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Heavy metals in three commercially valuable fish species from İskenderun Bay, Northern East Mediterranean Sea, Turkey

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Abstract

Concentrations of cadmium, iron, lead, zinc, copper, manganese, nickel, chromium, cobalt and aluminum were determined in three commercially valuable fish species, *Saurida undosquamis*, *Sparus aurata*, *Mullus barbatus*, from İskenderun Bay in August 2003. The concentration of metals was measured by atomic absorption spectrophotometry (AAS). Concentrations of the heavy metals in examined fish species ranged as follows: Cd 0.01–4.16; Fe 0.82–27.35; Pb 0.09–6.95; Zn 0.60–11.57; Cu 0.04–5.43; Mn 0.05–4.64; Ni 0.11–12.88; Cr 0.07–6.46; Co 0.03–5.61; Al 0.02–5.41 mg kg^{-1} dry weight, respectively. The concentration of metals was significantly affected by the sampling site and fish species. Heavy metals in the edible parts of the investigated fish were in the permissible safety levels for human uses.

Keywords: Heavy metals; Fish; İskenderun Bay; Turkey

1. Introduction

Fish are the major part of the human diet and it is not surprising that numerous studies have been carried out on metal accumulation in different fish species (Kucuksezgin, Altay, Uluturhan, & Kontaş, 2001; Lewis et al., 2002; Prudente, Kim, Tanabe, & Tatsukawa, 1997). Fish also have been popular targets of heavy metal monitoring programs in marine environments because sampling, sample preparation and chemical analysis are usually simpler, more rapid and less expensive than alternative choices such as water and sediments (Rayment & Barry, 2000). Industrial wastes, geochemical structure and mining of metals create a potential source of heavy metals pollution in the aquatic environment (Gümgüm, Ünlü, Tez, & Gülsün, 1994; Lee & Stuebing, 1990). Under certain environmental conditions, heavy metals might accumulate up to a toxic concentration and cause ecological damage (Güven, Özbay, Ünlü, & Satar, 1999; Jefferies & Freestone, 1984). Metals such as iron, copper, zinc and manganese, are essential metals since they play an important role in biological systems, whereas mercury, lead and cadmium are non-essential metals, as they are toxic, even in traces. The essential metals can also produce toxic effects when the metal intake is excessively elevated. Heavy metal discharges to the marine environment are of great concern all over the world, and have a great ecological significance due to their toxicity and accumulative behavior. Thus, it can both damage marine species diversity and ecosystems (Matta, Milad, Manger, & Tosteson, 1999). Heavy metals in aquatic environment can remain in solution or in suspension and precipitate on the bottom or be taken up by organisms. The analysis of metal

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concentrations in biota samples at the same locations can indicate the transfer of metals through food chains (Topcuoğlu, Kırbaşoğlu, & Güngör, 2002).

Along the coast of İskenderun Bay, there are many towns including İskenderun with approximate population of 700,000 to 800,000, agricultural lands, industrial plants (iron-steel plants, beverage, LPG plants, oil transfer docks, other industrial plants and cargo ship's ballasts water). Therefore mainly untreated agricultural, municipal and industrial wastes affect the bay direct or indirectly. The present study has been undertaken to determine cadmium, iron, copper, chromium, cobalt, zinc, lead, nickel, aluminum and manganese concentrations in the muscles of fish and to investigate the differences between the concentrations of heavy metal accumulated by the species in İskenderun Bay.

2. Materials and methods

Fresh samples of three fish species were as follows: brushtooth lizardfish (*Saurida undosquamis*, SU), gilthead seabream (*Sparus aurata*, SA) and red mullet (*Mullus barbatus*, MB). These species are commonly consumed by the local population in Turkey. Three selected sites along the approximate coastline of 120 km İskenderun Bay, Northeastern Mediterranean Sea of Turkey, were sampled as illustrated in Fig. 1. These stations are the Arsuz (ARZ), relatively clean area, İskenderun Harbour Area (IHA) and Petrotrans (PTS), intensively polluted areas by both industrial and domestic sources. Fish samples were trawled from three stations in the bay during August 2003. Fifteen samples from each fish species were obtained from each station. Samples were washed with clean sea water at the point of collection, separated by species, placed on ice, brought to the laboratory on the same day and then frozen at −20 °C until dissection. Total size and weight of the samples were measured to the nearest millimeter and gram before dissected with clean equipment. Size may be an important factor in heavy metal accumulation of aquatic animals (Canlı & Furnes, 1993). There was no significant difference regarding the size and weight of samples among and within stations ($p > 0.05$). Metal contents were expressed as mg kg^{-1} dry weight of fish.

