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Bioaccumulation of essential and toxic metals in four different species of bottom fish in the Marmara Sea, Tekirdag, Turkey: risk assessment to human health

Ayse Handan Dökmeci[a,*], Temine Sabudak[b], Veysi Dalmış[b]

[a]*Department of Emergency and Disaster Management, School of Health, Tekirdağ Namik Kemal University, Suleymanpaşa, Tekirdağ, Turkey, email: hdokmeci@nku.edu.tr (A.H. Dökmeci)*
[b]*Department of Chemistry, Faculty of Science and Letters, Tekirdağ Namik Kemal University, Suleymanpaşa, Tekirdağ, Turkey, email: tsabudak@nku.edu.tr (T. Sabudak), veysidalmis@gmail.com (V. Dalmış)*

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ABSTRACT

Due to the intensive industrial and agricultural activities in the Thrace region and the maritime transport that directly and/or indirectly affect Tekirdağ coastal areas, the pollution starts from the Tuna River and extends to the Marmara Sea through the Black Sea and the Bosphorus. Heavy metals are not only a threat to marine life, but also to the health of people who consume seafood. In order to investigate the presence of essential and toxic metals in Marmara Sea fish species, samples of 4 different bottom fish (sole, whiting, striped red mullet and angler fish) were collected from 4 different stations in the coastal line of Tekirdağ province in the spring of 2018. Bioaccumulation of essential and toxic metals (Cd, Ni, Cu, Pb, Cr, Mn, Hg, Fe, As, Zn) in the muscles of fish samples was measured by ICP-OES instrument. The accumulation pattern of the analyzed metals in all bottom fish muscles was as follows: Fe (58.8) > Zn (23.9) > Mn (3.3) > As (1.44) > Cu (0.8) > Cr (0.6) > Ni (0.3) > Cd (0.08) mg/kg dry weight. ANOVA analyses indicated significant variations ($p < 0.05$) of Fe, Zn, Mn, As, Cu, Cr, Ni, and Cd in the muscles of different fish species. Significant variations were observed in different stations ($p < 0.05$), and As, Pb and Hg were below the detection limit of ICP-OES. Except Cu, the levels of all essential and toxic metals in fish samples were found to be above the national and international legal limits recommended for human consumption. Since the THQ values of inorganic As were higher than 1 mg/kg d·w, it is likely to pose health risk for the consumers of the these fish species.

Keywords: Marmara Sea; Tekirdag; Essential and toxic metals; Bioaccumulation; Fish; Health risk assessment

1. Introduction

Metals can be classified as biologically essential [(copper (Cu), zinc (Zn), chromium (Cr), nickel (Ni), cobalt (Co), molybdenum (Mo) and iron (Fe)] and non-essential [(aluminum (Al), cadmium (Cd), mercury (Hg), tin (Sn) and lead (Pb)]. Heavy metals are found in both anthropogenic and different natural sources. If the concentration of non-essential heavy metals increases, their toxicity will rise in the environmental systems [1]. Aquatic ecosystems such as seawater, warm springs, groundwater, rivers and lakes contain arsenic (As) in different amounts. As comprises of arsenate and arsenite with arsenate being more dominant. Furthermore, As has inorganic and organic forms. Bioaccumulation of As occurs especially in liver and kidney of fishes [2]. Hg is one of the important heavy metals in aquatic ecosystems. Bioavailable form of Hg is methyl mercury (MeHg) which is highly toxic, stable and efficiently absorbed by diet [3]. MeHg causes some symptoms including impaired vision and hearing, headaches, paresthesia, movement difficulties and loss of coordination, fatigue, tremors and ataxia in intoxicated humans [4].

Parallel to the development of the industry, heavy metals remain as an alarming class of pollutants. These

*Corresponding author.

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toxic metals tend to accumulate in water spreading from domestic, industrial and natural sources and can reach people through food chain. Heavy metals have a particular significance in environmental toxicology because of their toxicity, hard degradation, long persistence, easy bioaccumulation, and bio-magnification in the food chain. The pollutant type, fish species, sampling location, trophic level, and their mode of feeding are key factors to assess pollution. Aquatic organisms are good indicators for the long-term monitoring of metal pollution in the aquatic ecosystems [5–9]. Some hazardous metals such lead (Pb), chromium (Cr), cadmium (Cd), mercury (Hg), and arsenic (As) that accumulate in fish pose a health risk to humans, even in trace amounts [10–14]. Therefore, the studies carried out are important as they reveal the level of heavy metals in the environment and determine their potential hazards to humans. While some metals are important as trace elements for living beings, they accumulate in the body after a certain concentration and cause toxic effects or they can be transformed into other compounds in their own environment, and sometimes they emerge as toxic and water-soluble compounds of a metal during this transformation [15–18].

