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Toxic Metals in Seven Commercial Fish from the Southern Black Sea: Toxic Risk Assessment of Eleven-Year Data Between 2009 and 2019

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Abstract

Concentrations of toxic metals (Cd, Hg and Pb) in the edible tissues of seven commercial fish species (*Mullus barbatus*, *Merlangius merlangus*, *Scophthalmus maximus*, *Mugil cephalus, Engraulis encrasicolus*, *Trachurus mediterraneus* and *Sarda sarda*) collected from Sinop coasts of the southern Black Sea were detected in 11 years. In several fish samples, the concentrations of elements (Cd, Hg and Pb) were not detected or were below the detection level. The present study showed that Pb was the most and Cd was the least accumulated metal in the studied fish species. The concentrations of those metals are below the international organizations' recommended limit. It was shown that the estimated metal dose values for daily average consumption and hazarded quotients (HI) in fish samples are below safety levels for human consumption (HI<1). From the human health point of view, this study showed that there was no possible health risk to people due to intake of any studied species under the current consumption rate in the country for 11 years. This study could be useful as a baseline data for metals exposure.

Keywords Toxic metals · Commercial fish · Black Sea · Estimated daily intakes · Hazard quotient

Introduction

Fish are healthy foods with high nutritional value for humans [1]. It is considered to be a good source of high biological value protein, which contains a good balance of good amino acids and fats, and highly useful polyunsaturated fatty acids. In addition, fish are easily digestible and a good source for most B-complex vitamins and contain important minerals. Due to their good taste and low cost compared to red meat, fish provide the opportunity for consumers to meet their daily nutritional needs as an animal protein source [2]. Despite their known benefits, fish can biologically accumulate non-essential toxic metals, which can pose potential people health risks to consumers even at small quantities.

It is well known that environmental problems come to the forefront in parallel with both industrial and population growth [3]. The amounts of toxic heavy metals in marine coastal environment and marine organisms especially fish have been of notably concern owing to their toxic effects which are threats in human beings. Heavy metals have the tendency to accumulate in fish, which in turn may enter the human metabolism due to consumption causing major health risks. Cd and Hg are the very toxic metals, followed by Pb. They are non-essential metals, as they are toxic, even in trace amounts, due to its high toxicity, bio-accumulative properties, and deleterious effects on biota [4, 5]. Fish can easily accumulate toxic metals, especially Cd, Hg and Pb, which are among the most important of these contaminants [6].

The Black Sea accumulates terrestrial contaminants, mostly through rivers. However, it accumulates many contaminants in the Black Sea by means of industrial, shipping, agricultural, domestic, touristic, harbour and fishing activities on its coasts. Fish are exposed to contaminants such as toxic metals in polluted waters. Toxic metals from the anthropogenic activities are continually entered into the marine ecosystem [7]. They are major health risks owing to of their toxicity, bioaccumulation, biomagnifications and long-persistence in

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the food chain. For example, mercury, cadmium and lead are highly toxic and may be reason for mental and central nervous system detriment. It is therefore necessary to detect and observe metal amounts in seafood, because metal ions can readily accumulate in fish more than those in other foodstuffs. Non-essential metals are toxic to fish even at very low concentrations. As a result of the risks associated with the consumption of metals, in order to assess the potential risks associated with the consumption of contaminated fish, it is necessary to periodically evaluate their level of concentration in the commercial fisheries of Turkey [8].

Although Sinop province is small, it is an important fishing centre [9, 10]. The ability of fishes to concentrate heavy metals in their muscles is well known. Considering the above information, we selected muscles as the primary site of metal accumulation in this study, and since fish are an essential component of human diet, they need to be carefully screened to ensure that excessive high heavy metal levels are not transmitted through ingestion to people. The present study is related to the monitoring of toxic metal distribution in the seven commercial fish from the southern Black Sea between 2009 and 2019 and to evaluate them in terms of human health. The daily intakes of the toxic metals from the consumption of the fish samples were also estimated for infant, children and adults, and hazard quotients (HIs) in fish samples were calculated to determine the health risks due to the consumption of commercial fish. In addition, the results were compared with values permitted by national and international organizations. In all Turkish seas particularly in the Black Sea, many studies have been conducted concerning metal accumulation in fish. The difference between this study and the others is that it is a regular monitoring study for 11 years.

Materials and Methods

Study Area

Sinop Peninsula is a natural port and an attractive centre for fishermen (Fig. 1). Since the coasts of Sinop have few settlements and no industrial activity, these coasts are regarded as clean region [11]. An important part of the Southern Black Sea fishing is obtained from here. Not only fishermen in Sinop but also fishermen in other Black Sea provinces catch fish from here. For 11 years, samples of seven fish species that went from commercial fishing boats for sale to fishing markets in Sinop of the southern Black Sea were obtained for toxic metal analysis. Fish samples were taken every year during the fishing season. T.R. the Ministry of Food, Agriculture and Livestock [12] has determined the minimum sizes of 9 cm for anchovy (*Engraulis encrasicolus*); 13 cm for red mullet (*Mullus barbatus*), Mediterranean horse mackerel (*Trachurus mediterraneus*) and whiting (*Merlangius merlangus*); 20 cm for flathead grey mullet (*Mugil cephalus*); 25 cm for Atlantic bonito (*Sarda sarda*) and 45 cm for turbot (*Scophthalmus maximus*) [13].

