

ACCUMULATION OF SOME HEAVY METALS IN DIFFERENT LIBYAN FISH SPECIES FROM FRESH WATER AND MARINE ENVIRONMENTS

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ABSTRACT

In this paper; the concentrations of Pb, Cd, Cr, Zn, Cu, Mn and Fe were determined in the muscle tissues of seven marine and one fresh water fish species. Marine fishes were collected from the Mediterranean sea of the shore of Misurata; while fresh water fishes were collected from Tawarga stream located at about 65 Km west of Misurata according to fish habitat and type of heavy metal itself. Different fish species were collected from different Libyan localities. The results showed that the highest levels of lead, cadmium, chromium, zinc, copper, manganese and iron were found in Bolty (1.64 mg / kg), Bouri (0.07 mg / kg), Sardine (0.93 mg / kg), Denis (5.84 mg / kg), Balameat (0.97 mg/kg), Sardine (0.57 mg / kg), and Maghazel (8.61 mg / kg) fish, respectively. The lowest levels of lead, cadmium , chromium , zinc and copper, were found in Balameat, (0.71 mg / kg), Sardine (0.03 mg / kg), Maghazel (0.06 mg / kg), Bolty (2.1 mg / kg) and Bolty (0.46 mg / kg) fish , respectively. With regard to manganese, the lowest levels were observed in Kawally (0.25 mg / kg), and Maghazel (0.24 mg / kg), whereas Denis and Tuna fish showed lowest levels of iron (1.17 mg / kg).

INTRODUCTION

Pollution as a serious problem facing the world, has received an increasing attention from earlier times. Heavy metals differ from other pollutants in that they are neither created nor destroyed by human . Nevertheless, utilization by human potentially influences health at least in two ways: by environmental transport and by altering the biochemical form of the element (Beijer and Jernelov, 1979 and Li, 1981).

Recently, some aquatic organisms are used as indicators of trace metals pollution. Due to the increasing human activities, the system nowadays, are considered the major receiver of metals from several drains. Heavy metal concentrations are extremely variable in various marine and freshwater organisms (Roperts, 1976 and Forstner and Prosi, 1979) depending on the geochemical background, the level of pollution in a given area, and fish activity (Bryan, 1973 and Ayling, 1974).

Although heavy metals are natural constituents of the aquatic environment, some of these metals are considered most hazardous from the public health aspect considering the characteristics of heavy metal pollution, toxicity to aquatic organisms, concentration within organisms to level greater than in the environment and persistent for long period of time which led to increase the metal concentration (Mcintyre and Mills, 1975 and Swaine, *et al.*, 1981). Some heavy metals such as Zn, Cu, Mn and Fe are essential for the growth and well-being of living organisms. However, they are likely to show toxic effects when organism are exposed to levels higher than normally

required . Other elements : Cd, Hg and Pb are not essential for metabolic activities and exhibit toxic properties. Most published data on the effects of metals on aquatic organisms, however, reported adverse effect at concentration higher than usually found in the environment (GESAMP, 1985).

Monitoring the level of heavy metals in economic fishes in different areas has high importance for the public health (Swaine *et al.*, 1981). Therefore, the objective of this work is to study the effect of fish type on the level of some heavy metal in some Libyan fishes with special references to its locality where these fishes normally grow which may shade some light on the type of certain locality.

MATERIALS AND METHODS

Materials

Samples collection

A total of 81 composites fish samples representing eight different fish type (Table 1) were collected from two different aquatic environments, the Mediterranean and Tawarga pond as well as fresh water environment. Samples were collected at random through the period from December 2009 to December 2010. All collected samples were analyzed to assess the level of heavy metals. The total each sample weighed 5 kg from which 1 kg were selected where the flesh was separated from the parts of the fish and homogenized to obtain 1 composite sample.

standard solutions:

Stock standard solutions of lead (Pb), cadmium (Cd), chromium (Cu), zinc (Zn), copper (Cu), manganese (Mn), and iron (Fe), were obtained from Merck company at concentrations of 1000 mg/l (Merck , Darmstadt, Germany).

