Determination of Heavy Metals in Some Commercial Fish from the Black Sea Coast of Turkey

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Abstract

In this study analysis of zinc, copper, lead and cadmium in six of the most commercially important fishes (*Trachurus trachurus*, *Belone belone, Pomatomus saltatrix, Mullus barbatus*, *Merlangius merlangus euxinus* and *Engraulis encrasicolus*) of the Black Sea coastal waters are presented. The results showed that the Zn concentrations were the highest and Cd concentrations the lowest in edible tissue of the six species. The muscle concentrations of Zn, Cu, Pb and Cd ranged from 6.95 - 34.33, 0.19 - 7.74, 0.02 - 1.51 and 0.02 - 0.18 ppm, respectively. Significant differences in metal concentrations were found between the species ($P < 0.05$). Higher muscle concentrations of Zn and Cd were found in *Merlangius merlangus euxinus* than the other species and the lowest concentration of Pb. However, *Mullus barbatus* contained the highest concentration of Cu and the lowest concentration of Zn and Cd. The highest concentration of Pb was found in *Pomatomus saltatrix.* In general, the concentrations of these heavy metals were mainly low and below the maximum permissible limit of the food regulations of Turkey and of international standards. Therefore, no public health problem would be raised in the consumption of these fishes.

Keywords: Heavy metals, Black Sea, commercial fishes, Turkish coast

Introduction

The pollution levels of the aquatic environment by heavy metals can be estimated by analyzing water, sediments and marine organisms [1,2]. The levels of heavy metals in marine organisms are often considerably higher than in other constituents of the marine environment. Because of their ability to concentrate heavy metals from their habitat it is crucial to know the changes in the metal levels that should be considered within a normal range, and how much their levels may be increased above these levels before commercial species become unsuitable as food [3].

The Black Sea has historically been one of the most biologically productive regions in the world [4,5]. Although it has 168 species of fish [6], there are only a few species of commercial importance and the supply of fishes is limited, because intensive fishing, industrialization and urbanization have caused the most favored species to decline [7]. The Black Sea itself has already been the victim of unmanaged fisheries, of unrestricted intense shipping activities, of mineral exploitation and the dumping of toxic waste [5]. As a result massive amounts of domestic wastewater and industrial effluents are transported by rivers and discharged into the Black Sea. Consequently, organic and inorganic pollutants are accumulated here [8]. These contaminants entering the aquatic ecosystem may not directly damage the organisms, but, they may be accumulated in aquatic organisms through the food chain process and eventually threaten the health of humans by fish consumption.

Most of the fishing industry in Turkey is based in the Black Sea waters. A total of 399,656 tons of sea fish were landed by Turkish fish vessels in 2010. The amount of fishing carried out in 2010 in the Black Sea consisted of 76 % of the total Turkish catch [9]. Of particular importance are landings of horse mackerel, garfish, whiting and anchovy were landed at the Black Sea ports including Sinop. Catches of fish from Turkish waters in 2010 are given in **Table 1** [9]. These data show that these species of fish are economically important for human consumption.

Table 1 Catches of fishes from Turkish waters in 2010 (in tons) (*From [9]).

Species	Total	Eastern Black Sea	Western Black Sea
Horse mackerel	14,392	7,968	2,879
Garfish	661	490	21
Bluefish	4,744	815	2,072
Red mullet	2,351	444	63
Whiting	13,555	9,278	2,616
Anchovy	229,023	173,059	29,967

The aim of the present study was to determine the concentrations of Zn, Cu, Pb and Cd in *Trachurus trachurus*, *Belone belone, Pomatomus saltatrix, Mullus barbatus*, *Merlangius merlangus euxinus* and *Engraulis encrasicolus* consumed in Turkey, with particular reference to public health. The fish were those which have commercial value and the heavy metals investigated in the present study are known to cause adverse health effects if consumed in sufficient quantities.