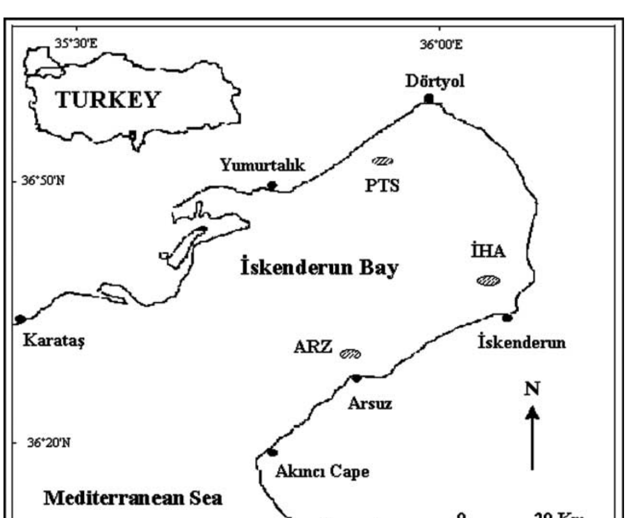

Fig. 1. Location map of sampling stations in İskenderun Bay.

The muscle samples of each fish were removed for metal analysis with a plastic knife, homogenized and weighed, and then individual samples were dried to constant weight at 60 °C in acid-washed petri dishes. The digestion was performed in a microwave digester to prepare the samples for analysis, (CEM MARS-5 Closed Vessel Microwave Digestion System) using the microwave digestion program. The advantages of microwave digestion against the classical methods are the shorter time, less consumption of acid and keeping volatile compounds in the solutions (Gulmini, Ostacoli, & Zelano, 1994; Krushevska, Barnes, & Chita, 1993; Sures, Tarschewski, & Haung, 1994). The completely digested samples were allowed to cool to room temperature, filtered and diluted to 50 ml in volumetric flasks with double distilled water. All digested samples were analysed three times for the metals Cd, Cu, Cr, Pb, Co, Zn, Fe, Ni, Al and Mn using AAS (Varian Spectraa 220 Fast Sequential Flame Atomic Absorption Spectrometry). Instrument was calibrated with standard solutions prepared from commercial materials. Analytical blanks were run in the same way as the samples and concentrations were determined using standard solutions prepared in the same acid matrix. The accuracy and precision of our results were checked by analyzing standard reference material (SRM, Dorm-2). The results indicated good agreement between the certified and the analytical values (Table 1), the recovery of elements being partially complete for most of them. All metal concentrations were quoted as mg kg^{-1} dry weight unless otherwise stated. All chemicals and standard solutions used in the study were obtained from Merck and were of analytical grade. Doubled distilled water was used throughout the study. All glassware and other containers were thoroughly cleaned and finally rinsed with double distilled water several times and air dried prior to use.

A logarithmic transformation was done on the data to improve normality. One-way analysis of variance (ANOVA) and Duncan's test ($p = 0.05$) were used to access whether heavy metal concentrations varied significantly between sites and species, possibilities less than 0.05 ($p < 0.05$) were considered statistically significant. All statistical calculations were performed with SPSS 9.0 for Windows.

Table 1
Concentrations of metals found in Standard Reference Material DORM-2 (dogfish muscle) from the National Research Council, Canada (all data as means ± standard errors, in mg kg^{-1} dry wt)

Value	Cd	Cu	Cr	Pb	Zn	Ni	Mn	Al	Fe	Co
Certified	0.043	2.34	34.7	0.065	26.6	19.4	3.66	10.9	142	0.182
SE	0.008	0.16	5.5	0.007	2.3	3.1	0.34	1.7	10	0.031
Observed[a]	0.047	2.39	33.8	0.067	24.8	18.3	3.88	10.4	136	0.186
SE	0.009	0.19	1.53	0.008	1.82	0.94	0.41	0.52	11.7	0.018
Recovery (%)	109	102	97	103	93	94	106	95	96	102

[a] Each value is the average of ten determinations.

