



Heavy Metal Concentrations in *Trachurus Mediterraneus* and *Merlangius Merlangus* Captured from Marmara Sea, Turkey and Associated Health Risks

Latife Köker¹ · Fatih Aydın¹ · Özcan Gaygusuz¹ · Reyhan Akçaalan¹ · Derya Çamur² · Hüseyin İlater³ · Ferruh Niyazi Ayoğlu⁴ · Ahmet Altın⁵ · Murat Topbaş⁶ · Meriç Albay¹

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Abstract

Rapid industrialization and excessive human population growth may cause deterioration in marine water quality and biodiversity. Heavy metals are one of the most common pollutants in the seas and marine organisms, including demersal and pelagic fish, can accumulate them from the environment. Assessment of the ecological risk of heavy metals from fish has an important role in public health. In this study, some heavy metal (Pb, Cd, As, Cr, Hg, Cu, Zn, and Fe) concentrations were determined in the muscle tissues of two commonly consumed fish species, *Trachurus mediterraneus* (Mediterranean horse mackerel) and *Merlangius merlangus* (Whiting), which are the fifth (14,222 tons/year) and sixth (6814 tons/year) highest commercial catches of marine fish species in Turkey, respectively. Heavy metal concentrations of samples collected from four sites (Adalar, İzmit Bay, Yalova, and Tekirdağ) in the Marmara Sea were determined using ICP-MS. Fish samples caught at Yalova station were found to have the highest heavy metal concentrations. According to the World Health Organization (WHO), in terms of the mean values, only As, and Cr were higher than permissible limits in *T. mediterraneus*, while Pb, Cd, As, and Cr were higher in *M. merlangus*. Arsenic concentrations were higher than maximum limits in both *T. mediterraneus* and *M. merlangus*. The estimated weekly intake (EWI) was calculated to assess the potential health impact. The EWI for arsenic, cadmium, lead, and mercury for some sites was above the provisional tolerable weekly intake.

Keywords Bioaccumulation · Heavy metals · Tissue concentration · Health risk · *Trachurus mediterraneus* · *Merlangius merlangus*

Introduction

Heavy metals are found in marine ecosystems as a consequence of daily anthropogenic activities. Even though they can naturally be found in aquatic ecosystems, particularly, over the last few decades, agriculture and industry have contributed further to the release of wastes including heavy metals into the environment (Gümgüm et al. 1994; Miri et al. 2017; Carolin et al. 2017). Heavy metal pollution has become a worldwide problem because of toxic effects and bioaccumulation in aquatic organisms (Censi et al. 2006; Zeitoun and Mehana 2014; Lee et al. 2019). Among aquatic organisms, fish are the most frequently exposed organisms to the harmful effects of these pollutants (Clarkson 1998; Olaifa et al. 2004). Due to its accumulation in fish, heavy metals can be transmitted to humans through the food chain and cause negative effects on human health and the ecosystem in the long term (Gao and Li 2012;

✉ Latife Köker
latife.koker@istanbul.edu.tr

¹ Department of Marine and Freshwater Resources Management, Faculty of Aquatic Sciences, Istanbul University, Istanbul 34130, Turkey

² Department of Public Health, Faculty of Gülhane Medicine, University of Health Sciences, Ankara, Turkey

³ Provincial Health Directorate, Ministry of Health, Ankara, Turkey

⁴ Department of Public Health, Faculty of Medicine, Bülent Ecevit University, Zonguldak, Turkey

⁵ Department of Environmental Engineering, Bülent Ecevit University, Zonguldak 67100, Turkey

⁶ Department of Public Health, Faculty of Medicine, Karadeniz Technical University, Trabzon, Turkey

Hu et al. 2013; Zhao et al. 2014). Heavy metals like Cu, Zn, and Fe are essential for fish metabolism while some others such as Hg or Pb have no known role in biological systems. While essential metals (Fe, Zn, Cu, Mg, Se, Co, and Vn) have taken up by fish, non-essential ones (Al, As, Cd, Sb, Sn, Pt, Hg, Pb, and Bi) are also taken and accumulate in their tissues (Munoz-Olivas and Camara 2001; Predescu et al. 2017). Furthermore, their concentrations varied greatly in different parts of the fish (Canlı and Atlı 2003; Alkan et al. 2016). Besides the type of heavy metal, there are many environmental, ecological, and physiological parameters which affect the bioaccumulation of heavy metals in fish such as temperature, pollutant type, feeding habitats, age/life span (Wagner and Boman 2003; Weber et al. 2013; Rejomon et al. 2010; Mziray and Kimirei 2016; Burada et al. 2017). Also, the sizes of fish play an important role in metal contents of tissue (Barak and Mason 1990; Monterio and Lopes 1990).