Metal Analysis

For toxic metal analysis, sampled fish individuals from each species were measured and rinsed in clean sea water. All

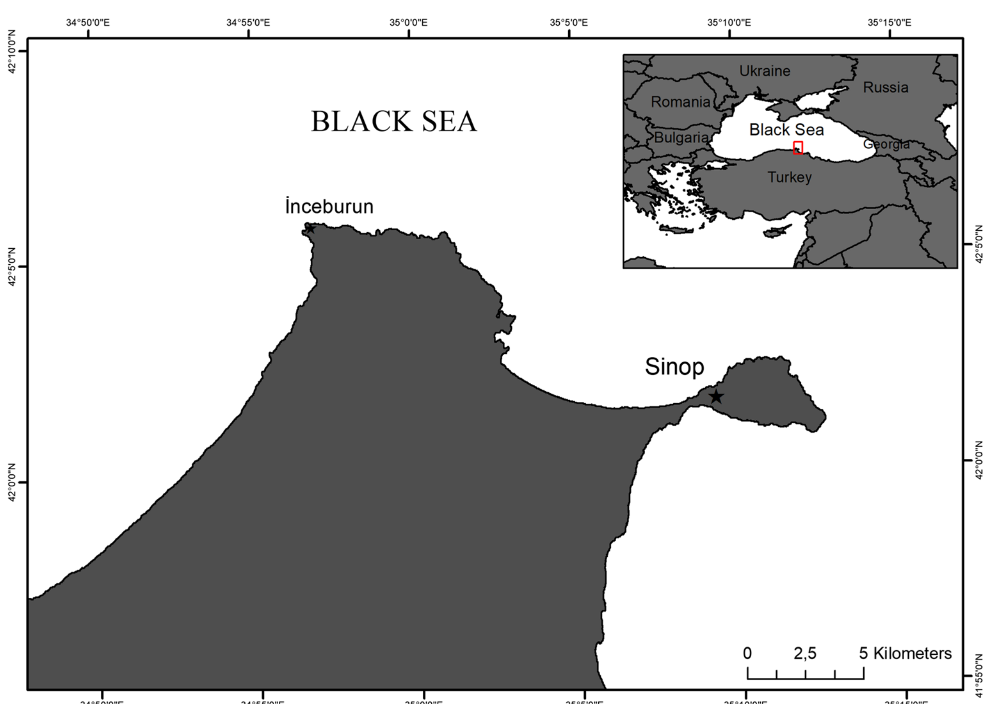


Fig. 1 Fish catching area

samples were stored deep frozen at −21 °C until their analysis. In each sampling, at least 20 specimens for anchovy, red mullet, Mediterranean horse mackerel and whiting; 10 for flathead grey mullet; 5 for Atlantic bonito and 3 for turbot were used. Metal analyses in fish tissues were measured by accredited laboratories with ICP/MS by applying TURKAK Test TS EN ISO/IEC 17025 NMKL 161, 170, and 186 methods. The measurement limit values of metals are 0.002 for cadmium, 0.001 for mercury and 0.005 for lead.

Health Risk Assessment

Risk from metal intake through ingestion may be characterized using a hazard index (HI) as the ratio of the estimated metal dose (EDI mg/kg of body weight per day) and the reference dose (Rf. D mg/kg year). The R_fD values for Hg and Cd are 0.0003 and 0.001 mg/kg day^{-1}, respectively [14]. R_fD is not available for Pb. The U.S. Department of Health and Human Services Public Health Service [15] pointed out that it would be unsuitable to develop a R_fD for inorganic Pb and its compounds because some of the sanitary impacts related with exposure to Pb happen at blood Pb levels as low as to be essentially without a threshold [16]. Therefore, the R_fD value for Pb in this study was 0.0035 as used by many researchers [17–19]. The HI was calculated by using the equation below:

$$\mathrm{HI} = \frac{\mathrm{EDI}}{\mathrm{Rf.D}}$$

If HI > 1.0, then the EDI of a particular metal exceeds the Rf. D, indicating that there is a potential risk associated with that metal. The estimated daily intake (EDI) depends on both the metal concentration and the amount of consumption of fish. The Turkish Statistical Institute reported that the average amount of fish consumed per capita between 2009 and 2019 was changed as 5.4 to 7.6 kg per year [12]. However, this amount is higher in people living in the cities on the Black Sea coast [1]. The EDIs of metals were determined using the following equation:

$$\mathrm{EDI} = \frac{C_{\mathrm{metal}} \times W}{m}$$

where C_{metal} is the concentration of metals in fish; W represents the daily average consumption of fish is given as: 0.013, 0.027 and 0.041 kg/day for infants, children and adults, respectively [20]; and m is the body weight of 10 kg for infants, 30 kg for children and 70 kg for adults.

