

Health risk assessment of heavy metals associated with fish consumption in Edku region, Egypt.

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ABSTRACT

Health risk assessment of exposure to heavy metals through fish consumption was carried out. Heavy metals concentrations were monitored in ventral muscles of boliti fish; *Tilapia sp.* collected seasonally during 2006/2007. Five geographic zones of Edku Lake were chosen for sampling, according to closeness to discharge points of pollutants effluents. The highest mean values of metal concentrations were 1202.9, 57.6 and 3465.8 ppb for Pb, Cd, and Zn, respectively, during autumn. While the levels were 1038.6 and 1036.0 ppb for Cu and Mn during winter and spring, respectively. Checked metal concentrations were in values not exceeding the WHO permissible limits except for Zn and Mn. Estimated risk was evaluated in correlation to fish consumption. Chronic daily intakes (CDI_s) based on 95th centile were 0.021, 0.0012, 0.032, 0.1117 and 0.0284 mg/kg/day for Pb, Cd, Cu, Zn and Mn, respectively. On the other hand, Target Hazard Quotient (THQ) reached the highest value in case of Mn which found to be values of 5.68 and 2.42 based on 95 and 90th centile of fish consumption. The lowest values, however, were 0.15 and 0.09 at 50 and 25th centile. Generally, THQ of measured metals was greater than unity suggesting that adverse health effects may be associated with high fish consumption.

Key words: Fish, heavy metals, risk estimation, Edku habitats, Egypt.

INTRODUCTION

Metals, particularly heavy metals such as lead, mercury, cadmium and others constitute a potential threat to human health, both occupationally and environmentally (Hu, 2000). Human is exposed to metals *via* different routes through food, water, air, soil. Additionally, U. S. Food and Drug Administration (USFDA, 1993) stated that fish and other seafood accounted for 90% of the total arsenic exposure in human.

Several authors reported different levels of heavy metals retained in fish during anthropogenic pressure (Olsson *et al.*, 1988; Mohamed *et al.*, 1990; Khallaf *et al.*, 1994; Gary *et al.*, 1997 and Chan *et al.*, 1999).

Contamination of aquatic ecosystems (e.g. lakes, rivers, streams) with heavy metals has been receiving increased attention worldwide (Prat, *et al.*, 1999; Bhattacharya and Sarkar, 1998; Davis and Bastian, 1990; Hakanson, 1984; Harms, 1975; and Wiener and Giesy, 1979). Such contaminant was, in most cases, the result of seepage of industrial wastes (PNUE, 1984).

In Egypt, samples collected from seven Governorates indicated the presence of some heavy metals (lead, cadmium, chromium, zinc, copper, manganese and iron) in different fish organs (Gomaa *et al.*, 1995 and Mansour and Sidky, 2002).

Lake Edku; Behira Governorate, with an area of 8000 hectares, has an economic importance for the surrounding region, where more than 2 thousands of the inhabitants work in the lake for fish catching and distribute in different areas inside and outside the Governorate. The boliti fish; *Tilapia* sp. is the common species in this Lake. The Lake is continuously exposing to different pollutants emerging from industrial, agricultural and domestic effluents originating from drains pour in the lake. There were lack of laws and management of fish catching and breeding programs in the lake. Most of surveyed people were in high consumption rates of fish in their diets where, fish consumption in coastal cities was about 10 times that of the average quantity of seafood consumed in rest of countries (FAO, 1980). Thus, the average of fish consumption rates was estimated to be 100-150 g/day/person. Historically, discharges of untreated effluents into the waters resulted in accumulation of pollutants and heavy metals in water columns, sediment and aquatic biota, especially fish tissues (Connell *et al.*, 1998a, b and Wu, 1988).

Therefore, the current study was aimed to determine the levels of some heavy metals in fish tissue samples collected seasonally from Edku Lake during 2006/2007 and assess the potential risks to which the inhabitants consuming these fish are exposed in relation to their state of contamination by heavy metals.

and Me'Adeyyah regions. The risk factors that were being tested through the survey revealed presence of untreated effluents into the Lake, decreasing in fish population and fish contamination.