Material and methods

Samples of fish were purchased in the fish market at Sinop port during the fishing season from the Black Sea between September 2010 and December 2010. **Figure 1** shows the fish sampling area. Sampling covered six edible, commercially exploited fish species (*Trachurus trachurus*, *Belone belone, Pomatomus saltatrix, Mullus barbatus*, *Merlangius merlangus euxinus* and *Engraulis encrasicolus*), all of which were obtained iced but not frozen. Twenty individuals from each species taken randomly were rinsed in clean sea water and then filleted. The fillets from each fish were thoroughly chopped and mixed and a subsample of about 50 g taken. All prepared fish samples were stored deep frozen at –21 °C until their analysis. The fish samples were thawed and then dried in an oven at 100 - 105 °C to a steady weight. Approximately 5 - 10 g of samples were introduced into acid cleaned jars and digested with hot concentrated nitric acid to release the heavy metals. All organic materials in each sample were completely digested. After cooling 1 ml of HNO_3 was added to the digested residue and the solution was transferred to a 25 ml volumetric flasks and diluted with double distilled water to 15 ml. The digestion of each sample was made in triplicate and in all cases 3 blanks were also performed in order to check for possible contamination. Before analysis, the fish samples were filtered through a 0.45 µm Millipore membrane filter. All determinations were made using Atomic Absorption Spectrophotometer (AAS) (modified from [10]). The values were used to plot a standard curve. The standards and blank were treated in the same way as the real samples to minimize matrix interferences during analysis. Metal contents were expressed as µg g^{-1} dry weight. All reagents used were of analytical reagent grade (Merck, Germany).

Results and discussions

The present study was conducted to ascertain the levels of heavy metals, namely zinc, copper, lead and cadmium, in edible portions in commonly consumed fresh fish (*Trachurus trachurus*, *Belone belone, Pomatomus saltatrix, Mullus barbatus*, *Merlangius merlangus euxinus* and *Engraulis encrasicolus*) and those that are used in delicacies. In all the fish samples, Zn is the most abundant of the metals examined, followed by Cu, Pb and Cd. The concentration ranges found for these heavy metals, expressed on a dry weight basis, were as follows: 6.95 - 34.33 ppm for total Zn, 0.19 - 7.74 ppm for total Cu, 0.02 - 1.51 ppm for total Pb, 0.02 - 0.18 ppm for total Cd (**Figure 2**). Zn is widespread among living organisms, due to its biological significance. Higher muscle concentrations of Zn and Cd were found in *Merlangius merlangus euxinus* than the other species and the lowest concentration of Pb. However, *Mullus barbatus* contained the highest concentration of Cu and the lowest concentration of Zn and Cd. The highest concentration of Pb was found in *Pomatomus saltatrix*. The essential metals including Zn and Cu are in higher concentrations, probably due to their function as co-factors for the activation of a number of enzymes and regulated to maintain a certain homeostatic status in fish. Whereas, the non-essential metals including Pb and Cd have no biological function or requirement and their concentrations in coastal fishes are generally low [11].

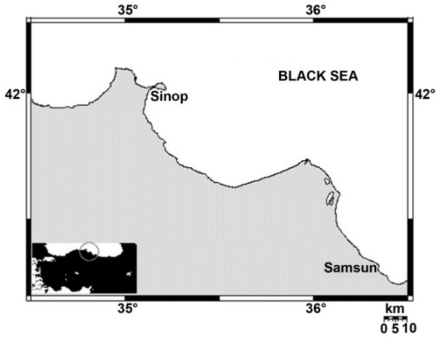

Figure 1 Fish sampling area from Sinop coasts of the Black Sea, Turkey.

