Detection of Heavy Metals Residues in Fish and Shellfish

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A B S T R A C T

The present study was conducted on 90 random samples of fish of *Claris gariepinus*, *Oreochromis niloticus* and *Mugil cephalus* and shellfish of Oyster, Shrimp and Crab (15 of each) were collected from different fish markets in Kalyobia governorate, Egypt, for determination of their content of mercury, lead and cadmium. The obtained results recorded that, the mean value of concentrations with mg/kg of mercury; lead and cadmium in fish samples were 1.62 ± 0.15, 1.29 ± 0.12, 0.95 ± 0.08;0.67 ± 0.09, 0.53 ± 0.06, 0.36 ± 0.05 and 0.25 ± 0.03, 0.19 ± 0.01, 0.14 ± 0.01 for *C. gariepinus, O. niloticus* and *M. cephalus,* respectively. Meanwhile, in shellfish samples they were 1.37 ± 0.16, 1.14 ± 0.10, 0.73 ± 0.09;0.56 ± 0.07, 0.48 ± 0.05, 0.40 ± 0.04 and 0.22 ± 0.02, 0.15 ± 0.01, 0.12 ± 0.01 for oyster, shrimp and crab, respectively. Moreover, the results revealed that,the highest mercury contamination was in *Claris gariepinus* followed by oyster; *O. niloticus*; shrimp; *M. cephalus* and finally crab. Whereas the highest lead contaminations were in *C. gariepinus* followed by oyster; *O. niloticus*; shrimp; crab and finally *M. cephalus*. Moreover, for cadmium the highest ratio in in *C. gariepinus* followed by oyster; *O. niloticus*; shrimp; *M. cephalus* and finally crab.

Key words: Shellfish, Residues, mercury, cadmium, lea.

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1. INTRODUCTION

Fish and fish products are important part of a healthy diet due to many nutritional benefits as they contain high-quality protein, low cholesterol level, omega-3 fatty acids, minerals as well as vitamins. Also, their lower costs potentiate their nutritive values. A well-balanced diet includes a variety of fish and shellfish can contribute to heart health and children's growth and development. The pollution of aquatic environment with heavy metals constitutes a public health hazard during recent years. Scientific researches have proven that, the presence of the residues in foods is harmful to human health which be associated with immune-suppression, hypersensitivity to chemical agents, breast cancer, reduce sperm count and infertility (Sharp, 1999). Some heavy metals such as mercury, cadmium and lead introduced into environmental water system may pose high toxicities on the aquatic organisms (Ambreen *et al*., 2015). Pollution enters fish and shell fishes through five main routes: via food or non-food particles, gills, oral consumption of water and the skin (Mitra *et al*., 2012). In general, heavy metals are not biodegraded and therefore, their bioaccumulation in fish, oyster, mussels, sediments and other components of aquatic ecosystems have been reported from all over the world (Kumar and Singh, 2010). In addition, it appears that problem of heavy metals accumulation in aquatic organisms including fish needs continuous monitoring and surveillance owing to biomagnifying potential of toxic metals in human food chain (Kumar *et al.,* 2009). The accumulation of metals in fresh water fish and marine shellfish has been proposed as a more sensitive and specific indicator of environmental contamination (Kavun and Podgurskaya, 2009). It is important to investigate the levels of heavy metals in these organisms to assess whether the concentration is within the permissible level and will not pose any hazard to the consumers (Cid *et al*., 2001). Scientific researches have proven that, the presence of heavy metals residues in foods is harmful to human health which be associated with immune-suppression, hypersensitivity to chemical agents, anaemia, chronic renal failure, encephalopathy, breast cancer, reduce sperm count and infertility (Rubin and Strayer ,2008 and Rhman *et al.* ,2011). Therefore, the present study was conducted to evaluate the contamination levels of fish and shellfish with heavy metals and their acceptability for human consumption.

2. MATERIAL AND METHODS

2.1. *Collection of samples*

A total of 90 random samples of fresh fish of *Claris gariepinus*, *Oreochromis niloticus* and Mugil *cephalus* (15 of each) and shellfish of Oyster, Shrimp and Crab (15 of each) were collected from different fish markets in Qaliobia governorate, Egypt, for determination of mercury, lead and cadmium levels.

2.2. Determination of heavy metals

All collected samples were examined for determination of heavy metals (mercury, lead and cadmium) levels on the basis of wet weight (mg/Kg).