Sample Preparation and Treatment: Fish samples were dissected and 25 gm of each were taken from ventral muscles for analysis by the dry ashing method (Crosby, 1977). The samples were oven dried at 105 °C for 24 hrs and powdered in a mixer grinder. Three powdered samples (5 gm, each) were accurately weighed and placed in crucibles and few drops of concentrated nitric acid (Merck) were added to the solid as an ashing aid. Dry-ashing process was carried out in a muffle furnace by stepwise increase of the temperature up to 550 °C and then left to ash at this temperature for 4 hrs. The ash was left to cool and then rinsed with 1 M nitric acid. The ash suspension was filtered into a 25 ml volumetric flask using Whatman filter paper No. 41, and the solution was completed to the mark with nitric acid (1 M).

Flame Atomic Absorption Analysis: Analysis of heavy metals was performed using a Thermo elemental type SOLAAR atomic absorption spectrophotometer. Measurements were made using the hollow cathode lamps for Pb, Cd, Mn, Zn and Cu. The limit of detection (LOD) of the analytical method for each metal was calculated as double the standard deviation of a series of measurements of a solution, the concentration of which is distinctly detectable above, but close to blank absorbance measurement (USEPA, 1983). These values were 0.001, 0.002, 0.01, 0.001 and 0.02 mg/kg and the limits of quantification (LOQ) were 0.016, 0.002, 0.030, 0.127 and 0.113 mg/kg for Pb, Cd, Mn, Zn and Cu, respectively.

Standards and Quality Assurance: Standard solutions of the tested heavy metals were provided by Merck (Darmstadt, Germany) and prepared from the individual metal 1000 mg/l standards in 0.1 N HNO₃. Working standards were prepared from the previous stock solutions. Appropriate quality assurance procedures and precautions were carried out to ensure reliability of the results. Samples were carefully handled to avoid contamination. Deionized water was used throughout the study. A recovery experiment was carried out by spiked a fish tissue powder which supplied by Oceanographic Museum, Monaco-Ville, France with each 10 and 20 µg/ml of metals standard solutions. The spiked samples were processed for the analysis by the dry-ashing method and analyzed as described above. The average recoveries obtained were 74.5, 49.5, 62.4, 92.5 and 98.4 % for Pb, Cd, Mn,

Zn and Cu, respectively. The blank was processed without tissue and the procedures were done as described before.

Risk estimation: The risk contaminated fish consumption was estimated based on guidelines of EPA (USEPA, 1985, 1999a and 2002), concentration of metals and data of Integrated Risk Information System (IRIS).

The chronic daily intake (CDI) ($\mu\text{g}/\text{kg}/\text{day}$) from fish ingestion was estimated from the following formula:

$$CDI_{ing} = \frac{C \cdot I_R \cdot E_D \cdot E_F}{B_W \cdot A_T}$$

Where C is the concentration of the metal expresses as mg/kg, I_R the ingestion rate (0.113 g/meal)(50th percentile for fin fish) Pao *et al.*, (1982), E_D is the average age (70 year; Age-specific values) USEPA, (1985, 1989a), E_F is the exposure frequency (48 day/year; EPA tolerance assessment system in USEPA, 1989b), B_W is the average body weight of the receptor over the exposure period (70 kg; adult, average; USEPA, 1989c), and A_T is the averaging time, the period over which exposure is averaged 30 years (USEPA, 1989c).

Furthermore, non-cancer toxic effect, i.e. Target hazard quotient (THQ) was also estimated using the following equation (USEPA, 1999 b):

$$THQ = \frac{CDI}{RfD}$$

Where RfD is the reference dose obtained from USEPA (USEPA, 2005). Total THQ (TTHQ) of heavy metals for individual's foodstuff (fish) is the sum of the following compositions:

$$TTHQ = THQ (\text{toxicant}_1) + THQ (\text{toxicant}_2) + \dots + THQ (\text{toxicant}_n).$$

Statistical Analysis: The obtained data were expressed as mean \pm S.E. and statistically analyzed using ANOVA to determine the significant differences between treatments (Steel and Torrie, 1980).

RESULTS AND DISCUSSION

Heavy metal levels in fish tissue samples: The concentrations of heavy metals found in fish tissue samples were presented in Table (1). Means concentrations of Cu, Zn and Mn were high in most of the seasons, where mean of Cu concentration recorded the highest value (1038.6 ppb) in winter and Zn recorded highest values (3244.5 and 3465.6 ppb) during summer and autumn seasons, respectively. In contrast, Cd was found in low concentrations (10.6, 35.5, 27.4 and 57.6 ppb) compared with the other examined metals during winter, spring, summer and autumn seasons, respectively. Lead was found in high concentrations (765.7 and 1202.0 ppb) during summer autumn seasons, respectively.

