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Composition and human health risk analysis of elements in muscle tissues of wild and farmed fish species from Northeast Mediterranean

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ABSTRACT

It was aimed to determine the concentrations of several heavy metals in muscle tissues of 18 wild and 2 farmed fish species obtained from 6 stations and 3 different fish farms, which were located between Hatay and Antalya provinces in the Northeast Mediterranean. The metal analysis of tissue samples was conducted using the Inductively-coupled Plasma Mass Spectrometry (ICP-MS) method. Given all the tissue samples, the mean metal concentrations were found to be as follows; Li: < 0.001–0.15 mg kg^{-1} ww, Al: < 0.0006–2.54 mg kg^{-1} ww, Cr: < 0.001–0.49 mg kg^{-1} ww, Mn: < 0.0002–1.05 mg kg^{-1} ww, Fe: < 0.001–0.02 mg kg^{-1} ww, Cu: < 0.006 – 1.30 mg kg^{-1} ww, Zn: 1.76–15.4 mg kg^{-1} ww, As: 0.52–35.1 mg kg^{-1} ww, Sn: < 0.0004–2.04 mg kg^{-1} ww. Co, Ni, Cd, and Pb were found to be lower than the detectable limits in all tissue samples. For human consumption estimated weekly intakes (EWI), target hazard quotients (THQ), and cancer risk (CR) levels of the metals were determined in muscle tissues of the wild and farmed fish species. The mean EWI, THQ, and CR levels for As were found 34.82, 3.97E+ 00 ± 4.84E+ 00 and 1.06–03 ± 1.30E-03 for wild species while these values for farmed species were as follows; 2.46, 2.80E-01 ± 1.18E-01 and 7.49E-05 ± 3.15E-05 respectively. When compared to the wild species, metal levels were significantly lower in farmed fishes and consuming these fishes was determined not to cause any health problems. For the wild samples, however, inorganic As concentrations were found to be risky from carcinogenic and non-carcinogenic aspects. Moreover, the mean metal concentrations in muscle tissues of fishes obtained from the Northeast Mediterranean were compared to the national/international codex limits and it was found that all the metal concentrations in tissue samples were lower than the upper limits set by Turkish Food Codex (TFC), European Union (EU), and World Health Organization (WHO).

1. Introduction

Fish is a unique nutrition source, which is rich in fundamental nutrients, protein, lipids, and minerals in comparison to other animal meats and can be easily digested by human populations (Miao et al., 2020). Fish meat is an essential component of a healthy and balanced diet for human populations and fish consumption gradually increases day by day, because of its advantages such as low cholesterol level, high vitamin diversity, and fatty acid content such as Omega-3 (Sidhu, 2003). Besides constituting 17% of animal protein sources, seafood meets 6% of the total protein intake of humans and the global annual fish consumption reached 20.5 kg per capita (Avigliano et al., 2019; Copat et al., 2013). Moreover, it is known that regular fish consumption decreases the risk of various diseases such as preterm birth, stroke, arrhythmia, thrombosis, heart attack, and asthma, hence it should be consumed a minimum of twice a week (Costa, 2007; Tanamal et al., 2021).

Because fish are at the top of aquatic ecosystems (Kwaansa-Ansah et al., 2019), they play a major role in delivering pollutants to humans (Verbeke et al., 2005). In aquatic ecosystems, heavy metals are one of the most important pollutants absorbed by fish and transferred to upper trophic levels through food or the environment (Miri et al., 2017). Much greater attention should be paid to heavy metals because they are not biologically degradable, remain in nature for a long time, and accumulate in the food chain at gradually increasing concentrations (Bettini et al., 2006). The disasters such as "Minamata", "İtai-itai", "Alamosa River", and "Ok Tedi Mine" had heavy metal origins and, despite the years that have passed, their social and environmental effects are still present (Castro-González and Méndez-Armenta, 2008; Hyndman, 2001; Imamura et al., 2007; Woody et al., 2010). Moreover, in recent studies, it was reported that As levels in muscle tissues of wild and marketed fish

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species obtained from the Northeast Mediterranean region were found in high levels and they pose risks in terms of carcinogenic and non-carcinogenic effects (Korkmaz et al., 2017, 2016; Korkmaz, Ay, Çolakfakıoğlu et al., 2019). In the same studies it was also stated that Zn concentrations increased and exceeded the limits set by international organizations.

Hatay, Adana, Mersin, and Antalya, which are crowded, industrialized, and urbanized cities with intense tourism and fishing activities, are located on the Northeast Mediterranean coasts of Turkey. With its 65 km-long shores, Iskenderun Gulf is situated within the borders of Hatay province, which hosts many iron and steel facilities (Sarihan et al., 2006). Mersin Gulf which has 320 km shoreline, is under the effects of anthropogenic (industrial, agricultural etc.) pollutants (Yılmaz et al., 2016). With its urbanized structure and 2-million population, Adana is one of the most crowded cities of Turkey (Say et al., 2017). Antalya is the most important tourist destination in Turkey, hosting 10–15 million tourists annually (Kökçen, 2019; Olguner et al., 2018). Besides its tourism potential, Antalya has the highest agricultural production level among the cities in the country (ADPAF, 2019). Moreover, the Northeast Mediterranean has recently become one of the most important locations in Turkey in terms of fish farming. According to the Republic of Turkey Ministry of Agriculture and Forestry, the farmed-fish production (*Sparus aurata* and *Dicentrarchus labrax*) in Mersin has reached 12000 tons (RTMAF, 2019a).). With the new licenses given for the region, it is foreseen that the fish production capacity of the aquaculture industry will increase to 100,000 tons soon (RTMAF, 2019b).

For these reasons, the present study aimed to determine the compositions of metals (Li, Al, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Cd, Sn, and Pb) in the muscle tissues of 18 fish species (*Sarda sarda*, *Mullus surmuletus*, *Sardina pilchardus*, *Boops boops*, *Scomber japonicus*, *Saurida lessepsianus*, *Trachurus trachurus*, *S. aurata* (wild and aquaculture), *Pagrus pagrus*, *Mullus barbatus*, *Sphyraena sphyraena*, *Scomber japonicus*, *Saurida undosquamis*, *D. labrax* (wild and aquaculture), *Mugil cephalus*, *Solea solea*, *Nemipterus randalli*, *Lithognathus mormyrus*) obtained from 6 stations and 3 different fish farms, which were located between Hatay and Antalya provinces in the Northeast Mediterranean. After the statistical analysis, the results obtained from the muscle tissues were compared to the standards set by Turkish Food Codex (TFC), European Union (EU), and World Health Organization (WHO) and by calculating the estimated weekly intakes (EWI), they were compared to the metals' provisional tolerable weekly intake (PTWI) limits. In addition, the target hazard quotients (THQ) and cancer risk (CR) levels of metals in muscle tissues of wild and farmed fishes were evaluated and risk assessments were conducted for human consumption.

2. Material and method

Fish samples were taken by fishing or harvesting from 6 different stations and 3 different fish farms between Hatay and Antalya in autumn 2020. The samples were taken to Mersin University, Faculty of Fisheries and Basic Sciences Procedure Laboratory via cold chain, and the muscle tissues were stored at − 20 °C temperature. From all the stations, 258 fish samples were taken in total and the coordinates of sampling stations are presented in Fig. 1.

After measuring the wet weights of muscle tissues, they were transferred to petri dishes and kept at 150 °C for 48 h until ready for weighing. After measuring the dry weights, the muscle tissues were taken to the test tubes and added with 2 ml nitric acid (HNO_3, % 65, SW: 1.40, Merck, Darmstadt, Germany) and 1 ml perchloric acid ($HClO_4$, % 60, SW: 1.53, Merck, Darmstadt, Germany) mixture and digested at 120 °C for 8 h (Korkmaz, Ay, Çolakfakıoğlu et al., 2019). Digested samples were taken to polyethylene tubes and added with deionized water until filled to 10 ml. The same procedure was conducted for the blank samples with no tissue specimen. Before the analysis, the samples were filtered through 0.45 μm membrane filter. Li, Al, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Cd, Sn, and Pb concentrations of tissue samples were determined using Agilent 7500ce (Octopole Reaction System, Agilent Technologies, Tokya, Japan) ICP-MS system.