Statistical Analysis

The samples were analysed in triplicate and the average results were used to represent the data. The Durbin-Watson test was used to measure of autocorrelation in residuals from regression analysis. Autocorrelation is the similarity of a time series over successive time intervals. The significance was set at 0.05 and statistical software was performed by IBM SPSS-21 to examine the effect of years and the metal concentrations in fish species. Microsoft Excel 2010 was used for conducting other calculations. All values were being expressed on mg/kg wet wt. basis.

Results and Discussion

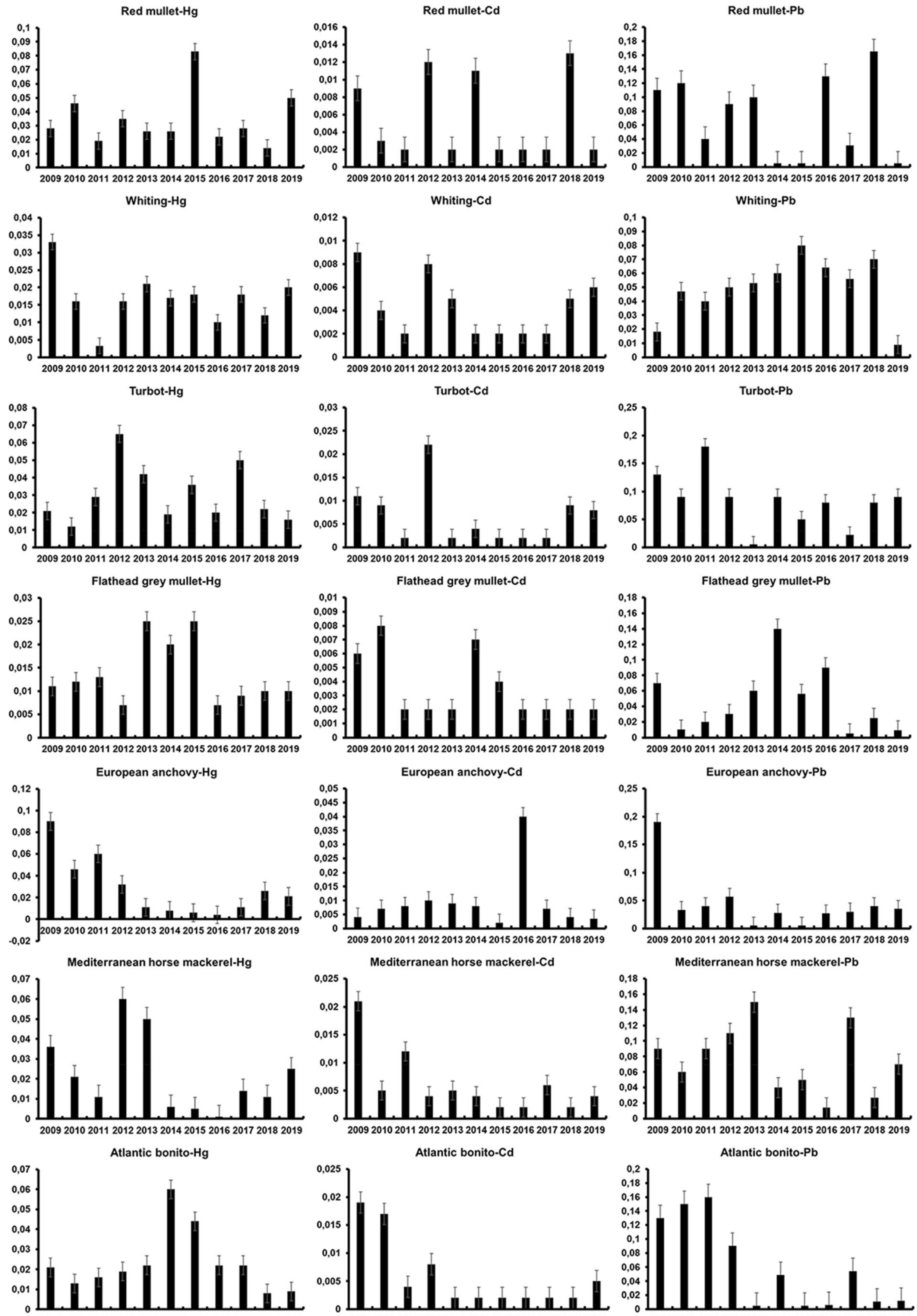
The concentrations of toxic metals (Hg, Cd and Pb) in muscle tissue of seven commercial fish species under this study for 11 years are presented in Fig. 2. There were significant differences in toxic metal levels between fish species. It revealed that the order of mean concentrations of the toxic metals in the muscles of all fish species is recorded as follows: Pb > Hg > Cd.