Methodology

1- Fish samples were prepared by using the method described in Association Official Analytical Chemists (AOAC, 1980) and U. S. Environmental protection Agency (USEPA, 1978) guideline.

2- Analytical methods

Perkin - Elmer 2380 Atomic absorption spectrophotometer was employed for the analysis. The maximum absorbance was obtained by adjusting the cathode lamps at specific slit and wave lengths.

3- Statistical Analysis

To detect the significance of differences of the tested heavy metals among the different types of collected fish samples, the data were analyzed using one way analysis of variance (ANOVA) according to the following model (Winer, 1971).

$$Y_{ij} = \mu + a_1 + \Sigma_{ij}$$

Were Y_{ij} : observation of element j under effect of a_1 .

μ : general population mean.

a_1 : fish type effect.

Σ_{ij} : experimental error.

General Linear Model of SAS (SAS, 1988) was used to perform the Analysis of variance. Duncan Multiple Range Test was used for means separation (Winer, 1971).

RESULTS AND DISCUSSION

Concentrations of Mn, Cu, Cd, Cr, Fe, Zn and Pb were determined in eight of more popular species of fish in Libya. Fish samples were collected from the Mediterranean sea and Tawarga pond. All determinations were carried out in fish flesh, being the most concern to man as it is the main tissue consumed as food. The different metals detected among the eight species of fish analyzed is given in Tables (2 – 3).

Lead

The results showed in Table (2) indicate that the mean level of lead in fresh water fishes (Bolty) and marine water fishes (Sardine) recorded higher mean value (1.64 and 1.18 mg/kg) and they were higher than those in marine fishes except for Tuna which recorded 1.55 mg/kg. The lowest mean level was found in Balameat, (0.71 mg/kg) also Denis of the Mediterranean recorded the second lower value. This result indicated that the effect of fish type is more important than the locality effect especially when no major pollution in particular area occurred. These mean values are within the maximum permissible limits for pb, being 2 mg/kg (USFDA, 1974).

The obtained results of the level of lead in Bolty collected from fresh water being 1.64 mg/kg which was higher than the level reported by Allam *et al.* (1993a) in Bolty collected from Tawarga pond which was 0.55 mg/kg wet weight.

Sardine in the current study contained a mean level of lead of 1.18 mg/kg wet weight which was higher than the level reported by Lowe *et al.*, 1985 (mean 0.51 mg/kg wet weight).

With regard to Egyptian environment, Emara *et al.* (1993) reported lower value of lead (0.53 mg/kg) in Balameat than that reported in the present study (0.71 mg/kg). Also Abdel-Moneim *et al.* (1994) reported lower value of lead (0.54 mg/kg) in Tuna than that detected in this study (1.55 mg/kg). Denis (Mediterranean) in the current study contained lower level of lead (0.81 mg/kg) than that recorded by Allam *et al.* (1993b) which was 1.7 mg/kg.

Concerning pb mean level in Bouri that reached 1.03 mg/kg wet weight in this study, was higher than those recorded by Emara *et al.* (1993) (0.75 mg/kg wet weight), Abdel-Moneim *et al.* (1994) (0.54 mg/kg) and Lowe *et al.* (1985) (0.16 mg/kg) and it was lower than that recorded by Allam *et al.* (1993) in Faiyum Bouri (3.2) and Abdel-Salam, (1981) (7.14 mg/kg). The lowest level was recorded in the study of Mendoza-Diaz *et al.* 2013 and Chahid *et al.* (2014) who found lead at maximum residual levels.

Cadmium

Table (2) showed that the highest mean level of Cd was detected in Bouri which was 0.07 mg/kg wet wt, while the lowest mean level was detected in Sardine (0.03 mg/kg). However, no significant differences in cd

concentrations were detected among all fish types. These values are lower than the maximum permissible limits for Cd being 0.5 mg/kg (USFDA, 1974).

