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Heavy Metal Concentration in Two Freshwater Fishes from Wadi Hanifah (Riyadh, Saudi Arabia) and Evaluation of Possible Health Hazard to Consumers

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Abstract.- The contamination of heavy metals, such as chromium (Cr), iron (Fe), cobalt (Co), nickel (Ni), zinc (Zn), cadmium (Cd) and lead (Pb) were evaluated in the water and tissues of African catfish *Clarias gariepinus* and Sabaki tilapia *Oreochromis spilurus* from Wadi Hanifah, Riyadh. The samples were collected from three sampling sites (Namar, Al-Masani and Al-Hair) of Wadi Valley of central Saudi Arabia during May-June 2013. The water and fish samples collected from Al-Masani, nearby a vehicle mechanical industrial area, were found to be more contaminated by Cr, Fe, Zn and Pb than those of other sampling sites. The high concentrations of Fe were detected in all the water and fish samples, followed by Zn, Pb, Cr, Co, Ni and Cd. However, none of the detected HM (heavy metals) concentrations were exceeded the permissible limits set by the different authorities. The estimated daily intake was found to be below the provisional tolerable daily intake (PTDI), established by the FAO and WHO, indicating that there was no carcinogenic risk for humans.

Key Word: Metal concentration, health hazard and consumer, provisional tolerable daily intake.

INTRODUCTION

Increasing environmental pollution by toxic substances is a growing concern throughout the world. A wide range of contaminant is continuously introduced into the European Food Safety Authority aquatic environment mainly due to increasing industrialization, technological development, growing human population, oil exploration and exploitation, and agricultural and domestic wastes run-off (Lima *et al*., 2008). Among these contaminants, heavy metals constitute one of the most dangerous groups because of their persistence in nature, toxicity, tendency to accumulate in organisms and biomagnifications potential, and non-degradable chemical properties (Fufeyin and Egborge, 1998). They could pose risk to the ecological balance of the environment, affecting bio-diversity of aquatic organisms (Vosyliene and Jankaite, 2006; Farombi *et al.*, 2007) and also enter into the human metabolism through consumption causing serious health hazards (Raja *et al*., 2009). Among animal species, fishes are the inhabitants that cannot escape themselves from the detrimental effects of these pollutants (Olaifa *et al.*, 2004). Fish are often considered as an important bioindicator of aquatic ecosystems, because they obtain a high trophic level and are an important source of balanced protein in the human diet (Rahman *et al*., 2012). Metal residue problems in fish tissues are serious, as reflected by the high metal concentrations recorded in waters and sediments (Pintaeva *et al.,* 2011). Fish are well known for their inherent potential to accumulate heavy metals in their muscles. For this reason, fish muscle is commonly analyzed to determine the concentration of contaminants.

African catfish and Sabaki tilapia are the most common and popular fish species in Wadi Hanifah. The purpose of this study was to determine the heavy metals (Cr, Fe, Ni, Co, Zn Cd and Pb) level in the tissues of two fishes and estimated daily intake (EDI) comparing with provisional tolerable daily intake (PTDI) recommended by FAO and WHO in order to evaluate possible alert regarding human health hazards and give a preliminary assessment of the river water pollution by these metals.

MATERIALS AND METHODS

Sampling site

Wadi Hanifah is a valley in the Nejd region

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of central Saudi Arabia. The valley runs for a length of 120 km from north to south, cutting through the city of Riyadh, the capital of Saudi Arabia. In the past, it was used as a source of water and now as a convenient means for disposing the city's wastewater (Abdel-Baki *et al.*, 2011). In addition to the industrial wastes, the valley also receives the municipal wastes and domestic sewage from the Riyadh City. Because of this scenario, three sampling sites (Namar, Al-Masani and Al-Hair) were selected for water and fish sampling, those of which are located between 24°25 N – 46°48 E and 24°34 N – 46°40 E (Fig. 1).