Toxic metals are not necessary for living metabolism at any level. Even the lowest levels of these metals can harm living organisms and indirectly humans through food. Toxic metals (Cd, Hg and Pb) therefore have been included in the hazardous metals' legislation of the European Union and Turkish Food Codex. The European Commission Regulation [21] and Turkish Legislation [22] indicate that maximum levels of Pb, Hg and Cd are 0.30, 0.50 and 0.05 mg/kg wet wt., respectively. In this study, no toxic metal value exceeded the allowable values in the fish species studied (Fig. 2).

Hg concentrations in the muscle tissues of seven commercial fish species from the southern Black Sea were detected from 0.0033 to 0.083 mg/kg wet wt. Maximum Hg levels were found in red mullet (0.083 mg/kg wet wt.) in 2015, followed by a turbot with 0.065 mg/kg wet wt. in 2012. Similarly, the mean Pb value was the highest in turbot and red mullet, whereas the lowest was found in anchovy. Based on the increasing Hg and Pb values in benthic species, it may indicate that the biggest difference occurs between pelagic and benthic species. Although they are non-migratory and benthic species, different Hg and Pb accumulations were observed in their tissues. These differences may have resulted from the variation in their diet and accumulation strategies. Generally, the benthic species accumulated higher concentrations of the Hg, due to the greater exposure to Hg-enriched sediment and interactions with benthic organisms in the enriched sediment.

Although there is no industrial pollution in Sinop, organic material originating from domestic wastes and land-based pollution is dumped on the shores [23]. Hg concentrations were found relatively higher enrichment values than other metals. It is assumed that the high enrichment factor values show an anthropogenic source of heavy metals [24]. They concluded that Hg may be transported from the atmosphere as vapour then to the coastal environment.

According to the mean metal values, the highest Cd level was found in anchovy followed by turbot and Mediterranean


Red mullet-Hg
Red mullet-Cd
Red mullet-Pb
Whiting-Hg
Whiting-Cd
Whiting-Pb
Turbot-Hg
Turbot-Cd
Turbot-Pb
Flathead grey mullet-Hg
Flathead grey mullet-Cd
Flathead grey mullet-Pb
European anchovy-Hg
European anchovy-Cd
European anchovy-Pb
Mediterranean horse mackerel-Hg
Mediterranean horse mackerel-Cd
Mediterranean horse mackerel-Pb
Atlantic bonito-Hg
Atlantic bonito-Cd
Atlantic bonito-Pb
2009 2010 2011 2012 2013 2014 2015 2016 2017 2018 2019

◀ **Fig. 2** Mean with standard deviations (vertical lines) toxic metal concentrations (mg/kg wet wt.) in the muscle tissues of the commercial fish species from the southern Black Sea by years

horse mackerel and the lowest in flathead grey mullet and whiting. This difference may be due to the pelagic fish feeding through different water bodies.

While fish are an important source of protein, rapid industrialisation and mechanized farming practices affect fish, leading to an increase in the concentration of toxic metals in fish. Depending on their feeding habits and habitats, age, and size of the fish, metal concentrations can differ between species. Ecological requirements, metabolism and feeding patterns, behaviour and body size are suggested to be the result of different metal levels in different species of fish.

In the study, the Durbin-Watson test was applied to show whether there is "autocorrelation" on 11-year data. This is done in 3 steps. In the first step, the normality test of the data was performed for homogeneous distribution. If the data is normal, Shapiro-Wilk (Sig.) is used; if not, a logarithmic transformation is made. Therefore, values less than 0.05 have been converted as Log. Then, ANOVA was applied for these data. In the second step, the regression test was performed and R^2 values greater than 0.4 were determined. In the 3rd step, the values that should be meaningful, that is, p^2 less than 0.05, were calculated. Results are presented in Table 1.

The Durbin-Watson (DW) test was applied to the data that provided the above three steps. If the values found are between $1.5 < DW < 2.5$, there is no "autocorrelation", i.e. no trend here. If DW results are $DW < 1.5$, there is a positive correlation between residues, meaning that trend metal levels have increased over the years. If the DW results are $2.5 < DW$, there is a negative correlation between residues, which means that trend metal levels have decreased over the years. Accordingly, annual decreases of levels were observed for Hg in anchovy, Cd in Mediterranean horse mackerel and Pb in Atlantic bonito (Table 1). No correlation could be found in trend analysis for other fish species.