Turkey is surrounded by four different seas and fishing is one of the biggest income sources for the country. The Marmara Sea, which is a semi-enclosed inland sea, has an area of 11,350 km². The rapid population growth, distorted urbanization, and the density of industry are the main factors for pollution in the Marmara Sea and different types of pollutants often reach the sea through river discharges (Pekey et al. 2004). The northeast coastline area of the Marmara Sea is the most industrialized region of Turkey, in which İzmit Bay has been especially affected by industrialization (Okay et al. 1998; Coelhan et al. 2006; Ergül et al. 2018). It is also heavily urbanized with 1/3 of Turkey's population living around the sea of Marmara. Heavy metals have also been discharged into the Marmara Sea for many years. Despite all these factors, the fisheries in the Marmara Sea are based on coastal fishing of benthic/demersal species and fishing of pelagic fish that rests upon seasonal migration fact. Pelagic fish, such as anchovy, bluefish, horse mackerel, and demersal fish, hake, whiting, striped red are the most consumed marine foods in Turkey (TUIK 2018). In 2018, *Trachurus mediterraneus* (Steindachner 1868) and *Merlangius merlangus* (Linnaeus 1758) were the fifth (14,222 tons/year) and sixth (6814 tons/year) most landed fish species in Turkey, respectively (TUIK 2018). In addition, these two species are also the most commercially caught fish species in the Marmara Sea (5000 tons/year and 1000 tons/year, respectively; TUIK 2018). Many studies have been focused on the determination of heavy metals in anchovy, horse mackerel, sea bass, and whiting which are commonly consumed in Turkey (Keskin et al. 2007; Türkmen et al. 2008a, b; Berik and Kahraman 2012; Özden and Erkan 2016; Güngör and Kara 2018). In some of these studies showed that the heavy metal concentrations of some fish species were below permissible limits while the others were over limits that indicating metal

pollution in the Marmara Sea (Keskin et al. 2007; Türkmen et al. 2008a; Özden and Erkan 2016). Türkmen et al. (2008c) reported that lead levels in muscles of anchovy in Yalova were just higher than the Turkish permissible limit. Cucu et al. (2019) found, during the sampling period (2013), that cadmium and lead were lower than Turkish limits for *T. mediterraneus* and *M. merlangus* on the Gebze Region in the Marmara Sea. In these previous studies in the Marmara Sea, fish samples were either obtained from local fish markets (Demirkol and Aktaş 2002; Topcuoğlu et al. 2003; Türksönmez and Diler 2019) or sampling was done only from one region (Kayhan et al. 2017; Güngör and Kara 2018; Cucu et al. 2019).

The main objective of the present study was to investigate the concentration of heavy metals (arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), lead (Pb), mercury (Hg) and zinc (Zn)) in the fish tissues of pelagic *T. mediterraneus* (Mediterranean horse mackerel) and demersal *M. merlangus* (Whiting), which are economically important fish species caught in the Marmara Sea. It is aimed to reveal the effects of heavy metal pollution in two fish species on public health by sampling from four stations that represent the Marmara Sea. Health risks posed by the heavy metals to humans consuming these fish were also evaluated.

Materials and Methods

Fish were sampled seasonally, four times from February to October in 2018, by bottom trawling at four different sites in the coastal waters of the Marmara Sea in 2018 (Fig. 1). Sampling areas were selected in metropolitan areas where the intensive commercial fishery is operating in the Sea of Marmara which were Adalar (1. St), the entrance of İzmit Bay (2. St), Yalova (3. St), and Tekirdağ (4. St).