Sample collection

Ten fishes of each species were collected from each sampling site along with ten water samples. Water samples were collected with 1 litre polyethylene bottles which were previously cleaned by washing with non-ionic detergent, rinsed with tap water and later soaked in 10% HNO_3 for 24 h and finally rinsed with deionized water prior to use. During the sampling, bottles were rinsed with sampled water three times and then filled to the brim at a depth of one meter below the water surface from each of the three designated sampling points. The water samples were digested and heavy metals were determined using Inductively Coupled Plasma Mass Spectrometry (ICP-MS) (Perkin Elmer, NexION 300D, USA).

Fish sample preparation

All the individuals (30 for each species) of the two fish species (*Clarias gariepinus* and *Oreochromis spilurus*) were young adults. Fishes were collected at three sampling sites of Wadi Hanifah using portable lift hand nets and traps as well directly from the fisherman on the spot. The fishes were killed with percussive stunning (Van de Vis *et al.*, 2003), and then transferred into a cooler packed with ice block in order to maintain the freshness and later brought to the laboratory. Ten catfishes with an average length of 25.47±2.23 cm and an average weight of 170.94±41.54 g were caught from each sampling area. Similarly, ten tilapia specimens having an average length and weight of 12.24±2.414 cm and of 33.36±21.45 g, respectively were collected from each sampling site. Each sample was carefully dissected for its muscle. To prevent metal contamination, special care was taken, and the tissues were dissected with special ceramic knife, scissors and plastic forceps (Miyako, California, USA). The muscle was then washed with distilled water and cut into small pieces (2.00–3.00 cm). Then the tissues were oven dried at 65^0C overnight and allowed to cool at room temperature. The dried samples were powdered using a glass mortar, sieved through 1 mm mesh and stored in airtight plastic vials inside the desiccators. Water sampling for the analysis of different elements was done at the same two sampling sites and in the same time.

Digestion

The dried fish samples were digested according to the method of Hanson (1973) as described by Rahman *et al.* (2012). A 0.5g of dried powdered fish tissues (having three replicates) was taken in a digestion apparatus and then 2.5 ml of concentrated H_2SO_4 and 4ml of concentrated HNO_3 were taken. The mixture was slowly heated using a hotplate for 20 min. at 130^{oC} and allow to cool at room temperature (Rahman, 2004). The content was deionized distil water (DDW) and filtered quantitatively into a 50 ml volumetric flask.

Analytical methods

After acid digestion, concentrations were determined according to APHA (1998) through Inductively Coupled Plasma Mass Spectrometry (ICP-MS) (Perkin Elmer, NexION 300D, USA). Analytical blanks were run in the same way as the samples and concentrations were determined using standard solutions prepared in the same acid matrix. Standards for the instrument calibration were prepared on the basis of mono element certified reference solution ICP Standard (Merck). All laboratory plastic and glassware were cleaned by soaking overnight in a 10% nitric acid solution and then rinsed with deionized water.

Calculation of Bioaccumulation Factor (BAF)

The bioaccumulation factor (BAF) consists of ratios of the concentration of a given contaminant in biota (a particular metal concentration in fish muscle) to that in an abiotic media (water and food).

