filtrating through qualitative filter paper diameter 9 cm. Before sampling, the bottles were rinsed three times with sample water before being filled with the sample, projecting the mouth of the container against the flow direction. Then, samples were transported in cooler boxes to the laboratory, water samples were preserved in 2% concentrated nitric acid before their use to prevent precipitation of metals (APHA, 2005).

Fish sampled with the help of local fishermen using a fishing net and purchased from them at the spot. The fish were killed by blow on head, and then transferred to the laboratory by using ice box on the same day. In order to minimize the difference in metal accumulation, the length were between 20 cm to 50> cm. The fish specimens dissected to separate organs (gills, intestine, muscle, skin and the entire liver) according to Food and Agriculture Organization of the United Nations

(FAO, 1983) method. After weighting dissection organs were kept frozen at -20°C. Then samples thawed at room temperature, about 1gm of these native organs put in petri dishes and dried at 105°C for 8 hrs in oven to reach constant weight. After that burned by Muffle furnace at 550^{o}C for 4 hrs, the ashes were cooled in desiccator, it's weight was recorded on four point sensitive balance. The ash samples were put into a beaker, Ten ml of concentrated HNO_3 (69%) added into beaker. After that they were left in a dark fume-hood for 2 hours, then stirred to accelerate the reaction of complete digestion. The productive mass was completed to 50 ml with de-ionized water and filtered by using qualitative filter paper diameter 9 cm. Finally the filtered samples were kept in polyethylene container.

Sediments samples were taken by hand from the surface layer (0-10 cm) from the areas always covered with water, placed in polyethylene bags and transported to laboratory in a cool box. The sediment samples were packed in polyethylene containers and stored below -20°C prior to analysis. At first, the sediments were put in Petri dishes and dried in oven at 150 °C for 12 hours, the water content was expressed as a percentage of wet weight. The dried sediments were powdered by using mortar and pestle, and then the powder of samples was sieved to remove larger particles. One gm of sieved sediment was ignited for 4 h at 550°C. After cooling in desiccator, their ashes were weighed and recorded. The ignited sediment was transferred to a beaker then digested with 10ml of concentrated HNO_3 which was slowly added to the sample. It was left in a dark fume-hood for 2 hours with continues stirring, to ensure that the sample is properly wetted. The beaker was covered with watch glass and leaved overnight in hood and then wormed. De-ionized water was added to complete 50 ml volume, and then filtered by qualitative filter paper diameter 9 cm (Binning and Baird, 2001). Finally the sediment solutions were analyzed by ICP-OES (spectro acros) (Inductively Coupled Plasma-Optical Emission Spectrometry) to determine the metal concentration.

Results and Discussion:

The concentration of different heavy metals was varied in different water sample which illustrated in Table (1) and the mean concentrations heavy metals (Cd, Zn, Ni, Cr, Cu and Pb) were below detection limit (1µg/L) except Zn, it was 8.18 µg/L in water. Recently, monitoring of water quality is very important in rivers that are affected by pollutants discharge from cities, atmospheric precipitation and industrial domestic sewage that represent the major source for water pollution (Lomniczi *et al*., 2007). In natural aquatic ecosystem, metals occur in low concentrations, normally at the nanogram to microgram per liter level. The toxicity of heavy metals can be listed in order of decreasing toxicity as Hg> Cd> Cu> Zn> Ni> Pb> Cr> Al> Co (Gray, 1999). The study of (Kassim *et al*., 1997) on Euphrates River recorded that the concentration values of dissolved Zn varied between 0.0026 to 0.0556 ppm, and this agrees with our results. Zinc metal concentrations also showed a general decrease as a result of dilution effect from run off as well as absorption by plants and sediments in the river (Kar *et al*., 2008).

Table 1. Concentration of heavy metals (µg/L) in water.

Heavy metals in water	Cd	Zn	Ni	Cr	Cu	Pb
First time	BDL	8.19[a]	BDL	BDL	BDL	BDL
Second time	BDL	8.26[a]	BDL	BDL	BDL	BDL
Third time	BDL	8.11[a]	BDL	BDL	BDL	BDL
Maximum		8.63				
Minimum		7.63				
Mean± SD		8.14±0.31				
SE		0.1				

*. A different letter means: there is significant at ($p<0.05$).
*. BDL: Below Detection Limit (1µg/L)

Table 2. Concentration of heavy metals (μg/L) in sediment.