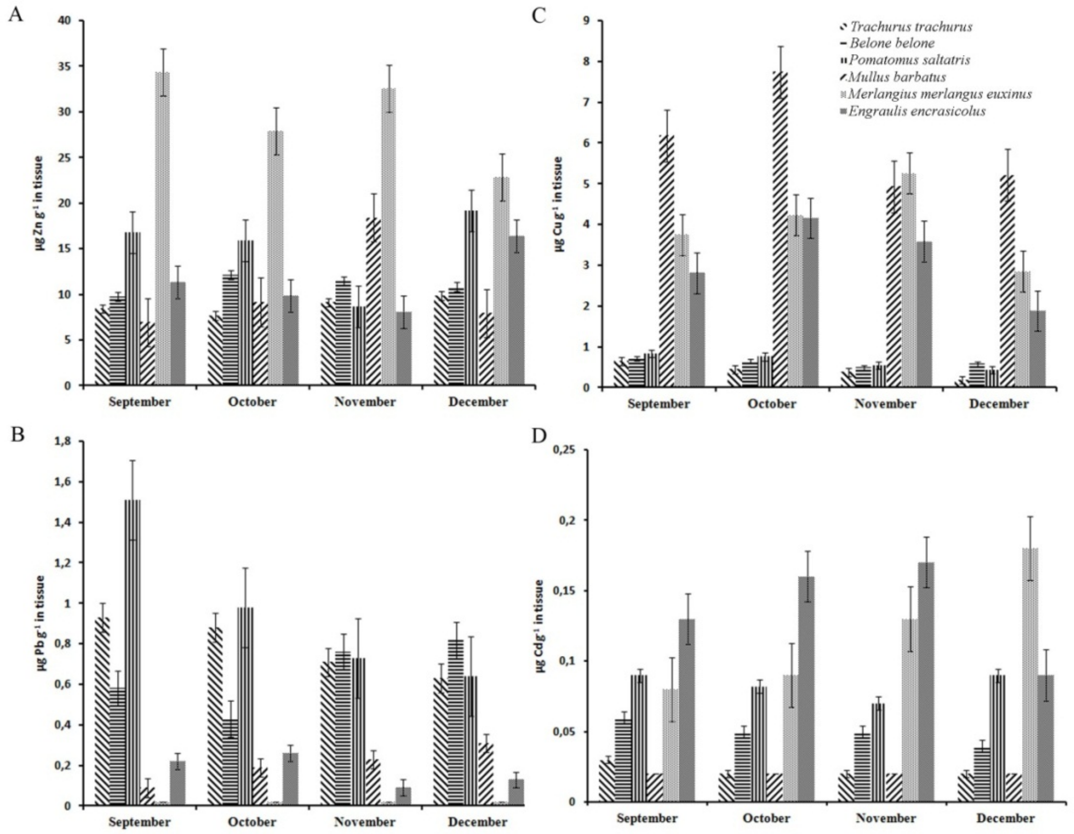

Figure 2 Zn, Cu, Pb and Cd concentrations (µg/g dry wt) in fish from Black Sea in 2010.

The presence of heavy metals in the marine environment is caused by both natural and anthropogenic factors. Heavy metals depending on their regions are found in different concentrations in marine biota. Some metals in their natural concentrations play a significant role in biochemical processes [12,13]. The uptake of such toxic heavy metals from sea water or food can be an important factor for the metal budget of biota [2,14-16]. In polluted marine ecosystems the transfer of heavy metals through food chains can be high enough to bring about harmful concentrations in the marine biota. Almost all metals are toxic at higher concentrations and some are lethal even at very low concentrations [1,17-19].

Zn in the fish samples accumulated in the order *Trachurus trachurus* < *Belone belone* < *Engraulis encrasicolus* < *Mullus barbatus* < *Pomatomus saltatrix* < *Merlangius merlangus euxinus*. Although Zn is an essential metal for marine organisms including fish it is known to be involved in most metabolic pathways and may regulate their body burden of this metal. Mean concentrations of Zn found in all fish samples studied are all less than 35 ppm, well below the guideline level of 50 ppm [20,21].

Cu concentrations in all fish samples were low (mainly less than 8 ppm) and far below the guideline level of 20 ppm [20,21]. Cu accumulation showed the order as *Mullus barbatus* < *Merlangius merlangus euxinus* < *Engraulis encrasicolus* < *Pomatomus saltatrix* < *Belone belone* < *Trachurus trachurus*.

The Pb level was highest in lipid-rich pelagic fish *Pomatomus saltatrix*, followed by *Trachurus trachurus, Belone belone, Mullus barbatus, Engraulis encrasicolus* and *Merlangius merlangus euxinus*. The maximum concentation was 1.51 ppm, this concentration was, however, well below the proposed maximum in the food safety regulations (2 ppm) [20].