Heavy metals and trace elements are classified as essential and non-essential according to their degree of participation in biological processes. Those identified as vital should have a certain concentration in the organisms. These metals are required to be taken on a regular basis via nutrients as they participate in biological reactions. For example, copper(Cu) is an indispensable part of red blood cells and many oxidation and reduction processes in animals and humans [19,20]. On the other hand, even very small concentrations of non-vital heavy metals can cause health problems by affecting the physiological structure. The best example to this group is mercury (Hg) which binds to sulfuric enzymes. Whether a heavy metal is necessary for the living body depends on the organism which is taken into consideration. For example, while nickel is toxic for plants, it must be present as a trace element in animals [21].

Located in the Thrace Region of Turkey, Tekirdag province has 1180 industrial enterprises today. Along with the increase in the facilities in sectors such as textile, leather, paper and chemistry in the region, the quality of surface water has started to deteriorate. This deterioration negatively affects the coasts of the Marmara Sea in Tekirdağ province.

The objectives of this study were:

1. To determine the concentration and bioaccumulation of trace elements (Zn, Cu, Fe and Mn) and heavy metals (As, Cd, Cr, Hg, Ni and Pb) (mg/kg dry weight) in muscles of bottom fish species *Mullussurmuletus*, *Merlangiusmerlanguseuxinus*, *Solea vulgaris* and *Lophiuspiscatorius* collected from coastal waters of Marmara Sea of Turkey around Tekirdağ.
2. To compare the estimated dietary intake (EDI) of heavy metals via fish consumption with the daily intake reference dose (RfD).
3. To perform an assessment of the potential health risk of essential and toxic metals in fish by using the target hazard quotient (THQ).

2. Materials and methods

2.1. Study area and sample collection

In this study, four different bottom fish samples (n = 160); sole (*Solea vulgaris*; n = 10), whiting fish (*Merlangiusmerlanguseuxinus*; n = 10), striped red mullet (*Mullussurmuletus*; n = 10), and angler fish (*Lophiuspiscatorius*; n = 10) were collected from four different stations along the coast of the Marmara Sea around Tekirdağ province. The first of these stations was Şarköy which has an international environmental award of blue flag. The rest of the three stations were determined to be the centre of Tekirdağ, Yenice town where one of the branches of the Ergene River flows into the Marmara Sea and the town of Marmara Ereğlisi (Fig. 1). Tekirdağ province, located in the Ergene basin, is one of the rare cities in Turkey with two separate sea shores and a maximum length of the coast (135 km). Urbanization along the coast of Tekirdağ province continues to increase rapidly because it is a coastal city and the industry is widespread. The population of the coastal strip of Tekirdağ is doubled in the summer, due to people coming from Tekirdağ province and outside. The coastal area of Tekirdag province has been strongly influenced by intense industrial and agricultural activities since 1960, by heavy ship traffic of Asiaport, by the first transit container port of Turkey started to operate in 2015, and by the pollution resulting from atmospheric deposition.

In this study, algarna method and bottom net method were used to hunt bottom fish at different depths. Fish samples collected from four different points were brought to laboratory in ice boxes. Before splitting the muscle tissue of the fish samples, each of them was washed with distilled water and packed in polyethylene bags and kept at –30°C until analysis after measured in length and width. After the frozen fish samples were thawed at room temperature, the muscle samples from each fish were dissected and each sample was dried in drying-oven for 24 h at 600°C. Plastic knives were used to prevent contamination when separat-

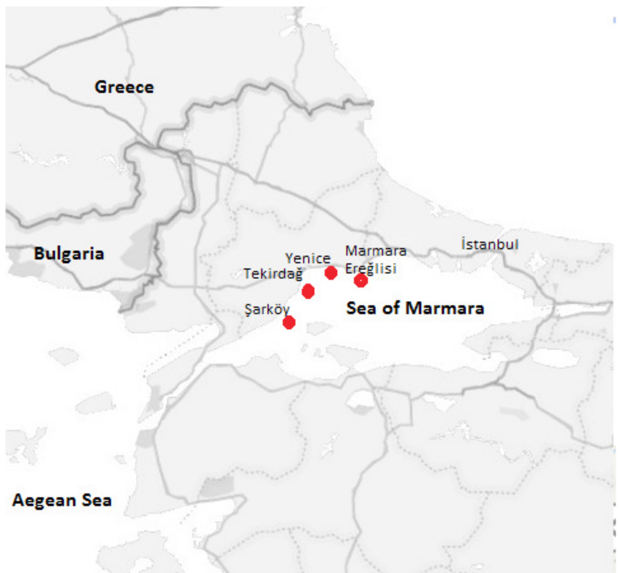

Fig. 1. Area of study.

ing the C tissues. After the dried fish samples were homogenized in the blender, 0.5 g of the homogenate was aliquated. After adding 6 mL of HNO_3 (65%) and 4 mL H_2O_2, they were digested in a microwave digestion system according to the USEPA 3051 method (CEM, Inc. Mars 6, Matthews, NC, USA). Cooled at room temperature, the digested muscle samples were diluted with Milli-Q water to 25 mL. A blank digest was carried out in the same way. All the results were expressed in dry mass.