It is important to investigate these yearly fluctuations. In the North Sea, the Sound and the Great Belt, the time trend analysis (*Pleuronectes platessa* and *Platichthys flesus*—11 years) showed a recent decrease in Pb concentrations, whereas the yearly variation among the Cd, Cu, Hg and Zn concentrations was high [25]. Polak-Juszczak reported that there was a downward trend in the contents of Pb, Hg and As, an upward trend which was noted in Zn and no change trends in Cu and Cd in Baltic fish between 1991 and 1997. It was stated that differences of the heavy metal content in tissue depend on the fish species as well as on their habitat [26].

Daily Intake of Metals and Hazard Quotient

This study was also conducted for the fish muscle as this tissue was the most important part consumed by people. According to Turkish the Ministry of Agriculture and Forest General Directorate of Fisheries and Aquaculture [12], estimates of fish consumption in Turkey indicated that the adult population consumes 21 g wet wt./person/day of fish species. This amount is higher for the coastal cities of the Black Sea, especially Sinop [8]. The EDI (estimated daily intake) values presented in Table 2 were estimated by assuming that infants, children and adults will consume 0.013, 0.027 and 0.041 kg fish per day, respectively. The result revealed that the EDI values for the examined fish samples were below the recommended values, indicating that health risk associated with the intake of these toxic metals through the consumption of seven commercial fish species was absent.

The daily intakes (EDI) of the metals were estimated as the means of Hg, Cd and Pb in commercial fish species and the mean consumption of fish per day for infants, children and adults, respectively, as reported by the UNSCEAR [20]. Table 2 shows that EDIs of toxic metals (Hg, Cd, Pb) are much below the recommended values of the FAO (2004) [27]. Estimated hazarded quotients (HI) of the metals suggest that these toxic metals in fish samples do not pose any apparent threat to the population, where the HIs of all the considered metals were below the value of 1 (US-EPA) as shown in Table 2.

Comparison of the Results with the Available Literature

Comparisons were made with heavy levels found in the literature for the studied commercial fish species from elsewhere, where possible, on those from different waters in the Black Sea (Table 3).

Table 1 Trend analysis chart

Fish (metal)	Normality test Shapiro-Wilk (Sig.)	R^2	ANOVA[a] p^2	Durbin-Watson	Metal	Trend
E. encrasicolus (Hg)	0.042	0.499	0.015	0.879	−0.006	↓
T. mediterraneus (logCd)	0.154	0.445	0.025	2.394	−0.065	↓
S. sarda (Pb)	0.025	0.504	0.014	1.095	−0.001	↓

Table 2 Estimated daily intakes (EDI) and hazard index (HI) of elements in edible tissues of fishes from the southern Black Sea

Species	Toxic metals	EDI (2009–2019) mg/day/kg body wt.			HI		
		Infants	Children	Adults	Infants	Children	Adults
Engraulis encrasicolus	Hg	0.00003822	0.00002646	0.00001722	0.1274	0.0882	0.0574
	Cd	0.000012883	0.000008919	5.80443E–06	0.012883	0.008919	0.005804429
	Pb	0.00005915	0.00004095	0.00002665	0.0169	0.0117	0.007614286
	Total				0.157183	0.108819	0.070818714
Trachurus mediterraneus	Hg	0.00002795	0.00001935	1.25929E–05	0.093166667	0.0645	0.04197619
	Cd	0.00000819	0.00000567	0.00000369	0.00819	0.00567	0.00369
	Pb	0.00009893	0.00006849	4.45729E–05	0.028265714	0.019568571	0.012735102
	Total				0.129622381	0.089738571	0.058401293
Sarda sarda	Hg	0.00003211	0.00002223	1.44671E–05	0.107033333	0.0741	0.04822381
	Cd	0.0000078	0.0000054	3.51429E–06	0.0078	0.0054	0.003514286
	Pb	0.0000858	0.0000594	3.86571E–05	0.024514286	0.016971429	0.011044898
	Total				0.139347619	0.096471429	0.062782993
Mullus barbatus	Hg	0.00004251	0.00002943	1.91529E–05	0.1417	0.0981	0.063842857
	Cd	0.00000754	0.00000522	3.39714E–06	0.00754	0.00522	0.003397143
	Pb	0.00010348	0.00007164	4.66229E–05	0.029565714	0.020468571	0.013320816
	Total				0.178805714	0.123788571	0.080560816
Merlangius merlangus	Hg	0.000021359	0.000014787	9.62329E–06	0.071196667	0.04929	0.032077619
	Cd	0.00000533	0.00000369	2.40143E–06	0.00533	0.00369	0.002401429
	Pb	0.00006994	0.00004842	3.15114E–05	0.019982857	0.013834286	0.009003265
	Total				0.096509524	0.066814286	0.043482313
Scophthalmus maximus	Hg	0.00004108	0.00002844	1.85086E–05	0.136933333	0.0948	0.061695238
	Cd	0.00000845	0.00000585	3.80714E–06	0.00845	0.00585	0.003807143
	Pb	0.00010621	0.00007353	4.78529E–05	0.030345714	0.021008571	0.013672245
	Total				0.175729048	0.121658571	0.079174626
Mugil cephalus	Hg	0.00001807	0.00001251	8.14143E–06	0.060233333	0.0417	0.027138095
	Cd	0.00000481	0.00000333	2.16714E–06	0.00481	0.00333	0.002167143
	Pb	0.00006578	0.00004554	2.96371E–05	0.018794286	0.013011429	0.008467755
	Total				0.083837619	0.058041429	0.037772993