The metal concentrations in tissue samples were calculated in dry weight (mg kg^{-1} dw) and, by considering the water content of each

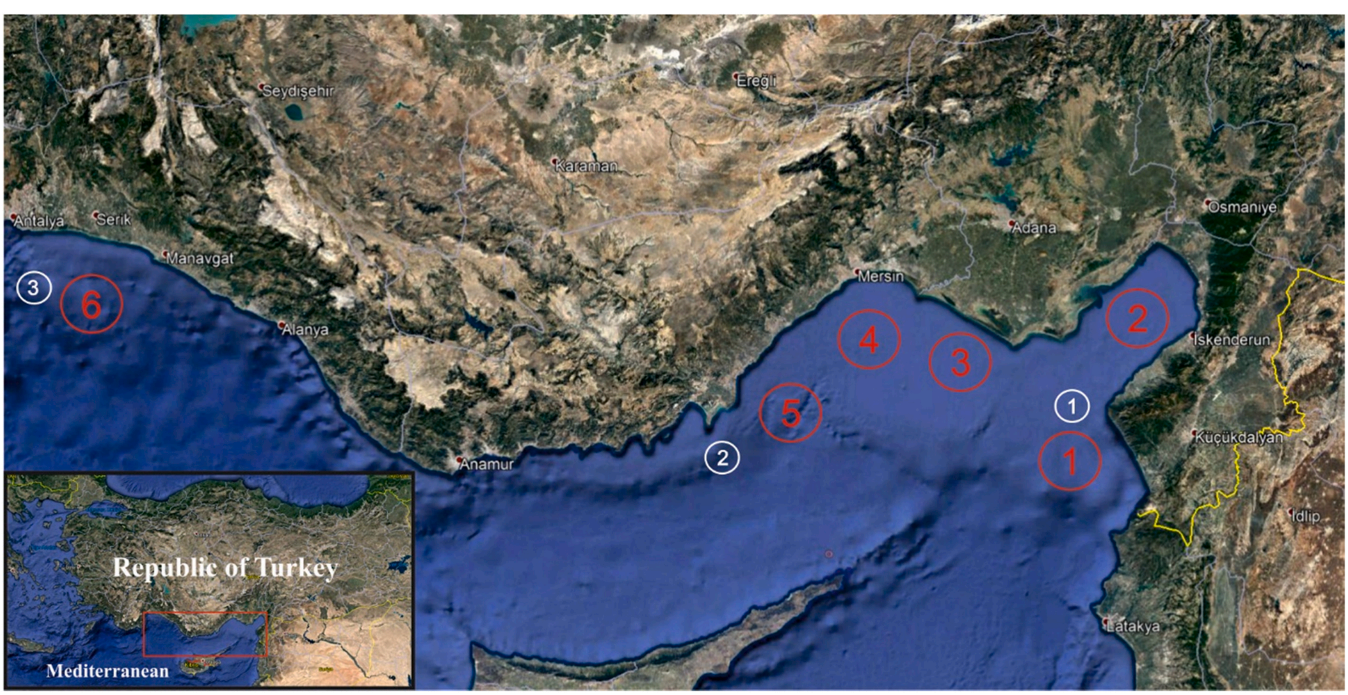

Fig. 1. Locations of sampling stations (red circles refer to the sampling stations of wild species and white circles refer to the fish farming facilities). The coordinates of sampling stations for wild species: 1- Samandağ (36°04'04.6"N 35°43'41.6"E), 2- Iskenderun (36°40'02.4"N 35°58'14.2"E), 3- Adana – Karataş (36°29'42.1"N 35°24'27.9"E), 4- Mersin (36°43'57.5"N 34°38'55.3"E), 5- Silifke (36°16'09.6"N 33°52'46.2"E), 6- Antalya (36°37'13.2"N 30°57'00.7"E). The coordinates of stations for farmed species are not provided in order not to advertise the commercial companies.

tissue, the values were converted to wet weight (mg kg^{-1} ww) (Korkmaz, Ay, Çolakfakıoğlu et al., 2019). The analyses were triplicated. International Atomic Energy Agency (IAEA-407, Vienna, Austria) fish homogenate was used for recovery studies. The same procedures were conducted for the IAEA-407 samples. Recovery values of reference material were given in Table 1. The detection limits for Li, Al, Cr, Mn, Fe, Ni, Cu, Zn, As, Cd, Sn and Pb were as follows; 0.001, 0.0006, 0.001, 0.0002, 0.001, 0.0006, 0.0006, 0.002, 0.005, 0.0004, 0.0004, and 0.0003 mg kg^{-1}.

The results obtained from tissue samples were adjusted for human consumption by making use of the formulas below and it was determined if the fish species obtained from the Northeast Mediterranean region are suitable for human consumption in terms of metal content.

2.1. 2.1 Human risk assessment

The amounts of daily and weekly metal intake (EDI and EWI) by consumption of sampled fish species were calculated with the given formulas below;

$$\text{EDI}(\mu\text{g/day/70kg body}\textit{weight}) = (C\ x\ FIR) \quad (1)$$

$$\text{EWI }(\mu\text{g/week/70kg body}\textit{weight}) = EDI\ x\ 7days \quad (2)$$

C (Concentration); Mean metal level of muscle tissue (mg kg^{-1} ww).

FIR (Fish consumption ratio); Daily consumption of fish is approximately 17 g per capita according to Turkish Minister of Agriculture and Forestry (RTMAF, 2020). BW (Body weight); Average bodyweight of Turkish people was announced by WHO to be 70 kg (WHO, 2018).

The health risk of non-carcinogenic and carcinogenic effects due to the consumption of fish were evaluated based on the THQ and CR equations (Gu et al., 2018).

$$THQ = \frac{\text{EF x ED x EDI}}{AT\ x\ RfD\ x\ BW}\ x10^{-3} \quad (3)$$

$$CR = \frac{EF\ x\ ED\ x\ EDI\ x\ CSF}{ATx\ BW} x10^{-3} \quad (4)$$

$$Total\text{THQ(TTHQ)} = \text{THQ(metal1)} + \text{THQ(metal2)} + \ \ldots\ + \text{THQ(metal}n) \quad (5)$$

PTWI (µg/week/70 kg bodyweight) = RfD for each metal (Table 2) x BW x 7 (days) (6)

Table 2 defines the parameters used in Eqs. (3)–(4), as well as data for Turkish people used in the calculations. Table 2.

2.2. Statistical analysis

The statistical analyses of the results obtained from the tissue samples were conducted using t-test, one-way variance analysis (ANOVA), and Student-Newman Keuls (SNK) tests by making use of SPSS 16.0 package software. Data normality were checked by boxplot graphs before statistical analysis (Ghasemi and Zahediasl, 2012).

3. Results and discussion

The mean metal concentrations in the muscle tissues of the fishes from the Samandağ, Iskenderun, Adana (Karataş), Mersin, Silifke, and Antalya stations are presented in Table 3. The concentrations of Co, Ni, Cd, and Pb were found to be lower than the detectable limits in all the samples. Considering all the fish tissues, the mean metal concentrations (mg kg^{-1} ww) were found to range between < 0.001–0.15 for Li, < 0.0006–2.54 for Al, < 0.001–0.49 for Cr, < 0.0002–1.05 for Mn, < 0.001–0.02 for Fe, < 0.006 – 1.30 for Cu, 1.76–15.4 for Zn, 0.52–35.1 for As, and < 0.0004–2.04 for Sn.

The mean metal concentrations found in the muscle tissues of wild and farmed species obtained from Northeast Mediterranean are illustrated in Fig. 2. The mean metal concentrations in muscle tissues of wild and farmed fish species were 0.02 ± 0.05 mg kg^{-1} ww, 0.01 ± 0.03 mg kg^{-1} ww (Li); 0.11 ± 0.61 mg kg^{-1} ww, 0.06 ± 0.27 mg kg^{-1} ww (Al); 0.05 ± 0.18 mg kg^{-1} ww, < 0.001 mg kg^{-1} ww (Cr); 0.25 ± 0.42 mg kg^{-1} ww, 0.07 ± 0.20 mg kg^{-1} ww (Mn); 0.01 ± 0.01 mg kg^{-1} ww, 0.00 ± 0.01 mg kg^{-1} ww (Fe); 0.36 ± 0.52 mg kg^{-1} ww, 0.19 ± 0.32 mg kg^{-1} ww (Cu); 6.58 ± 4.33 mg kg^{-1} ww, 4.40 ± 1.74 mg kg^{-1} ww (Zn); 8.36 ± 10.17 mg kg^{-1} ww, 0.59 ± 0.25 mg kg^{-1} ww (As), and 0.27 ± 0.79 mg kg^{-1} ww, 0.19 ± 0.98 mg kg^{-1} ww (Sn), respectively. Co, Ni, Cd, and Pb concentrations were found to be lower than the detectable limits in all the muscle tissues of wild and farmed fish species.

3.1. Lithium

USA Environmental Protection Agency (US-EPA) reported that RfD value of lithium is 2 µg kg^{-1} day^{-1} and any intake higher than this level might cause central nervous system impairment, renal damages and

Table 1
Recovery values of reference material IAEA-407.