3. Results

The mean and comparison of heavy metal concentrations for the selected three fish, *S. aurata*, *M. barbatus*, *S. undosquamis*, in mixed sites were given in Table 2. Iron showed the highest levels (concentrations for *S. aurata* and *M. barbatus* were 13.166 and 9.682 mg kg^{-1}) in examined species except *S. undosquamis*. Following Fe; Zn showed the second highest levels (concentrations for *S. aurata* and *M. barbatus* were 4.873 and 4.078 mg kg^{-1}, respectively) in examined species except *S. undosquamis*. Ni had the highest value (6.531 mg kg^{-1}) in *S. undosquamis*. Al (0.831), Zn (3.025) and Fe (4.175) were minimum in *S. undosquamis*, Co (0.953) was minimum in *M. barbatus*, Cu (1.239) and Mn (1.266) were minimum in *S. aurata*. In general, the differences between the mean metal concentrations of species were statistically significant ($p < 0.05$) for most of the measured metals, such as Fe and Ni.

The average heavy metal concentrations of same species from different sites were given in Table 3. In *Saurida undosquamis*, Cr level in IHA was higher than that in other sites, Al, Cd, Zn, Fe Cu, Mn, Co, Pb and Ni levels in PTS were higher than those in other sites. In *Mullus barbatus*, although Cu, Mn, Co, Cd, Zn, Fe and Pb levels in PTS were higher than those in other sites, Cr and Ni levels in ARZ, and Al level in IHA were higher than those in other sites. In *Sparus aurata*, Al, Cu, Mn and Co levels in PTS were higher than those in other sites, Cr and Ni levels in ARZ were higher than those in other sites, Cd, Pb, Zn and Fe levels in IHA were higher than those in other sites.

Heavy metal concentrations (mg kg^{-1} dry weight) and comparison of different species from the same sites were summarized in Table 4. In ARZ; Cd (1.441) and Fe (9.272) in *S. aurata*, Cr (3.289) in *M. barbatus*, Ni (4.973) in *S. undosquamis* were higher than those in other species. Al (0.512), Cr (1.142), Cu (0.639), Zn (1.854) and Fe (2.383) in *S. undosquamis*, Co (0.528) in *M. barbatus*, Mn (0.734) in *S. aurata* were lower than those in other species.

In IHA; Ni (6.465) in *S. undosquamis*, Fe (18.20) in *S. aurata* were higher than those in other species. Cu (1.191), Zn (2.585) and Fe (3.009) in *S. undosquamis*, Ni (1.183) and Pb (1.332) in *M. barbatus*, Al (0.691)

Table 2
Means and comparison of heavy metal concentrations in mixed sites of examined species*

Metals and species	N	Heavy metal concentrations (mg kg^{-1} dry weight)		
		Mean	Lower	Upper
Aluminum				
S. undosquamis	45	0.831[a]	0.07	2.64
M. barbatus	45	2.228[b]	0.47	5.41
S. aurata	45	0.919[a]	0.02	3.15
Chromium				
S. undosquamis	45	1.654[a]	0.21	3.13
M. barbatus	45	2.719[b]	0.69	6.46
S. aurata	45	1.309[a]	0.07	3.48
Copper				
S. undosquamis	45	1.318[a]	0.04	3.56
M. barbatus	45	2.201[b]	0.24	5.43
S. aurata	45	1.239[a]	0.08	3.46
Manganese				
S. undosquamis	45	1.361[a]	0.33	4.01
M. barbatus	45	2.151[b]	0.75	4.58
S. aurata	45	1.266[a]	0.05	4.64
Cobalt				
S. undosquamis	45	2.156[a]	0.36	5.32
M. barbatus	45	0.953[b]	0.03	2.84
S. aurata	45	1.295[b]	0.05	5.61
Nickel				
S. undosquamis	45	6.531[a]	2.46	12.88
M. barbatus	45	1.359[b]	0.11	3.98
S. aurata	45	2.537[c]	0.25	7.59
Cadmium				
S. undosquamis	45	1.310[a]	0.08	4.16
M. barbatus	45	0.831[b]	0.02	2.92
S. aurata	45	1.341[a]	0.09	3.70
Lead				
S. undosquamis	45	3.474[a]	1.12	6.95
M. barbatus	45	1.808[b]	0.32	4.83
S. aurata	45	2.314[b]	0.19	6.23
Zinc				
S. undosquamis	45	3.025[a]	0.60	8.70
M. barbatus	45	4.078[b]	0.72	9.85
S. aurata	45	4.873[b]	0.86	11.57
Iron				
S. undosquamis	45	4.175[a]	0.82	11.28
M. barbatus	45	9.682[b]	2.45	17.92
S. aurata	45	13.166[c]	4.59	27.35

* Post-hoc: Mean metal concentrations of species from all sites sharing a common letter for a particular metal are not significantly different, $p > 0.05$.