2.2. Reagents

All the reagents used were of ultrapure grade (Merck Suprapur, Darmstadt, Germany). Tap water was deionized and further purified using an 18 MΩ cm Milli-Q system (Millipore, Milford, MA, USA) and used to prepare all the reagents and standard solutions along with analytical grade HNO_3 and HCl (Merck, Darmstadt, Germany) when needed. All glass and polyethylene materials, were firstly kept in dilute nitric acid solution (1/9, v/v) for 48 h and were rinsed by distilled deionized water prior to use.

2.3. Instrumentation

Being essential and toxic metals in muscle tissues of fish; Cd, Ni, Cu, Pb, Cr, Mn, Hg, Fe, As, and Zn measurements were performed with an Inductively Coupled Plasma Optical Emission Spectroscopy (SPECTRO, Spectroblue model ICP-OES) [22,23] (Tables 1 and 2). The recoveries of the elements ranged from 95% to 104%. All elements of heavy metals were expressed in mg kg^{-1} dry weight. Data were expressed as mean ± standard deviation, and each measurement was performed in triplicate.

2.4. Risk assessment to human health

The estimated daily and weekly intake levels (EDI and EWI, respectively) of heavy metals and trace elements were calculated to determine whether the essential and toxic metal accumulation levels detected in fish muscles were safe for human consumption (Table 4). EDI was calculated on the basis of the average concentrations determined in the muscle tissues of fish samples for each metal and the amount of daily fish consumption (Eq. (1)). The annual per capita consumption of fishery products in Turkey is 6.0 kg for a 70 kg person [24]. Fish consumption in Turkey which is surrounded by seas on three sides, as much as only the half of the world average and only one-third of the European Union average. The Marmara region is the leading areas as far as the amount and variety of fish consumption are concerned [25]. Turkey's average daily per capita consumption of fish is reported to be 20 g [26]. This amount is 140 g/person/week (for 70 kg adults). EDI and EWI values are given in Table 4.

The estimated daily intake (EDI) (μg kg^{-1} day^{-1}) was calculated based on the following assumption [27,28];

1) The human weight is 70 kg
2) The human intake from species per day is 20 g.

$$EDI = [C_{fish} \times IR_{fish}]/BW \quad (1)$$

Table 1
Instrumental parameters for Cd, Ni, Cu, Pb, Cr, Mn, Hg, Fe, As, and Zn elements determination by ICP-OES

Parameter	Condition
RF power (W)	1400
Plasma gas flow rate (L min^{-1})	12
Auxiliary gas flow rate (L min^{-1})	1.00
Nebulizer gas flow rate (L min^{-1})	1.00
Nebulizer	Cross-flow

Table 2
Instrumental detection limit (ppb) for Cd, Ni, Cu, Pb, Cr, Mn, Hg, Fe, As, and Zn elements determination by ICP-OES

Metals	Inst. detection limit (ppb)
Cr	0.007
Cu	0.014
Fe	0.07
Mn	0.5
Ni	0.05
Cd	0.001
Pb	0.05
Zn	0.05
As (total)	0.05
Hg	0.06

C_{fish}: the average heavy metal concentration in bottom fish muscle (μg g^{-1} d.w.); IR_{fish}: Daily fish consumption (g d^{-1}); BW: The average body weight (kg) of the target population.

In addition, the potential non-cancer risk for individual heavy metals was performed using the target hazard quotient (THQ) [29,30].

The target hazard quotient (THQ) was calculated by the following equation:

$$THQ = EDI/RfD \quad (2)$$

RfD: The daily intake reference dose (μg kg^{-1} day^{-1}). If the THQ value exceeds 1, there would be concerns for potential non-cancer effects. If the THQ value is less than 1, there would be no obvious risk.

2.5. Statistical analysis

Statistical analysis was performed using SPSS 17.0 for Windows (Chicago: SPSS Inc., Chicago, USA). The one-way analysis of variance (ANOVA) and the comparative analyses were conducted and used to test for differences in essential and toxic metal concentrations (Cd, Ni, Cu, Pb, Cr, Mn, Hg, Fe, As, and Zn) of four different species by using Tukey's multiple range test. The level of significance was acceptable at $p < 0.05$.