Analyte	Certified value	Unit	95% Confidence interval	Std Deviation	Observed value
Li	0.685	mg kg^{-1}	0.62 – 0.74	0.094	0.63
Al	13.8	mg kg^{-1}	12.4 – 15.2	3.7	12.9
Cr	0.73	mg kg^{-1}	0.67 – 0.79	0.22	0.70
Mn	3.52	mg kg^{-1}	3.44 – 3.60	0.32	3.38
Fe	146	mg kg^{-1}	143 – 149	14	137
Co	0.1	mg kg^{-1}	0.09 – 0.11	0.02	0.09
Ni	0.6	mg kg^{-1}	0.55 – 0.65	0.18	0.59
Cu	3.28	mg kg^{-1}	3.20 – 3.36	0.40	3.20
Zn	67.1	mg kg^{-1}	66.3 – 67.9	3.8	61.88
As	12.6	mg kg^{-1}	12.3 – 12.9	1.2	12.1
Cd	0.189	mg kg^{-1}	0.185 – 0.193	0.019	0.181
Sn	0.1	mg kg^{-1}	0.06 – 0.13	0.05	0.09
Pb	0.12	mg kg^{-1}	0.10 – 0.14	0.06	0.11

Table 2
Parameters and values used in the human health risk models (Korkmaz, Ay, Ersoysal et al., 2019).

Factor	Definition	Unit	Value
BW	Average bodyweight	Kg	70
EF	Exposure frequency	Days/ year	365
ED	Exposure duration	Years	70
AT	Average time	Days	25550 (BW x EF)
RfD	Oral reference dose	mg kg^{-1} day^{-1}	2.0E-03 (Li), 1E+ 00 (Al), 1.5E+ 00 (Cr^{+3}), 3.0E-03 (Cr^{+6}), 1.4E-01 (Mn), 7E-01 (Fe), 3.0E-04 (Co), 2.0E-02 (Ni), 4E-02 (Cu), 3E-01 (Zn), 3E-04 (As), 1E-03 (Cd), 6.0E-01 (Sn), 4E-03 (Pb)
CSF	Cancer slop factor	mg kg^{-1} day^{-1}	1.5E+ 00 (As)

Table 3
Levels of metals^{+} in muscle tissues of fish species collected from North-East Mediterranean (mg kg^{-1} ww).

Station	Species	Li*	Al*	Cr*	Mn*	Fe*	Cu*	Zn*	As*	Sn*
Samandağ	S.s	0.01 ± 0.01^{a}	BDL^{a}	BDL^{a}	0.11 ± 0.15^{a}	0.01 ± 0.01^{a}	0.63 ± 0.61^{a}	4.38 ± 0.83^{a}	1.62 ± 0.26^{a}	0.62 ± 0.27^{a}
	M.s	0.01 ± 0.01^{a}	0.17 ± 0.28^{a}	BDL^{a}	0.53 ± 0.33^{ab}	BDL^{a}	0.06 ± 0.14^{a}	3.97 ± 1.40^{a}	11.21 ± 4.15^{b}	BDL^{b}
	S.p	0.06 ± 0.03^{ab}	BDL^{a}	BDL^{a}	0.74 ± 0.42^{b}	0.01 ± 0.01^{a}	0.23 ± 0.21^{a}	6.69 ± 2.59^{a}	4.56 ± 1.58^{a}	0.42 ± 0.44^{ab}
	B.b	0.08 ± 0.03^{ab}	BDL^{a}	BDL^{a}	0.22 ± 0.31^{ab}	0.01 ± 0.01^{a}	BDL^{a}	7.58 ± 3.13^{a}	5.06 ± 0.77^{a}	BDL^{b}
	S.j	0.09 ± 0.09^{b}	BDL^{a}	BDL^{a}	0.08 ± 0.17^{a}	0.01 ± 0.01^{a}	0.54 ± 0.58^{a}	5.23 ± 3.46^{a}	3.08 ± 1.94^{a}	0.51 ± 0.38^{a}
	S.l	0.02 ± 0.04^{ab}	BDL^{a}	BDL^{a}	0.20 ± 0.45^{ab}	BDL^{a}	BDL^{a}	4.50 ± 1.83^{a}	4.73 ± 2.67^{a}	BDL^{b}
	T.t	0.02 ± 0.01^{ab}	BDL^{a}	BDL^{a}	0.25 ± 0.25^{ab}	BDL^{a}	0.34 ± 0.35^{a}	4.43 ± 1.09^{a}	3.22 ± 0.41^{a}	BDL^{b}
İskenderun	S.a^{Aq}	0.07 ± 0.04^{ab}	0.38 ± 0.65^{a}	BDL^{a}	BDL^{a}	BDL^{a}	0.24 ± 0.41^{a}	6.2 ± 1.9^{abc}	0.69 ± 0.10^{a}	BDL^{a}
	M.s	0.15 ± 0.05^{b}	2.54 ± 1.85^{b}	BDL^{a}	0.51 ± 0.42^{ab}	0.01 ± 0.01^{a}	0.74 ± 0.15^{ab}	7.37 ± 1.67^{bc}	35.1 ± 7.1^{b}	2.04 ± 1.93^{b}
	P.p	0.10 ± 0.08^{b}	0.48 ± 1.18^{a}	BDL^{a}	BDL^{a}	0.01 ± 0.01^{a}	0.27 ± 0.42^{a}	5.38 ± 0.95^{ab}	25.9 ± 11.1^{c}	1.05 ± 1.43^{ab}
	S.l	BDL^{ab}	BDL^{a}	BDL^{a}	BDL^{a}	BDL^{a}	BDL^{a}	3.48 ± 0.68^{a}	5.58 ± 0.47^{a}	0.19 ± 0.29^{a}
	S.p	0.09 ± 0.05^{b}	BDL^{a}	BDL^{a}	0.92 ± 0.49^{b}	0.01 ± 0.01^{a}	0.82 ± 0.49^{ab}	6.80 ± 0.97^{bc}	1.71 ± 0.24^{a}	BDL^{a}
	S.s	BDL^{ab}	BDL^{a}	BDL^{a}	0.09 ± 0.21^{a}	BDL^{a}	BDL^{a}	5.25 ± 0.66^{ab}	2.31 ± 1.06^{a}	BDL^{a}
	S.j	BDL^{ab}	0.01 ± 0.03^{a}	BDL^{a}	0.11 ± 0.17^{a}	0.01 ± 0.01^{a}	1.29 ± 0.93^{b}	8.85 ± 3.36^{c}	5.27 ± 0.76^{a}	BDL^{a}
Adana (Karataş)	S.p	BDL^{a}	BDL	BDL^{a}	0.38 ± 0.43^{a}	0.02 ± 0.01^{a}	1.16 ± 0.34^{ac}	8.82 ± 3.44^{ab}	12.6 ± 6^{a}	1.95 ± 2.18^{a}
	B.b	BDL^{a}	BDL^{a}	BDL^{a}	0.15 ± 0.23^{a}	0.01 ± 0.00^{a}	0.28 ± 0.32^{b}	15.4 ± 7.9^{b}	4.97 ± 2.02^{b}	0.23 ± 0.26^{b}
	S.j	BDL^{a}	BDL^{a}	0.22 ± 0.49^{a}	BDL^{a}	0.01 ± 0.01^{a}	1.20 ± 0.44^{ac}	10.5 ± 1.8^{ab}	3.56 ± 0.73^{b}	BDL^{b}
	S.s	BDL^{a}	BDL^{a}	BDL^{a}	BDL^{a}	BDL^{a}	0.45 ± 0.05^{ab}	7.55 ± 0.29^{ab}	1.48 ± 0.07^{b}	BDL^{b}
	D.l	BDL^{a}	BDL^{a}	0.09 ± 0.15^{a}	0.15 ± 0.34^{a}	0.00 ± 0.01^{a}	0.49 ± 0.34^{ab}	12.9 ± 6.1^{ab}	1.90 ± 0.62^{b}	BDL^{b}
	S.a	BDL^{a}	BDL^{a}	0.04 ± 0.08^{a}	0.16 ± 0.16^{a}	BDL^{a}	0.42 ± 0.34^{ab}	6.36 ± 2.22^{a}	4.18 ± 2.20^{b}	BDL^{b}
	M.c	BDL^{a}	BDL^{a}	0.26 ± 0.27^{a}	0.69 ± 0.60^{a}	0.01 ± 0.01^{a}	1.30 ± 0.74^{c}	12.4 ± 3.2^{ab}	1.32 ± 0.48^{b}	BDL^{a}
Mersin	S.j	BDL^{a}	BDL^{a}	BDL^{a}	BDL^{a}	0.01 ± 0.01^{a}	0.34 ± 0.54^{ab}	4.36 ± 2.44^{a}	3.47 ± 1.49^{a}	BDL^{a}
	M.c	0.01 ± 0.02^{a}	BDL^{a}	BDL^{a}	0.19 ± 0.43^{a}	BDL^{a}	0.61 ± 0.58^{b}	12.7 ± 5.9^{b}	0.83 ± 0.19^{a}	BDL^{a}
	B.b	0.01 ± 0.02^{a}	BDL^{a}	0.13 ± 0.20^{a}	BDL^{a}	BDL^{a}	BDL^{a}	8.05 ± 5.85^{ab}	4.11 ± 1.14^{a}	BDL^{a}
	S.p	BDL^{a}	BDL^{a}	BDL^{a}	0.73 ± 0.48^{b}	BDL^{a}	0.25 ± 0.31^{ab}	7.42 ± 5.36^{ab}	4.30 ± 2.68^{a}	BDL^{a}
	M.s	0.05 ± 0.03^{b}	1.29 ± 1.85^{a}	BDL^{a}	0.87 ± 0.44^{b}	0.01 ± 0.01^{a}	BDL^{a}	4.57 ± 1.91^{a}	16.8 ± 2.6^{b}	1.64 ± 1.52^{b}
	P.p	0.01 ± 0.01^{a}	BDL^{a}	BDL^{a}	0.33 ± 0.57^{ab}	0.01 ± 0.03^{a}	BDL^{a}	2.02 ± 0.64^{a}	30.8 ± 11.7^{c}	0.59 ± 0.64^{c}
Silifke	S.a^{Aq}	BDL^{a}	BDL^{a}	BDL^{a}	BDL^{a}	BDL^{a}	BDL^{a}	2.98 ± 0.86^{a}	0.57 ± 0.08^{a}	BDL^{a}
	D.l^{Aq}	BDL^{a}	BDL^{a}	BDL^{a}	0.24 ± 0.35^{a}	0.01 ± 0.01^{a}	0.10 ± 0.23^{a}	4.62 ± 1.88^{a}	0.52 ± 0.37^{a}	1.25 ± 1.63^{a}
	M.s	BDL^{a}	BDL^{a}	BDL^{a}	0.39 ± 0.33^{a}	BDL^{a}	0.20 ± 0.31^{a}	2.90 ± 0.54^{a}	18.17 ± 5.92^{b}	BDL^{a}
	S.s	BDL^{a}	BDL^{a}	0.01 ± 0.02^{a}	0.36 ± 0.56^{a}	BDL^{a}	BDL^{a}	3.83 ± 1.10^{a}	19.4 ± 5.0^{b}	BDL^{a}
	S.a	BDL^{a}	BDL^{a}	BDL^{a}	BDL^{a}	BDL^{a}	0.13 ± 0.20^{a}	4.92 ± 2.14^{a}	5.67 ± 2.09^{a}	BDL^{a}
	S.l	BDL^{a}	BDL^{a}	0.04 ± 0.06^{a}	BDL^{a}	BDL^{a}	BDL^{a}	3.13 ± 0.97^{a}	3.54 ± 1.13^{a}	BDL^{a}
	M.c	BDL^{a}	BDL^{a}	0.08 ± 0.14^{a}	BDL^{a}	BDL^{a}	0.25 ± 0.42^{a}	2.24 ± 1.10^{a}	1.26 ± 0.33^{a}	BDL^{a}
	B.b	BDL^{a}	BDL^{a}	0.29 ± 0.50^{a}	BDL^{a}	BDL^{a}	0.08 ± 0.19^{a}	4.90 ± 3.65^{a}	2.06 ± 1.58^{a}	BDL^{a}
	N.r	BDL^{a}	BDL^{a}	BDL^{a}	BDL^{a}	BDL^{a}	0.10 ± 0.24^{a}	1.76 ± 0.92^{a}	4.78 ± 2.55^{a}	0.32 ± 0.50^{a}
Antalya	S.a^{Aq}	BDL^{a}	BDL^{a}	BDL^{a}	BDL^{a}	BDL^{a}	0.43 ± 0.41^{a}	4.51 ± 1.25^{ab}	0.61 ± 0.31^{a}	BDL^{a}
	S.j	0.01 ± 0.01^{a}	BDL^{a}	BDL^{a}	BDL^{a}	0.01 ± 0.01^{ab}	0.43 ± 0.54^{a}	7.2 ± 2.3^{bc}	1.58 ± 0.70^{a}	0.69 ± 0.59^{b}
	S.a	BDL^{a}	BDL^{a}	BDL^{a}	BDL^{a}	BDL^{a}	0.22 ± 0.35^{a}	6.80 ± 1.15^{bc}	34.0 ± 12.4^{b}	BDL^{a}
	B.b	0.13 ± 0.07^{b}	BDL^{a}	0.08 ± 0.15^{a}	BDL^{a}	BDL^{a}	0.12 ± 0.28^{a}	9.53 ± 4.20^{c}	3.27 ± 0.56^{a}	BDL^{a}
	L.m	BDL^{a}	BDL^{a}	BDL^{a}	BDL^{a}	BDL^{a}	BDL^{a}	6.01 ± 0.94^{ab}	8.76 ± 4.41^{a}	BDL^{a}
	P.p	BDL^{a}	BDL^{a}		BDL^{a}	BDL^{a}	0.09 ± 0.22^{a}	3.27 ± 0.78^{a}	6.42 ± 2.46^{a}	BDL^{a}