Table 3
Mean heavy metal concentrations of examined species and comparison of different species within same station (ARZ: Arsuz, IHA: İskenderun Harbour Area, PTS: Petrotrans, *SU*: *Saurida undosquamis*, *MB*: *Mullus barbatus*, *SA*: *Sparus aurata*)*

Site	Species	Mean heavy metal concentrations (mg kg^{-1} dry weight)									
		Al	Cr	Cu	Mn	Co	Ni	Cd	Pb	Zn	Fe
ARZ	SU	0.512^{a}	1.142^{a}	0.639^{a}	0.915^{ab}	1.493^{ab}	4.973^{a}	0.515^{ac}	2.992^{ac}	1.854^{a}	2.383^{a}
	MB	1.608^{b}	3.289^{c}	1.416^{b}	1.706^{b}	0.528^{c}	2.069^{b}	0.338^{ab}	1.477^{b}	2.352^{ab}	6.081^{b}
	SA	0.839^{a}	1.437^{a}	0.961^{ab}	0.734^{a}	1.032^{ac}	3.231^{ab}	1.441^{c}	1.985^{ab}	3.131^{ab}	9.272^{b}
IHA	*SU*	0.757^{a}	2.215^{a}	1.191^{a}	1.394^{ab}	2.131^{ab}	6.465^{a}	0.927^{ac}	3.280^{ac}	2.585^{a}	3.009^{a}
	MB	2.925^{b}	2.763^{c}	2.210^{b}	2.111^{b}	0.911^{c}	1.183^{b}	0.513^{ab}	1.332^{b}	4.066^{ab}	9.536^{b}
	SA	0.691^{a}	1.158^{a}	1.281^{ab}	1.348^{a}	1.336^{ac}	2.684^{ab}	1.783^{c}	2.291^{ab}	6.303^{ab}	18.20^{b}
PTS	*SU*	1.234^{a}	1.578^{a}	2.124^{a}	1.773^{ab}	2.853^{ab}	8.153^{a}	2.488^{ac}	4.149^{ac}	4.637^{a}	7.131^{a}
	MB	2.151^{b}	2.107^{c}	2.976^{b}	2.637^{b}	1.421^{c}	0.824^{b}	1.642^{ab}	2.616^{b}	5.815^{ab}	13.43^{b}
	SA	1.229^{a}	1.332^{a}	1.474^{ab}	1.696^{a}	1.517^{ac}	1.697^{ab}	1.404^{c}	2.667^{ab}	5.184^{ab}	12.03^{b}

* Post-hoc: Mean metal concentrations of different species from same sites sharing a common letter for a particular metal are not significantly different, $p > 0.05$

and Mn (1.348) in *S. aurata* were lower than those in other species.

In PTS; Ni (8.153), Pb (4.149) and Cd (2.488) in *S. undosquamis* were higher than those in other species. Zn (4.637) and Fe (7.131) in *S. undosquamis*, Co (1.421) and Ni (0.824) in *M. barbatus*, Al (1.229), Cu (1.474) and Mn (1.696) in *S. aurata* were lower than those in other species.

4. Discussion

This study examined the concentrations of Cd, Fe, Cu, Zn, Cr, Co, Mn, Ni, Pb and Al in *S. undosquamis*, *M. barbatus* and *S. aurata* from three sites in İskenderun Bay. Multiple comparison method was applied to determine which fish species are significantly different from others.

Kalay, Ay, and Canlı (1999) studied the heavy metal concentrations in different fish species from İskenderun Bay in 1996. Compared to the results of Kalay et al. (1999), in general, our results were lower, except Cr (Table 2). Similarly, Canlı and Atlı (2003), studied the heavy metals in different fish species from İskenderun Bay in 2000. Present results generally were lower for Fe and Zn than theirs. On the other hand, Cd, Cr and Pb concentrations were agreed well with the present results (Table 2). Yılmaz (2003) determined the heavy metals in the muscle, skin and gonads of *Mugil cephalus* and *Trachurus mediterraneus* caught from İskenderun Bay in 2001. The major findings of this study are that heavy metals concentrations in gonads, skin and muscle tissue of *M. cephalus* and *T. mediterraneus* from İskenderun Bay were very high compared with our findings. This situation indicated that the heavy metal concentrations in İskenderun Bay were likely decreased from 1996 to 2003. The decrease in regional mercantile marine, and consequently, gradual decrease in industrial business by the start of Gulf War in early 1990s, might be the reason for this slight decrease in metal concentrations. On the other hand, concentration of Cr stayed stable, probably because of the active Cr mines in the region.