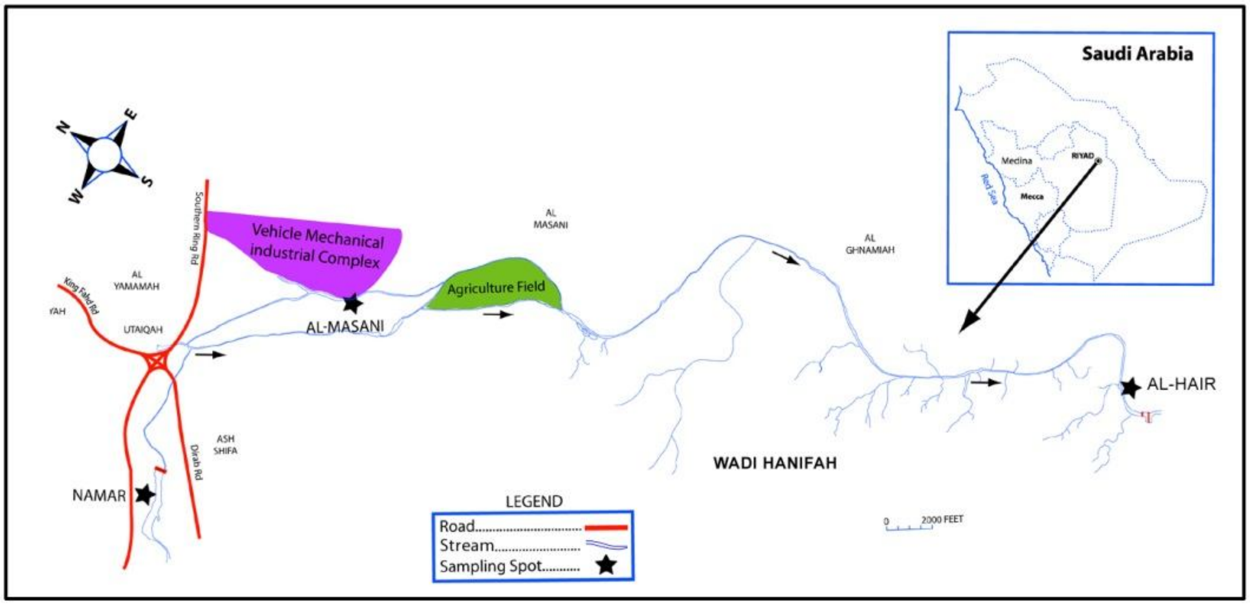

Fig. 1. Riyadh City showing Wadi Hanifah. Asterisk indicates sampling sites.

The accumulation factor was calculated in several studies (Rashed, 2001; Abdel-Baki *et al.*, 2011) according to the formula:

$$\text{Bioaccumulation factor} = \frac{\text{Conc. of metal in fish muscle}}{\text{Conc. of metal in abiotic media}}$$

Risk assessment

The risk for human health as a result of eating these species was evaluated by calculating estimated daily heavy metal exposure Em (US-EPA 2000, Copat *et al.* 2012) as follows:

$$E_m = (C_m * IR_d)/BW$$

where C_m represents the metal concentration in fish muscle (µg/g), IR the daily ingestion rate (µg/d) of freshwater fish. US-EPA risk analysis, considering an adult average body weight of 70 kg (US-EPA, 2000). The average freshwater fish consumption estimated suggested by FAOSTAT (Demo faostat3.fao.org 2013) is only 7 g per day for Saudi.

RESULTS

Heavy metals in water and fish

Average concentrations of Cr, Fe, Co, Ni, Zn, Cd and Pb in water and fish were varied among the sampling sites and fish species (Table I). In water, Fe was the dominant metal at all sampling area followed by Ni, Zn, Cr, Co, Pb and Cd (Table I). Among the three sampling sites, the high level of heavy metal (HM) concentrations in water was found at Al-Masani, where mechanical industrial complex are located.

The average HMs concentration in *C. gariepinus* and *O. spilurus* followed the order Fe> Zn> Pb > Cr > Ni > Co > Cd, and Zn > Fe > Pb > Cr > Co > Ni > Cd respectively (Table I). The bioaccumulation of Fe in *C. gariepinus* was the highest (7300.00 ug/l) at Al-Masani, and that of Cd was the lowest (0.28 ug/l) in *C. gariepinus at* Namar.

Bioaccumulation factor (BAF)

It was found that the concentration of HMs (Cr, Fe, Co, Ni, Zn, Cd and Pb) in fish tissues were several times higher than that obtained in water (Table I). The order of average BAFs in *C. gariepinus* and *O. spilurus* were found to be Pb (119.25) >Zn (81.08) >Cd (50.50) >Cr (22.70) >Co (11.67) >Ni (1.76) >Fe (1.28), and Zn (132.31) >Pb (120.79) >Cd (25.50) >Cr (23.55) > Co (17.37) >Ni (1.43) > Fe (0.29) respectively (Table I).

Table I.- Heavy metals (HM) concentrations in water and tissues of two fish species at different sampling sites of Wadi Hanifah and bioaccumulation factor (BAF). All values represent mean±SD.

