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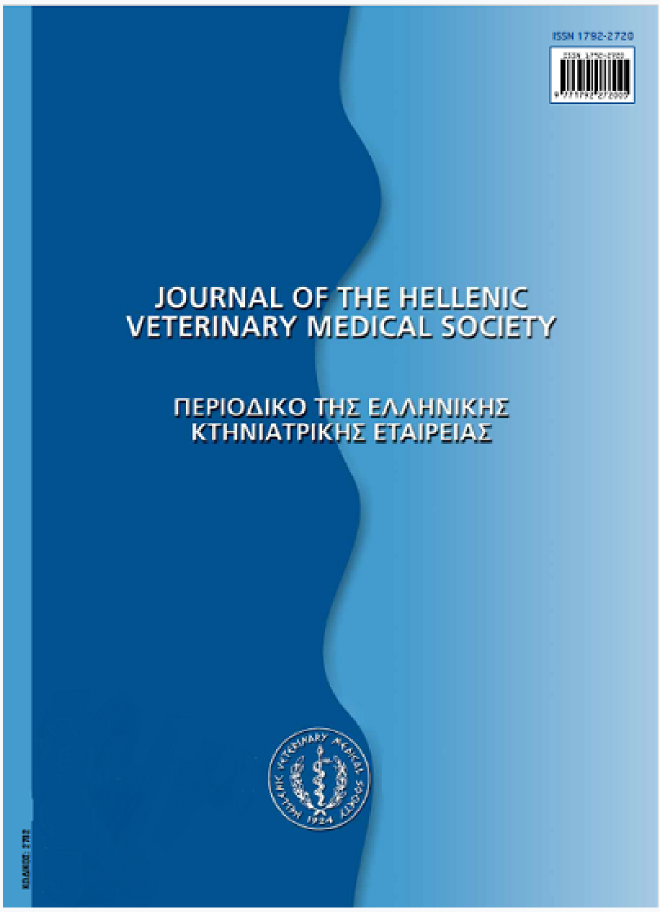

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Determination of Metal Contamination in Seafood from the Black, Marmara, Aegean and Mediterranean Sea Metal Contamination in Seafood

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ABSTRACT. Seafood is one of the most important components of a healthy diet due to its composition. With the Black, Marmara, Aegean and Mediterranean Sea, Turkey has substantial sources of seafood. Seas are highly impacted by environmental pollution. Among these, heavy metal pollution has long been recognized as a serious problem for seafood. As heavy metals cannot be degraded, they are deposited, assimilated or incorporated in water, sediment and aquatic animals. By these properties, they can be transferred to humans through the food chain especially by the consumption of fish and shellfish. The aim of this study is to determine the concentrations of Cd, As, Pb and Hg levels in selected fish species and marine animals from all of the 4 seas of Turkey by using the ICP-MS technique, and to compare the results with the legislations safe limits. For this purpose, 13 different fish species, mussels and shrimps have been obtained from the Black, Marmara, Aegean and Mediterranean Seas. According to the results, metal concentrations decrease in the order As>Pb>Hg>Cd. In all the seas, the same order was found. Statistically significant differences were observed in the metal levels between fish species and the shellfish in all regions. Except for the two samples, all the results was found compatible with the Turkish Food Codex and European Commission Regulation limits. Arsenic levels were detected between 0,076-4,230 mg/kg within the samples. Cadmium levels were detected as higher than the limits in two samples obtained from the Mediterranean Sea, *Scophthalmus maximus* and *Mullus barbatus* species as 0,076 mg/kg and 0,064 mg/kg, respectively. The highest and the lowest levels of mercury and lead were measured as 0,005-0,405 and 0,015-0,405 mg/kg, respectively. The results obtained from this study revealed that, except for a few cases, the selected heavy metal concentrations in most samples were below the limits. Also, besides the mussels and the shrimps, there was no single type of fish that was consistently high in all metals. The examined seas and the seafood were found to be safe for human consumption.