Fish samples were stored in cold conditions (−18 °C) immediately after being caught, then transferred to the Limnology laboratory for further analyses. Total length (TL) was measured to the nearest 0.1 cm and body weight (W) was weighed to the nearest 0.1 g. Dissected muscle samples were homogenized and ~1.0 g muscle tissue sample from each fish specimen was taken and washed with distilled

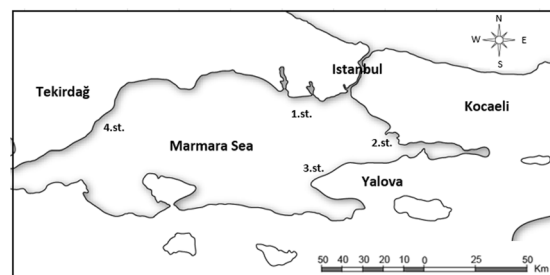


Fig. 1 Fish sampling sites in the Marmara Sea, Turkey

water. Samples were digested with nitric acid and hydrogen peroxide (2:1) in a microwave digestion system (MWS 4, Berghof) (Guhathakurta and Kaviraj 2000) and were transferred to 50 ml flasks. After the samples were filtered through a 0.45 µm nitrocellulose membrane filter, they were introduced into the ICP-MS (Perkin Elmer ELAN-DRC-e). All reagents used were of analytical grade (Merck). All the analyses were performed in triplicate by ICP-MS (Perkin Elmer ELAN-DRC-e). The operational parameter settings used for the ELAN 6000 Perkin Elmer were shown in Table 1 (Chamberlain et al. 2000). Multi-element (Merck) standards were used for standard solutions prepared from stock solutions. For mercury analyses, a trace amount of gold solution added to HNO₃ preserved all forms of mercury.

The accuracy of the analysis was verified by analyzing the ERM-BB422 certified reference material (Fish Muscle European Reference Material). Instrumental limits of detection (LOD), the instrumental limit of quantification (LOQ), and relative standard deviation (RSD) were calculated (Mataveli et al. 2013). The calculation of the detection limit is based on the 3x of SD of the blank solution. Recoveries between 98 and 105% were accepted to validate the calibration. %RSD values were varied

between 0.63 and 8.15%. Values were expressed as the mean of three analyses for each sample. LOD and LOQ of elements were Pb 0.001–0.003 µg L⁻¹; Cd 0.002–0.007 µg L⁻¹; As 0.004–0.013 µg L⁻¹; Cr 0.009–0.030 µg L⁻¹; Zn 0.010–0.033 µg L⁻¹; Cu 0.006–0.012 µg L⁻¹; Hg 0.005–0.017 µg L⁻¹ and Fe 0.012–0.040 µg L⁻¹, respectively.

Prior to statistical analyses, data sets were tested for normality and homogeneity. Because the data showed not normal distribution considering the Shapiro–Wilk test, non-parametric tests (Mann–Whitney *U* and Kruskal–Wallis) were selected. Non-parametric Mann–Whitney *U* test was performed to determine the differences, and the Kruskal–Wallis test (KW) (with Tamhane’s as a post-hoc test) was performed to determine differences of body size groups (small, medium, large) for each fish species, in terms of heavy metal concentrations (As, Cd, Cr, Cu, Fe, Hg, Pb, and Zn). The level of significance was set at $p < 0.05$.

The estimated weekly intake (EWI) values of heavy metals were calculated. Since, the annual quantity of fish consumed was 5.5 kg/person in 2017 (TUIK 2017), the EWI and EDI (Estimated Daily Intake) values were assumed as being for a 70 kg person who would consume 105.5 g fish/week. Furthermore, Provisional Tolerable Weekly Intake (PTWI) was calculated to represent the maximum amount of pollutant that a person could be exposed to per week throughout a lifetime without risk of an unacceptable health impact (National Academy of Sciences 1989; WHO 1996; Council of Europe 2001; FAO/WHO 2010; EFSA (2010, 2012a, b)).

Table 1 Instrumental parameters of ICP-MS

Characteristics	Instrument conditions
RF Power	1400 W
Nebulizer gas	Argon
Plasma gas flow	14.0 L/min
Auxiliary gas flow	1.4 L/min
Nebulizer gas flow	0.99 L/min
Sweeps/reading	10
Number of replicates	3
Sampler & skimmer cone	Nickel

Table 2 Individual numbers (*n*), body weight and total length distributions of *T. mediterraneus* and *M. merlangus* in each sampling site in the Marmara Sea