The current results of Cd in Bolty (0.04 mg/kg) was similar to that recorded by Khidr *et al.* (2008) (0.03 mg/kg).

While the level in Sardine in this study (0.03 mg/kg) was coincided with Lowe *et al.*, (1985) and lower than that by Khidr *et al.* (2008) (0.14 mg/kg).

Higher value of Cd was reported in Tuna (0.45) by Abdel-Moneim., *et al.* (1994) and in Balameat (0.17 mg/kg, Emara *et al.*, 1993) than that reported in the present study (0.04 mg/kg). Cd mean level in Bouri (0.07 mg/kg) in this study was higher than that reported by Lowe *et al.* (1985) (0.01 mg/kg) and lower than of Abdel-Moneim., *et al.* (1994) (0.54 mg/kg) Cd was found upper the safe limits in tissue of *Cyprinus carpio* fish by Javed and Usmani 2013 (0.3 mg/kg) .

Chromium

The maximum mean value of Cr (0.93 mg/kg wet wt) was recorded in Sardine (0.7 mg/kg) Mendoza-Diaz *et al.* (2013) while the minimum (0.06 mg/kg) was recorded in Maghazel. However, no significant differences were detected among Sardine, Kawally and Tuna while significant difference was detected between Sardine and Maghazel (Table 2). Also, the variations among fish types were more pronounced than the variations among fishes of Mediterranean environment which contained the lowest mean level (Maghazel) and one of the highest mean level (Tuna) and (Sardine) and safe limits was found in *Cyprinus carpio* fish reported by Iqbal and Shah (2013).

Zinc

In Table (2), the highest mean level of Zn (5.84 mg/kg) was recorded in Denis whereas the lowest mean was in Bolty (2.10 mg/kg). No significant difference was detected between Denis and Maghazel as well as among Bolty, Tuna, and Kawally.

Zn mean level in this study in Sardine (3.46 mg/kg) was lower than that of Lowe *et al.* (1985) (59.7 mg/kg) and close to that reported by Allam *et al.* (1993b) (2.0 mg/kg). Zn level in Bolty (2.1 mg/kg) was also close to that of Allam *et al.* (1993a) (4.64 mg/kg).

Regard to Balameat, Zn level in this study was 3.81 mg/kg which is similar to level reported by Emara *et al.* (1993) (3.37 mg/kg) and El-Deek, *et al.* (1994) (4.57 mg/kg). Although Zn in Denis was the highest level in the current study (5.84 mg/kg) but it was still lower than that recorded by Allam *et al.* (1993) (9.0 mg/kg).

Bouri in this study contained Zn level of 4.84 mg/kg which is similar to that reported by Allam *et al.* (1993b) in cultures Bouri (3.35 mg/kg) and Faiyum Bouri (5.94 mg/kg); where it was lower than that noticed by Abdel-Moneim *et al.* (1994) (12.28 mg/kg), Lowe *et al.* (1985) (13.7 mg/kg) and Abdel-salam (1981) (15.56 mg/kg), the most lower level was found in some atlantic sea fishes (0.3 mg/kg) as registered by Javed and Usmani (2013) and safe limits was found in *Cyprinus carpio* fish as in Iqbal and Shah study (2013).

Copper

Cu concentration was more pronounced in Balameat (0.97 mg/kg) where the lowest mean level (0.46 mg/kg) was in Bolty. Table (3) showed that variation was clear for Cu concentration, where the highest levels were detected in the Mediterranean fish types (Balameat and Tuna) and the lowest level was in fresh water types (Bolty).

Level of Cu in the current study in Sardine (0.52 mg/kg) was lower than that reported by Lowe *et al.* (1985) (1.33 mg/kg) and Allam *et al.* (1993a) in cultures carp (0.05 mg/kg).

