HEAVY METAL CONCENTRATIONS IN TISSUES OF EDIBLE FISH (*MULLUS BARBATUS* L., 1758) FROM THE ÇANDARLI BAY (TURKEY)

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ABSTRACT

Heavy metal contents were investigated in different organ tissues of *Mullus barbatus* (L., 1758) from Çandarlı Bay in the northeast Agean Sea in May 2004. Muscle, liver and gill tissue were analyzed for copper, lead, chromium, zinc and iron by Atomic Absorption Spectrophotometer (AAS). The concentrations of copper in the muscle tissue ranged between 0.11 and 1.25, in liver 0.62 – 2.08, in gill 0.32 – 1.26 ; chromium in muscle 4.21 - 9.02, in gill 10.49 – 17.49, in liver 15.50 – 26.37 ; lead in muscle 1.20 - 9.74, in gill 3.18 - 11.82, in liver 5.30 - 12.52; zinc in muscle 0.36 - 0.97, in gill 1.16 - 2.57, in liver 2.08 - 6.19; iron in muscle 4.16 - 9.32, in gill 8.23 - 25.63, and in liver 5.43-58.45 ($\mu g\ g^{-1}$ wet weight). The order of the metal concentrations found in *M. barbatus* was Cu < Zn < Pb < Cr < Fe. According to statistical analysis, there were no significant relationship between individual lengths and weights of *Mullus barbatus* and bioaccumulation of heavy metals (ANOVA, $p > 0.05$). In this study, the mean values of Pb and Cr in muscle tissues were exceeded the legislation limits of FAO (Food and Agriculture Organization of the United Nations) and MARA (T.C. Ministry of Agriculture and Rural Affairs). In addition, Pb and Cr values in excess of permissible limits may be harmful to human health.

KEYWORDS:
Heavy metals, Fish, Çandarlı Bay, Turkey

1. INTRODUCTION

Metals are introduced into the aquatic ecosystems such as lakes, rivers and seas in many ways. They may be accumulated by aquatic organisms such as fish and may be a potential risk to health of ecosystems and organisms. The risk carried on humans in terms of heavy metal toxicity is determined by analyzing metal concentrations in the most consumed and high economic value species [1, 2]. Fish is widely used as monitoring organisms in the aquatic environment. Metals can be taken up by fish from water, food, sediment and by suspended particulate material [3]. They may also be accumulated via the general body surface, across the gills and through the digestive system, to be then carried to tissues and organs by the blood circulatory system to bind on carrier proteins. They can reach high concentrations as a consequence of being connected to metal binding proteins [4]. Some heavy metals, especially cadmium, mercury, lead and chromium, can penetrate a living organism via the food chain and can accumulate. Then, due to excessive concentrations in the body structure, they cannot be discharged by natural physiological mechanisms producing toxic effects [5]. Gill, liver and muscle were chosen as target organs for assessing metal accumulation. The concentrations of metals in gills reflect those in their surrounding waters. The metal content in the dorsal muscle was analyzed, because of its importance related to human consumption, and the liver was analyzed as this organ has a tendency to accumulate metals [6].

Metal concentrations have previously been investigated for many organisms in the Aegean Sea. [7-16] in selected marine fishes. In addition, target organs, such as liver, gonads, kidney and gills, have a tendency to accumulate heavy metals in high values, as shown in many species of fish in different areas: in *M. barbatus* and *Sparus aurata* in the İskenderun Gulf [17] in *M. barbatus* in the Eastern Adriatic Sea [18], in *M. barbatus* and *Mullus surmuletus* in the Izmir Bay [19], in *Thunnus thynnus* in Antalya Gulf [20], in *Liza ramada* in Mersin Gulf [21], in *Sardinella aurita*, *L. ramada*, *Diplodus annularis*, *Solea lutea* and *Umbrina cirrosa* in Mersin Gulf [22], in *M. barbatus*, *Mugil cephalus*, *Trachurus trachurus*, *Pagellus acarne*, *Dicentrarchus labrax*, *Sparus auratus*, *S. aurita*, *Boops*

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boops, *Scomber japonicus*, *Solea solea* in Antalya Bay [23], *Mugil capito, Siganus rivulatus, S. auratus* and *Sardina pilchardus* in Mediterranean Sea [24], in *S. auratus, Atherina hepsetus, M. cephalus, Trigla cuculus, S. pilchardus* and *Scomberesox saurus* in the northeast Mediterranean Sea [25]. It is generally accepted that muscle is not an organ in which metals accumulate [17, 26-28]. Similar results have been reported for a number of fish species showing that muscle is not an active tissue in accumulating heavy metals [17, 29]. Analyzes are also needed for muscle tissues, as metals are consumed by humans and carry health risks [30].