Keywords: Fish Species, Shellfish, ICP-MS, Heavy metal, Contamination

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INTRODUCTION

Seafood is one of the most important components of a healthy diet due to its proteins of high biological value, vitamin D, vitamin E, iodine, and the long chain omega-3 polyunsaturated fatty acids (Sioen et al., 2007). With the Black, Marmara, Aegean and Mediterranean Sea, Turkey has substantial sources of seafood. According to the last data's, seafood production in Turkey is 537,345 tons, and 392,972 tons of it is obtained from the seas by fishing. For many coastal cities in Turkey fishing is a major economic activity and a means of subsistence. Because of its geographical location and unique habitat, Turkey's seas are characterized by a rich marine biodiversity (Anon, 2016a).

Seas are highly impacted by environmental pollution. Among these, heavy metal pollution has long been recognized as a serious problem for seafood (Caçador et al., 2012). Although adverse health effects of heavy metals have been known for a long time, exposure to them continues. The sources of heavy metals can be anthropogenic or natural. Draining of sewage, dumping of wastes, recreational, mining and other industrial activities are generally the causes of the anthropogenic contamination. Naturally heavy metals may enter into aquatic system by ore-bearing rocks, windblown dust, forest fires and vegetation in small amounts (Fernandez-Leborans and Herrero, 2000; Jarup, 2003). To avoid the pollution of the marine waters, countries taking measures. For example in the EU the main goal of the Marine Directive is to achieve Good Environmental Status (GES) of EU marine waters by 2020. GES was defined by the directive as the environmental status of marine waters where these provide ecologically diverse and dynamic oceans and seas which are clean, healthy and productive (Anon., 2016c). Cadmium, mercury, zinc, lead, arsenic and manganese are some of the heavy metals that can be toxic, persistent and bioaccumulative in aquatic environments. As heavy metals cannot be degraded, they are deposited, assimilated or incorporated in water, sediment and aquatic animals. With these properties, heavy metals are transferred to humans through the food chain especially by the seafood (Malik et al., 2010).

Besides the ecological damage, heavy metals cause carcinogenic and other adverse effects on human health. Some of the heavy metals are essential for the body but some are toxic even in small amounts. Also, essential metals can have toxic effects when they are consumed in excessive amounts (Ubillus et al., 2000). Cadmium (Cd) was classified as human carcinogen by Environmental Protection Agency (EPA). Cd causes kidney failure and fragile bones in human. Lead (Pb) affects the nervous system. Mercury (Hg) causes lung damage and brain function loss. High levels of arsenic (As) can be fatal while small amounts affect the cardiovascular system. Chronic effects after long exposure to heavy metals are more important but the acute cases have also been witnessed in Minimata and Itai-itai diseases (Martin and Griswold, 2009).

Heavy metal pollution in the aquatic environment is identified by measuring its concentration in water, sediment and living organisms (Boran, 2010). Especially marine animals such as fish, accumulate metals higher than present in water and sediment so it is crucial to determine the level of heavy metals in fish and the other sea products to evaluate the possible risk to human health (Bat, 2012). The metal accumulation, changes according to the location, distribution, habitat, feeding habits, age and size of the organism (Velusamy, 2014).

There are different methods like Inductively Coupled Plasma Atomic Emission Spectrometric Method (ICP-MS), Flame Atomic Absorption Spectrometric (FAAS), Atomic Absorption Spectrometric with Graphite Furnace (GFAAS), Electro-Thermal Evaporation Inductively Coupled Plasma Mass Spectrometry (ETV-ID-ICP-MS), Inductively Coupled Plasma Optical Spectrometry (ICP-OES), Inductively Coupled Plasma Spectrometry Having Isotope (ID-ICP-MS), Inductively Coupled Plasma Flame Emission Spectrometry (ICP-AES) to detect metal levels in fish and the other marine animals (Aygun and Abanoz, 2011, Caçador et al., 2012, Hussein and Khaled, 2014, Uysal et al., 2008).