Hg in Fish Species

Hg is very well known as one of the most toxic metals to humans and fishes. The average levels of Hg revealed in the edible tissues of the seven commercial fish studied were found to be low. Compared to other metals, Hg has been less studied as it is more expensive to measure and takes a lot of work. The amount of Hg in *Engraulis encrasicolus* was found to be less in the current study compared to other studies [28, 69]. The Hg levels in *Trachurus mediterraneus* and *Sarda sarda* were found to be much higher on the Bulgarian coasts [48, 50].

Although Hg levels in *Merlangius merlangus* were the same in this study [46, 57, 58, 62], it was higher in other studies [28, 55, 57, 63, 65].

Hg levels in *Mullus barbatus* were found to be high in the studies conducted by Ergül and Aksan, Özden et al. [43, 55].

Hg levels in *Mugil cephalus* were found to be the lowest in the present study and the highest was found in the study conducted by Stancheva et al. [53] in Varna Lake.

Hg levels in *Scophthalmus maximus* were found to be the lowest in the study conducted by Bat et al. [46] and the highest in the study conducted by Tüzen [28].

Cd in Fish Species

Cd is also known to be one of the most dangerous metals to people. The mean levels of Cd revealed in this study are different but low in all fish species (Fig. 2). In the scientific literature in Table 1, similar Cd values were recorded in the muscles of *E. encrasicolus* from the Black Sea coasts [30, 33, 35], while low levels of Cd were found in the muscles of *E. encrasicolus* from Rize coasts in the Black Sea [40]. On

Table 3 Comparison of the amounts (mg/kg wet wt.) of toxic metals in the edible tissues of seven commercial species from the southern Black Sea coasts between 2009 and 2019

Sp.	Location	Metals			References
		Pb	Cd	Hg	
Engraulis encrasicolu	BS	0.30±0.02	0.27±0.02	0.055±0.003	[28]
	İstanbul	0.0658	0.0248	–	[29]
	Western BS	0.14	0.007	ND	[30]
	Samsun	0.08	0.04	–	[31]
	BS	0.004	–	–	[32]
	Sinop	0.09–0.26	0.09–0.17	–	[33]
	Southeastern BS	0.03	0.3	–	[34]
	Sinop	0.082	0.0074	–	[35]
	Southeastern BS	ND	ND	ND	[36]
	Crimea	0.003–3.4	–	–	[37]
	Samsun	4.07±0.70	0.06±0.01	–	[38]
	Sinop	2.98±0.63	0.19±0.17	–	[38]
	Kocaeli	4.58±0.54	0.08±0.06	–	[38]
	BS	0.078–0.284	0.004–0.054	–	[39]
	Rize	0.0028–0.006	0.0004–0.0006	–	[40]
	Romania	0.0184–0.1492	0.0148–0.0716	–	[41]
	Giresun	4.87±1.04	0.15±0.05	–	[42]
	Trabzon	3.85±0.57	0.04±0.02	–	[42]
	Rize	2.99±0.37	0.31±0.15	–	[42]
	Sinop	0.055	0.033	0.09	[19]
Scophthalmus maximus	BS	0.28±0.02	0.10±0.01	0.045±0.002	[28]
	Southeastern BS	0.146	0.004	–	[30]
	BS	0.004	0.002	–	[43]
	Kocaeli	–	0.002	–	[43]
	Sinop	0.084	0.0042	–	[35]
	Sinop	0.08	0.03	–	[44]
	Sinop	<0.05	<0.02	–	[45]
	Sinop	0.07±0.005	0.011±0.004	0.017±0.003	[46]
Trachurus mediterraneus	Sinop	0.17–0.23	0.043–0.048	–	[47]
	BS	<0.001	–	–	[32]
	Southeastern BS	0.25	0.02	–	[34]
	Bulgaria	0.06±0.01	0.008±0.001	0.16±0.02	[48]
	Sinop	<0.05	<0.02	<0.05	[24]
	Romania Constanta	<LOD 0.1023±0.0452	0.0163±0.0075 0.0338±0.0004	–	[49]
	Bulgaria	0.06±0.01	0.008±0.001	0.16±0.02	[50]
Sarda sarda	BS	0.61±0.04	0.13±0.01	0.025±0.002	[28]
	BS	0.056	0.07	–	[51]
	Western BS	0.18	0.005	ND	[30]
	Sinop	0.13–0.19	0.023–0.028	–	[47]
	Kocaeli	–	0.004	–	[43]
	Samsun	0.35±0.07	0.05±0.01	–	[38]
	Sinop	0.29±0.04	0.13±0.03	–	[38]
	Kocaeli	0.25±0.06	0.11±0.02	–	[38]
	Kastamonu	5.48±0.56	0.20±0.01	–	[52]
	Bulgaria	0.06±0.01	0.015±0.002	0.130±0.02	[50]
	Giresun	0.52±0.23	0.04±0.00	–	[42]