Çandarlı Bay is an important area for the trawling in the Agean Sea where the red mullet fishery is well developed. This bay has been heavily impacted by man-made modifications and is surrounded by urbanization, industrial and agricultural zones. In addition, this region contains many petrochemical industries, as well as 10 factories in metal production. Seven companies are involved in fertilizers, paper, mining and chemicals, and 15 businesses are engaged in shipyard repairs [31]. Industry is more developed in the Aliağa district. The Bakırçay River which flows into the Çandarlı Bay has no reliable canalization or sewage treatment in the residential zones of its passageway. Thus, these untreated waters are distributed to the river and its branches [32]. The aim of this study was to determine heavy metal (Cu, Pb, Cr, Zn and Fe) concentrations in the muscle, gill and liver of *Mullus barbatus* (L.1758) from four trawl areas (T1, T2, T3 and T4) in the northeast Agean Sea.

2. MATERIALS AND METHODS

The red mullet *Mullus barbatus* (18.00±0.36 cm and 65.02±4.02 g) used in this study were sampled at four stations (Table 1) in the Çandarlı Bay in the northern part of the Aegean Sea in May 2004 (Figure 1). Twenty fish were collected from each station, and then brought to the laboratory on ice, at the same day. Total size and weight of the animals were measured and their sex determined. They were then kept freezing at -20 ºC until analysis. The numbers of samples and main biometric parameters are represented in Table 2. The gill, liver and muscle tissues of the animals were dissected using clean stainless steel equipment. Then all tissues were pooled, mixed and homogenized. The triplicate analysis was conducted for all tissues and each sampling location. The tissues homogenates were prepared according to international standard methods. The composite samples of each tissue were weighed and digested with conc. HNO_3: $HClO_4$ (5:1) (extra pure Merck) under reflux and filtered [33-35]. Metal samples were analyzed by Atomic Absorption Spectrophotometer using a SOLAR-UNICAM 929 (AAS). The detection limits of the AAS were 0.01, 0.001, 0.125, 0.05 and 0.05 μg g^{-1} wet weight for Cu, Zn, Pb, Cr and Fe, respectively [36].

TABLE 1 - Coordinates and depths of trawling zones

Trawling Stations	Initial-final coordinates	Depths (m)	Bottom
1	38º54´71″N 26º53´41″E 38º55´06″N 26º55´02″E	61-54	muddy-sandy
2	38º54´48″N 27º01´17″E 38º54´09″N 26º59´40″E	36-47	muddy-sandy
3	38º53´08″N 26º57´90″E 38º52´06″N 26º56´36″E	56-58	muddy-sandy
4	38º47´63″N 26º52´68″E 38º44´30″N 26º52´08″E	94-92	muddy-sandy

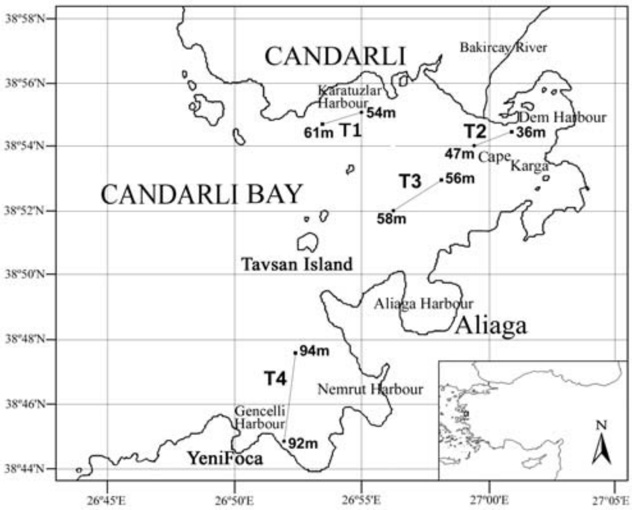

FIGURE 1 - The map of sampling locations in Çandarlı Bay, Turkey.