3.Results and discussion

3.1. Concentration and bioaccumulation of heavy metals in bottom fish muscle samples

Metal concentrations in coastal ecosystems of regions with high industrial and agricultural activity may come up high. Aquatic organisms tend to accumulate these metals in their bodies [21]. In this study, the contents of toxic metals and essential elements were detected in four fish species collected from four different points of Marmara Sea coast of Tekirdağ province. The analysis results (in mg/kg d.w) of the selected essential elements (Mn, Zn, Fe and Cu) and toxic metals (As, Cd, Cr, Hg, Ni and Pb) in muscles of *M. surmuletus*, *M. merlangus*, *S. vulgaris* and *L. piscatorius* fish species from coastal waters of Marmara Sea around Tekirdağ are summarized in Table 3. The results revealed that accumulation pattern of the analyzed metals in all bottom fish muscles was as follows: Fe (58.8) > Zn (23.9) > Mn (3.3) > As (1.44) > Cu (0.8) > Cr (0.6) > Ni (0.3) > Cd (0.08) mg/kg d.w. However, Pb and Hg metals were not determined in the fish muscle samples. Both of them were below detection limit of ICP-OES. ANOVA analyses indicated significant variations ($p < 0.05$) of Fe, Zn, Mn, As, Cu, Cr, Ni, and Cd in the muscles of different fish species. Significant variations ($p < 0.05$) of As were observed in different stations.

The minimum and maximum Fe accumulation in bottom fish muscle samples were 8.2 and 202.7 mg kg^{-1} in the *M. merlangus* and *L .piscatorius*, respectively. When an average ratio was taken into consideration among the fish species, the minimum Fe was (32.31 mg kg^{-1}) in the Şarköy and the maximum was (77.93 mg kg^{-1}) in the Marmara Ereğlisi. The levels of Fe concentration in the *L. piscatorius* muscle tissue samples collected from the sea by Tekirdağ province and in the *S. vulgaris* collected from Marmara Ereğlisi were higher than the permissible limit of 151 mg kg^{-1} d.w recom-

Table 3
Concentrations of essential and toxic metals (mg kg^{-1} d.w) in muscle tissue of bottom fish in the Tekirdağ coastal area

Location	Fe	Cu	Mn	Zn	As*	Cd	Cr	Ni	Pb	Hg
M. surmuletus										
Şarköy	63.75^{c}±0.35	1.30^{a}±0.00	1.25^{a}±0.07	23.25^{c}±0.07	13.95^{b}±0.36	0.03^{b}±0.02	0.23^{c}±0.08	0.06^{b}±0.03	ND	ND
Tekirdağ	16.05^{d}±0.21	0.95^{b}±0.07	0.85^{b}±0.07	27.05^{b}±0.07	5.93^{c}±0.11	0.62^{a}±0.08	4.44^{a}±0.10	4.48^{a}±0.24	ND	ND
Yenice	98.65^{a}±0.07	1.15^{ab}±0.07	1.45^{a}±0.07	100.10^{a}±0.28	31.00^{a}±1.17	0.09^{b}±0.06	0.88^{b}±0.22	0.26^{b}±0.05	ND	ND
Marmara Ereğlisi	65.20^{b}±0.28	1.30^{a}±0.00	1.00^{b}±0.00	18.75^{d}±0.35	8.05^{c}±0.18	0.07^{b}±0.01	0.38^{bc}±0.14	0.28^{b}±0.00	ND	ND
M. merlangus										
Şarköy	8.20^{d}±0.00	0.50^{b}±0.00	1.20^{b}±0.01	17.35^{a}±0.07	9.88^{b}±0.78	0.01^{a}±0.00	0.27^{a}±0.02	ND^{b}	ND	ND
Tekirdağ	14.70^{c}±0.14	0.50^{b}±0.00	0.95^{c}±0.07	16.30^{b}±0.14	10.83^{b}±0.43	0.02^{a}±0.00	0.14^{a}±0.05	ND^{b}	ND	ND
Yenice	58.80^{a}±0.14	0.75^{a}±0.07	0.60^{d}±0.00	15.15^{c}±0.21	9.31^{b}±0.24	0.04^{a}±0.01	0.20^{a}±0.06	ND^{b}	ND	ND
Marmara Ereğlisi	29.05^{b}±0.35	0.65^{ab}±0.07	1.75^{a}±0.07	16.65^{b}±0.07	18.68^{a}±0.78	0.03^{a}±0.01	0.12^{a}±0.00	0.10^{a}±0.01	ND	ND
S. vulgaris										
Şarköy	46.70^{b}±0.42	0.50^{c}±0.00	6.45^{c}±0.07	18.80^{c}±0.28	24.10^{a}±0.45	0.04^{a}±0.02	0.34^{a}±0.15	ND^{b}	ND	ND
Tekirdağ	9.90^{d}±0.28	1.55^{a}±0.07	8.60^{b}±0.14	27.00^{a}±0.42	12.31^{c}±0.55	0.04^{a}±0.02	0.60^{a}±0.06	0.08^{a}±0.03	ND	ND
Yenice	41.45^{c}±0.21	0.55^{c}±0.07	3.40^{d}±0.00	16.10^{d}±0.14	20.41^{b}±0.12	0.02^{a}±0.01	0.44^{a}±0.14	0.05^{ab}±0.03	ND	ND
Marmara Ereğlisi	183.55^{a}±1.48	0.95^{b}±0.07	23.20^{a}±0.42	25.20^{b}±0.28	20.62^{b}±0.25	0.07^{a}±0.02	0.54^{a}±0.08	0.04^{ab}±0.03	ND	ND
L. piscatorius										
Şarköy	10.60^{d}±0.14	0.20^{c}±0.00	0.40^{b}±0.01	13.70^{b}±0.01	26.17^{a}±0.24	0.03^{a}±0.01	0.21^{a}±0.00	ND^{b}	ND	ND
Tekirdağ	202.70^{a}±1.41	0.70^{a}±0.00	0.75^{a}±0.07	16.40^{a}±0.00	7.25^{b}±0.12	0.10^{a}±0.03	0.18^{a}±0.07	0.02^{a}±0.01	ND	ND
Yenice	58.05^{b}±1.20	0.70^{a}±0.00	0.30^{bc}±0.00	13.60^{b}±0.28	6.74^{b}±0.07	0.07^{a}±0.06	0.30^{a}±0.05	ND^{b}	ND	ND
Marmara Ereğlisi	33.90^{c}±0.00	0.35^{b}±0.07	0.20^{c}±0.00	16.95^{a}±0.07	4.63^{c}±0.02	0.01^{a}±0.02	0.20^{a}±0.09	ND^{b}	ND	ND
Guideline										
[36]	–	20	–	50	–	0.05	–	–	0.2	
[6]	–		–	30	–	2	1	–	1–6	
[31]	151	3	–	–	–	–	0.15	–	2	
[73]	43	–	5.5		0.26	0.2		1	0.6	
[41]	–	1.3	0.5	–	–	–	0.15	0.5	2	
[42]	–	–	–	–	2	0.05	–	–	0.3	
[74]	–	–	–	–	0.15	–	–	–	–	
gB 2736-94	–	10	–	50	0.5	0.1	0.5	–	–	