	Number <i>n</i>	Body weight (W, g)		Total length (TL, cm)	
		Min.–max.	Mean	Min.–max.	Mean
1.st					
<i>T. mediterraneus</i>	15	5.3–23.3	12.9 ± 5.2	9.4–14.8	11.9 ± 1.6
<i>M. merlangus</i>	30	11.2–55.1	22.8 ± 9.9	11.4–18.9	14.4 ± 1.8
2.st					
<i>T. mediterraneus</i>	19	5.4–28.4	15.75 ± 4.4	9.6–15.2	12.5 ± 1.1
<i>M. merlangus</i>	14	10.8–29.6	13.8 ± 1.8	11.1–16.3	13.1 ± 0.9
3.st					
<i>T. mediterraneus</i>	29	4.0–16.0	11.7 ± 1.7	9.0–13.1	11.5 ± 0.6
<i>M. merlangus</i>	16	11.0–14.1	12.5 ± 4.6	11.9–3.5	12.9 ± 1.4
4.st					
<i>T. mediterraneus</i>	48	4.4–24.2	9.8 ± 4.5	8.6–14.1	10.7 ± 1.4
<i>M. merlangus</i>	30	7.9–31.6	14.9 ± 6.3	10.7–16.4	12.8 ± 1.5

Results and Discussion

A total of 201 fish specimens ($n = 111$ for *T. mediterraneus*; $n = 90$ for *M. merlangus*) were sampled during the

Table 3 Mean heavy metal concentrations (mg kg⁻¹) in each length class of *T. mediterraneus* and *M. merlangus* caught from the Mediterranean Sea

	Total length (cm)	Heavy metal concentrations (mg kg ⁻¹)							
		Cr	Fe	Cu	Zn	As	Cd	Pb	Hg
<i>T. mediterraneus</i>									
Small	8.5–10.9 cm	0.22	23.0	1.26	23.35	16.09	0.24	0.40	0.03
Medium	11–12.9 cm	0.15	20.63	1.89	17.40	17.53	0.26	0.30	0.13
Large	13–15.5 cm	0.11	15.72	0.93	15.34	13.22	0.13	0.04	0.08
<i>M. merlangus</i>									
Small	10–12.9 cm	0.41	23.22	0.41	16.16	23.47	0.93	0.28	0.02
Medium	13–15.9 cm	0.34	17.99	0.31	12.10	24.33	1.38	0.39	0.01
Large	16–19 cm	0.31	14.68	0.30	10.60	21.28	0.84	0.21	0.15

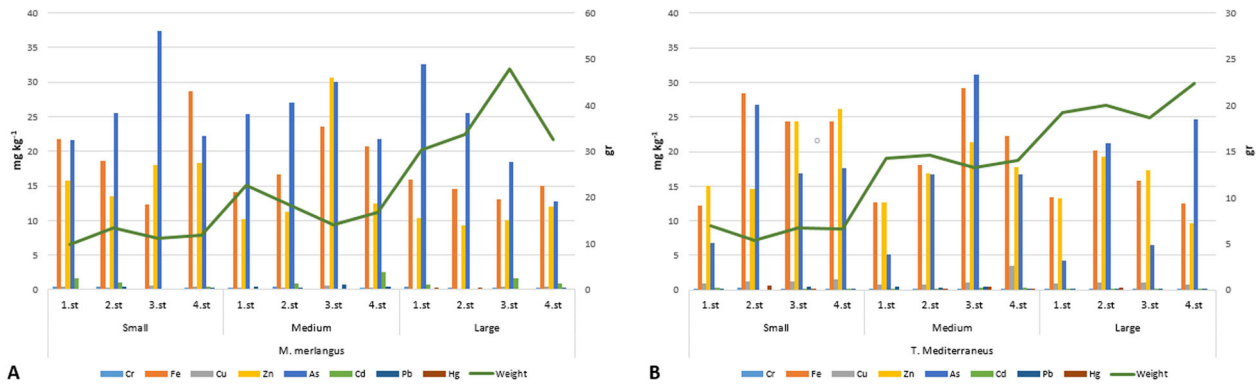


Fig. 2 Mean values of heavy metals in different size of **a** *T. mediterraneus* and **b** *M. merlangus*

study period and all of them were analysed for heavy metal concentrations. The TL and body-weight of the specimens varied between 8.6–15.2 cm and 4.0–28.4 g for *T. mediterraneus* and 10.2–18.9 cm and 6.4–55.1 g for *M. merlangus* (Table 2).