In general, the mean concentrations of heavy metals were not exceeding the WHO permissible limits except for Zn and Mn (Table 2). Muscles usually have the lowest accumulation potential of heavy metals, and metal concentrations are usually highest in parenchymatous organs (liver and kidney)(Lafaurie *et al.*, 1981; Lafaurie, 1982; Hilmy *et al.*, 1983; Dallinger, *et al.*, 1987 and Chen and Chen, 1999). Indeed the metabolism of these metals involves metallothioneines (proteins with a high affinity for toxic metals), which intervene in detoxification processes. Rashed (2001) showed that the transfer factors (Tf) of some heavy metals from Nasser Lake water, aquatic plants and sediments to fish were greater than 1, and this means that the fish undergo bioaccumulation of these elements from water.

Agricultural runoff was the important sources of pollution in the Lake, especially during spring and summer seasons. Also, continuous discharging of solid wastes into the Lake led to sedimentation and decreasing the depth of water in some areas and continuously causing low movement of water between the Lake and Mediterranean Sea via Me'Adyyeha strait. All such conditions led to accumulation of heavy metals in water media and biomagnifications in fish tissues. However, further continuous monitoring of fish is recommended to ensure protection of consumers from the contamination with metals.

Table (1) Seasonal concentrations of heavy metals (ppb) dry weight in fish samples collected from Edku Lake during 2006/2007.

Zones	Winter					Spring				
	Pb	Cd	Cu	Zn	Mn	Pb	Cd	Cu	Zn	Mn
El-Morda	104.9 ±7.48	23.1 ± 3.99	1731.9 ±29.4	2750.2 ±37.1	705.3 ±18.8	38.3 ±4.37	16.1 ±2.83	799.0 ±19.9	2186.7 ±33.1	847.0 ±20.6
Me'Adeyyah st.	85.1 ±6.52	11.6 ± 2.41	527.5 ±16.2	2040.0 ±31.9	274.2 ±11.7	38.2 ±4.38	16.7 ±2.89	349.4 ±13.2	3433.4 ±41.4	1252.9 ±25.0
Middle of lake	58.8 ±5.42	0.01 ±0.05	467.4 ±15.2	2394.5 ±34.6	632.7 ±17.8	108.3 ±7.35	18.7 ±3.06	607.2 ±17.4	2547.0 ±35.7	1053.3 ±22.9
Barseq pumps	312.7 ±12.5	7.4 ±1.92	704.0 ±18.7	1982.0 ±31.5	487.0 ±15.6	98.3 ±7.00	11.8 ±2.44	180.2 ±9.49	1891.3 ±30.8	984.2 ±22.2
Kom Belage	66.4 ±5.76	10.9 ±2.34	1762.4 ±29.6	4623.4 ±48.1	1319.2 ±25.6	37.7 ±4.35	114.0 ±7.55	510.2 ±15.9	2354.3 ±34.3	1042.6 ±22.8
Mean	125.7 ±5.60	10.6 ±1.63	1038.6 ±16.1	2758.0 ±26.3	683.7 ±13.1	64.2 ±4.00	35.5 ±2.98	489.2 ±20.0	2480.7 ±24.9	1036.0 ±16.1
LSD 0.05 (Zones x Season)			225.5					23.85		

Each value is the mean of three replicates ±S.E.

Continued;