Sampling area	HM in water (ug/l) / species (ug/g)	Cr	Fe	Co	Ni	Zn	Cd	Pb
Namar	Water	1.74±0.08	4121.46±158.98	1.18±0.13	9.75±0.09	11.94±1.34	0.02±0.01	0.66±0.05
	Clarias gariepinus	25.00±6.26	4250.31±165.85	23.93±5.37	20.55±4.26	475.75±15.27	0.28±0..36	78.65±3.07
	Oreochromis spilurus	9.25±2.31	766.3±8.46	22.42±4.58	16.23±10.29	674.28±19.37	0.52±0.26	57.09±2.68
Al-Masani	Water	2.76±3.11	4836.37±120.76	0.91±0.20	7.91±1.25	13.87±4.09	0.03±0.02	0.94±0.81
	Clarias gariepinus	45.90±5.21	7300±145.45	12.53±2.28	16.71±3.26	1275.68±5.27	1.25±0.54	125.92±12.54
	Oreochromis spilurus	58.27±4.56	1795.44±66.25	24.99±3.41	14.65±2.19	2000.68±11.23	0.5±0.28	127.09±4.68
Al-Hair	Water	1.04±0.03	4643±93.41	1.44±0.02	12.06±0.14	11.77±1.40	0.02±0.2	0.21±0.37
	Clarias gariepinus	31.23±2.42	5125.32±178.32	9.42±1.41	19.71±4.24	831.68±5.27	1.5±0.25	72.85±11.89
	Oreochromis spilurus	37.57±2.78	1211.59±62.60	20.85±4.29	15.59±5.22	1540.54±7.84	0.5±0.36	55.26±6.33
Mean±Sd	Water	1.5±1.09	4351.63±679.04	1.31±0.35	10.79±2.50	10.62±3.95	0.02±0.01	0.44±0.43
	Clarias gariepinus	34.04±10.73	5558.54±1570.32	15.29±7.64	18.99±2.02	861.04±400.77	1.01±0.64	52.47±63.67
	Oreochromis spilurus	35.03±24.61	1257.78±516.12	22.75±2.10	15.49±0.79	1405.17±673.48	0.51±0.01	53.15±64.04
Average BAF	*Clarias gariepinus*	22.70	1.28	11.67	1.76	81.08	50.5	119.25
	Oreochromis spilurus	23.35	0.29	17.37	1.43	132.31	25.5	120.79

Risk assessment

The estimated daily intake (EDI) of HM from fresh water fish was in the following manner: Fe, Zn, Pb, Cr, Ni, Co, Cd, and Zn, Fe, Pb, Cr, Co, Ni, Cd for *C. gariepnius* and *O. spilurus* respectively (Table II). The estimated daily intake of HM in both fishes were almost similar except that Zn and Fe interchanged their trend at positions 1 and 2 as well as for Co and Ni interchanged their positions 5 and 6 respectively (Table II).

DISCUSSION

Ezemonye (1992), Egborge (1991) and Edema (1993) have linked the concentration of heavy metals (HMs) in the aquatic ecosystems with effluents from industries, refuse and sewage. This study indicated high level of iron concentrations in all sampling areas (Table I). Result showed an obvious contamination in the water samples taken from Al-Masani, where mechanical industrial complex of vehicles are located. However, the level of HMs concentration in water was generally lower than maximum permissible concentrations (MPC) of various metals in natural water recommended by the Environmental Protection Agency (US-EPA, 1987). Nevertheless, the concentrations of Fe, Zn, Ni and Co in water are however higher when compared with concentrations in non-oil exploration locations of various studies (Ideriah *et al.*, 2010). Concentration of Fe is higher than values obtained from River Benu, Central Niger (Eneji *et al.*, 2011) and Fe, Zn and Co concentrations are higher than that of the Lake Nasser, Egypt (Mohammed, 2008). Increasing contamination of HMs in water and their bioaccumulation in aquatic organisms may have long term adverse implication on human health and ecosystems (Fernandes *et al.* 2007).

As expected, fish tissues that were directly exposed to metals in water, had higher levels at

Table II.- Comparison of average heavy metal in tissue and daily intake from studied fishes with recommended dietary intake.