Cu in Balameat in this study (0.97 mg/kg) was higher than the levels reported by Emara *et al.* (1993) (0.59 mg/kg) and El-Deek, *et al.* (1994) (0.55 mg/kg). Also higher value of Cu in Denis (0.68 mg/kg) was recorded in this study compared with that of Allam *et al.* (1993b) (0.15).

Abdel-Salam, (1981) reported higher value of Cu in Bouri (6.75 mg/kg) than what reported in the current study (0.77 mg/kg). While it was similar to that noticed by Abdel-Moneim *et al.* (1994) (0.59 mg/kg) and it was higher than that of Allam *et al.* (1993b) in cultured Bouri (0.08 mg/kg) and Faiyum Bouri (0.20 mg/kg). Safe limits were found in *Cyprins carpio* fish as in Iqbal and Shah (2013).

Manganese

Although Sardine recorded the highest value of Mn (0.57 mg/kg), while Balameat, Kawally, and Maghazel recorded the lowest level (0.24 mg/kg), however no significant differences were detected among all fish types (Table 3).

Iron

Maghazel recorded the highest mean level of Fe (8.61 mg/kg wet wt) where the lowest was in Denis and Tuna (1.17 mg/kg). Significant differences were detected between the highest and lowest mean levels. Allam *et al.* (1993a,b) reported similar levels of Fe in Bolty (1.79 mg/kg) and in Denis (1.08 mg/kg) to those of the current study (1.75 mg/kg) and (1.17 mg/kg) respectively where in cultured Bouri (0.71 mg/kg) and Faiyum Bouri (0.59 mg/kg) the levels were lower than that of the this study (2.03 mg/kg).

Allam *et al.* (1993a,b) reported levels of Fe in Bolty (1.79 mg/kg) and Denis (1.08 mg/kg) which was similar to those found in the preset study which were (1.75 mg/kg) and (0.17 mg/kg) respectively, where in cultures Bouri (0.71 mg/kg) and Faiyum Bouri (0.59 mg/kg).

From the fore mentioned presentation, it can be concluded that the mean values of pb, Cd and Cr reported by Hernandez *et al.* (1990) from Spain (Mediterranean sea) are extremely higher than those reported in the current study which is due to that Mediterranean sea is comparatively subjected to heavy discharges of pollutants from numerous industrial process on the Spanish shores Programme Des Nations Unies Pour L'Environnement (1984). On the other hand, the given values by Ramelow *et at.*, (1989); and those by Allam *et al.* (1993) for Zn, by Winger, *et al.*, (1990); Ramelow *et al.*, (1989) and Lowe *et al.*, (1979) for Cu, Mn, Cd; and by Eisenberg *et al.*, (1986) for Cr were close to those reported in the current study. In the same regard, the maximum and minimum mean values given in this study

were lower than those reported by Eisenberg *et al.* (1986); Dallinger, (1985); Lowe *et al.*, (1985); Wiener and Giesy, (1979); for Zn; by Eisenberg *et al.*, (1986); by Dallinger, (1985) for Cu; by Dallinger, (1985); Wiener and Giesy, (1979); for Mn and by Dallinger, (1985) for Cr. Higher values of Pb was recorded in this study than these levels reported by Winger, *et al.* (1990) and Ramelow *et al.* (1989).

The different species of fish under investigation represent both Mediterranean and the fresh water (Tawarga pond) environments. The presence of metal in both environments is partly due to natural processes such as corrosion but is mostly the result of industrial process. The Mediterranean sea and Tawarga pond are subjected to heavy discharges of pollutants from numerous industrial process. Among these industries are leather tanning and metallic transformation in Tawarga pond as well as oil refineries petroleum and organic chemical industries in Mediterranean.