Within these methods, ICP-MS has become more common in food analysis. Compared to the other techniques ICP-MS has some advantages like simultaneous multielement measurement capability, coupled with very low detection limits. Also this technique offers a wider linear dynamic range and provides simpler spectral interpretation and isotopic information (Nardi et al., 2009).

The aim of this study to determine the concentrations of Cd, As, Pb and Hg levels in selected fish species and marine animals from all of the 4 seas of Turkey by using the ICP-MS technique, and compare the results with the Turkish Food Codex according to the safety limits.

MATERIALS AND METHODS

Sample collection

For this study different fish species, mussels and shrimps were obtained from the Black, Marmara, Aegean and Mediterranean seacoast. Among the Black Sea mussels (*Mytilus galloprovincialis*) and 13 fish species were selected; mullet (*Mugil soiuy*), mackarel (*Trachurus trachurus*), seabass (*Dicentrarchus labrax*), shad (*Alosa fallax*), red mullet (*Mullus barbatus*), turbot (*Scophthalmus maximus*), anchovy (*Engraliusencrasicolus*), whitting (*Merlangius euxmus*), blue fish (*Pomatomus saltatrix*), bonito (*Sarda sarda*), grey mullet (*Mugil cephalus*), garfish (*Belone belone*), hake (*Merluccius merluccius*) . From the Marmara Sea mullet (*Mugil soiuy, Mugil cephalus*), mackarel (*Trachurus trachurus*), seabass (*Dicentrarchus labrax*), shad (*Alosa fallax*), red mullet (*Mullus barbatus*), turbot (*Scophthalmus maximus*), anchovy (*Engralius encrasicolus*), whitting (*Merlangius euxmus*), blue fish (*Pomatomus saltatrix*), bonito (*Sarda sarda*), grey mullet (*Mugil cephalus*), garfish (*Belone belone*), hake (*Merluccius merluccius*), seabream (*Sparus auratus*), 14 fish species were obtained along with mussel (*Mytilus galloprovincialis*) and shrimp (*Penaeus indicus).* Mussel (*Mytilus galloprovincialis*), shrimp (*Penaeus indicus)* and selected 9 species selected from the Aegean and Mediterranean Sea are; mullet (*Mugil cephalus*), shad (*Alosafallax*), hake (*Merluccius merluccius*), whitting (*Merlangius euxmus*), seabass (*Dicentrarchus labrax*), turbot (*Scophthalmus maximus*), red mullet (*Mullus barbatus*), blue fish (*Pomatomus saltatrix*), seabream (*Sparus auratus*).

One and a half kg from each fish species, shrimp and mussel, were purchased from 3 different fishermen or fish markets of each seacoast in 3 fishing seasons. Samples were washed in clean water and 1 kg of muscles with skin from the each party fish, mussel and shrimp were homogenized. All the prepared samples were stored in the freezer at -20°C until the analysis.

Standards and Reagents

All reagents were of analytical reagent grade. Ultrapure water was used for all dilutions. Nitric acid was not less than 65 %, with a density of approximately 1,4 g/mL. The element standard solutions from Merck Millipore were used for the calibrations and prepared by diluting stock solutions of mg/L.

Standard Preparation

The concentrations of the stock solutions were 20 mg/L for As, 10 mg/L for Cd, 10 mg/L for Pb. Mercury stock solution was prepared by diluting 1 mL of Hg and 1 mL of nitric acid in a 100 ml volumetric flask.