TABLE 2 - Main biometric parameters of *Mullus barbatus.*

Locations	Total length (cm)		Body weight (g)		Sex	
	Mean±SD	Range	Mean±SD	Range	♂	♀
T1	18.85±0.42	17-22	71.41±5.27	17-137	-	20
T2	15.01±0.40	12-17	36.61±3.43	17-58	1	19
T3	20.08±0.40	17-23	88.38±6.57	57-137	-	20
T4	18.13±0.59	16-21	65.90±7.83	36-111	2	18

Analysis of variance (one-way ANOVA, $p<0.05$) was performed to evaluate the differences in *Mullus barbatus* between stations, total length and total weight for each heavy metal. In order to find the linear relationship between fish length, fish weight and heavy metal concentration, Pearson's correlation coefficient (*r*) was conducted. All statistical analysis was performed according to Zar [37].

3. RESULTS AND DISCUSSION

3.1. Physico-chemical water quality results

The variability in metal concentrations of marine organisms depends on many factors, either environmental (metal concentrations in seawater, temperature, salinity, dissolved oxygen, pH, etc.) or purely biological (species, tissues, organs, feeding conditions, etc.) [38]. Average values of some physico-chemical environmental parameters, which are related to trace metal accumulation from four different locations in the Çandarlı Bay, are presented in Table 3.

3.2. Heavy metal results

The minimum, maximum and mean concentrations of heavy metals for each organ collected from four different locations along the Çandarlı Bay are summarized in Table 4. The order of the metal concentrations found in *M. barbatus* was Cu < Zn < Pb < Cr < Fe. The minimum and maximum copper concentrations varied from 0.11 to 1.25 µg g^{-1} wet weight (ww) in the muscle, from 0.32 to 1.26 µg g^{-1} ww in the gill, with the highest level at station T3, and from 0.62 to 2.08 µg g^{-1} ww in the liver, with the highest level at station T1. Copper concentrations vary significantly ($p<0.05$) in the gill and liver tissues, although copper concentrations did not vary significantly ($p>0.05$) in the muscle tissues of the *Mullus barbatus* in all locations (Table 4).

Several authors have noted the concentrations of heavy metals in fish vary with the length of the fish [38, 39]. However, no relationship was observed between individual lengths and weights of *Mullus barbatus* and bioaccumulation of heavy metals in this study ($p>0.05$).

Various researchers have addressed measurements of Cu, Zn, Pb, Cr and Fe in fish from different regions of the world, and some of these data are summarized in Table 5. There were differences between metal concentration in this study and those of previous studies. Copper concentrations in muscle tissues found in this study were higher than those of the Aegean Sea [14] and Izmir Bay [15, 19, 42], but lower than those of İskenderun Bay [43, 45] (Table 5). Lead concentrations in muscle tissues were higher than all other studies but showed similar values in the study reported in Izmir Bay [42] (Table 5). Zinc concentrations varied depending on the study areas. Zn levels detected in all previous researches were found to be much lower in all types of tissues compared with this study, except with the maximum 2.30 µg g^{-1} ww of zinc in the Eastern Adriatic Sea [18]. Cr concentrations have been less studied when compared to other metals in *M. barbatus*. Chromium concentrations found in this study were higher than those reported by [19, 43, 46] in liver and muscle tissues (Table 5). However, Fe concentrations in the liver and gill tissues were lower than those reported in previous literature, but those in the muscle tissues showed more or less similar values to those studies done in the Izmir Bay [15, 19, 42] (Table 5). Concentrations of heavy metals detected in various tissues were classified as follows: liver > gill > muscle. The reason for lower heavy metal concentrations in muscle tissue than those of gill and liver is related to lower metabolic activities of muscle. Higher values found in the liver are related to its role as a storage organ [48]. According to the declaration of trace heavy metals in Fisheries and Microbiological Control, the permissible tolerance limits of Pb, Zn and Cu in fresh fish are 1, 50 and 20 µg g^{-1} ww, respectively [49]. In addition, FAO Legislation limits are 0.5 µg g^{-1} for Pb, 30 µg $g^{-1}$for Zn, 30 µg g^{-1} for Cu and 1 µg g^{-1} for Cr [24]. In this study, the mean values in muscle tissues were found to be 5.79 µg g^{-1} ww for lead and 0.56 µg g^{-1} ww for zinc; 0.54 µg g^{-1} ww for copper and 6.30 µg g^{-1} ww for chromium. The mean values of Pb

TABLE 3 - Average values of some physico-chemical environmental parameters from Çandarlı Bay.