While evaluating the relationship between length and heavy metals, both species were classified and grouped according to their size. *T. mediterraneus* and *M. merlangus* were grouped as small (8.5–10.9 cm; 10–12.9 cm), medium (11–12.9 cm; 13–15.9 cm) and large (13–15.5 cm; 16–19 cm), respectively (Table 3). Heath (1995) reported that because of the large surface to the volume ratio, small fish tend to accumulate a higher quantity of heavy metals. Our results also showed that generally, the smaller sized specimens of both species had higher heavy metal concentrations. For *T. mediterraneus*, the Kruskal–Wallis test revealed significant differences between small and large size group and was found for Cr ($p = 0.000$), Fe ($p = 0.001$), Cu ($p = 0.002$), Zn ($p = 0.008$), Cd ($p = 0.002$) and Pb ($p = 0.042$). For *M. merlangus*, Cr ($p = 0.000$), Fe ($p = 0.000$) and Cu ($p = 0.044$) showed significant differences (Kruskal–Wallis, $p < 0.05$). Metal metabolism and metabolic activity in fish played a key role in metal accumulation (Heath 1995; Roesijadi and Robinson 1994). The studies examining the relationship between length and heavy metals showed that the accumulation of heavy metals

decreased with an increase in the length of fish (Liang et al. 1999; Nussey et al. 2000; Widianarko et al. 2000; Canlı and Athi 2003). The reason for negative correlations between length and metal concentrations could be explained by the fact that young individuals had higher metabolic activity than older ones (Elder and Collins 1991; Canlı and Furness 1993) (Fig. 2). There is a general view that Zn and Cu concentrations, in particular, decrease with increasing body size due to different metabolic rates of accumulation (Canlı and Athi 2003; Endo et al. 2008; Barone et al. 2013) and this view was supported by the results of the present study.

The results of the concentration of seven elements (Pb, Cd, As, Cr, Hg, Zn, Cu, and Fe) in the muscle of two species from the Marmara Sea are shown in Table 4. The concentration of As, Zn, and Fe were the highest in both species. On the other hand, generally the lowest concentrations for Cd, Pb, and Cr were found during the study period (Table 4).

The order of mean heavy metals concentrations from all stations was found to be Fe > Zn > As > Cu > Pb > Hg > Cd > Cr for *T. mediterraneus*. Although this pattern depending on the environment of the fish, this general order of monitored heavy metal bioaccumulation in pelagic fish is similar to that found in other studies (Bilandzic et al. 2011; Afandi et al. 2018). The concentrations of all heavy metals determined in *M. merlangus* were low from all stations, except

Table 4 Mean heavy metal contents with standard error in the tissues of the examined species (mg kg^{-1} wet wt)

Heavy metals	<i>T. mediterraneus</i>				<i>M. merlangus</i>			
	1.st	2.st	3.st	4.st	1st	2.st	3.st	4.st
Cd	0.2 ± 0.06	0.2 ± 0.03	0.3 ± 0.01	0.2 ± 0.05	0.5 ± 0.05	ND	0.2 ± 0.06	1.4 ± 0.25
Pb	0.3 ± 0.03	0.3 ± 0.01	0.8 ± 0.04	0.2 ± 0.03	0.3 ± 0.06	1.2 ± 0.03	0.9 ± 0.13	0.3 ± 0.08
As	5.4 ± 0.55	14.2 ± 1.15	28.1 ± 8.01	15.1 ± 2.83	24.2 ± 1.33	14.4 ± 2.15	35.5 ± 6.95	21.2 ± 4.03
Cr	0.2 ± 0.03	0.2 ± 0.02	0.2 ± 0.02	0.2 ± 0.03	0.3 ± 0.02	0.2 ± 0.02	0.3 ± 0.01	0.4 ± 0.08
Cu	0.9 ± 0.21	0.9 ± 0.25	1.0 ± 0.17	2.0 ± 0.38	0.3 ± 0.02	0.4 ± 0.15	0.6 ± 0.04	0.4 ± 0.01
Zn	14.1 ± 2.41	15.7 ± 1.71	21.0 ± 3.14	20.0 ± 5.31	10.6 ± 1.73	11.8 ± 2.33	19.6 ± 2.33	17.2 ± 2.31
Fe	12.9 ± 1.38	58.1 ± 11.9	83.0 ± 10.75	19.8 ± 4.64	15.9 ± 2.29	51.5 ± 9.43	63.1 ± 12.57	25.0 ± 3.64
Hg	ND	0.3 ± 0.02	0.4 ± 0.15	0.1 ± 0.01	0.1 ± 0.02	0.03 ± 0.01	0.4 ± 0.02	0.1 ± 0.01

ND not detected

As, Zn, and Fe. The order of mean heavy metals concentrations from all stations was found to be $\text{Fe} > \text{As} > \text{Zn} > \text{Cd} > \text{Pb} > \text{Cu} > \text{Cr} > \text{Hg}$ for *M. merlangus*.