*Values with different superscript lowercase letters are significantly different ($P < 0.05$).

mended by ANZFA [31]. Fe amounts of fish muscles displayed significant differences ($P < 0.05$).

The level of Fe has been reported to range from 9.52 to 32.4 mg kg^{-1} d.w in fish muscle samples of the middle Black Sea, Turkey [32], 68.6–163 mg kg^{-1} d.w for fish muscle samples from Aegean Sea and Black Sea, Turkey [33], and 9.18–136 mg kg^{-1} w.w in fish species from Aegean and Mediterranean seas [34]. Iron ore is found in nature as oxides. The use of iron in industry (metallurgy, foundry, steel) is quite common. Mineral iron salts are used as pigments, catalysts and herbicides. They exist in the structure of magnetic bands, drugs, and semiconductors. Iron carbonate is found as a trace element in mammals (90–140 µg/100 mL in human blood). In addition, many plants (wheat, oat, green vegetables, fig, walnut, hazelnut, etc.) contain 3 valuable irons (ferric). Iron reaches into the aquatic environment via iron rich wastewater which is released when chemical substances are being cleaned. Therefore, it accumulates in the body of aquatic organisms [21]. The levels of Fe (8.2–202.7 mg kg^{-1}) recorded in the bottom fish muscle samples under this investigation were higher than the maximum recommended limits of 151 mg kg^{-1} in seafood [31]. Since metals are not found alone in nature, it is important to assess the state of organisms and environmental conditions under the influence of metal mixtures. For example, Ca and Fe deficiency in an organism increases the absorption of Pb [35].

Zn amounts of the bottom fish muscle samples were determined to be 13.6–100.1 mg kg^{-1} d.w. When an average ratio was taken into consideration among fish species, the minimum and maximum Zn values were in the Şarköy and Yenice, respectively. Zn amounts of fish muscles were significantly different ($P < 0.05$). The maximum Zn level permitted for fish species is 50 mg kg^{-1} according to Turkish Food Codex [36]. The level of Zn concentration in the *M. surmuletus* muscle tissue samples collected from Yenice station was higher than the permissible limit of 50 mg kg^{-1} d.w recommended by Turkish Food Codex. The level of Zn has been reported to range from 5.2 to 11 mg kg^{-1} d.w in fish muscle samples from the Black Sea, Bulgaria [37], 38.8–93.4 mg kg^{-1} d.w for fish samples from the Aegan Sea and Black Sea, Turkey [38], 1.45–8.07 mg kg^{-1} d.w for fish muscle samples from Hainnan, China Sea [27], and 6.63–14.8 mg kg^{-1} w.w for fish muscle tissues caught from Paradeniz Lagoon, the Mediterranean coastal area, Turkey [39]. Zn is mostly present in the form of zinc sulfide (ZnS) and carbonate ($ZnCO_3$) in nature. Various Zn compounds are widely used in the industry for galvanizing and to produce glass, varnish, paint, paper and protective materials. Among the trace elements necessary for mammals, one could count Zn as well (normal value being 80–145 µg/100 mL in blood). Seafood is very rich in terms of Zn and Fe which are crucial for the body. Despite the fact that these minerals are beneficial for the body, when they are too abundant, they can be toxic for the same body [21]. The levels of Zn (13.6–100.1 mg kg^{-1}) recorded in bottom fish muscle samples within this investigation were higher than the maximum recommended limits of 50 mg kg^{-1} in seafood [36].