The role of the environment on the incidence of metals in fish clearly shows the maximum concentrations of the present data Tables (2-3) of Cr (0.93 mg/kg), Pb (1.64 mg/kg), and Mn (0.57 mg/kg) which were detected in fresh water fish (Tawarga pond) while the marine fish from the Mediterranean sea show the highest mean values of Zn (5.84 mg/kg), Fe (86.11 mg/kg) and Cu (0.97 mg/kg). The maximum Cd levels (2.1 mg/kg) were detected in Mediterranean fish type samples. From this presentation, it is clear that the maximum concentrations of the majority of heavy metals detected were shown in the marine fish which may due to its higher bioaccumulation ability which generally depends on exposure time and metal concentration in water. Therefore, metal level in marine fish organisms can reach higher values than those of the fresh water (Bryan, 1984 and Mance, 1987).

Accumulation patterns of heavy metals in fish are dependent on both uptake and elimination rates. The uptake of metals is influenced by many factors including fish species, type of fish organ studied and various environment factors (Badsha and Goldspink, 1982 and Hakanson, 1984). In the regard, gills and guts are involved in the uptake of heavy metals by fish (Segner and Back, 1985), gills playing an important role in the uptake of dissolved elements (Hughes and Flos, 1976), whereas particulated metal fractions can only, if at all be absorbed through the alimentary tract. On the other hand, it was reported that the passage of metal along the specific food chain into a fish is hard to demonstrate. The reason for these are that in most cases it is difficult to distinguish between uptake via the alimentary tract and absorption through the gills (Dallinger and Kautzky, 1985). Also, the elimination of metals is an active biochemical and physiological process (Barak and Mason, 1990). In this respect, it is worthy to report that some metals, are very strong binding (permanent strong), and that strong bound metals in fish flesh are not easily influenced by environment changes (Boyden, 1977).

Table 1: Different fish species analyzed for the presence of heavy metals

Common name	English name	Scientific name	Areas	Number of samples
Sardine	Sardines	<i>Clupeidae spp</i>	Mediterranean sea	11
Bolty	Tilapia	<i>Tilapia zillii</i>	Tawarga pond	12
Balameat	Plain bonito	<i>Orcynopsis unicolor</i>	Mediterranean sea	8
Maghazel	Barracuda	<i>Sphyraena spp</i>	Mediterranean sea	10
Tuna	Tuna	<i>Scombridae spp</i>	Mediterranean sea	8
Denis	Sea bream	<i>Sparus aurata</i>	Mediterranean sea	10
Kawally	Chub mackerel	<i>Scomber japonicus</i>	Mediterranean sea	11
Bouri	Leaping mullet	<i>Liza saliens</i>	Mediterranean sea	11

Table 2: The concentrations of lead, cadmium, chromium, and zinc in fish samples collected from different localities.

Name	Concentration (mg/Kg wet weight)							
	Lead		Cadmium		Chromium		Zinc	
	Range	Mean ±S.E	Range	Mean ±S.E	Range	Mean ±S.E	Range	Mean ±S.E
Balameat	0.03- 2.09	0.71± 0.19	0.2-0.09	0.04±0.007	0.05-0.79	0.24±0.09	0.06-6.56	3.81±0.55
Bolty	0.37-2.77	1.64±0.32	0.02-0.07	0.04±0.007	0.01-1.36	0.43±0.17	0.20-5.72	2.10±0.66
Bouri	0.04-2.69	1.03±0.29	0.01-0.23	0.07±0.020	0.05-1.52	0.62±0.18	0.24-9.78	4.84±1.27
Denis	0.02-2.96	0.81±0.36	0.01-0.09	0.05±0.012	0.02-1.51	0.35±0.15	0.43-14.16	5.84±2.67
Kawally	0.07-2.55	0.97±0.38	0.01-0.08	0.04±0.010	0.07-1.51	0.89±0.20	0.06-5.31	2.88±0.60
Sardine	0.48-2.57	1.18±0.48	0.01-0.06	0.03±0.009	0.74-1.16	0.93±0.12	1.30-4.30	3.46±0.73
Maghazel	0.13-257	1.00±0.48	0.01-0.07	0.04±0.010	0.01-0.13	0.06±0.05	1.02-8.05	5.15±1.11
Tuna	0.53-2.49	1.55±0.25	0.03-0.05	0.04±0.004	0.08-1.79	0.81±0.21	0.14-4.47	2.35±0.56

Table 3: The concentrations of Copper, Manganese, and Iron in fish samples collected from different localities.