(continued on next page)

Table 3 (*continued*)

Station	Species	Li*	Al*	Cr*	Mn*	Fe*	Cu*	Zn*	As*	Sn*
	S.p	0.02 ± 0.02[a]	BDL[a]	0.21 ± 0.29[a] 0.49 ± 0.42[b]	1.05 ± 0.60[b]	0.02 ± 0.01[b]	0.67 ± 0.77[a]	13.4 ± 2.1[d]	5.06 ± 0.67[a]	BDL[a]

* $\overline{X} \pm S_{\overline{X}}$ = Mean ± Standard error

[+] Co, Ni, Cd and Pb levels were not given in Table 2, hence the levels of metals were found below detectible limits in all samples.

SNK; Letters a, b, c and d show differences of a given metal between the species of that station. Data shown with different letters are significant at 0.05 level.

S.s = Sarda sarda; M.s = Mullus surmuletus;; S.p = Sardina pilchardus; B.b = Boops boops; S.j = Scomber japonicus; S.l = Saurida lessepsianus; T.t = Trachurus trachurus; S.a[Aq] = Sparus aurata (Aquaculture); P. p = Pagrus pagrus; M.b = Mullus barbatus; S.p = Sphyraena sphyraena; S.j = Scomber japonicus; S.u = Saurida undosquamis; D.l = Dicentrarchus labrax; S.a = Sparus aurata; M.c = Mugil cephalus; D.l[Aq] = Dicentrarchus labrax (Aquaculture); S.s = Solea solea; N.r = Nemipterus randalli; L.m= Lithognathus mormyrus

BDL[a] = Below detection limit (The detection limits for Li, Al, Cr, Mn, Fe, Ni, Cu, Zn, As, Cd, Sn and Pb were as follows; 0.001, 0.0006, 0.001, 0.0002, 0.001, 0.0006, 0.0006, 0.002, 0.005, 0.0004, 0.0004 and 0.0003 mg kg^{-1}.)

even death (Kjølholt et al., 2003; USEPA, 2018).

In the present study, the mean lithium concentrations in muscle tissues of fishes obtained from the Northeast Mediterranean region were found to range between < 0.001 and 0.15 mg kg^{-1} ww. The highest lithium concentration was found in the muscle tissues of *M. surmuletus* in Iskenderun station.

Li concentrations in the muscle tissues of wild fish species from the Mediterranean Sea, which have high economic value, were reported to range between 0.05 and 0.25 mg kg^{-1} ww (Guérin et al., 2011), 5.46–13.7 mg kg^{-1} ww (Yılmaz et al., 2010), 0.010–0.011 mg kg^{-1} ww (Marengo et al., 2018), and 2.58–3.20 mg kg^{-1} dw (Turan et al., 2009).

There are only a few studies on the lithium concentrations in farmed fishes from the Mediterranean Sea and researchers reported the Li concentrations to vary between 0.013 and 0.017 mg kg^{-1} ww (Marengo et al., 2018) and < 0.05–0.01 mg kg^{-1} ww (Kalantzi et al., 2016).

When compared to the wild species, the lithium concentrations in muscle tissues of farmed fish species were found to be lower by 50%.

3.2. Aluminum

USEPA (2018) reported the RfD value of aluminum to be 1000 μg kg^{-1} day^{-1} and the intakes exceeding beyond this level might cause many health problems such as forgetfulness, depression, dementia, osteomalacia, anemia, and motor disorders (Crisponi et al., 2013).

In the present study, the mean aluminum concentrations in muscle tissues of fishes obtained from the Northeast Mediterranean were found to range between < 0.0006–2.54 mg kg^{-1} ww. The highest aluminum concentration was found in the muscle tissues of *M. surmuletus* in Iskenderun station.

The aluminum concentrations in muscle tissues of fishes, which have high economic values, hunted from Mediterranean were found to range between 2.23 and 4.93 mg kg^{-1} ww (Yılmaz et al., 2010), 0.02–5.41 mg kg^{-1} dw (Türkmen et al., 2005), 8.38–95.31 mg kg^{-1} dw (Turan et al., 2009), 0.46–0.65 mg kg^{-1} ww (Marengo et al., 2018) and 0.56 – 7.49 μg g^{-1} ww (Soliman et al., 2021).

The aluminum concentrations in muscle tissues of farmed fishes from the Mediterranean Sea were found to range between 0.07 and 0.74 mg kg^{-1} ww (Squadrone et al., 2016), 0.41–0.97 mg kg^{-1} ww (Marengo et al., 2018), and < 13.58–1.92 mg kg^{-1} ww (Kalantzi et al., 2016).