Heavy metals in sediment	Cd	Zn	Ni	Cr	Cu	Pb
First time	5.60[a]	110.0[a]	114.83[a]	126.83[a]	30.96[b]	76.76[a]
Second time	7.86[a]	180.43[a]	190.17[a]	190.75[a]	37.86[ab]	105.28[a]
Third time	7.24[a]	148.45[a]	149.39[a]	167.65[a]	47.75[a]	95.43[a]
Maximum	7.93	188.82	192.44	193.16	48.18	106.05
Minimum	5.56	104.21	113.52	125.01	30.47	76.47
Mean± SD	6.91±1.01	146.31±31.32	151.47±32.70	161.75±28.09	38.86±7.33	92.50±12.56
SE	0.34	10.44	10.90	9.36	2.44	4.19
*. A different letter means: there is significant at (p<0.05).						

Agatha (2010) observed that the water was contaminated with Zn and could be toxic to other aquatic fauna and poisonous to human consumers and recommended constant monitoring of levels of contamination to assess the impact of the heavy metal in the aquatic system. Concentrations of Ni were below detection limit (1μg/L) from the three times respectively in water is likely to be of health concern in environments. As is the case with other essential elements Ni is also toxic to fish when present in high enough concentrations (Pickering 1974). Salman (2006) found that the HMs (Cd, Cr, Cu, Ni, Pb, and Zn) in the Euphrates River were in the permissible range of values reported by the WHO, but they were more than that recorded by Al-Taee (1999) in Al-Hilla River. Özkan, (2012) studied assessment of Cu, Cd, Zn, Pb, and Cr by enrichment factor in sediments of inner Izmir Bay and found the values of Cu, and Cd were especially less than 5 while Zn, Pb, and Cr were very heavily enriched in studied area.

The range of our study for Pb were below detection limit (1μg/L) respectively from the three times, a general decrease in the mean concentration of Pb was evident down the river and this could be due to dilution effect from runoff as well as absorption by plants and sediments in the river (Kar *et al*., 2008). The average concentrations of heavy metals Pb, Cd and Cu in the water samples of Al-Masab Alamm were 0.2, 0.05 and 0.034 μg/L respectively; Pb content was the highest and that of Cu was the lowest in water, the order of heavy metals accumulation in water was Pb>Cd>Cu (Al-khafaji *et al*., 2012). Concerning the mean concentrations of heavy metals Cd, Zn, Ni, Cu, Cr and Pb (6.91, 46.29, 151.46, 161.74, 38.86 and 92.49 μg/L respectively), the levels were below the permissible limits reported by Ontario Ministry of the Environment (ANZECC, 2000).

Comparing the heavy metals levels in sediments of the northern delta Lakes with other areas of the world, it was found that similar higher levels of Zn, Cu, Mn, Cd and Pb (13–150, 0.7–36, 160–760, 0.1–0.7 and 2.4–160 μg/g dry wt., respectively) were reported in sediment of Lake Balaton in Central Europe (Nguyen *et al*., 2005), Also showed strong positive correlation among most heavy metals concentrations in sediment, this means that the metals came from the same sources. The best correlation relationships were observed for Pb-Cd, Cd-Ni and Ni-Pb, the low correlation relationships were observed for Zn with the other heavy metals. Based on these fractionation studies of the metals in sediments and their mobility and bioavailability, the elements under study can be arranged as follows (from more bioavailable to less bioavailable), Cr>Ni>Zn>Pb>Cu>Cd.

The concentration of the studied elements in sediments of Lake Srebarna decreased in the order Zn> Pb = Cu and according to (Hristov, 2010), the results of analyses of heavy metals in the one-meter layer of sediments of Lake Srebarna (Cu, Pb, Zn, Cd, and Ni) evidence of increased concentrations over a period of 10 years. Nickel mean concentrations recorded between 0.15-1.12mg/kg DW range, the results showed that the accumulating order of different HMs were as following: Ni> Zn> Pb> Cd and all metals concentrations seemed that they were stable (Eggleton and Thomas, 2004). The Pb concentrations recorded in the sediments from 76.76 to 105.28 µg/L. While these Pb concentrations were below the recommended limits of 35 mg/kg by (WHO, 2011). Zn concentrations ranged from 110.01-180.43 µg/L. Higher Zn concentrations were close to values reported for Martil River, Oum er Rbia River and Sebou River in China and slightly higher than that of Bouregreg River, compared to regional studies, the Zn ranges were lower than those of Lianshan River (Zheng *et al.*, 2008).