Extraction and Clean-up

Heavy metal analysis was done according to the method of the Nordic Committee on Food Analyses No.186 2007 (Anon, 2007). All the glassware and plastics were held overnight in 10% (v/v) nitric acid. Before its use, it was rinsed with distilled water and deionized water and dried. Boneless muscle tissues (with skin for fish samples) were removed using stainless steel knife and were digested to a strong acid digestion. From each individual sample, 2 aliquots of 1 g homogenized specimen were taken for extraction. Microwave system was used for the extraction of the samples. A digestion solution was prepared with 5 ml of 65 % nitric acid and 5 ml ultra-pure water, with this solution samples were digested under 600 W power, 10 min. Ramp time, 450 Psi pressure at 180 °C in 10 minutes. Then the content was decanted to the falcon tubes and ultra-pure water was added up to 50 ml for the quantification in ICP-MS (Agilent 7500c ICP-MS). For the calibration 3 different concentrations of the metal solutions were used. 1 μg/L, 5 μg/L and 20 μg/L were used for As, while 0,5 μg/L, 2,5 μg/L and 10 μg/L for the Cd, Hg and Pb. During the test of mercury, gold was added in order to stabilize the Hg. Analytical blanks were run in the same way.

Table 1. The settings for the ICP-MS

Parameter	Setting
RF-Power (W)	1500
Carrier gas flow l min -1	1,2
Plasma gas flow l min –1	15
Auxiliary gas flow l min –1	1,0
Nebuliser	Babington
Spray chamber	Water cooled double pass
Spray chamber temperature °C	2
Lens voltage	4,5
Mass resolution	0,8
Integration time points/ms	3
Points per peak	3
Replicates	3

Table 2. Instrumental detection limits (LOD) and limits of quantification (LOQ)

Metal	LOD (µg/g)	LOQ (mg/kg)
As	0,5	0,023
Pb	0,3	0,0013
Hg	1	0,047
Cd	0,5	0,033

Icp-Ms

Mass spectrometer with inductively coupled argon plasma was operated in a mass range from 5 – 240 AMU. The settings of the ICP-MS are illustrated in Table 1. The recovery of the calibration was ranged within 10% . Instrumental detection limits (LOD) and limits of quantification (LOQ) was listed in Table 2. The calculation of the detection limit is based on the 3 times of SD of the blank solution. After calibration of the instrument, the test solutions were analyzed. The samples obtained by pressure digestion were diluted before measurement in order to avoid interference by high concentrations of matrix elements. Within suitable short intervals (after 10 samples) the blank solution and one calibration solution were checked regularly. Calculation of the concentration was done automatically by the software of the ICP-MS instrument.

Statistical analyses

Statistical analyses was conducted with one-way ANOVA test for the difference between seas and Student-t test for the difference between fish and shellfish, $p<0,05$ applied as the minimum level of significance. Statistical software SPSS 14.01 was used for the analyses.

RESULTS

Mean concentrations of all analyzed metals are shown in Table 3. The metal concentrations decrease in the order As>Pb>Hg>Cd, and are the same in all seas. The results of mussels and shrimps were found higher than the other fish species for every metal, in every region. Statistically significant differences were found in the metal levels between fish species and the shellfish in all regions. Between the fish species, *Merluccius merluccius* has the highest concentrations of arsenic in the Marmara, Aegean and Mediterranean Sea; also this species has the second highest concentration of arsenic in the Black Sea. In Turkish Food Codex, there are no specific limits for the As in foods.

For Cd, from all the regions, highest levels were detected in mussels. *Engralius encrasicolus* in the Black and Marmara Sea; *Scophthalmus maximus* in the Aegean and Mediterranean Sea has the highest levels within the fish species. Cadmium levels were

Table 3. Mean concentrations of all analyzed metals.