Although Cd was detectable in fish samples, the maximum concentation was 0.18 ppm, this concentration was, however, well below the expected values (< 0.2 ppm) [20]. Cd accumulated in the order of *Merlangius merlangus euxinus* < *Engraulis encrasicolus* < *Pomatomus saltatrix* < *Belone belone* < *Trachurus trachurus* < *Mullus barbatus*.

The level of contamination in fish by these heavy metals is relatively low and is able to meet the legal standards set by the Ministry of Agriculture, Fisheries and Food (MAFF), the Turkish Food Codex, Commission Regulation (EC). Regional comparison for results must be made with caution because of variations in both the quality of analytical data and in sampling procedure.

Turkish Food Codex [22] and Commission Regulation (EC) [23] indicate that maximum levels are 0.30 mg.kg^{-1} wet weight for Pb and 0.05 (0.10 for *Engraulis encrasicolus* and *Trachurus trachurus*) mg.kg^{-1} wet weight for Cd. It should be noted that metal contents were expressed as μg.g^{-1} dry weight in the present study. In general the fish on sale in Sinop markets are low in metals when compared with the levels found in same fish species studied in other parts of the Black Sea (**Table 2**). Therefore, it may be concluded that all species examined here indicate that the metal levels are not dangerous for the health of consumers.

The heavy metal levels in fish from the Black Sea have been investigated by several researchers (**Table 2**). When the metal concentrations were compared among the Black Sea coasts, Zn and Cu concentrations were found to be highest in Bartin ([24], **Table 2**). Pb concentrations were found to be highest in Trabzon followed by Bartin ([24,25], **Table 2**). Cd concentrations were found to be highest in Trabzon followed by Samsun ([26,27], **Table 2**).

Table 2 Heavy metal concentrations (µg metal g^{-1} wet wt.) in fish from the Black Sea coasts. (- : not measured *: expressed in µg metal g^{-1} dry wt. BS: Black Sea (modified from [40]).