The results in Table (3) show the concentration of studied heavy metals according to captured fish length which ranges from (10-20cm) to (41-50cm), a significant differences observed in Cd, Zn, Cu and Pb and no significant differences recorded in each of Ni and Cr.

Table 3. Concentration (µg/L) of studied heavy metals for *C. carpio* according to length groups

Length ranges	Cd	Zn	Ni	Cr	Cu	Pb
G1 (20-30)	BDL	4007.31[a]	6.92[a]	4.96[a]	32.49[a]	BDL
G2 (31-40)	BDL	3211.05[b]	18.26[a]	5.99[a]	24.46[b]	BDL
G3 (41-50)	BDL	2940.25[b]	4.14[a]	7.13[a]	30.37[ab]	0.56[a]
G4 (50>)	0.28[a]	3666.31[ab]	5.70[a]	7.81[a]	22.21[b]	1.07[a]
Maximum	4	15105	422	36	133	34
Minimum	BDL	56	0.1	0.01	4	BDL
Mean± SD	0.23±2.06	3425.07±3091.54	10.58±39	6.12±5.27	27.90±18.54	0.6±15.71
SE	0.166	248.319	3.133	0.424	1.489	1.262

*. A different letter means: there is significant at ($p<0.05$).
*. BDL: Below Detection Limit (1µg/L)

The results of our study showed significance differences among studied organs in which the liver Cd has higher significance than other organs while in each of muscle, skin and intestine were minimum than the blank which means that is below 1 µg/L as shown in Table (4).

Table 4. Concentration (µg/L) of studied heavy metals in different *C. carpio* organs

Organs	Cd	Zn	Ni	Cr	Cu	Pb
Liver	0.69[a]	2228.31[b]	11.21[a]	2.93[c]	42.18[a]	BDL
Gills	0.02[b]	6260.17[a]	10.91[a]	9.25[a]	20.13[b]	11.44[a]
Muscle	BDL	208.53[c]	2.93[a]	7.43[ab]	18.85[b]	BDL
Skin	BDL	2075.81[b]	16.52[a]	5.37[b]	21.29[b]	BDL
Intestine	BDL	6352.84[a]	11.25[a]	5.81[b]	37.15[a]	BDL
Maximum	4.00	15105.00	422.00	36.00	133.00	34.00
Minimum	BDL	56.00	BDL	BDL	4.00	BDL
Mean± SD	0.35±2.07	3425.07±3091.54	10.58±39.00	6.12±5.28	27.90±18.54	5.7±15.72
SE	0.17	248.32	3.13	0.42	1.49	1.26

*. A different letter means: there is significant at ($p<0.05$).
*. BDL: Below Detection Limit (1µg/L)

Target organs such as gills and intestine are metabolically active parts that can accumulate heavy metals in higher levels, as shown in various fish species in *C. carpio* and *Tinca tinca* from lake Beysehir, Turkey (Chouba *et al.*, 2007), in *Oreochromis mossambicus* and *Clarias gariepinus* from Olifant River, South Africa. The metal may be in high concentrations in gill and digestive gland because of relatively high potential for metal accumulation. Many authors reported that tissue is not an active organ in accumulation of heavy metals (Chouba *et al.,* 2007), as proved by our results in which it contain most less value of Ni, Zn and Cu as shown in Table (4). It is clear that gills, liver had higher tendency to accumulate heavy metals more than muscles. Moreover, the present results indicated that the order of metal distribution in fish organs followed the concentration pattern: Zn> Cu> Ni>Cr> Cd> Pb. It could be concluded that the concentrations of heavy metals in organs of the studied fish depended mainly on the metal, organ and species, this is in agreement with that reported by (Abdel-Moneim and Iskander 1995). The present results showed that Pb concentrations in organs of the fish ranged from (BDL to 11.44) as it is very low levels but with different significant respectively (Table 4).

This shows the maximum acceptable concentrations level of heavy metals in fish muscles (µg/g wet wt.) that were described by many international organizations as WHO (Table 5).

Table 5. limit concentrations (µg/g) of heavy metals in fish muscles

Organizations	Cd	Zn	Ni	Cu	Pb	Cr
WHO 2004	1.00	100	0.5-1.0	30	2.00	1.00

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