Table 3 (continued)

Sp.	Location	Metals			References
		Pb	Cd	Hg	
	Trabzon	0.37±0.20	0.04±0.01	–	[42]
	Rize	0.29±0.09	0.04±0.00	–	[42]
Mugil cephalus	BS	0.68±0.05	0.35±0.03	0.070±0.004	[28]
	Sinop	0.09–0.19	0.02–0.03	–	[47]
	Varna Lake	0.07±0.01	0.024±0.002	0.08±0.01	[53]
	Nessebar	0.05±0.01	0.012±0.002	0.05±0.01	[53]
	Bulgaria	0.05±0.01	0.012±0.002	0.05±0.01	[48]
	Bulgaria	0.05±0.01	0.012±0.002	0.05±0.01	[50]
	Giresun	0.02±0.01	0.57±0.27	–	[42]
	Rize	0.90±0.32	0.30±0.17	–	[42]
Merlangius merlangus	Sinop-Samsun	<0.05	<0.02	<0.05	[54]
	İstanbul	0.1	0.038	–	[29]
	BS	0.53±004	0.21±0.02	0.084±0.005	[28]
	Southeastern BS	0.092	0.036	–	[51]
	Southeastern BS	0.116	0.0004	ND	[30]
	İstanbul	0.004–1.581	0.001–0.151	0.003–0.491	[55]
	Samsun	ND-0.18	ND-0.04	–	[31]
	Western BS	6.80±5.88	0.40±0.29	–	[56]
	Sinop	0.004	0.016–0.036	–	[47]
	Terkos	3	0.07	0.014	[57]
	Sakarya	2.4	0.048	<0.01	[57]
	Bafra	3	0.014	0.018	[57]
	Ordu	2.6	0.044	0.1	[57]
	Trabzon	0.016	0.008	0.01	[58]
	BS	0.002	–	–	[32]
	BS	0.01	0.006	0.066	[43]
	Eastern BS	0.0048	0.0062	–	[59]
	Sinop	0.138	0.0054	–	[35]
	Trabzon	0.02±0.00	4.05±0.14	–	[60]
	Sinop	<0.05	<0.02	<0.05	[61]
	Samsun	1.41±0.23	0.06±0.02	–	[38]
	Sinop	0.63±0.06	0.05±0.003	–	[38]
	Kocaeli	0.69±0.12	0.06±0.01	–	[38]
	Kastamonu	6.12±1.45	0.24±0.02	–	[52]
	Giresun	0.05±0.00	0.66±0.08	–	[42]
	Southwestern BS	0.36±0.42	0.02±0.01	0.01±0.01	[62]
	Trabzon	1.30±0.31	0.12±0.03	–	[42]
	Rize	1.29±0.21	0.08±0.02	–	[42]
	BS	0.099	0.013	0.081	[63]
	Sinop	0.03–0.09	0.007–0.0085	0.01–0.017	[46]
	Sinop	1.17±1.01	0.02±0.01	–	[64]
	Kastamonu	1.18±0.45	0.03±0.01	–	[64]
	Zonguldak	0.86±0.34	0.03±0.01	–	[64]
	Sinop	0.19±0.02 0.90±0.28	0.03±0.00 0.22±0.03	0.13±0.01 0.23±0.00	[65]
Mullus barbatus	İstanbul	0.145	0.0416	–	[29]
	BS	0.36±0.03	0.17±0.02	0.036±0.002	[28]

Table 3 (continued)

Sp.	Location	Metals			References
		Pb	Cd	Hg	
	Romania	–	–	0.035±0.011 (0.021–0.072)	[66]
	Sinop	0.0105	<0.02	<0.05	[54]
	Samsun	0.0163	<0.02	<0.05	[54]
	Southeastern BS	0.08	0.046	–	[51]
	Southeastern BS	0.184	0.004	–	[30]
	Southeastern BS	0.025–0.355	0.002–0.266	0.066–1.592	[55]
	Western BS	1.11±1.60 (0.09–7.00)	0.11±0.13 (0.02–0.55)	–	[56]
	Trabzon	0.02	0.004	0.022	[58]
	Sinop	0.018–0.062	0.004	–	[47]
	BS	0.004	0.004	0.094	[43]
	Sinop	0.164	0.007	–	[35]
	Eastern BS Turkey	0.004	0.0036	–	[59]
	Turkey	<LOD	3.38±0.06	–	[60]
	Romania	0.32±0.25	0.026±0.001	–	[67]
	Sinop	<0.05	<0.02	<0.05	[24]
	Samsun	1.76±0.40	0.20±0.11	–	[38]
	Sinop	2.94±0.81	0.07±0.02	–	[38]
	Kocaeli	0.88±0.12	0.06±0.005	–	[38]
	Kastamonu	7.21±1.56	0.28±0.03	–	[52]
	Sinop	0.022–0.09	0.006–0.038	–	[39]
	Southwestern BS	0.03–1.70	0.02–0.05	0.01–0.03	[62]
	Giresun	0.45±0.05	0.04±0.00	–	[42]
	Trabzon	1.03±0.10	0.12±0.03	–	[42]
	Rize	1.30±0.16	0.09±0.02	–	[42]
	Ordu	0.162–0.308	0.16–0.182	–	[68]
	BS	0.165	0.016	0.032	[63]
	Sinop	0.025–0.06	0.007–0.011	0.015–0.021	[46]