According to comparisons between two species for heavy metal concentrations, performed by the non-parametric Mann–Whitney test, there were significant differences in levels of all heavy metals Cr, Cu, Zn, Cd, Pb, and As (Mann–Whitney, $p < 0.05$), except for iron and mercury ($p > 0.05$). The cadmium, iron, arsenic, chromium, and lead concentrations were greater in *M. merlangus* than *T. mediterraneus*. However, copper, zinc, and mercury were higher in *T. mediterraneus*. Similar to other studies in the Marmara Sea, Fe was the highest in both species in this study, followed by As. On the other hand, Cd was generally the lowest (Türkmen et al. 2008a, 2009). Especially at Yalova, for both species, As, Zn, Fe, and Hg showed the highest values and Pb displayed the highest value in *M. merlangus* at Site 2, the entrance to İzmit Bay.

The concentrations of Cr, Cu, As, Pb, Cd, Hg, Zn, and Fe in *T. mediterraneus* and *M. merlangus* from different sites of the Marmara Sea were compared with national and international standards. Values concerning permissible detected heavy metals for Turkish Fisheries Laws and Regulations (TKB), World Health Organization (WHO) and Environmental Protection Agency (EPA) are given in Table 5 (WHO 1989; EPA 1989; Anonymous 2005). *Trachurus mediterraneus* and *M. merlangus* results were averaged for all stations.

Demersal fishes can have a higher heavy metal accumulation potential, because they are exposed to more heavy metals due to their habitats, feeding habits and placing in high trophic levels (Stergiou and Karpouzi 2002; Naccari et al. 2015). According to permissible limits of national and international standards, our data show that pelagic fish had lower Pb ($p = 0.000$), Cd ($p = 0.008$), As ($p = 0.000$), and Cr ($p = 0.000$) concentrations than demersal fish (Mann–Whitney, $p < 0.05$), (Table 5). TKB has very low permissible limits and both fish exceeded the limits for Cd,

Table 5 Permissible content of heavy metals in fish (mg kg^{-1})

	mg kg^{-1}				
	TKB	EPA	WHO	<i>T. mediterraneus</i>	<i>M. merlangus</i>
Pb	0.3		0.5	0.4	0.7
Cd	0.05	1.4	0.5	0.2	0.7
As	1		1.8	15.7	23.9
Cr		4.1	0.15	0.2	0.3
Hg	0.5		0.5	0.3	0.2
Cu		54	30	1.2	0.4
Zn		410	30	17.7	14.8
Fe		410	100	43.5	38.9

TKB Turkish Fisheries Laws and Regulations, EPA Environmental Protection Agency, WHO World Health Organization

As, and Pb. According to the WHO, only As and Cr were higher than permissible limits in *T. mediterraneus*, while Pb, Cd, As, and Cr were higher in *M. merlangus*. Arsenic concentrations were higher than the maximum limits in both *T. mediterraneus* and *M. merlangus*.

The cadmium concentrations in *T. mediterraneus* ranged from 0.3 to 2.8 mg kg^{-1} and 0.2 to 22.3 mg kg^{-1} in *M. merlangus*. These results correspond with the results of Pekey (2006). In his study, he found As, Cd, Pb and Zn elements in high concentrations in the northern part of İzmit Bay. However, Cucu et al. (2019) reported that in 2013 the mean values for Cd from the Gebze region near 2.st were 2.2 $\mu\text{g kg}^{-1}$ and 0.22 $\mu\text{g kg}^{-1}$ for *T. mediterraneus* and *M. merlangus*, respectively which is very low when compared to our findings. It was determined that in the Gulf of Gemlik which is one of the most industrialized and contaminated regions in Turkey, *T. mediterraneus* and *M. merlangus* both had high Cd concentrations with 1 mg kg^{-1} and this exceeded the permissible limits determined by WHO (Ünlü et al. 2008; Kayhan et al. 2017). In the Aegean Sea, these concentrations were lower than 0.01–0.04 mg kg^{-1} (Dalman et al. 2006). It is widely known that Cd is found in trace concentrations in marine environments and aquatic