Region	Sample	Arsenic (mg/kg)	Cadmium (mg/kg)	Mercury (mg/kg)	Lead (mg/kg)
Black	*Mugil soiuy*	0,374	0,034	0,068	0,159
	Trachurus trachurus	0,915	0,024	0,035	0,182
	Dicentrarchus labrax	0,168	0,012	0,023	0,113
	Alosa fallax	1,56	0,021	0,043	0,096
	Mullus barbatus	2,44	0,016	0,032	0,165
	Scophthalmus maximus	0,637	0,027	0,065	0,191
	Engralius encrasicolus	0,896	0,063	0,007	0,071
	Mytilus galloprovincialis	4,23	0,097	0,405	0,375
	Merlangius euxmus	0,463	0,013	0,081	0,099
	Pomatomus saltatrix	0,484	0,016	0,046	0,085
	Sarda sarda	0,257	0,008	0,064	0,072
	Mugil cephalus	0,269	0,011	0,008	0,045
	Belone belone	0,209	0,018	0,023	0,086
	Merluccius merluccius	2,43	0,014	0,049	0,128
Marmara	*Mugil cephalus*	0,335	0,015	0,036	0,055
	Alosa fallax	0,689	0,017	0,037	0,087
	Merluccius merluccius	2,09	0,009	0,062	0,115
	Engralius encrasicolus	0,565	0,032	0,005	0,044
	Dicentrarchus labrax	0,173	0,006	0,026	0,143
	Scophthalmus maximus	0,482	0,014	0,039	0,152
	Belone belone	0,303	0,007	0,013	0,069
	Mullus barbatus	0,942	0,009	0,016	0,098
	Merlangius euxmus	0,324	0,011	0,043	0,054
	Sarda sarda	0,197	0,005	0,028	0,061
	Mytilus galloprovincialis	2,85	0,087	0,341	0,267
	Sparus auratus	0,264	0,012	0,025	0,078
	Trachurus trachurus	0,707	0,016	0,022	0,136
	Penaeus indicus	6,93	0,037	0,061	0,173
	Mugil soiuy	0,279	0,026	0,032	0,093
	Pomatomus saltatrix	0,345	0,006	0,018	0,047

Aegean	*Mugil soiuy*	0.175	0,037	0,014	0,078
	Alosa fallax	0,403	0,015	0,028	0,046
	Merluccius merluccius	1,15	0,025	0,034	0,075
	Merlangius euxmus	0,118	0,009	0,063	0,059
	Dicentrarchus labrax	0,093	0,005	0,008	0,04
	Scophthalmus maximus	0.613	0,108	0,045	0,139
	Mytilus galloprovincialis	1,96	0,143	0,079	0,405
	Mullus barbatus	0,551	0,028	0,016	0,099
	Pomatomus saltatrix	0,182	0,011	0,025	0,049
	Penaeus indicus	2,76	0,051	0,094	0,015
	Sparus auratus	0,118	0,027	0,019	0,077
Mediterranean	*Pomatomus saltatrix*	0,076	0,008	0,012	0,027
	Merlangius euxmus	0,272	0,012	0,048	0,071
	Sparus auratus	0,193	0,035	0,086	0,101
	Alosa fallax	0,298	0,044	0,013	0,086
	Merluccius merluccius	0,843	0,038	0,035	0,108
	Mugil soiuy	0,316	0,012	0,008	0,051
	Mytilus galloprovincialis	1,38	0,112	0,203	0,366
	Mullus barbatus	0,773	0,064	0,032	0,104
	Penaeus indicus	1,82	0,091	0,014	0,068
	Dicentrarchus labrax	0,116	0,015	0,006	0,032
	Scophthalmus maximus	0,443	0,076	0,018	0,045

detected as higher than the limits in two samples obtained from the Mediterranean Sea, *Scophthalmus maximus* and *Mullus barbatus* species as 0,076 mg/kg and 0,064 mg/kg, respectively.

Results of Hg showed that mussels are the most contaminated samples in the Black, Marmara and Mediterranean Sea, while the shrimp was in the Aegean Sea. Within the fish species *Merlangius euxmus* in the Aegean and Black Sea, *Merluccius merluccius* and *Sparus auratus* in the Marmara and Mediterranean Sea has the highest levels of Hg, respectively. It was found that, none of the samples have higher levels than the Turkish Food Codex and European Commission Regulation limits.