2.2.1. Washing procedures following (Lars, 2003).

2.2.2. Digestion technique following (Staniskiene et al., 2006).

2.2.3. Preparation of blank and standard solutions following (Andreji et al., 2005).

2.2.4. Quantitative determination of heavy metals:

Absorbency of mercury, lead and cadmium was directly recorded from the digital scale and their concentrations were calculated according to the following equation: C=R x (D/W). Where, C= Concentration of the element (wet weight) R= Reading of digital scale of AAS.D= Dilution of the prepared sample. W= Weight of the sample.

N. B. The concentration of each element in the blank solution was also calculated and subtracted from each analysed sample.

3. RESULTS

The obtained results in Table (1), revealed that, the minimum and the maximum mercury concentrations "mg/Kg " in the examined samples of C. *gariepinus, O. niloticus* and *M. cephalus* were ranged from 0.38 to 2.45; 0.31to 2.07 and 0.22 to 1.51 with mean values of 1.62 ± 0.15; 1.29 ± 0.12 and 0.95 ± 0.08, respectively. Meanwhile, in shellfish samples of oyster, shrimp and crab were ranged from 0.35 to 2.24; 0.26 to 1.85 and 0.17 to 1.33 with mean values of 1.37 ± 0.16;1.14 ± 0.10 and 0.73 ± 0.09, respectively. Moreover,73.3% samples ofC.*gariepinus;*66.7% of *O. niloticus*; 46.7% of *M. cephalus;*9oyster (60 %)*;*53.3% of shrimp and 40% of crab, were unaccepted, as they were exceeded the maximum permissible limit of mercury in fish and shellfish that should not exceed 0.50 mg/kg (EOS, 2010).

Results in Table (2) declared that, the minimum and the maximum lead concentrations "mg/Kg " in the examined samples of C. *gariepinus, O. niloticus* and *M. cephalus* were ranged from 0.12 to 1.04; 0.09 to 0.82 and 0.05 to 0.60 with mean values of 0.67 ± 0.09; 0.53 ± 0.06 and 0.36 ± 0.05, respectively. Meanwhile, for shellfish samples oyster, shrimp and crab were ranged from 0.10 to 0.89;0.08 to 0.74 and 0.07 to 0.68 with mean values of 0.56 ± 0.07;0.48 ± 0.05 and 0.40 ± 0.04, respectively. In addition; 42 samples, 10C.*gariepinus* (66.7%)*;* 8 *O. niloticus* (53.3%)*;* 4 *M. cephalus* (26.7%)*;*8oyster (53.3%)*;*7shrimp (46.7%) and 5 crabs (33.3%), were unaccepted, as they were exceeded the maximum permissible limit of lead in fish and shellfish that should not exceed 0.30 mg/kg (EOS, 2010).

As shown in Table (3) results revealed that, the minimum and the maximum cadmium concentrations "mg/Kg " in the examined samples of fish were ranged from 0.02 to 0.53; 0.02 to 0.37 and 0.01 to 0.24with a mean value of 0.25 ± 0.03; 0.19 ± 0.01 and 0.14 ± 0.01 in C. *gariepinus, O. niloticus* and *M. cephalus*, respectively. Also, in shellfish samples of oyster, shrimp and crab were ranged from 0.02to 0.41; 0.01 to 0.30 and 0.01 to 0.19 respectively, with mean values of 0.22 ± 0.02;0.15 ± 0.01 and 0.12 ± 0.01, respectively. Moreover; 27 samples, 7C. *gariepinus* (46.7%)*;* 5 *O. niloticus* (33.3%)*;* 2 *M. cephalus* (13.3%)*;* 6oyster (40%)*;* 4shrimp (26.7%) and 3 crabs (20%), were unaccepted, as they were exceeded the maximum permissible limit of cadmium in fish and shellfish that should not exceed 0.05 mg/kg (EOS,2010).

Table (1): Mean values of mercury concentrations "mg/Kg" in the examined samples of fish and shellfish (n=15).