BS, Black Sea

the other hand, some studies have shown high levels of Cd in the muscles of anchovy in the Black Sea coasts [28, 34, 42].

Cd levels were generally found to be low in the muscles of *T. mediterraneus*. The lowest values were obtained from this study, followed by the Bulgarian coasts [48, 50].

The findings of this study were found to be the least in Cd levels in the muscles of *S. sarda* and were similar to those of Nisbet et al. [30] and Ergül and Aksan [43]. The highest value was found in Atlantic bonito with 0.20 ± 0.01 mg/kg wet wt. on the Kastamonu coasts of the Black Sea [52].

Cd levels in the muscles of *M. merlangus*, which are frequently used in monitoring studies, were found to be very different in different studies. The highest value was found in whiting with 4.05 ± 0.14 mg/kg wet wt. on the coasts of Trabzon [60], followed by Giresun coasts with 0.66 ± 0.08 mg/kg wet wt. [42] and Amasra coasts with 0.40 ± 0.29 mg/kg wet wt. [56], respectively.

M. barbatus is the other fish most commonly used in monitoring studies and the highest Cd value was obtained from Trabzon coasts with 3.38 ± 0.06 mg/kg wet wt. [60], followed by Kastamonu coasts 0.28 ± 0.03 mg/kg wet wt. [52] and Samsun coasts with 0.20 ± 0.11 mg/kg wet wt. [38], respectively.

The lowest Cd level in *M. cephalus* was found in this study. However, the high values were determined in flathead grey mullet from Giresun and Rize coasts of the Black Sea [42].

In *S. maximus*, Cd levels were generally found to be very low except for a study of Tüzen [28].

Pb in Fish Species

Another important toxic metal is lead. In this study, Pb values were higher than both Cd and Hg as expected. Pb levels differ widely in other studies. Comparing the results of the present study with previous studies, Pb levels in *E. encrasicolus* were found to be high on the coasts of Giresun, Trabzon and Rize [42].

Although Pb levels in *T. mediterraneus* were below the allowed values (0.3 mg/kg wet wt.) in all studies, it was found to be the highest value (0.25 mg/kg wet wt.) in the study conducted by Alkan et al. [34].

Pb levels in *S. sarda* were found above the permissible values (0.3 mg/kg wet wt.) in many studies [28, 38, 42, 52]. Likewise, Pb levels in *M. merlangus* and *M. barbatus* were found to be much higher than allowed values in many studies [28, 38, 42, 52, 56, 57, 64].

Pb levels in *M. cephalus* were below the allowed values, except for Tüzen [28] and Türkmen and Akaydın [42]. Pb levels in *S. maximus* were below the accepted values (0.3 mg/kg wet wt.) in all studies (Table 3).

Conclusion

Toxic metals are among the contaminants which could pose a danger to marine coastal environments in general and fish fauna in particular. Owing to their extremely high toxicity, even at low concentrations, their sustained persistence in the environment and their propensity to bioaccumulate in fish species, these elements are very harmful because they are not biodegradable and thus concentrate large amounts of them in their tissues. Therefore, for the evaluation of the possible health risks associated with the consumption of fish from the coasts of the Black Sea, awareness of the toxic metal values in these fish is of great importance.

In this study, the toxic metal concentrations for seven commercial fish species have been analysed. The most Pb was found in all studied fish muscles, followed by Hg and Cd. The results of toxic metal levels in the fish samples did not exceed the permissible limits set for these toxic metals by the European Commission Regulation [21] and Turkish Legislation [22].

The daily intakes (EDIs) of the metals were estimated as the means of Hg, Cd and Pb in all fish samples and the mean consumption of fish per day for infants, children and adults, respectively. These results are significantly lower than the recommended values. The estimated hazarded quotients (HIs) of the considered metals were below the value of 1. Therefore, metals in fish samples do not pose an apparent threat to the population and these fishes are safe for consumption.

Declarations

Conflict of Interest The authors declare no competing interests.

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