Common name	Concentration (mg/Kg wet weight)					
	Copper		Manganese		Iron	
	Range	Mean ±S.E	Range	Mean ±S.E	Range	Mean ±S.E
Balameat	0.17-4.10	0.97±0.34	0.02-0.80	0.24±0.09	1.13-4.37	2.02±0.79
Bolty	0.03-0.95	0.46±0.09	0.02-0.11	0.38±0.12	0.11-3.08	1.75±0.33
Bouri	0.1-1.47	0.77±0.2	0.04-0.79	0.52±0.10	0.21-3.69	2.03±0.45
Denis	0.42-1.31	0.68±0.16	0.01-0.81	0.34±0.10	0.03-3.09	1.17±0.36
Kawally	0.19-1.58	0.70±0.16	0.02-0.76	0.25±0.08	0.33-2.26	1.34±0.33
Sardine	0.14-0.84	0.52±0.15	0.42-0.76	0.57±0.10	0.56-3.28	1.58±0.60
Maghazel	0.63-1.03	0.76±0.08	0.02-0.45	0.24±0.22	ND-13.31	8.61±3.08*
Tuna	0.08-1.73	0.84±0.27	0.04-0.88	0.35±0.13	0.61-2.22	1.17±0.28

ND: Not Detected

* : Significant

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تراكم بعض المعادن الثقيلة في أنواع مختلفة من الأسماك الليبية في كل من بيئة المياه العذبة والبيئة البحرية
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أصبح تراكم المعادن الثقيلة في الأسماك لدرجة قد تكون سامة للإنسان يمثل تهديدا متزايدا ومرتبيا بزيادة التخلص من النفايات الصناعية في النظام المائي. ويختلف نوع السمك في مدى القابلية التراكمية ومعدل التراكم في وحدة الزمن باختلاف البيئة المحيطة وطريقة التغذية الخاصة بكل نوع وكذلك باختلاف المعادن الثقيلة وصورة تواجدها في النظام المائي.

وقد قامت هذه الدراسة علي تجميع أنواع مختلفة من الأسماك من البيئة البحرية (البحر الابيض المتوسط) والبيئة المياه العذبة من (بحيرة تاورغاء) ثم دراسة تأثير نوع السمك ونوع البيئة المحيطة على مستوى تواجد المعادن الثقيلة في لحوم الأسماك العينات المجمعة.

وقد أظهرت النتائج أن أعلى مستوى للرصاص كان في اسماك البلطي (١.٦٤ مجم/كجم) وأدنى مستوى له كان في اسماك البلامييط (٠.٧١ مجم/كجم). وكان أعلى مستوى للكاديوم في اسماك البوري (٠.٠٧ مجم/كجم) وكان أدنى مستوى له في أسماك السردين (٠.٠٣ مجم/كجم). وكان أعلى مستوى للكروميوم في اسماك السردين ٠.٩٣ مجم/كجم وأدنى مستوى له كان في اسماك المغازل (٠.٠٦ مجم/كجم). وكان أعلى مستوى للزنك في اسماك الدنيس (٥.٨٤ مجم/كجم) وأدنى مستوى له كان في اسماك البلطي (٢.١ مجم/كجم). أما أعلى مستوى للنحاس كان في اسماك البلامييط والكواللي والمغازل بمستويات ٠.٢٤ ، ٠.٢٥ و ٠.٢٤ مجم/كجم على الترتيب. وكان أعلى مستوى للحديد في أسماك المغازل (٨.٦١ مجم/كجم) وأدنى مستوى له كان في اسماك الدنيس والتونة (بمستوى ١.١٧ مجم/كجم).



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