According to the lead data, mussels were found to have the highest contamination levels in all regions. Highest levels of lead were detected in *Scophthalmus maximus* species from the Black, Marmara and Aegean Sea. *Merluccius merluccius* was found the most contaminated fish species in the Mediterranean Sea. None of the samples showed higher levels than the Turkish Food Codex and European Commission Regulation limits.

DISCUSSION

Studying the concentration of heavy metals in marine organisms, provides an opportunity to observe the mechanism of bioaccumulation, environmental pollution and to assess the effects on public health. It is known that, muscles of the marine organisms are not the active parts of accumulation. But because they are edible parts and showed exceeded levels in the studies, we preferred muscle for the analysis. In our study toxic elements like As, Cd, Pb and Hg was detected in some fish species, mussels and shrimps obtained from 3 seas of Turkey.

The metal concentrations was found in the same order in all seas as, As>Pb>Hg>Cd. In compliance with the other studies, Bat et al. (2012), Damino et al. (2011), Monikh et al. (2013) and Turkmen et al. (2009). Cd was reported as the lowest contaminant.

Statistically significant differences were found in the metal levels between fish species and the shellfish in all regions, according to the student t test ($p<0,05$). It is known that mollusks are filter feeders, they adsorb high doses of contaminants from the environment they lived in. Also, especially mussels were lived near to the coastal so they are affected much more by coastal contamination such as boat sewage and draining of sewerage. Similarly, Culha et al. (2007) detected a significant difference in the metal levels between mussels, sea snails and the fish species. Also in Copat et al. (2013) study Cd, Cr, Pb, Mn, Ni and V levels found higher in *Donax trunculus* then the other fish species. Hussein and Khaled (2014) compared the levels of metals of two mussels (*Pinctada radiate* and *Paphia textile*) with the other fish species they detected. They reported the levels of all metals they analyzed higher in bivalves than the fish species. Their study also revealed that the large bivalves recorded higher values for Fe, Zn, Mn, Cu, and Cd; whereas the small ones recorded highest levels for Cr, Ni and Pb.

Arsenic is a naturally occurring ubiquitous, highly mobilized element and mainly cycled by water in the environment. Because of this character fish and the other marine organisms are possible contamination routes for food chain. According the results of our study, As was the highest contaminated metal in all samples, in all regions. It is well known that, factors such as seasonal cycle of absorption, temperature and salinity might enhance the large bioaccumulation of As. Also, previous findings suggest that non-toxic As was accumulated more in marine organisms then the toxic form coming from anthropogenic activities. Because of the complexity in differantiating the toxic and non-toxic forms, no specific limits can be determined for As. There is still not a limit for seafood, in Turkish Food Codex and European Commission Regulations (Anon 2016b). In Bilandzic et al. (2011) study, four fish species from the Adriatic Sea, detected for arsenic and the concentrations ranged from 0.01 to 70.9 mg/kg. Tuzen (2009) reported the arsenic level in muscles of fishes in the Black Sea, ranged from 0.11 to 0.32 mg/kg. In Spain, lower levels of arsenic were reported in the following ranges (μg/g): 15.39–17.77 in red mullet, 3.93–5.42 in anchovy and 1.73–7.47 in mackerel (Falcó et al., 2006). Gorur et al. (2012) revealed the highest and lowest mean arsenic concentrations of *T. mediterranus* and *M. merlangus* as 4.40 μg/g and 1.32 μg/gdw.