Fish and shellfish species	Min.	Max.	Mean ± SEM*	Unaccepted Samples Maximum Residual Limit (mg/Kg) ** No.	%
Fish:					
C. Gariepinus	0.38	2.45	1.62 ± 0.15	11	73.3
O. Niloticus	0.31	2.07	1.29 ± 0.12	10	66.7
M. Cephalus	0.22	1.51	0.95 ± 0.08	7	46.7
Shellfish:					
Oyster	0.35	2.24	1.37 ± 0.16	9	60
Shrimp	0.26	1.85	1.14 ± 0.10	8	53.3
Crab	0.17	1.33	0.73 ± 0.09	6	40

SEM* = standard error of mean

** Maximum Residual Limit of mercury (0.50 mg/Kg) stipulated by Egyptian Organization of Standardization "EOS" (2010).

Table (2): Mean values of lead concentrations "mg/Kg" in the examined samples of fish and shellfish(n=15).

Fish and shellfish species	Min.	Max.	Mean ± SEM*	Unaccepted Samples Maximum Residual Limit (mg/Kg) ** No.	%
Fish:					
C. Gariepinus	0.12	1.04	0.67 ± 0.09	10	66.7
O. Niloticus	0.09	0.82	0.53 ± 0.06	8	53.3
M. Cephalus	0.05	0.60	0.36 ± 0.05	4	26.7
Shellfish:					
Oyster	0.10	0.89	0.56 ± 0.07	8	53.3
Shrimp	0.08	0.74	0.48 ± 0.05	7	46.7
Crab	0.07	0.68	0.40 ± 0.04	5	33.3

S.E.M* = standard error of mean

** Maximum Residual Limit of lead (0.30 mg/Kg) stipulated by Egyptian Organization of Standardization "EOS" (2010).

Table (3): Mean values of cadmium concentrations "mg/Kg" in the examined samples of fish and shellfish (n=15).

Fish and shellfish species	Min.	Max.	Mean ± SEM*	Unaccepted Samples Maximum Residual Limit (mg/Kg) **	
				No.	%
<u>Fish:</u>					
C. Gariepinus	0.02	0.53	0.25 ± 0.03	7	46.7
O. Niloticus	0.02	0.37	0.19 ± 0.01	5	33.3
M. Cephalus	0.01	0.24	0.14 ± 0.01	2	13.3
<u>Shellfish:</u>					
Oyster	0.02	0.41	0.22 ± 0.02	6	40
Shrimp	0.01	0.30	0.15 ± 0.01	4	26.7
Crab	0.01	0.19	0.12 ± 0.01	3	20

SEM* = standard error of mean

** Maximum Residual Limit of cadmium (0.05 mg/Kg) stipulated by Egyptian Organization of Standardization "EOS" (2010).

4. DISCUSSION

The recorded results for mercury concentrations revealed that, the highest mercury contamination was in *C. gariepinus* followed by oyster; *O. niloticus*; shrimp; *M. cephalus* and finally crab. The results for mercury concentrations in fish samples were nearly similar to that obtained by Love *et al.* (2003) which was with mean level was 1.10mg/kg. But, they were disagreed with those of Stancheva *et al*. (2013) who found the mean mercury residual level in muscle samples of *M. cephalus* fish collected from two different Black sea areas- Varna Lake and Nesebar were (0.08 ± 0.01 and 0.05 ± 0.01) ppm, respectively and with those of and reji *et al*. (2005) who detected higher levels (2.85±1.22 µg/kg, wet weight). Meanwhile, for shellfish samples, the results came in accordance with those obtained by Balfour *et al.*, (2012) who reported that the mercury levels were varied from 86.67 ±2.66 –98.60 ± 12.16%. (µg g−1). But disagreed with those of Falusi and Olanipekun (2007) who detected mercury in shellfish samples with lower concentrations (0.665mg/kg). Moreover, such variations in mercury concentration among the examined fish and fish products may be attributed to variation in feeding habits, and type of sea fish (Morgan, 1999). Mercury is highly toxic to human health; it is a particular threat to the development of child in utero and early life. Methyl mercury affects the kidneys and also the central nervous system, particularly during development, as it crosses both the blood –brain barrier and placenta (Clarkson, 2002).