In the present study, the aluminum levels were found to be lower than in the previous studies and it was determined that the Al concentrations found in the muscle tissues of farmed fishes were 45% lower than in the wild species.

3.3. Chromium

Chromium is one of the most widely found elements in seawater and naturally exists in three forms: Cr^0, Cr^{+3}, and Cr^{+6} (Cefalu and Hu, 2004). Oral RfD of Cr^{+3} and Cr^{+6} were reported to be 1500 and 3 μg kg^{-1} day^{-1}, respectively, and intakes higher than these values might cause medical problems such as liver and kidney damages, allergic reactions, skin rash, ulcer, and gene mutations (USEPA, 2018).

The mean chromium concentrations in muscle tissues of fishes obtained from the Northeast Mediterranean were found to range between < 0.001 and 0.49 mg kg^{-1} ww and the highest concentration was found in muscle tissues of *S. pilchardus* from Antalya station.

The chromium concentrations in muscle tissues of fishes caught from the Mediterranean Sea were found to range between 0.07 and 6.46 mg kg^{-1} dw (Türkmen et al., 2005), 0.04–1.75 mg kg^{-1} dw (Türkmen et al., 2008), 2.15–7.10 mg kg^{-1} ww (Korkmaz, Ay, Ersoysal et al., 2019), 0.07–0.09 mg kg^{-1} ww (Iamiceli et al., 2015), 0.10–1.893 μg g^{-1} dw (Turan et al., 2009) and 0.14 – 0.43 μg g^{-1} ww (Mutlu et al., 2011).

The chromium levels in muscle tissues of farmed fishes in the Mediterranean Sea were found to range between 0.001 and 0.310 mg kg^{-1} ww (Lourenço et al., 2012), 0.011–0.014 mg kg^{-1} ww (Marengo et al., 2018), and 0.13–0.91 mg kg^{-1} dw (Castritsi-Catharios et al., 2015).

The chromium concentrations found in the present study were similar to those reported in previous studies and it was determined that the chromium accumulation in farmed species was 4 times lower when compared to the wild species ($p < 0.05$).

3.4. Manganese

Manganese, is one of the most abundant metals on earth and it is the cofactor of various enzymes including pyruvate carboxylase and superoxide dismutase (Goldhaber, 2003). USEPA (2018) reported the RfD of manganese to be 140 μg kg^{-1} day^{-1} and stated that the intake of manganese higher than this value might cause a psychological and neurological disorder called "manganism", which is very similar to Parkinson's disease (Andruska and Racette, 2015).

In this study, the manganese concentrations in muscle tissues of fishes were found to range between < 0.0002 and 1.05 mg kg^{-1} ww and the highest manganese concentration was found in the muscle tissues of *S. pilchardus* from Antalya station.

The manganese concentrations in muscle tissues of wild species hunted from the Mediterranean Sea were found to range between 0.30 and 0.60 mg kg^{-1} dw (Minganti et al., 2010), 0.08–0.10 mg kg^{-1} dw (Percın et al., 2011), and 0.10–0.99 mg kg^{-1} dw (Türkmen et al., 2008).

The manganese concentrations in muscle tissues of farmed fishes were reported to range between 0.07 and 0.42 mg kg^{-1} ww (Rubio et al., 2011), 0.3–1.1 mg kg^{-1} dw (Minganti et al., 2010), 0.12–0.40 mg kg^{-1} ww (Lourenço et al., 2012), and 0.11–0.14 mg kg^{-1} dw (Percın et al., 2011).

When compared to the previous studies, the manganese levels in the present study were found to be lower and farmed fish were found to have 72% less manganese accumulation in comparison to the wild fish

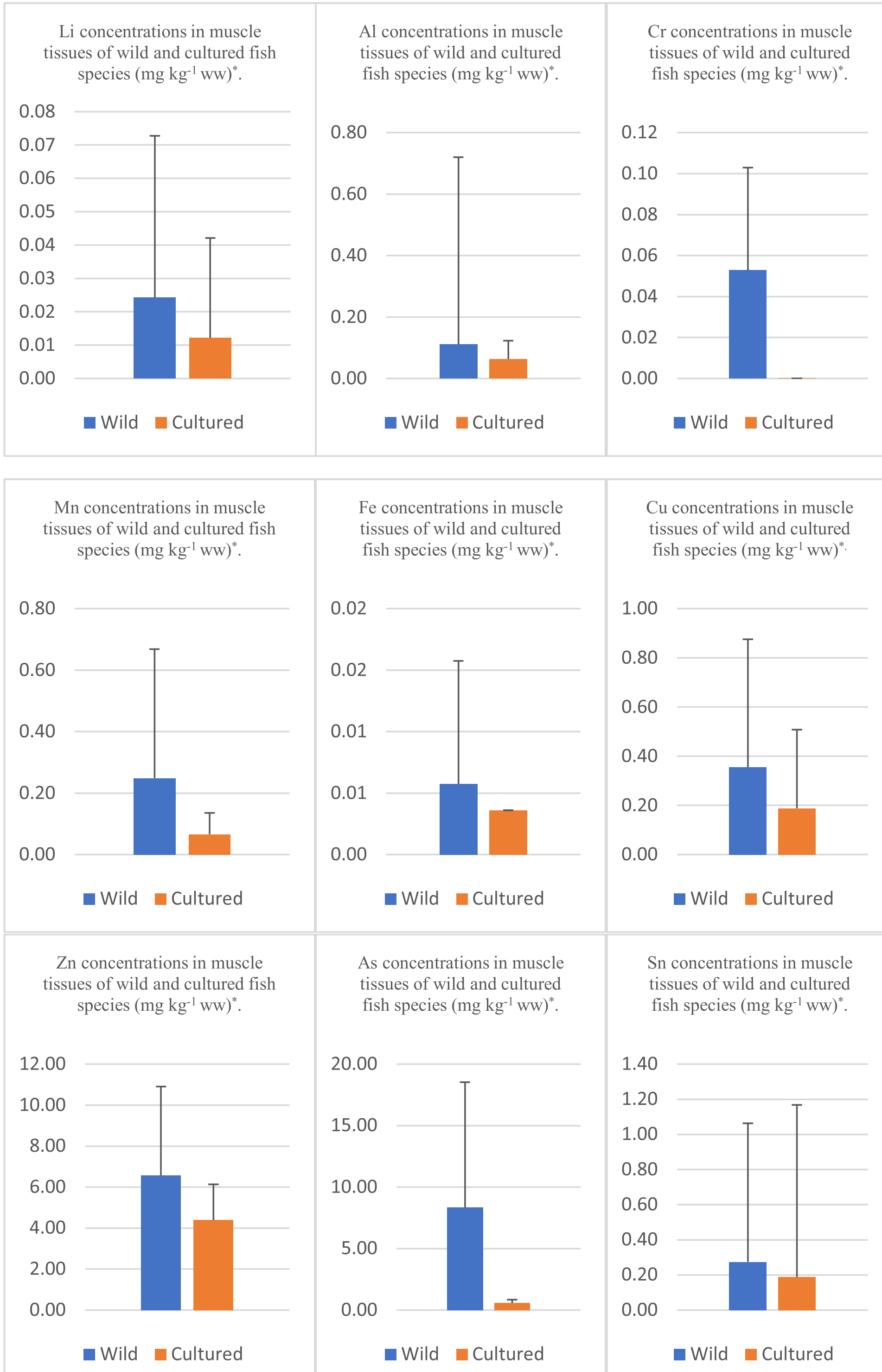

Fig. 2. Metal levels of farmed and wild fish species obtained from North-East Mediterranean (Mean Co, Ni, Cd and Pb levels were not given in Fig. 2, hence the levels of metals were found below detectible limits in all samples). *One sample t-test; Letters a and b show difference of given metal between farmed and wild fish species obtained from North-East Mediterranean at $p < 0.05$ level.

($p < 0.05$).

3.5. Iron

Iron is an essential element, which is a combination of hemoglobin, myoglobin, and a series of enzymes. Iron deficiency results in anemia, whereas the surplus amount of iron may cause various health problems such as cardiovascular and central nervous system disorders and renal and liver function disorders (Anderson, 1994). USEPA (2018) reported that oral RfD of iron is 700 μg kg^{-1} day^{-1}.

The mean iron concentrations in muscle tissues of fishes obtained from the Northeast Mediterranean Sea were ranged between < 0.001–0.02 mg kg^{-1} ww, and the highest level of iron was found in muscle tissues of *S. pilchardus* taken from Antalya and Karataş stations.

The iron levels in muscle tissues of wild fishes hunted from the Mediterranean Sea were reported to be 7.67–9.22 mg kg^{-1} ww (Percın et al., 2011), 0.82 – 27.35 mg kg^{-1} dw (Türkmen et al., 2005), 7.46 – 40.1 mg kg^{-1} dw (Türkmen et al., 2008), 0.001 – 19.02 mg kg^{-1} ww (Korkmaz, Ay, Ersoysal et al., 2019), 41.84 – 70.28 mg kg^{-1} ww (Yilmaz, 2003), 8.80 – 19.00 mg kg^{-1} dw (Minganti et al., 2010) and and 4.64 – 32.82 µg g^{-1} ww (Mutlu, 2021).