Fish species	Area	Zn	Cu	Pb	Cd	References
Belone belone	Sinop	7.76±1.37	0.54±0.05	0.51±0.08	0.05±0.007	[28]
*Belone belone**	Sinop	9.75-12.13	0.49-0.71	0.43-0.82	0.04-0.06	Present study
Engraulis encrasicolus	Inebolu	-	0.68-1.33	0.06-0.06	-	[29]
*Engraulis encrasicolus**	BS	50.7±8.3	3.39±0.49	2.51±0.09	0.27±0.06	[30]
Engraulis encrasicolus (liver)	Sinop	7.30±1.12	1.76±0.08	1.87±0.08	0.112±0.009	[31]
Engraulis encrasicolus	Sinop	3.55±0.68	0.69±0.06	0.78±0.04	0.025±0.005	[31]
*Engraulis encrasicolus**	Amasra	35.7±0.4	2.21±0.11	< 0.05	0.10±0.01	[33]
*Engraulis encrasicolus**	Samsun	17.38±2.01	1.94±0.10	0.38±0.02	0.20±0.03	[27]
Engraulis encrasicolus	Trabzon	10.8±1.29	0.88±0.10	0.12±0.03	0.03±0.01	[24]
Engraulis encrasicolus (liver)	Trabzon	14.1±2.31	1.08±0.20	0.47±0.13	0.07±0.02	[24]
Engraulis encrasicolus	Sinop	10.6±0.88	1.12±0.16	0.27±0.05	0.02±0.00	[24]
Engraulis encrasicolus (liver)	Sinop	12.5±0.96	1.27±0.20	0.74±0.19	0.06±0.01	[24]
Engraulis encrasicolus	Bartin	45.6±22.1	8.58±2.15	0.87±0.40	0.06±0.02	[24]
Engraulis encrasicolus (liver)	Bartin	145±38	30.7±7.54	3.38±0.55	0.24±0.09	[24]
*Engraulis encrasicolus**	Samsun, Sinop, Terme, Fatsa, Ordu	26.25±1.67	2.73±0.21	0.70±0.07	0.035±0.005	[34]
*Engraulis encrasicolus**	BS	40.2±3.2	0.95±0.08	0.33±0.01	0.65±0.04	[43]
*Engraulis encrasicolus**	BS	38.8±3.2	1.96±0.14	0.30±0.02	0.27±0.02	[44]
*Engraulis encrasicolus**	Sinop	8.12-16.41	1.88-4.16	0.09-0.26	0.09-0.17	Present study
Merlangius merlangus euxinus	Sinop	-	0.12-2.00	0.033-1.76	-	[35]
Merlangius merlangus euxinus (liver)	Sinop	9.18±1.98	1.87±0.11	1.81±0.07	0.110±0.009	[31]
Merlangius merlangus euxinus	Sinop	4.36±0.71	0.88±0.08	0.74±0.06	0.025±0.004	[31]
*Merlangius merlangus euxinus**	Trabzon	15.322	2.71	1.078	0.601	[26]
*Merlangius merlangus euxinus**	Samsun	-	-	< 0.05	< 0.02	[32]
*Merlangius merlangus euxinus**	Sinop	-	-	< 0.05	< 0.02	[32]
Merlangius merlangus euxinus	Sinop	9.46	4.56	0.44	0.087	[36]
*Merlangius merlangus euxinus**	Sinop	8.862-163.277	0.913-8.952	-	-	[37]
Merlangus merlangus	Kastamonu	-	0.62-3.25	0.02-0.11	-	[29]
Merlangius merlangus	Zonguldak	-	0.37-7.72	0.05-2.26	-	[29]
*Merlangius merlangus**	Perşembe	43.1±0.1	1.86±0.04	< 0.05	< 0.02	[33]
*Merlangius merlangus**	Rize	30.2±0.1	4.54±0.11	< 0.05	< 0.02	[33]
*Merlangius merlangus**	BS	3.3	1.3	0.088	0.0131	[38]
Merlangius euxinus *	Samsun, Sinop, Terme, Fatsa, Ordu	31.34±1.61	3.72±0.59	0.58±0.03	0.002±0.000	[34]
*Merlangius merlangus**	BS	48.6±3.9	1.25±0.10	0.93±0.07	0.055±0.04	[43]
*Merlangius merlangus**	BS	65.4±4.2	1.32±0.11	0.53±0.04	0.21±0.02	[44]
*Merlangius merlangus euxinus**	Sinop	22.82-34.33	2.85-5.26	0.02	0.08-0.18	Present study
*Mullus barbatus**	Trabzon	11.5±3.5	9.10±5.9	6.86±0.26	< 0.1	[25]
Mullus barbatus (liver)	Sinop	3.79±0.90	1.49±0.10	0.89±0.23	0.070±0.006	[31]