Metals	Water (µg/l)	Mean HM concentration in tissue (µg/kg ww)		Recommended concentration (µg/kg ww)		Estimated daily intake µg/kg bw/d		Recommended daily intake µg/kg bw/d	
		C. gariepinus	*O. spilurus*	Water (µg/L) (EPA)	Tissue (µg/kg ww) (IAEA)	*C. gariepinus*	*O. spilurus*	µg/kg bw/d	Reference
Cr	1.74	34.04	35.03	50000	730	0.0034	0.0035	0.71-2.9	WHO (1996)
Fe	4121.46	5558.54	1257.78	300000	146000	0.5558	0.1258	800	FAO/WHO*
Co	1.18	15.29	22.75	-	100	0.0015	0.0023	-	-
Ni	9.75	18.99	15.49	13400	600	0.0019	0.0015	300	WHO (1996)
Zn	11.94	861.04	1405.17		67000	0.0851	0.1405	300-1000	FAO/WHO*
Cd	0.02	1.01	0.51	10000	189	0.0001	0.0000	0.83	FAO/WHO*
Pb	0.66	52.47	53.15	5000	120	0.0052	0.0053	3.57	EFSA*

International Atomic Energy Agency (IAEA),
*Tolerable intake, *World Health Organization (FAO/WHO 2010),
*European Food Safety Authority (EFSA, 2010)

Al-Masani than those found in Namar and Al-Hair. As shown in Table I, the sequence of HM concentration in *C. gariepinus* and *O. spilurus* were Fe > Zn > Pb > Cr > Ni > Co > Cd, and Fe > Zn > Pb > Cr > Co > Ni > Cd respectively. This order has followed due to the variations in bioavailability, intrinsic fish processes, and trophic structure (Eneji *et al.* 2011). Fe was the most abundant metal in both fishes and in particular, it was significantly elevated in fish muscle in all samplings dates. Chan (1995) opined that the trace metal in fish tissue was increased proportionally to their level in water. The bioaccumulation of Fe is comparatively high in *C. gariepinus* than *O. spilurus.* The high accumulation of Fe in fish tissues can be attributed to the large quantities of Fe detected in water that agrees well with the findings of Zyadah (2005) and Mohammed (2008). The high levels of Fe and Zn recorded in the present study might be due to the disposal of industrial and domestic wastes from residential areas which may contain higher levels of Fe and Zn. Similar observation has also been reported by Alex *et al.* (2013).

Olaifa *et al.* (2004) reported that fish species can accumulate HMs above the levels of abiotic environment to incur bioaccumulation. Differences in heavy metals bioaccumulation could be linked to the differences in feeding habits and behaviour of the species (Altindag and Yigit, 2005). Heavy metals are also known to be accumulated in fish tissues, reaching to the concentrations up to 20000 fold higher than surrounding water environment (Popek *et al.*, 2006). Generally the BAF of two fish species from Wadi Hanifah were found to be low compared to the other works from the River Niger at Jebba (Lawani and Alawode, 1987); Lagos lagoon (Okoye, 1991); River Niger, Onitsha (Obodo, 2002), River Benue, North and Central Nigeria (Eneji *et al.*, 2011). BAF was found to be inversely proportional to metal toxicity levels in fish tissue (Bu-Olayan and Thomas, 2008). It also gives an indication about the accumulation efficiency of any particular pollutant in any fish. However, BAF of the studied metals indicate that the elevated concentrations of the HMs in both fish were derived from water, which is also supported by Soltan *et al.* (2005) and Rashed (2001). It is clear that the tissues of *C. gariepinus* and *O. spilurus* accumulated metals, which are more or less similar to each other. It could therefore be concluded that the concentrations of metals in the studied fish tissues are dependent upon the types of metal and surrounding water environment.

The estimated daily intake (EDI) values indicate the low concentration of all HMs, which are several fold lower than that in fish muscle recommended by several international organizations (WHO, 1996, 2010) indicating a lesser contamination of fish in this area.

In conclusion, the level of HM in water and fish tissue were detectable but presumably below any toxic risk for consumers. There appears to be no human health risk of ingesting HMs from two fish muscles collected from Wadi Hanifah, Riyadh, Saudi Arabia. However, more research investigation should be required including monitoring with other pelagic as well as benthic species, considering that the later are more reliable for this task. Furthermore, it is quite evident that there was accumulation of HMs in fish tissues, and if no preventive measures are taken, the condition may get worse in time to come.

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