The iron concentration in muscle tissues of farmed fishes from the Mediterranean Sea was reported between 2.46 and 2.47 mg kg^{-1} ww (Marengo et al., 2018), 3.00 – 13.00 mg kg^{-1} ww (Lourenço et al., 2012), 5.2 – 15.2 mg kg^{-1} mg kg^{-1} dw (Minganti et al., 2010), 0 – 5.49 µg kg^{-1} ww (Kalantzi et al., 2016), and.64 – 3.30 mg kg^{-1} ww (Squadrone et al., 2016).

In the present study, the farmed fish were found to have 47% less iron accumulation in comparison to the wild species.

3.6. Cobalt

Cobalt plays a role as the metal component of Vitamin-B12, which is the only known biological function of it known as cyanocobalamin (Strachan, 2010). Oral RfD value of cobalt was specified to be 0.3 µg kg^{-1} day^{-1} (USEPA, 2018) and it was emphasized that the intakes, which exceed this level, might cause diseases such as allergic reactions, chronic bronchitis, alveolar and bronchial tumors, acute inflammation, alveolar epithelial hyperplasia, bronchial necrosis, and lung cancer (Kartal et al., 2004).

The concentration of cobalt in muscle tissues of wild fish hunted in the Mediterranean Sea was reported to be 0.03 – 5.61 mg kg^{-1} dw (Türkmen et al., 2005) and 0.04 – 0.41 mg kg^{-1} dw (Türkmen et al., 2008), whereas the concentration in muscle tissues of farmed fish from Mediterranean Sea was reported to be < 0.04 – 0.01 µg kg^{-1} ww (Kalantzi et al., 2016) and 0.01 mg kg^{-1} ww (Squadrone et al., 2016).

In the present study, it was determined that the cobalt concentrations were lower than the detectable limits in both wild and farmed fishes.

3.7. Nickel

USEPA (2018) reported the RfD value of nickel to be 20 µg kg^{-1} day^{-1} and stated that the intakes higher than this value may cause chronic nickel poisoning, cardiovascular and respiratory system disorders, and multiple organ failures (Denkhaus and Salnikow, 2002).

The nickel concentrations in the fishes obtained from the Mediterranean Sea were reported to range between 0.10 and 0.12 mg kg^{-1} ww (Percın et al., 2011), 0.11–12.88 mg kg^{-1} dw (Türkmen et al., 2005), 0.02–3.97 mg kg^{-1} dw (Türkmen et al., 2008), 0.94–1.22 µg g^{-1} ww (Yilmaz, 2003), 87.6–124 mg kg^{-1} dw (Iamiceli et al., 2015) and 0.01 – 0.05 µg g^{-1} ww (Kılıç et al., 2021).

The nickel concentrations in muscle tissues of farmed fishes from the Mediterranean Sea were reported to range between 0.07 and 0.09 mg kg^{-1} ww (Percın et al., 2011), 0.01–0.07 mg kg^{-1} ww (Lourenço et al., 2012), 0.04–0.05 mg kg^{-1} ww (Squadrone et al., 2016), 4.52 - s211.20 µg kg^{-1} dw (Rubio et al., 2011), and < 0.77–0.19 mg kg^{-1} ww (Kalantzi et al., 2016).

The nickel concentrations in the present study were found to be lower than the detectable limits.

3.8. Copper

Copper is an essential material for the utilization of iron to form hemoglobin. Copper deficiency causes deterioration in iron absorption, which develops anemia in humans. Copper is also a component of cytochrome oxidase enzyme and plays a role in the development of bones and elastic tissues (Nabrzyski, 2002). The oral RfD level of copper set by USEPA (2018) is 40 µg kg^{-1} day^{-1} and it was reported that an intake higher than this level might cause vascular diseases, liver, kidney, and central nervous system damages (Gaetke and Chow, 2003).

In this study, the mean copper concentrations in muscle tissues of fish species obtained from the Northeast Mediterranean Sea were found to range between < 0.0006 and 1.30 mg kg^{-1} ww. The highest copper concentration was detected in muscle tissues of *M. cephalus* taken from Karataş station.

The copper concentrations found in wild species hunted from the Mediterranean Sea were stated between 0.18 and 0.40 mg kg^{-1} ww (Lounas et al., 2021), 0.20 – 0.33 mg kg^{-1} ww (Marengo et al., 2018), 1.30–2.20 mg kg^{-1} dw (Minganti et al., 2010), 0.79–0.59 mg kg^{-1} ww (Percın et al., 2011) and 0.06 – 0.65 µg g^{-1} ww (Soliman et al., 2021).

The copper concentrations in the muscle tissues of farmed fishes from the Mediterranean Sea were reported to be 0.28 – 0.98 mg kg^{-1} ww (Lounas et al., 2021), 0.44 – 0.50 mg kg^{-1} ww (Marengo et al., 2018), 0.12 – 0.97 mg kg^{-1} ww (Lourenço et al., 2012), 0.21 – 1.03 mg kg^{-1} dw (Castritsi-Catharios et al., 2015), and 0.80 – 2.00 mg kg^{-1} dw (Minganti et al., 2010).

In the present study, the copper concentrations in farmed fish species were found to be 90% less than in the wild species.

3.9. Zinc

Zinc is a cofactor of more than 300 enzymes, which have important functions in RNA and DNA metabolism (Castillo-González et al., 2018). Zinc deficiency causes loss of appetite, weariness, diminished taste, delay in wound healing, growth and development retardation (Akdeniz et al., 2016). Oral RfD of zinc was reported to be 300 µg kg^{-1} day^{-1} and any intake exceeding this level might cause Alzheimer's disease, cancer, early aging, diabetes, depression, and major neurological diseases such as Wilson's disease (Chasapis et al., 2012; USEPA, 2018).

In the present study, the mean zinc concentrations in the muscle tissues of fish species obtained from the Northeast Mediterranean region were found to range between 1.76 and 15.38 mg kg^{-1} ww, while the highest zinc concentration was found in the muscle tissue of *B. boops* taken from Karataş station.

The zinc concentrations detected in wild species hunted from the Mediterranean Sea were reported to be 4.79–5.58 mg kg $^{-1}$ ww (Percın et al., 2011), 0.60 – 11.57 mg kg^{-1} dw (Türkmen et al., 2005), 3.82 – 55.05 mg kg^{-1} ww (Korkmaz, Ay, Ersoysal et al., 2019), and 19.55 – 38.23 mg kg $^{-1}$ ww (Yilmaz, 2003).

The zinc levels of farmed fishes were found between 4.10 and 9.40 mg kg^{-1} ww (Lourenço et al., 2012), 3.39 – 4.63 µg g^{-1} ww (Percın et al., 2011), < 0.50 – 7.2 mg kg^{-1} dw (Dalman et al., 2006), 3.61 – 5.56 mg kg dw (Yigit et al., 2019), and 2.63 – 4.08 mg kg^{-1} ww (Dugo et al., 2006).

In the present study, the zinc levels in farmed fishes were found to be 33% less than in wild fishes ($p < 0.05$).

3.10. Arsenic

Arsenic is found in nature with both organic and inorganic forms and is used as an additive for bactericides, herbicides, fungicides, animal feeds, corrosion inhibitors, veterinary medicines, and wood protectants (Kowalska et al., 2020). USEPA (2018) specified the RfD value arsenic to be 0.3 µg kg^{-1} day^{-1} and reported that any intake of arsenic higher than this level might cause kidney and liver damages, anemia, deterioration in digestion system functions, and various types of cancers (Korkmaz et al., 2017).

In this study, the mean arsenic concentrations in fish species were found to range between 0.52 and 35.10 mg kg^{-1} ww, while the highest concentration of arsenic was found in muscle tissues of *M. surmuletus* taken from Iskenderun Gulf.

The mean arsenic concentrations in muscle tissues of wild fishes hunted from the Mediterranean Sea were reported to be 1.08 – 15.06 mg kg^{-1} ww (Korkmaz, Ay, Ersoysal et al., 2019), 1.88 – 5.02 mg kg^{-1} ww (Lounas et al., 2021), 1.53 – 11.02 mg kg^{-1} ww (Copat et al., 2013), and 18.40 – 39.60 mg kg^{-1} ww (Minganti et al., 2010).

The mean arsenic concentrations in muscle tissues of farmed fishes in the Mediterranean Sea were reported to be 1.03 – 5.49 mg kg^{-1} ww (Marengo et al., 2018), 3.32–3.68 mg kg^{-1} ww (Lounas et al., 2021), 0.42 – 2.99 mg kg^{-1} ww (Kalantzi et al., 2016), 2.10 – 7.6 µg g^{-1} ww (Minganti et al., 2010), and 0.07 – 0.56 mg kg^{-1} ww (Squadrone et al., 2016).