Table 6 Estimated weekly intakes (EWI) of *T. mediterraneus* and *M. merlangus*

	Cr	Fe	Cu	Zn	As ^a	Cd	Pb	Hg	
<i>T. mediterraneus</i> (EWI mg/week/ 70 kg bw)	1.st	0.28	19.5	1.33	21.3	0.3	0.35	0.34	ND
	2.st	0.11	23.5	1.23	24.5	0.8	0.23	0.24	0.09
	3.st	0.20	41.6	1.57	32.7	1.6	0.45	0.64	0.64
	4.st	0.30	35	3.10	32.8	0.9	0.36	0.53	0.05
<i>M. merlangus</i> (EWI mg/week/ 70 kg bw)	1.st	0.45	23.97	0.48	16.02	1.3	0.43	0.39	0.02
	2.st	0.30	77.65	0.54	17.71	0.8	ND	1.81	0.05
	3.st	0.39	40.50	0.67	28.56	0.96	0.73	0.17	0.03
	4.st	0.69	28.63	0.48	20.37	1.2	3.27	0.62	0.07
Guidelines for PTWI (mg/week/kg)	0.02	5.6	3.5	7	0.02	0.01	0.03	0.004	
PTWI (mg/week/ 70 kg bw)	1.63	392	245	490	1.05	0.49	1.75	0.28	

EWI estimated weekly intake, PTWI provisional tolerable weekly intake

^aTotal arsenic concentrations were converted to inorganic arsenic

organisms. In this study, mean cadmium concentrations for both species were found to be higher than Turkish Fisheries Laws and Regulations and lower than the WHO values. Castro-González and Méndez-Armenta (2008) stated that Cadmium negatively affects vital organs: brain, kidney, lung, bones, and the central nervous system. Previous studies showed that in the Marmara Sea cadmium concentrations in *Merluccius merluccius* varied between 0.01 and 2.14 mg kg⁻¹, considerably lower than our results. Although the heavy metal concentrations in fish are affected by their diet, the age of fish and physicochemical properties of water, these results may be associated with increased cadmium pollution in the Marmara Sea (Afshan et al. 2014; Pandey et al. 2008).

T. mediterraneus and *M. merlangus* had a lower concentration of Cu than national and international limits. For fish samples from Saroz Bay, Güngör and Kara (2018) also detected Cu levels of 0.73 and 0.28 mg kg⁻¹, respectively. At 4. St near Saroz Bay, the mean Cr concentration in *T. mediterraneus* was 2.0 mg kg⁻¹. In Tekirdağ province, urbanization and industry are widespread and the coastal area is heavily affected by pollution (Dökmeci et al. 2019).

To assess the human health risk from heavy metals in fish, it is important to have knowledge of the dietary intake of metals and their synergistic effects. For this reason, the EWI for a 70 kg body weight of an adult person was determined based on the results of the present study in comparison with Provisional Tolerable Weekly Intake (PTWI) which were established by The Joint FAO/WHO Expert Committee on Food Additives (Table 6; FAO/WHO 2004). The results indicated that for Cr, Fe, Cu, and Zn, there is no adverse health risk from the consumption of these two species. Although As, Cd, Pb, and Hg were at relatively higher levels than the PTWI, this could potentially cause adverse effects and toxicity (Marcotrigiano and Storelli 2003; Castro-González and Méndez-Armenta 2008). Lead, cadmium, and mercury are heavy metals that

do not naturally occur in living organisms and their function in human body is still unknown (Lenntech Water Treatment and Air Purification 2004). Therefore uptake of these metals at low concentrations can be very harmful. These heavy metals could be toxic, neurotoxic, carcinogenic, or mutagenic (Duruibe et al. 2007).

The FAO/WHO Expert Committee on Food Additives reported that due to the long half-life of Cd, it does not have a significant effect concerning the overall exposure from daily food intake (Saei-Dehkordi and Fallah 2011). Inorganic arsenic compounds are more acutely toxic than organic forms, and EWI values calculated as inorganic arsenic and especially from Site 3, were higher than the guidelines (1.05 mg/week/70 kg bw).

Lead concentrations in *M. merlangus* ranged from 0.01 to 3.1 mg kg⁻¹ and 0.05 to 8.14 mg kg⁻¹ for *T. mediterraneus*. Mean lead concentrations for both species were found to be higher than Turkish Fisheries Laws and Regulations. In 2013, lead concentrations were reported at 4.5 µg kg⁻¹ for *T. mediterraneus* and 0.23 µg kg⁻¹ for *M. merlangus* in the Marmara Sea (Cucu et al. 2019). Compared to the Black, Aegean, and Mediterranean seas, *T. mediterraneus* had lower Pb concentrations (Türkmen et al. 2008b). *Merlangius merlangus* also had higher lead concentrations than WHO guidelines. Aksu et al. (2011) determined that in *M. merluccius* which is a demersal fish like *M. merlangus*, Pb concentrations ranged between 3.23 and 10.5 mg kg⁻¹ but our results were higher than the Southern Black Sea Shelf which was found to be 0.05–5.75 mg kg⁻¹ for *M. merluccius* (Aksu et al. 2011).