Locations	Water temperatures (ºC)		Dissolved oxygen (mg/l)		pH		Salinity (‰)	
	2003	2004	2003	2004	2003	2004	2003	2004
T1	20.1	18.1	8.30	7.74	7.45	7.43	38.35	36.03
T2	20.2	18.3	8.30	8.12	7.47	7.42	38.13	36.17
T3	19.9	19.7	8.19	8.13	7.49	7.41	37.67	36.32
T4	20.0	19.7	8.31	8.16	7.48	7.43	38.37	36.80

TABLE 4 - Minimum, maximum and mean values of the metal concentrations in different organs of *Mullus barbatus* (µg g^{-1} wet weight).

Tissue	Station	Cu		Zn		Pb		Cr		Fe	
		Min.-Max.	Mean	Min.-Max.	Mean	Min.-Max.	Mean	Min.-Max.	Mean	Min.-Max.	Mean
Muscle	T1	0.44-0.64	0.54^{a*}	0.38-0.51	0.44^{a}	5.87-7.98	6.79^{ab}	5.42-6.76	6.27^{a}	4.16-8.70	6.98^{a}
	T2	0.16-0.35	0.26^{a}	0.47-0.97	0.63^{a}	5.25-9.74	8.07^{a}	5.73-9.02	6.62^{a}	6.28-8.94	7.88^{a}
	T3	0.12-1.25	0.79^{a}	0.38-0.78	0.60^{a}	4.66-7.66	5.98^{b}	4.42-8.74	6.82^{a}	5.23-9.32	7.06^{a}
	T4	0.11-1.11	0.56^{a}	0.36-0.84	0.55^{a}	1.20-2.76	2.30^{c}	4.21-7.90	5.49^{a}	4.71-7.93	6.13^{a}
Gill	T1	0.65-0.99	0.78^{b}	1.16-1.93	1.72^{a}	8.41-9.59	9.17^{c}	11.22-17.49	14.23^{ab}	11.06-17.37	13.90^{bc}
	T2	0.32-0.65	0.46^{c}	1.40-2.57	1.98^{a}	10.43-10.97	10.57^{b}	14.15-17.28	16.07^{a}	8.23-12.36	10.63^{c}
	T3	1.17-1.26	1.23^{a}	1.96-2.24	2.14^{a}	11.54-11.82	11.66^{a}	11.45-16.19	13.38^{b}	16.06-25.63	19.57^{a}
	T4	1.11-1.12	1.12^{a}	2.01-2.39	2.18^{a}	3.18- 4.86	3.97^{d}	10.49-13.36	11.71^{b}	12.17-16.95	15.06^{b}
Liver	T1	1.03-2.08	1.59^{a}	2.08-6.19	3.72^{ab}	10.33-11.34	10.83^{a}	21.64-25.53	23.26^{a}	19.76-27.36	22.46^{bc}
	T2	0.62-0.73	0.67^{b}	3.36-5.64	3.92^{a}	10.79-12.52	11.59^{ab}	19.28-24.12	21.66^{a}	15.41-21.34	18.52^{c}
	T3	1.28-1.59	1.41^{a}	3.25-6.08	4.27^{b}	12.14-12.52	12.27^{a}	17.42-26.37	21.87^{a}	26.06-58.45	36.17^{ab}
	T4	1.49-1.71	1.59^{a}	2.44-6.08	3.72^{c}	5.30 - 6.53	5.83^{c}	15.50-17.93	16.54^{b}	22.15-46.06	36.39^{a}

* Data shown with different letters are statistically significant at the $P<0.05$

TABLE 5 - Levels of heavy metals in *Mullus barbatus* from the different regions of the world (µg g^{-1} wet weight).