In the present study, the arsenic concentrations found in muscle tissues of wild fishes were 14 times higher when compared to the farmed fishes ($p < 0.05$).

3.11. Cadmium

Oral RfD of cadmium specified by USEPA (2018) is 3 µg kg^{-1} day^{-1} and it was reported that the intakes higher than this level might cause kidney and liver damages, reproduction and circulatory system disorders, bone diseases, hypertension, and some kinds of tumors (Czeczot and Skrzycki, 2010; Satarug et al., 2017).

The cadmium concentrations in muscle tissues of wild fishes from Mediterranean Sea were reported to be < 0.07 – 9.30 µg kg^{-1} ww (Rubio et al., 2011), 0.001 – 0.002 mg kg^{-1} ww (Marengo et al., 2018), < 0.002 – 0.004 mg kg^{-1} ww (Lounas et al., 2021), 0.0004 – 0.0053 mg kg^{-1} ww (Copat et al., 2013), 0.01 – 4.16 mg kg^{-1} ww (Türkmen et al., 2005), 0.02–0.37 mg kg^{-1} (Türkmen et al., 2008), 0 – 0.81 mg kg^{-1} ww (Ramon et al., 2021), and 0.001 – 0.005 µg kg^{-1} ww (Mahjoub et al., 2021).

The cadmium concentrations found in farmed fishes from Mediterranean Sea were stated to be < 0.01 mg kg^{-1} ww (Squadrone et al., 2016), 0.002 – 0.004 mg kg^{-1} ww (Marengo et al., 2018), < 0.002 – 0.0078 mg kg^{-1} ww (Lounas et al., 2021), and 0.01 – 0.13 µg kg^{-1} ww (Dugo et al., 2006).

In this study, the cadmium concentrations were found to be lower than the detectable limits in all the wild and farmed fishes.

3.12. Tin

Tin is used as biocides and plastic stabilizers and conveyed to human body via toothpaste and canned foods (Rüdel, 2003). The intake of tin at high concentrations might cause stomach and bowel irritations, liver and kidney damage, anemia, diarrhea and, RfD of tin was reported at 600 µg kg^{-1} day^{-1} (Ikem and Egiebor, 2005; USEPA, 2018).

The tin concentrations in muscle tissues of wild fishes hunted from the Mediterranean Sea were reported to be 0.002 – 0.040 mg kg^{-1} ww (Marengo et al., 2018) and < 0.0004 – 0.92 mg kg^{-1} ww (Korkmaz, Ay, Ersoysal et al., 2019), while the concentrations in farmed fishes were reported to be 0.004 – 0.056 mg kg^{-1} ww (Marengo et al., 2018) and < 0.01 mg kg^{-1} ww (Squadrone et al., 2016).

The results achieved here were found to be similar to those reported in previous studies and the tin concentrations in farmed fishes were 33% less than in wild fishes.

3.13. Lead

USEPA (2018) specified the oral RfD of lead to be 4 µg kg^{-1} day^{-1} and reported that intakes higher than this level might cause various health problems including liver and kidney damage (Lee et al., 2011), mental retardation and death in humans (Ab Latif Wani and Usmani, 2015).

The lead concentrations in muscle tissues of fishes from the Mediterranean Sea were found to be 0.003 – 0.006 mg kg^{-1} ww (Marengo et al., 2018), 0.09 – 6.95 mg kg^{-1} ww (Türkmen et al., 2005), 0.02 – 0.37 mg kg^{-1} dw (Türkmen et al., 2008), < 0.0003 – 0.06 mg kg^{-1} ww (Korkmaz, Ay, Ersoysal et al., 2019), 1.03 – 7.45 mg kg^{-1} ww (Yilmaz, 2003), 0 – 0.18 mg kg^{-1} ww (Ramon et al., 2021) and 0.06 – 0.24 µg g^{-1} ww (Kılıç et al., 2021).

Moreover, the lead concentrations in farmed fishes in Mediterranean Sea were found to be 0.003 – 0.013 mg kg^{-1} ww (Marengo et al., 2018), < 0.02 – 0.40 mg kg^{-1} dw (Dalman et al., 2006), 0.08 – 0.35 mg kg^{-1} ww (Dugo et al., 2006), and 0.002 – 0.004 mg kg^{-1} ww (Lounas et al., 2021).

In the present study, the lead concentrations in all the wild and farmed fishes obtained from the Northeast Mediterranean Sea were lower than the detectable limits.

3.14. Risk analysis

Various organizations have made regulations for the presence of some metals in food products. The upper tolerable limits for some metals and the results obtained from this study are presented in Table 4 for making a comparison.

Table 4 shows that the mean metal concentrations detected in this present study are lower than the upper acceptable limits set by TFC, EU, and WHO. However, in recent studies carried out in the same region, Zn and Pb levels were reported not to comply with the limits set by TFC (Korkmaz et al., 2017; Korkmaz, Ay, Ersoysal et al., 2019). These two studies were conducted in the spring months, when the agricultural activities are intense and anthropogenic pollutants are highly conveyed to the coastal regions but the present study was carried out in the autumn season, which is arider. Future studies should investigate if the differences between the present and past results arise from the seasonal effects.

EWI's of metals were calculated for each stations and the data obtained were compared with the PTWI values of these metals (Table 5). Tolerable daily intake is an estimate of an additive, contaminant, or chemical in food products that can be taken daily over a lifetime without appreciable health risk to the consumers. The tolerable daily intake of a metal varies depending on the metal's concentration in the food, the amount of food product that is consumed and the body weight of consumer. In Turkey, according to the data published by RTMAF (2020), the daily and weekly mean fish consumptions per capita were reported to be 17 and 119 g, respectively. EWI's of metals were calculated by assuming that a person weighing 70 kg and consuming 119 g of fish weekly. Table 5 shows that the EWI's of metals in muscle tissues of fishes were much lower than the PTWI values of those fishes. However, the As concentrations in muscle tissues of fishes obtained from Iskenderun, Mersin, and Antalya stations were found to be very close to the PTWI values. Moreover, EWI's of farmed fishes were found much lower than the metal concentrations of wild species.

In recent years, the studies carried out on food safety adopted the calculation of THQ and CR values as a new approach. Hence, the THQ and CR values of the metals were calculated and the further risk analysis is presented below. THQ values of wild and farmed fishes obtained from Northeast Mediterranean Sea were found to be 1.73E-03 ± 3.47E-03, 8.66E-04 ± 2.14E-03 (Li); 1.57E-05 ± 8.72E-05, 9.02E-06 ± 3.81E-05 (Al); 2.54E-04 ± 4.25E-04, 6.70E-05 ± 2.05E-04 (Mn); 1.17E-06 ± 1.84E-06, 7.35E-07 ± 1.49E-06 (Fe); 1.27E-03 ± 1.87E-03, 6.72E-04 ± 1.14E-03 (Cu); 3.13E-03 ± 2.06E-03, 2.10E-03 ± 8.27E-04 (Zn); 3.97E+ 00 ± 4.84E+ 00, 2.80E-01 ± 1.18E-01 (As), and 6.52E-05 ± 1.88E-04, 5.29E-05 ± 2.33E-04 (Sn), respectively. TTHQ values were also found to be 3.98E+ 00 and 2.82E-01 for wild and farmed species. Analyzing the data, it can be seen that THQ values of metal concentrations for farmed fishes were lower than the wild species and the consumption of these fishes would not cause health problems. For the wild species, it was determined that THQ and TTHQ values of As were higher than 1 and they pose risk in terms of non-carcinogenic effects (Fig. 3). Since different values of Cr did not determine separately and Co, Ni, Cd, and Pb concentrations were found to be lower than the detectable limits in all the samples, THQ values were not calculated for these metals.

The element As is classified in the carcinogenic metal category by US-EPA. For this reason, among the metals examined, CR analysis could be conducted only for As. The mean CR values for As in the muscle tissues of wild and farmed fishes obtained from the Northeast

Table 4
Maximum acceptable levels of some heavy metals in fish muscle tissue according to national / international standards (mg kg^{-1} w.w.).

Organization/ Country	Metals												Reference	
	Li	Al	Cr	Mn	Fe	Co	Ni	Cu	Zn	As	Cd	Sn	Pb	
FAO (1983)	–	–	–	–	–	–	–	30	30	–	0.05	–	0.5	(Korkmaz et al., 2017)
WHO Limits	–	–	–	–	–	–	–	30	40	–	0.5	–	0.5	
European Commission	–	–	–	–	–	–	–	–	–	–	0.05	–	0.2	
England	–	–	–	–	–	–	–	20	50	–	0.2	–	2.0	
EU Limits	–	–	–	–	–	–	–	10	–	–	0.1	–	0.1	
TFC	–	–	–	20	–	–	–	20	50	–	0.1	–	1	
Present Study (Mean Interval)	BDL* - 0.15	BDL* - 2.54	BDL* _0.49	BDL* – 1.05	BDL * – 0.02	BDL*	BDL*	BDL* – 1.30	1.76–15.38	0.52 – 35.10	BDL*	BDL* – 2.04	BDL*	

- No data given
*Below detection limit ((The detection limits for Li, Al, Cr, Mn, Fe, Ni, Cu, Zn, As, Cd, Sn and Pb were as follows; 0.001, 0.0006, 0.001, 0.0002, 0.001, 0.0006, 0.0006, 0.002, 0.005, 0.0004, 0.0004 and 0.0003 mg kg^{-1}.)