Mn accumulation in the bottom fish muscle samples were recorded as 0.4 (min) and 23.2 mg kg^{-1} d.w (max) in *L. piscatorius* and *S. vulgaris*, respectively. While average Mn values of fish muscles collected from Şarköy was 2.35 mg kg^{-1}, values of fish muscle from the Marmara Ereğlisi had a mean of 6.5 mg kg^{-1}. Mn amounts of fish muscles were significantly different ($P < 0.05$). There is no information about maximum Mn levels in fish muscle samples in Turkish Food Codex [36]. The level of Mn has been reported to range from 1.28 to 7.4 mg kg^{-1} d.w in fish muscle samples of the Black and Aegean Seas [33], 0.38–1.06 mg kg^{-1} w.w for fish samples from Paradeniz Lagoon, Mediterranean coastal area, Turkey [39], and 0.79–1.26 mg kg^{-1} d.w for fish muscle samples from Eastern Black Sea, Turkey [40]. Mn is found in nature in the form of some mineral ores such as *manganosite* (MnO) and *braunite* (Mn_2O_3 $MnSiO_3$). In medicine, *potassium permanganate* ($KMnO_4$) is utilized for its astringent and antiseptic effects. Mn is one of the essential trace elements for the organisms. It plays an important role as a cofactor in many metabolic reactions such as the synthesis of fatty acids, phosphorylation and cholesterol. Serum Mn level is normally 2.5 µg/L. Mn is released into the aquatic environment by using various compounds of it in the industry (such as their usage in the construction of batteries, ceramics, electrical materials, glass and animal feed additives) [21]. The levels of Mn (0.4–23.2 mg kg^{-1}) recorded in fish muscle samples in this investigation were higher than the maximum recommended limits of 0.5 mg kg^{-1} in seafood [41].

The As values of the bottom fish muscle samples were determined to be 4.63–31 mg kg^{-1} d.w. When an average ratio was taken into consideration among fish species, the As value of *S. vulgaris* was higher than that of the other fish species; it was 12.31–24.10 mg kg^{-1} d.w. The As amounts of fish muscles were significantly different ($P < 0.05$). The maximum As level permitted for fish species is 2 mg kg^{-1} according to European Commission [42]. The level of As has been reported to range from 0.38 to 1.9 mg kg^{-1} d.w in fish muscle samples of the Black Sea, Bulgaria [37], 0.48–6.99 mg kg^{-1} w.w for fish samples from Hainan South China Sea, [27], 0.03–80.2 mg kg^{-1} d.w for fish muscle samples from the Three Gorges, Yangtze River, China [43], and 0.64–1.10 mg kg^{-1} w.w for fish muscle tissues from the Straits of Malacca [44]. In other reports, the As values of red mullet and whiting fish muscles were found to be 1.330–2.375 and 0.580–1.085 mg kg^{-1} d.w, respectively [45]. Our results are quite high comparing to this study [45].

The International Agency for Research on Cancer has classified inorganic As in their Group 1 of substances that are carcinogenic to humans [46]. Pure metal As and inorganic compounds (sulfides) are insoluble in water and are theoretically devoid of toxic effects, yet they can convert into toxic compounds (As_2O_3) by oxidation in humid environments. Organic As does not accumulate in the human body as it is rapidly excreted from the body. Water-soluble inorganic compounds (arsenious anhydride, arsenides, and alkali arsenides) are very toxic [47]. Inorganic As compounds are often used in agriculture as insecticides, herbicides, fungicides, desiccants, defoliants, and animal feed additives [48]. Inorganic As is the most toxic form of As compounds because it is stable and soluble and is abundant in high levels in drinking water. Among nutrients, marine products are the main source of As intake. As may be present in very high concentrations in some seafood (up to 100 ppm). This measured As is the total As concentration instead of its toxic inorganic form (arsenite). About 90% of the As in fish muscle tissues is in the form of non-toxic arsenobetain [49–51]. The levels of As (4.63–31 mg kg^{-1}) recorded in bottom fish muscle samples in this investigation were higher than the maximum recommended limits of 2 mg kg^{-1} in seafood [42].