One of the most important properties of Cd is to be biomagnified in the environment. From the upper parts of the food chain, the level of Cd accumulation getting higher. More developed species accommodate more Cd in their systems (Velusamy et al., 2014). Trace metal content of nine fish species harvested from the Black and Aegean Sea were determined in Uluozlu et al. (2007) study, the lowest cadmium content was found 0.45 μg/g in *M. cephalus* and *M. barbatus*, while the highest cadmium content was found 0.90 μg/g in *S. sarda*. Cd concentrations have been reported as 0.02–0.24 mg/kg for muscles of fish from the Black Sea coasts (Topcuoglu et al., 2002). Damino et al. (2011) reported that, the average concentrations did not exceed 0.30 μg/g in fishes from the Mediterranean and Atlantic area. In our study levels were detected as higher than the limits in two samples, *Scophthalmus maximus* and *Mullus barbatus* species obtained from the Mediterranean Sea. The Mediterranean Sea is surrounded by some of the most populated and industrialized countries in the world, and it is almost a closed basin. This cause a regular and constant pollution of the sea by toxic compounds. These may be the reasons that the Mediterranean fishes tend to exhibit high levels of heavy metal than those of populations inhabiting other areas (Storelli et al., 2005). Also in Mendil et al. (2010) study, Cd levels was found higher than the recommended levels in Atlantic bonito red mullet, mackerel and whiting. Tuzen et al. (2009) also have compatible results with our study, they reported high levels of

Cd than the legal limits, from the different fish species caught from the Black Sea. In Turkmen et al. (2008) study, Cd levels of twelve fish species from the Mediterranean, Marmara, Aegean Sea's was also found higher than the permissible limits.

In our study in the Black and Marmara Sea, *Engraluis encrasicolus* has the highest Cd levels inside the fish species. This is considerable cause, in Turkey this species has the big percentage of the fishing with 179 615 tons (Anon, 2016a), and being cheap and widespread makes this species the most preferred one. Similarly in Nisbet et al. (2010) study, *Engraluis encrasicolus* was found to have the higher level for Cd.

Mercury is one of the most toxic heavy metal in the environment and generally human exposure to mercury through the consumption of seafood that may contain methyl-mercury in their tissues. We found that, none of the samples detected in this study, have higher Hg concentrations than the Turkish Food Codex and European Commission Regulation limits. Differently from our study, Nisbet et al. (2010) reported that, they were not detected Hg from the fishes they analyzed from the Black Sea. But in Tuzen et al. (2009) study, mercury levels were found between 0.025-0.084 µg/kg in the fish species obtained from the Black Sea. Harmelin-Vivien et al. (2009) indicated the level of mercury in red mullets from the Mediterranean and Black Sea's as 0.16 µg/kg. These differences can be the result of the methods that used for the analyses, as we mentioned above there were different methods which have different detection limits and sensitivity.

Lead is a toxic element causes carcinogenic effects in marine organisms and humans. Uluozlu et al. (2007) reported the levels of Pb between 0.33-0.93 mg/kg for the fishes they analyzed from the Black and Aegean Sea. In another study levels was found to be in a range of 0.22-0.85 mg/kg for muscles of fish from the Black Sea (Tuzen et al., 2003). From a study conducted in Portugal 0.01-0.15 mg/kg Pb determined in the muscles of fish (Cid et al. 2001). In contrast with these results, in Mendil et al. (2010) and Tuzen et al. (2009) studies, lead levels were reported above the legal limits. In our study, *Merluccius merluccius* was found the most contaminated fish species with Pb, in the Mediterranean Sea. Likewise, in Findik and Cicek (2011) study, it was determined that *Merluccius merluccius* has a higher mean of metal concentration when compared with the other species. But in the study authors indicated that, the levels of Pb was at least 6 times higher than the Turkish Food Codex for *M. barbarous* and 34 times for *M. merluccius*. Copat et al. (2012) was reported that the amount of Pb found in *E. encrasicolus* from the Mediterranean Sea exceeded the limits set by the EC Regulation.

CONCLUSION

The results obtained from this study revealed that, except in a few cases, the selected heavy metal concentrations in most samples were below the limits. Also, instead of the mussels and the shrimps, there was no single type of fish that was consistently high from all metals. The examined seas and the seafood were found to be safe for human consumption.

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