Table 5
Comparison of EWI's of metals in muscles tissues of fishes with PTWI's of metals.

Metal	PTWI[a]	Samandağ (EWI[b])	İskenderun (EWI[b])	Adana (EWI[b])	Mersin (EWI[b])	Silifke (EWI[b])	Antalya (EWI[b])	Wild (EWI[b])	Farmed (EWI[b])
Li	980	1.19 – 10.71	< 0.001 – 17.85	< 0.001	< 0.001 – 5.95	< 0.001	< 0.001 – 15.47	2.38	1.19
Al	490000	< 0.0006 – 20.23	< 0.0006 – 302.26	< 0.0006	< 0.0006 – 153.51	< 0.0006	< 0.0006	13.09	7.14
Cr^{+3}[c]	735000	< 0.001	< 0.001	< 0.001 – 30.94	< 0.001 – 15.47	< 0.001 – 34.51	< 0.001 – 58.31	5.95	< 0.001 –
Cr^{+6}[c]	1470	< 0.001	< 0.001	< 0.001 – 30.94	< 0.001 – 15.47	< 0.001 – 34.51	< 0.001 – 58.31	5.95	< 0.001 –
Mn	68600	9.52 – 88.06	< 0.0002 – 109.48	< 0.0002 – 82.11	< 0.0002–103.53	< 0.0002 – 46.41	< 0.0002 – 124.95	29.75	8.33
Fe	343000	< 0.001 – 1.19	< 0.001 – 1.19	< 0.001 – 2.38	< 0.001 – 1.19	< 0.001 – 1.19	< 0.001 – 2.38	1.19	0.00
Co	196	< 0.0003	< 0.0003	< 0.0003	< 0.0003	< 0.0003	< 0.0003	< 0.0003	< 0.0003
Ni	9800	< 0.0006	< 0.0006	< 0.0006	< 0.0006	< 0.0006	< 0.0006	< 0.0006	< 0.0006
Cu	19600	< 0.0006 – 74.97	< 0.0006 – 153.51	33.32 – 154.70	< 0.0006 –72.59	< 0.0006 – 29.75	< 0.0006 – 79.73	42.84	22.61
Zn	147000	472.43 – 902.02	414.12 – 1053.15	756.84 – 1830.22	240.38 – 1508.92	209.44 – 585.48	389.13 – 1589.84	783.02	523.6
As[d]	147	6.75 – 46.69	2.87 – 146.19	5.50 – 52.52	3.46 – 128.20	2.17 – 80.97	2.54 – 141.69	34.82	2.46
Cd	490	< 0.0004	< 0.0004	< 0.0004	< 0.0004	< 0.0004	< 0.0004	< 0.0004	< 0.0004
Sn	294000	< 0.0004 – 73.78	< 0.0004 – 242.76	< 0.0004 – 232.05	< 0.0004 – 195.16	< 0.0004 – 148.75	< 0.0004 – 82.11	32.13	22.61
Pb	1960	< 0.0003	< 0.0003	< 0.0003	< 0.0003	< 0.0003	< 0.0003	< 0.0003	< 0.0003

[a] PTWI value for an adult man of 70 kg weight (µg/week/70 kg body weight).
[b] EWI, Mean amount of metal uptake by fish consumption (µg/week/70 kg body weight).
[c] Cr ions in muscle tissues were calculated assuming Cr is in the form of Cr+ 3 or Cr+ 6.
[d] As concentrations were calculated as total As and 3.5 conversion factor was used to convert them into inorganic arsenic (Korkmaz et al., 2017).

Mediterranean Sea were found to be 1.06E-03 ± 1.30E-03 and 7.49E-05 ± 3.15E-05, respectively. For food products, CR values between 1×10^{-6} and 1×10^{-4} are considered to be tolerable. Given the results achieved, it was determined that farmed species obtained from the Northeast Mediterranean Sea do not pose risk in terms of As concentrations but, even if at low incidence, wild species pose carcinogenic risks.

4. Conclusion

In aquatic environments the factors such as species, season, age, habitat, and diet play an important role in the metal accumulation. Minganti et al. (2010) reported that mercury and arsenic levels in *S. aurata,* farmed in the Mediterranean Sea were lower than in the wild species. Marengo et al. (2018) stated that *S. aurata*, farmed in the Mediterranean Sea do not pose risk in terms of consumption by humans but the wild species obtained from the same regions should not be consumed at high amounts. In another study, it was emphasized that the farmed species in the Mediterranean Sea have much higher metal accumulations in comparison to the wild species (Kalantzi et al., 2013). In this study, it was found that the mean Li, Al, Cr^{+3}, Cr^{+6}, Mn, Fe, Co, Ni, Cu, Zn, Cd, Sn, and Pb concentrations in muscle tissues of farmed species and THQ and CR values were much lower than the wild species. This is thought to be related to these facts: 1) The farmed species have shorter harvesting time (12–14 months), 2) They are younger than wild species, 3) They are fed on non-toxic feeds, 4) They are farmed in regions distant from industrial, agricultural, and domestic wastes.

CRediT authorship contribution statement

Cengiz Korkmaz: Conceptualization, Methodology, Software, Writing – original draft preparation, Visualization. **Gülsemin Şen Ağılkaya:** Writing – original draft preparation, Data curation, Methodology, Investigation. **Sahire Karaytuğ:** Data curation, Methodology, Visualization, Investigation. **Özcan Ay:** Data curation, Methodology, Supervision, Validation.

Declaration of Competing Interest

The authors declare that they have no known competing financial

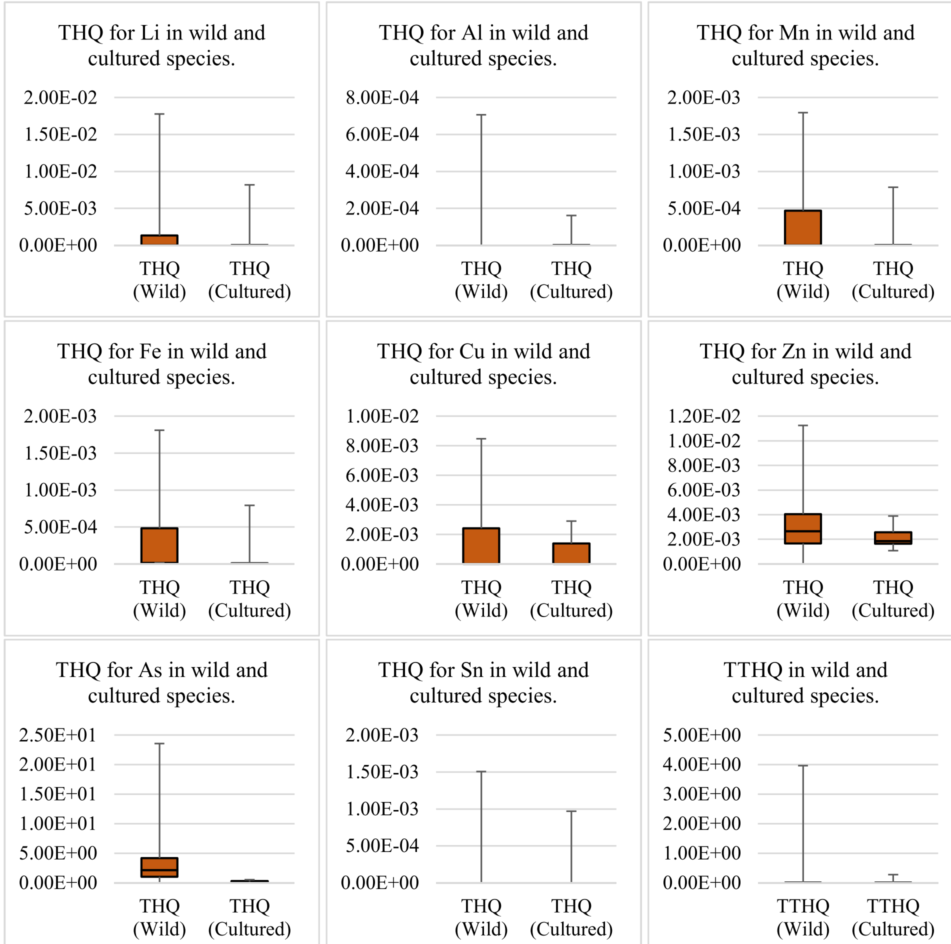

Fig. 3. THQ and TTHQ values of metal concentrations in muscle tissues of wild and farmed species.

interests or personal relationships that could have appeared to influence the work reported in this paper.

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