In general, the concentrations of Mn, Cu and Ni in fish collected from the Black Sea coast were agreed well with the present results (Topcuoğlu et al., 2002). On the other hand, Cd, Co, Cr and Pb concentrations were lower, but Fe and Zn concentrations were higher than our results (Table 4). Similarly, Tüzen (2002) measured the concentrations of heavy metals in the muscles of fish from Middle Black Sea (Table 4). While concentrations of Cd and Pb were low comparing the data obtained from the present study. Fe and Zn concentrations, however, were higher than our findings. This is perhaps due to the heavy metal levels in different species depend on feeding habits (Amundsen et al., 1997; Mormede & Davies, 2001; Romeoa, Siaub, Sidoumou, & Gnassia-Barelli, 1999; Watanabe, Desimone, Thiyagarajah, Hartley, & Hindrichs, 2003), age, size and length of fish (Al-Yousuf, El-Sahahawi, & Al-Ghasis, 2000; Linde et al., 1998) and their habitats (Canlı & Atlı, 2003).

Data from the open literatures showed that metal concentrations in muscles of fish varied widely depending on where the animals caught (Table 4). Bustamante, Bocher, Chérel, Miramand, and Caurant (2003), found that different fish species Kerguelen Islands contained 0.01–0.1 Cd, 0.5–2.5 Cu, 9.2–32.2 Zn mg kg^{-1} dry weight in the muscle. Kwon and Lee (2001), measured the concentrations of heavy metals in the muscles of different fish species from Masan Bay, and reported 0.01 Cd, 0.02–0.05 Cr, 0.18–0.25 Cu, 0.02 Cu, 0.04–0.15 Pb, 6.33–12.9 Zn mg kg^{-1} dry weight. Their results generally were higher than present results, except Zn. The results reported by Tamira, Shane, and Ambrose (2001) from California Lagoons were generally higher than present results, except Cd, Co and Pb.

Table 4
Comparison of heavy metal concentrations in fish with values taken from the open literature*

Sample area	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn	References
Middle Black Sea	0.09–0.48			1.28–2.93	9.52–32.4	1.06–3.76		0.22–0.85	9.5–22.9	Tüzen (2002)
Black Sea Coast	<0.02–0.24	<0.05–0.40	<0.06–0.84	1.01–4.54	30–60	0.69–3.56	<0.01–2.04	<0.05–0.06	25.7–44.2	Topcuoğlu et al. (2002)
Kerguelen Islands	0.01–0.1			0.5–2.5					9.2–33.2	Bustamante et al. (2003)
Masan Bay, Korea	0.01		0.02–0.05	0.18–0.25			0.02	0.04–0.15	6.33–12.9	Kwon and Lee (2001)
California Lagoons	0.1–0.3	1.6–1.7	1.9–24	1.9–7.5			0.61–12	0.8–4.1	36–150	Tamira et al. (2001)
Mediterranean Sea, 1996	1.07–1.43		1.28–1.60	3.40–5.88	59.6–73.4		4.25–6.07	7.33–9.11	16.1–31.4	Kalay et al. (1999)
Mediterranean Sea, 2000	0.37–0.79		1.24–2.42	2.19–4.4	19.60–78.4			2.98–6.12	16.5–37.4	Canlı and Atlı (2003)
İskenderun Bay, 2001**			1.03–1.79	0.66–1.98	29.10–93.6		0.32–1.72		8.99–42.18	Yilmaz (2003)
İskenderun Bay 2003	0.95	1.42	1.69	1.57	10.2	1.71	2.90	2.32	4.36	Present study

* Values represent the ranges or mean expressed as mg kg^{-1} dry wt.
** Values represent the ranges or mean expressed as μg g^{-1} wet wt.

5. Conclusion

Present study provides new information on the distribution of heavy metals in fish from the İskenderun Bay. Based on the samples analyzed, metal concentrations found in the edible parts of fish are not heavily burdened with metals, and the concentrations are below the legal values for fish and fishery products proposed by Nauen (1983) and EPA (2002). On the other hand, when compared with previous results, present results indicated that heavy metal concentrations in fish from the bay slightly decreased from 1996 to 2003.

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