Tissue	Area	Cu	Pb	Zn	Cr	Fe	Reference
Liver	Agean Sea, 1999	1.30-6.75	0.40-1.80	18.50-62.50	-	-	[14]
	Northeast Mediterranean, 1999	-	-	35.3-55.3	-	135.0-258.0	[41]
	Izmir Bay, 2001	1.10-3.20	2.40-9.43	42.00-62.25	-	44.00-114.71	[42]
	Eastern Adriatic Sea, 2007	0.15-0.68	-	0.28-2.30	-	0.35-1.68	[18]
	Turkish Seas, 2008	1.11-26.7	0.66-5.20	17.4-34.9	0.20-1.79	49.9-328.0	[46]
	Çandarlı Bay	0.62-2.08	5.30-12.52	2.08-6.19	15.5-26.37	15.41-58.45	This study
Gill	Agean Sea, 1999	0.70-1.90	0.70-3.05	11.00-99.00	-	-	[14]
	Northeast Mediterranean, 1999	-	-	30.5-57.0	-	263.0-299.0	[41]
	Izmir Bay, 2001	0.49-1.05	3.83-11.44	19.00-34.89	-	90.85-170.11	[42]
	Çandarlı Bay	0.32-1.26	3.18-11.82	1.16-2.57	10.49-17.49	8.23-25.63	This study
Muscle	Izmir Bay, 1982	0.48	2.17	3.02	1.08	12.02	[19]
	Agean Sea, 1999	0.03-0.50	0.07-0.85	5.00-17.00	-	-	[14]
	Northeast Mediterranean, 1999	-	-	16.1-25.8	-	32.2-103.1	[41]
	Izmir Bay, 2001	0.11-0.48	0.80-2.55	6.59-10.99	-	2.12-10.83	[42]
	Agean Sea, 2001	-	0.04-0.20	-	-	-	[11]
	Izmir Bay, 2002	-	0.01-0.91	-	-	-	[12]
	Izmir Bay, 2004	0.11-0.50	0.80-2.60	6.59-11.21	-	2.12-13.25	[15]
	Iskenderun Bay, 2005	0.24-5.43	0.32-4.83	0.72-9.85	0.69-6.46	2.45-17.92	[43]
	Iberian Peninsula Coast, 2007	0.33-0.44	0.03-0.12	3.30-5.06	-	-	[44]
	Iskenderun Bay, 2008	1.10	-	18.23	-	42.93	[45]
	Turkish Seas, 2008	0.57-5.06	0.13-1.00	5.73-12.9	0.13-0.27	21.9-160.0	[46]
	Black Sea, 2009	-	0.515-0.081	-	-	-	[47]
	Çandarlı Bay	0.11-1.25	1.20- 9.74	0.36- 0.97	4.21-9.02	4.16-9.32	This study

and Cr in muscle tissues were exceeded the legislation limits of FAO and MARA. Due to the presence of factories involved in metal production, fertilizers, paper, mining, chemicals and shipyard repairs around the Çandarlı Bay, chromium detected in the wastes of these activities could have adverse effects on living organisms. Similarly, as lead levels were a higher than the maximum permissible value in fresh fish, as well as, the presence of Pb wastes in fertilizers and pesticides used in agricultural activities, especially, in oil refineries and the petrochemical industry located in Aliağa, suggests a negative impact in the region. Therefore, it is suggested that consumption of these fish as a food resource which showed higher. Pb and Cr values in excess of permissible limits may be harmful to human health. However, in terms of the other metals, they have not yet shown such a danger like this.

The average heavy metal concentrations in muscle tissue were examined between the stations, and accordingly, all metals were detected at their highest levels, either in T2 station along the Bakırçay River, or in T3 station offshore the Aliağa, compared to others. In particular, lead, chromium and iron concentrations were found higher in T2 station. This may show that Pb, Cu, Cr and Zn found in fertilizers and pesticides used in agricultural activities in the region, could be transported by the Bakırçay River to the Çandarlı Bay. Specifically, the reason the highest lead concentrations were found at T2 station compared to other stations may be related to the high inputs from Soma thermal power plant. Previous studies have shown high concentrations of Cu and Pb at the power plant exit point of the Bakırçay River. The reasons considered for these were gas flue emissions from the thermal power plant, coal washing water, and industrial activities in the region [50]. In another study which examined the accumulation of heavy metal concentrations in sediment, it reported the highest levels of Pb and Zn concentrations in the same location as that where the Bakırçay River flows into the region [51].

4. CONCLUSION

According to results, organic and inorganic substances resulting from domestic and industrial activities in the Bakırçay basin, pesticides and artificial fertilizers used in agricultural areas, harmful chemical substances spread over thermal power plant, discharge waters from industrial companies such as oil and dairy products etc. in the region, are discharged by the Bakırçay River, and this may play a major part in threatening the Çandarlı Bay.

In the ports of Aliağa especially, the petrochemical industry can be a high source of pollution. In addition, it is considered that several industrial companies located around the bay (chemical, paper, metal), ship dismantling and heavy ship traffic could affect negatively on the Çandarlı Gulf over time. Therefore, regular measurements of heavy metals in those aquatic species with a high economic value, consumed as food, are very important for those people carrying out health risk checks.

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