Zones	Summer					Autumn				
	Pb	Cd	Cu	Zn	Mn	Pb	Cd	Cu	Zn	Mn
El-Morda	403.9	21.2	1075.2	2970.1	515.6	2333.5	80.0	828.4	2860.0	191.4
	± 14.2	± 3.25	± 23.2	± 38.6	± 16.1	± 28.6	± 6.31	± 20.4	± 37.8	± 9.79
Me'Adeyyah st.	320.8	15.9	805.1	2538.5	360.7	1204.5	49.0	771.4	3009.2	587.0
	± 12.7	± 2.81	± 20.1	± 35.6	± 13.4	± 24.5	± 4.95	± 19.6	± 38.8	± 17.1
Middle of lake	358.5	16.4	997.1	3438.5	577.8	1317.1	77.5	1133.0	4445.5	942.0
	± 13.4	± 2.86	± 22.3	± 41.5	± 17.0	± 25.7	± 6.22	± 23.8	± 47.1	± 21.7
Barseq pumps	1001.8	52.2	1061.5	3852.8	906.4	860.5	40.0	1014.0	3680.0	995.5
	± 22.4	± 5.11	± 23.1	± 43.9	± 21.3	± 20.7	± 4.48	± 23.8	± 42.9	± 22.3
Kom Belage	1743.3	31.4	930.9	3422.7	548.9	299.3	41.5	872.5	3334.1	986.0
	± 29.5	± 3.96	± 21.6	± 41.4	± 16.6	± 12.2	± 4.55	± 22.5	± 40.8	± 22.2
Mean	765.7	27.4	973.9	3244.5	581.9	1202.9	57.6	923.9	3465.8	740.4
	± 13.8	± 2.62	± 17.1	± 28.5	± 12.1	± 17.3	± 3.79	± 20.9	± 29.4	± 13.6
LSD 0.05			365.7					364.5		
(Zones x Season)										

Each value is the mean of three replicates ±S.E.

Table (2). Average of seasonal heavy metals concentration (ppb) in fish tissue of Edku Lake during 2006/2007.

Metals	Mean (ppb)	Permissible limits (ppb), FAO (1983)
Pb	552.0	40000.0
Cd	33.0	2000.0
Cu	860.0	30000.0
Zn	2000.9	-
Mn	761.0	500.0

- = Not listed

Risk estimation: Percent cumulative probability was investigated against concentrations of each metal. The CDI equation described the relationship obtained by linear regression, from which corresponding exposure from 95th to 25th centiles were calculated to provide measures of exposure (Solomon *et al.*, 2000). Chronic daily intake of metals *via* fish consumption and THQ were presented in Table (3). The values of CDI were variable for all metals according to consumption rates and duration periods. Mn and Zn showed the highest values (0.0284 and 0.1117 mg/kg/day) respectively, based on 95th centile. In contrast, Cd was reached the lowest values in all probable percents to be 0.00053 and 0.00049 mg/kg/day based on 90 and 50th centile, respectively. These values were lower than that values reported by the joint FAO/WHO (1999), where set a limit for heavy metals intake based on body weight. For an average adult (60 kg body weight), the provisional tolerable daily intake (PTDI) for lead, iron, copper and zinc are 214 µg, 48, 3 and 60 mg, respectively.

Table (3). Slope factors (S.F.) and reference doses (RfD_s) of heavy metals, (USEPA, 1999).

Metals	S.F. oral (mg/kg/day) ⁻¹	RfD mg/kg/day
Pb	Missing slope	Missing RfD
Cd	Missing slope	0.0005
Cu	Missing slope	Missing RfD
Zn	Missing slope	0.3
Mn	Missing slope	0.005

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Pb	Missing slope	Missing RfD
Cd	Missing slope	0.0005
Cu	Missing slope	Missing RfD
Zn	Missing slope	0.3
Mn	Missing slope	0.005

On the other hand, THQ was exceeded the recommended value of EPA in case of Mn. The highest value was 5.68 based on 95th centile, while the lowest was 1.30 at 25th centile. In contrast, not exceeded 1 in case of Zn. TTHQ was exceeded 1 to be 8.45 based on 95th centile and 1.95 at 25th centile as illustrated in Figure (2) and Table (4). All values were greater than unity suggesting that adverse health effects may be associated with high fish consumption.

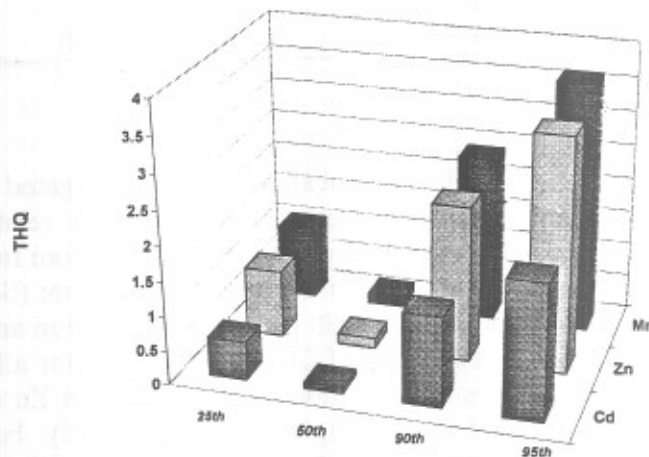


Fig. 2 Target hazard quotient (THQ) of heavy metals with different percentiles.