Mercury concentrations in *T. mediterraneus* ranged from 0.007 to 1.24 mg kg⁻¹ and 0.008 to 0.51 mg kg⁻¹ in *M. merlangus*. Mean mercury levels were found to be lower than permissible limits. The results showed that the mean values of Hg were the lowest at sites 1 and 4 for *T. mediterraneus*. In general, although the results were below permissible limits, Site 3 results were very close to

the limit values for both fish species. As it is well known, Hg may have toxic effects on the digestive, immune, and nervous systems (Bhardwaj et al. 2009; Li et al. 2010; Rice et al. 2014).

Arsenic concentrations in both species were found to be very high. In *T. mediterraneus* total As varied between 3.2 and 49.9 mg kg⁻¹ and in *M. merlangus* 8–52.9 mg kg⁻¹. In a study by Güngör and Kara (2018), in Saroz Bay, As concentrations for *T. mediterraneus* and *M. merlangus* were found to be 1.23 and 3.6 mg kg⁻¹, respectively. Dökmeci et al. 2019 reported that in Tekirdağ province As concentrations in sediment samples were determined as being highly polluted in accordance with the EPA sediment quality guidelines (EPASQG). High As concentrations in fish samples could be the result of heavy metal loads in sediment. For both species, the maximum levels were determined at Site 3 where the pollutants were related to anthropogenic inputs from İzmit Bay. Studies have shown that exposure to arsenic is caused by cancer, cardiovascular disease, neurological disorders, and dermal effects (Miller et al. 2002; Vahidnia et al. 2007; De Vizcaya-Ruiz et al. 2009).

The lowest and highest iron levels in *T. mediterraneus* were 8.6–102.9 mg kg⁻¹ and 9.5–51.5 mg kg⁻¹ in *M. merlangus*. Iron showed the highest metal level in both species and Site 3 had the highest concentrations of Fe. Due to being a pelagic fish *T. mediterraneus* feed at higher trophic levels and these predatory fish contained significantly higher concentrations of Fe and Zn (Afandi et al. 2018). In previous studies, the Fe concentrations in *M. merlangus* in Marmara, Mediterranean, and Black seas were 5.0, 6.8, and 4.5 mg kg⁻¹, respectively, which were 10 times lower than our results (Turan et al. 2009). Similarly, with Zn concentrations, *M. merlangus* in the Mediterranean and Black seas were 5.3 and 6.5 mg kg⁻¹, respectively which were lower than our results (Turan et al. 2009; Güngör and Kara 2018).

Generally, environmental contaminants are present together (Borgmann et al. 2008; Naz and Javed 2013). Toxicity tests indicate that metal combinations are usually more toxic than single metals (Marr et al. 1998; Verma et al. 1982). Studies showed that some fish species are less sensitive to heavy metal mixtures such as Cu, Zn, Cr, and Fe according to the stage of fish development (Kazlauskiene and Stasiunaite 1999; Vosylienė et al. 2003). Batool and Javed (2015) reported that a combination of heavy metals revealed a synergistic effect. Heavy metal combinations affect public health as well as fish species. Groten et al. (1997) demonstrated that arsenic-lead combination could cause changes in rats. Also, lead-mercury mixtures act synergistically which are extremely neurotoxic and exhibit more toxic than the single one (Wildemann et al. 2015). While conducting the risk assessment for public health, because of all these adverse effects of heavy metal

combinations, toxicity tests and synergistic interactions should be investigated.

Conclusion

This study provides additional data on the heavy metal content of two economic fish in Turkey. Results are essential in risk assessment of consumer exposure to heavy metals. The Provisional Tolerable Weekly Intake of As, Cd, Pb, and Hg was higher than the safe limits. Results showed that both pelagic and demersal fish species contain heavy metals above recommended limits and precautions need to be taken urgently to reduce pollution in the Marmara Sea. Heavy metal accumulation in fish muscle tissue should be considered as an important warning both for fish health and human health via consumption. To assess the risk to human health based on heavy metal concentrations in fish species, periodical monitoring is necessary.

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Compliance with Ethical Standards

Conflict of Interest The authors declare that they have no conflict of interest.

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