Cu accumulation in the bottom fish muscle samples were found to be 0.2–1.6 mg kg^{-1} d.w. Average Cu values of fish muscle samples were 0.63, 0.93, 0.79 and 0.81 mg kg^{-1} d.w in the Şarköy, Tekirdağ, Yenice and Marmara Ereğlisi, respectively. Cu levels are permitted for fish species up to 20 mg kg^{-1} according to Turkish Food Codex [36]. Cu amounts of fish muscles were significantly different ($P < 0.05$). The level of Cu has been reported to range from 1.96 to 5.61 mg kg^{-1} w.w in fish muscle samples of Tigris River, Turkey [52], 0.34–1.4 mg kg^{-1} w.w in fish samples from Black Sea, Bulgaria [37], 0.07–0.94 mg kg^{-1} w.w in fish samples from Hainan South China Sea [27], and 0.65–2.78 mg kg^{-1} d.w in fish species from the Black Sea, Turkey [33], and 0.034–0.037 mg kg^{-1} w.w in fish muscle samples from Taihu lake, China [53]. Copper is a metal which is red, bright, very conductive, easily oxidized when exposed to air and capable of making alloys with other materials (e.g. bronze, brass). It is widely used in the electrical and lead industry. Copper exists as an essential trace element in the organisms. Copper deficiency leads to hypochromic microcytic anemia due to hemoglobin synthesis disorder [21]. The levels of Cu (0.2–1.6 mg kg^{-1}) recorded in bottom fish muscle samples in this investigation were lower than the maximum recommended limits of 20 mg kg^{-1} in seafood [36].

Cr amounts of fish muscle samples were 0.14 mg kg^{-1} (*M. merlangus*) and 4.44 mg kg^{-1} d.w (*M. surmuletus*). Cr amount of *M. surmuletus* was significantly different ($P < 0.05$). However, in the other fish muscles, significant differences ($P > 0.05$) were not observed. The level of Cr has been reported to range from 0.63 to 1.74 mg kg^{-1} d.w in fish species caught from the Black Sea, Turkey [38], 0.07–1.48 mg kg^{-1} w.w for fish samples from Aegean and Mediterranean Sea, Turkey [34], 1.38–4.36 mg kg^{-1} w.w in fish muscle samples from Hainan South China Sea [23], 24.37–130.27 µg kg^{-1} d.w in fish species from the Caspian Sea [54], and 0.032–0.092 mg kg^{-1} w.w in fish muscle samples from Taihu lake, China [53]. Cr is used as toner for photocopiers, and it is used in smaller quantities in various industries such as drilling mud and textile. Humans are exposed to Cr (particularly to Cr III) via food, drinking water and breathing the air that contains chemicals. Cr III is an important element for adults who need to take 50–200 µg of it per day. Acute animal tests have shown that CrIII has moderate toxicity from oral exposure. CrVI is much more toxic than Cr III for both acute and chronic exposures [21,47]. The levels of Cr (0.14–4.44 ppm) recorded in fish muscle samples were higher than the maximum recommended limits of 0.15–1.0 ppm in seafood [41,55].

Ni accumulation in fish muscle samples were determined to be <0.5 mg kg^{-1} (0.02–0.26 mg kg^{-1}) except for *M. surmuletus* which had a value of 4.48 mg kg^{-1} d.w. The maximum Ni level permitted is 0.5–0.6 mg/kg by WHO/FEPA [41,55]. The level of Ni has been reported to range from 0.19 to 1.81 mg kg^{-1} d.w in fish species caught from Tigris River, Turkey [52], 1.14–3.60 mg kg^{-1} d.w for fish species in the Black Sea, Turkey [38], <0.01–31 mg/kg for wet weight (w.w) in aquatic organisms in coastal areas of South China [56], 0.12–0.18 mg kg^{-1} d.w for fish species in the Gulf of Paria, Venezuela [57], and 1.37–2.56 mg kg^{-1} d.w for fish species in Ogun Coastal Water, Nigeria [58]. Ni is a natural element of earth crust; therefore, it is found in small quantities in food, water, soil and air. Food is the main source of Ni exposure. The average intake level for adults is approximately 100–300 µg per day. The result in the present study (0.02–4.48 mg kg^{-1}) was higher when compared to the maximum recommended limits of 0.5–0.6 mg kg^{-1} in seafood [41,55].

Cd values of fish muscle samples varied from 0.01 (for *L. piscatorius)* to 0.62 (for *M. surmuletus)* mg kg^{-1} d.w. Significant differences ($P < 0.05$) were observed in the Cd amount of *M. surmuletus*. The maximum limits for Cd level permitted for marine fish samples were 0.05 mg kg^{-1} w.w by European Community [42]. The level of Cd has been reported to range from 0.006 to 0.015 mg kg^{-1} w.w for fish samples from the Black Sea, Bulgaria [37], 0.10–0.35 mg kg^{-1} d.w for fish species from the Black Sea, Turkey [38], 0.002–0.012 mg/kg w.w for fish samples from Hainan South China Sea [27], and 0.023–0.042 mg kg^{-1} w.w for fish muscle tissue samples from Taihu lake, China [53]. Usero et al. [59] determined 0.08–0.43 and 0.01–0.028 (mg/kg wet mass) Cd values in *S. vulgaris* livers and muscles in the southern Atlantic coast of Spain, respectively. Cd accumulation of *Mullus barbatus ponticus* and *Merlangiusmerlanguseuxinus* in the Southeastern Black Sea were 0.02 and 0.04 mg kg^{-1} d.w, respectively [60]. In some other studies, Cd amounts of *Merlangiusmerlanguseuxinus* fish were found to be 0.001 [61], 0.192 [62] and 0.55 mg kg^{-1} d.w [63]. Cd is found in nature in the form of yellow cadmium sulfide (CdS) together with zinc ores. Since metallic Cd is resistant to corrosion, it is widely used in various forms. Metallic Cd is often used for alloys, coating, lacquer, paint production etc. Cd intoxications are included in the list of occupational diseases in EU countries. Cd levels in some foods may increase due to the application of phosphate fertilizer or the use of sewage sludge in agricultural areas. Cd is also toxic to aquatic invertebrates for which toxicity depends on the parameters of the water quality [21,47,64]. The levels of Cd (0.01–0.62 mg kg^{-1}) recorded in fish muscle samples were higher than the maximum recommended limits of 0.05 mg kg^{-1} in seafood [42].