Table (4). Risk estimation of metals exposure associated with fish consumption.

Probability risk values	CDI _f (mg/kg/day)					THQ			TTHQ
	Pb	Cd	Cu	Zn	Mn	Cd	Zn	Mn	
95 th percentile	0.021	0.0012	0.032	0.1117	0.0284	2.40	0.37	5.68	8.45
90 th percentile	0.011	0.00053	0.014	0.0477	0.0121	1.06	0.16	2.42	3.64
50 th percentile	0.008	0.00049	0.0127	0.0444	0.0113	0.98	0.15	2.26	3.39
25 th percentile	0.005	0.00028	0.00015	0.0256	0.0065	0.56	0.09	1.30	1.95

Generally, metals and their salts exert toxic effects in the organisms at tissue, cellular, subcellular, and molecular levels. Toxicity at the cellular levels causes deranged reproduction, differentiation, and maturation

resulting in teratogenesis. While at the molecular levels, some metals interact with proteins, leading to denaturation, precipitation, allosteric effects, or enzyme inhibition. At individual statements, lead poisoning as example causes edema, a direct cytotoxic effect, and neuromuscular, gastrointestinal, renal and hematological effects. On the other hand, cadmium causes different toxic effects such as inhibition of α -antitrypsin, urinary β micro globulin, and causes of hypertension in cardiovascular tract (Abou-Donia, 1992).

Total risks for residents: Results showed that people have high risk through fish consumption in Edku region. Based on the assumption that over 50,000 (CAPMAS, 2006) people living in Edku have 70-year lifespan could get more risk in the future through fish consumption from the lake. Thus, we recommended that, remediation programs must be carried out to minimize the risk factors in this region.

CONCLUSION

In the present study, the risk assessment of exposure to heavy metals through fish consumption was carried out. The concentration levels of examined metals in bolti tissue reached the highest values of 1202.9, 57.6 and 3465.8 ppb for Pb, Cd and Zn, respectively, during autumn season. Most values of metal concentrations not exceeded the WHO permissible limits except for Zn and Mn. TTHQ was exceeded 1 to be 8.45 based on 95th centile and 1.95 at 25th centile. These values were greater than unity suggesting that adverse health effects may be associated with high fish consumption. Generally, individual or multi-exposure of metals contamination cause different health effect such as edema, direct cytotoxic effect, neuromuscular effects...etc. However, continuous monitoring of fish is recommended to ensure protection of consumers against these metals.

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تقييم المخاطر الصحية الناتجة عن استهلاك الأسماك الملوثة بالعناصر الثقيلة في ادكو- مصر

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تم دراسة المخاطر الناتجة عن استهلاك اسماك البلطي المأخوذة من بحيرة ادكو خلال عام 2007. خمس مناطق جغرافية تم اختيارها تبعا لمستوى التلوث الناتج من المصارف التي تصب في البحيرة. أعلى تركيز كان 1202.9 ، 57.6 ، 3465.8 جزء في البليون لكل من الرصاص، الكاديوم، الزنك على التوالي خلال فصل الخريف بينما كان أعلى تركيز 1038.6 ، 1036.0 ، جزء في البليون لكل من النحاس والمنجنيز خلال فصلي الشتاء والربيع على التوالي. كان معدل الاستهلاك اليومي عند أعلى احتمال (95%) 0.021 ، 0.0012 ، 0.032 ، 0.1117 ، 0.0284 مجم/كجم/يوم لكل من الرصاص ، الكاديوم ، النحاس ، الزنك ، المنجنيز على التوالي. على الجانب الآخر معامل التأثير الغير سرطاني THQ وصل أعلى قيمة 5.68 ، 2.42 للمنجنيز عند احتمالات 95 ، 90 % من معدل الاستهلاك وهذا يبين أن هناك مخاطر متزايدة مع تزايد استهلاك الأسماك من هذه المنطقة.