The poisoning with lead in human leading to brain dysfunction impairment in learning abilities as it affects 3 main neurotransmission systems: dopaminergic, cholinergic and glutaminergic so, it causes neurodegenerative disease. Also, it affects humeral immunity (perhaps through interference with macrophage function), may or may not affect cellular immunity and increases the frequency of chromosomal aberrations (Xu *et al*., 2009). The obtained results for lead concentrations revealed that, the highest lead contamination was in *C. gariepinus* followed by oyster; *O. niloticus*; shrimp; crab and finally *M. Cephalus*. The results for lead concentrations in fish samples were nearly similar to that recorded by Ayeloja *et al.* (2014) who mentioned that, the mean residual level of lead in muscle samples of *Tilapia nilotica* and Catfish collected from Eleyele reservoir Ibadan Oyo State South-western, Nigeria, were (0.393 ± 0.04 and 0.463 ± 0.12) ppm and Badr *et al*. (2014) who evaluated the mean value of lead in muscle samples of *O. niloticus* fish collected from two areas in River Nile which were (0.662 ± 0.058 ppm and 0.833 ± 0.057) ppm. Meanwhile, the results were disagreed with those of Olusola *et al.*, (2012) and Stancheva *et al*., (2013) who detected lead in fish samples with lower concentrations which were 0.039±0.004 ppm and 0.07±0.01 and 0.05±0.002ppm for them, respectively. And with those of Turkmen *et al.*, (2011) who detected higher levels (0.63±0.17 ppm) and with those of Tajiri *et al.*, (2011) and Adeosun *et al.*, (2015) who failed to detect it in all examined samples of fish. In addition, for shellfish samples, the results came in accordance with those obtained by Bat *et al*., (2013); Olgunoğlu *et al*., (2015) and Olgunoğlu and Olgunoğlu (2016) which were0.291-0.491 µg. g-1 wet weight,0.43 µg/g, and 0.02± 0.01 mg/kg for them, respectively. But, these results came in contrast to those of Falusi and Olanipekun (2007) who detected lead in shellfish samples with lower concentrations which were 0.195mg/kg; and those of Vazquez-Boucard *et al*., (2014) who detected higher levels (7.2 to 9.9 µg/g) and those of Olowu *et al.*, (2010) who failed to detect it in all examined samples of shellfish.

The contamination of the environment with heavy metals associated directly with public health hazard due to bioaccumulation and bio magnification nature in food chain (Hussain *et al*., 2012). The obtained results for cadmium concentrations cleared that, the highest cadmium contamination was in *C. gariepinus* followed by oyster; *O. niloticus*; shrimp; *M. cephalus* and finally crab. The results for cadmium concentrations in fish samples were nearly like that recorded by Olusola *et al* (2012); Ayeloja *et al.*, (2014) and Badr *et al.*, (2014) which were 0.020±0.006 ppm,0.123±0.04 ppm and0.024 ± 0.005ppm, respectively. But these results were disagreed with those of Tajiri *et al.* (2011); Stancheva *et al*., (2013) and Shreadah *et al.*, (2015) who detected cadmium in fish samples with lower concentrations (0.011 µg. g^{-1},0.012±0.002 and 0.08 ppm) for them, respectively and with those of Turkmen *et al.,* (2011) who detected higher levels (0.49 ± 0.05 ppm) and with those of Adeosun *et al.,* (2015) who failed to detect it in all examined samples of fish. Meanwhile, for shellfish samples, the results came in accordance with those obtained by Olgunoğlu *et al*., (2015) and Olgunoğlu and Olgunoğlu

(2016) which were144.12±20.42µg/g and0.50±0.02mg/kg, respectively. But, they were disagreed with those of Ayas and Ozogul (2011) who detected cadmium in shellfish samples with lower concentrations with 0.4 mg/kg and with those of Falusi and Olanipekun (2007) who determined the cadmium concentrations crab which were 3.375 mg/kg; Olowu *et al.,* (2010) who investigated the level of cadmium in crabs and prawns and found that, the mean concentration of cadmium was 1.66±1.82 µg/g and 0.07±0.08 µg/g respectively, and Balfour*et et al.*, (2012) who found the cadmium level in shrimp were varied from 95.53 ±0.60-95.77 ±2.06%.

Finally, the present study proved that the highest contamination for fish was in *C. gariepinus* and the lowest one was in *M. cephalus* but for shellfish the highest contamination was in *oyster* and the lowest one was in *crab.* So, these have a public health importance as the heavy metal residual levels (mercury; lead and cadmium) in them, might be exceeded the recommended safe permissible limits stipulated by Egyptian Organization for Standardization (EOS, 2010) and must be controlled to prevent or minimize them and improve the sanitary status of fish and shellfish.

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