Fish species	Area	Zn	Cu	Pb	Cd	References
Mullus barbatus	Sinop	2.42±0.27	0.76±0.07	0.28±0.06	0.023±0.002	[31]
*Mullus barbatus**	Samsun	-	-	0.0815±0.003	< 0.02	[32]
*Mullus barbatus**	Sinop	-	-	0.0515±0.0005	< 0.02	[32]
Mullus barbatus	Sinop	9.90	8.968	0.424	0.076	[36]
Mullus barbatus (viscera)	Sinop	11.47	1.365	1.276	0.230	[36]
*Mullus barbatus**	BS	4.3	0.01	0.077	0.017	[38]
*Mullus barbatus**	Sinop	1.424-63.290	0.380-2.714	-	-	[37]
*Mullus surmelutus**	Sinop	28.0±9.0	4.20±1.8	< 0.5	0.42±0.09	[25]
*Mullus barbatus**	Samsun, Sinop, Terme, Fatsa, Ordu	23.71±0.71	3.14±0.31	0.92±0.12	0.020±0.002	[34]
*Mullus barbatus**	BS	106±9.1	0.98±0.07	0.84±0.07	0.45±0.04	[43]
*Mullus barbatus**	BS	75.5±5.3	0.96±0.08	0.36±0.03	0.17±0.02	[44]
*Mullus barbatus**	Sinop	6.95-18.43	4.93-7.74	0.09-0.31	0.02	Present study
Pomatomus saltatrix	Sinop	9.40±1.48	0.58±0.08	0.55±0.08	0.05±0.004	[28]
Pomatomus saltatrix	Sinop	15.39	6.666	0.322	0.064	[36]
*Pomatomus saltatrix**	Samsun, Sinop, Terme, Fatsa, Ordu	25.51± 0.92	2.86±0.58	1.26±0.21	0.025±0.002	[34]
*Pomatomus saltatrix**	BS	35.4± 3.2	1.83± 0.10	0.38± 0.02	0.6± 0.05	[43]
*Pomatomus saltatrix**	BS	93.4±5.8	2.78± 0.21	0.87± 0.07	0.23± 0.02	[44]
*Pomatomus saltatrix**	Sinop	8.64-19.2	0.43-0.83	0.64-1.51	0.07-0.09	Present study
Trachurus trachurus	Igneada	-	0.36-0.68	-	-	[29]
Trachurus trachurus	Inebolu	-	1.24-2.8	0.02-0.06	-	[29]
Trachurus trachurus	Sakarya	-	0.06-0.24	0.27-0.66	-	[29]
Trachurus trachurus (liver)	Sinop	4.16±1.09	1.38±0.09	1.36±0.38	0.050±0.007	[31]
Trachurus trachurus	Sinop	3.28±0.66	0.79±0.06	0.74±0.21	0.028±0.002	[31]
*Trachurus trachurus**	Samsun	12.05±2.30	1.52±0.35	0.85±0.16	0.47±0.10	[27]
*Trachurus trachurus**	Samsun, Sinop, Terme, Fatsa, Ordu	27.70±1.00	1.79±0.12	0.60±0.07	0.012±0.002	[34]
*Trachurus trachurus**	BS	37.4±2.9	0.95±0.04	0.68±0.05	0.50±0.03	[43]
*Trachurus trachurus**	BS	52.7±4.9	0.65±0.05	0.82±0.06	0.32±0.03	[44]
*Trachurus trachurus**	Sinop	7.66-9.12	0.19-0.65	0.63-0.93	0.02-0.03	Present study

Altas and Büyükgüngör [39] investigated the impact of marine activities on heavy metal pollution. Although, the Turkish coastal regions of the Black Sea are relatively poor in metal releasing industrial activities, mining and agricultural activities on land may be an important source in the delivery of some metal pollutants. However, the Black Sea receives large quantities of unregulated and uncontrolled fresh water withdrawal for irrigation purposes, hydro and thermal power generation and the use of coastal areas for permanent human settlements; shipping; and untreated domestic, industrial and agricultural waste drain into the sea via the rivers or directly [40]. In terms of metal pollutants along the Turkish coast of the Black Sea Trabzon, Bartin and Samsun contain higher levels than other cities of the Turkish coast of the Black Sea (**Table 2**). Boran and Altinok [41] concluded that heavy metal pollution in living organisms of the Black Sea has attracted considerable research attention since last 20 years.

Accumulations of heavy metals were generally found to be species specific and may be related to their feeding habits and the bio-concentration capacity of each species. Topping [42] suggested that mainly plankton feeding fish contain much higher concentrations of some heavy metals than bottom feeding fish. In the present study the highest concentration of Zn and Cd were found in *Merlangius merlangus euxinus*. The species is commonly found near the bottom in waters and moves into midwater in the pursuit of its prey. We found anchovy in the stomachs of whiting. The highest concentration of Cu was found in *Mullus barbatus* in the present study. The species prefers muddy bottoms and feeds on mainly benthic organisms such as shrimps, amphipods, polychaete worms, molluscs and small fishes. The highest concentration of Pb was found in *Pomatomus saltatrix* which feeds mainly on fish.

Fish have been considered good indicators for heavy metal contamination in aquatic ecosystems because they occupy different trophic levels. Meanwhile, fish are widely consumed in many parts of the world by humans.

The results of the present study revealed that consuming fish from the Sinop coast of the Black Sea may not have harmful effects to human health because levels of heavy metals are below the permissible limits. Bat and Öztürk [28] point out the local Black Sea environment especially the Sinop coasts is not facing a heavy metal pollution problem. Bat *et al.* [40] pointed out that Turkey is a developing country where industrial and urban developments mostly occur in coastal areas resulting in increased input of waste imposing a further stress on the Turkish coasts of the Black Sea. However it is better to continue the studies on the metal pollution effects on food chain organisms comparatively before reaching any definitive conclusion.

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