3.2. Risk assessment to human health

Because of heavy metals' toxicity and their threat to human health, different methods have been established to assess the potential health risk of them to those who consume them [27,65]. In our study, average bioaccumulation levels (C_{fish}) of fish muscle tissues of bottom fish caught in the seashore in Tekirdağ province in the Marmara Sea and consumed widely, were taken as a basis, and EDI and EWI quantities for adults along with THQ were evaluated. (Table 4). A dry-weight to wet-weight conversion factor of 4.8 for fish was used in this study [66,67].

The EDI was compared to the RfD proposed by the EPA (2013). The EDI values of each metal in the studied bottom fish muscles were less than the corresponding recommended values [47] (Table 4). The EDI values of each heavy metal (except for inorganic As) and each essential metal in the studied fish muscles were less than that of the previous

Table 4
Estimated dietary intake (µg kg^{-1} day^{-1} and week^{-1}) of Fe, Cu, Mn, Zn, As, Cd, Cr and Ni of fish muscle samples (based on 70 g of human body)

	Fe	Cu	Mn	Zn	As*	Cd	Cr	Ni
Average concentration (mg/kg d.w)	58.8	0.8	3.3	23.9	1.44	0.08	0.6	0.3
Average concentration (mg/kg w.w)	12.25	0.16	0.7	5.0	0.3	0.02	0.13	0.07
EDI dry (µg/kg^{-1} d^{-1})	16.8	0.22	0.94	6.8	0.4	0.02	0.17	0.1
EWI (µg/kg^{-1} d^{-1})	117.6	1.54	6.6	47.6	2.9	0.14	1.2	0.7
EDI wet (µg/kg^{-1} d^{-1})	3.5	0.05	0.2	0.19	0.08	0.006	0.04	0.02
EWI wet (µg/kg^{-1} d^{-1})	24.5	0.35	1.4	1.3	0.6	0.042	0.28	0.14
RfD (µg/kg^{-1} d^{-1})	700	40	140	300	0.3	1	3	20
THQdry	0.02	0.005	0.007	0.02	1.36	0.02	0.06	0.005
THQwet	0.005	0.001	0.001	0.0006	4.7	0.006	0.01	0.001

Conversion factor (wet weight to dry weight): 4.8

*Inorganic As was estimated by using a value of 10 % of total arsenic an accordance with the United States Food and Drug Administration (USFDA).

reports [37,68–70]. No health problem would be explored as a result of consumption of these fish caught in the Marmara Sea coast of Tekirdağ.

THQ parameters are useful for evaluating the risk assessment of heavy metal ingestion associated with the consumption of contaminated seafood [14,71,72]. Ranging from 0.005 to 0.06 except for inorganic As, the mean THQ of the metals was low in the fish species. The THQ values of inorganic As were higher than 1 which indicated that inorganic As would likely pose health risks to seafood consumers. Another study also reported that THQ value for arsenic was higher than 1 [27]. The fact that THQ values of other metals were lower than 1 apart from As, suggested that average fish consumption in Turkey is low. The levels of heavy metals (other than Cu) and trace element levels were higher than the recommended national and international legal limits. This finding suggests that if consumption of these fish species rises, there might be an increase in the health risks of the consumers.

4.Conclusion

Many studies have been carried out on the bioaccumulation of heavy metals and essential metals in fish muscle tissues which are the part of seafood consumed by humans. The determination of accumulation is of great importance in the risk assessment for human health. In our study Pb and Hg were below the detection limit of ICP-OES. The levels of all heavy metals and trace elements except for Cu in fish muscle tissue samples were above the recommended national and international legal limits for human consumption. Although THQ values calculated for other heavy metals except for As were less than 1, the reason for this is the lack of fish consumption in Turkey. In addition, taking other chemicals into account, studies on mixture toxicity should be carried out and risk assessment results should be supported. In conclusion, as long as they are consumed in reasonable and small amounts, deep water fish caught from Tekirdağ province of Marmara Sea are safe for human consumption with respect to heavy metal accumulation except for inorganic As which is likely to pose health risks to seafood consumers.

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