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Assessment of Accumulation and Potential Health Risk of Cr, Mn, Fe, Cu, and Zn in Fish from North-Eastern Mediterranean Sea

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ABSTRACT: Heavy metal accumulation in aquatic organisms has been an important issue due to environmental pollution resulting from anthropogenic activities. In this study, Cr, Mn, Fe, Cu, and Zn in the selected fish species (*Mullus barbatus, Solea solea, and Siganus rivulatus*) from three consecutive bays (İskenderun, Mersin, and Antalya from North-Eastern Mediterranean Sea) were considered to provide some information on heavy metal accumulation level and assessment of their health risk on both general and fishermen populations. There were some significant differences inter- and intra- species/ tissues/bays. The stability in heavy metal accumulation in fish tissues varied and the most stable tissue for Cr, Cu, Fe, Mn and Zn were determined as skin, muscle, liver, liver and, muscle, respectively. In general, the lowest heavy metal values were observed in the fish muscle. The Target Hazard Quotients (THQ) and Total Target Hazard Quotients (TTHQ) values based on muscle were not exceeded 1.00. Therefore, these results suggest that both general and fishermen populations are not subjected to the significant potential health risk from those bays.

Keywords: heavy metal, marine pollution, Turkey, consecutive bays

INTRODUCTION

Fisheries products are known as the most important nutritious foods with essential proteins- polyunsaturated fatty acids and liposoluble vitamins (Pal et al., 2018). But, in recent years, heavy metal accumulation in aquatic organisms has been a major issue due to environmental pollution resulting from anthropogenic activities (Yılmaz et al., 2017). For that reason, there has been a growing interest in "food safety" and keeping food quality within acceptable levels for human beings, worldwide. The accumulation rate and amount of heavy metals may vary depending on the fish species, quality of some environmental parameters such as salinity, temperature, pH, hardness, heavy metal concentration, exposure period, sex and size of fish, etc. (Jitar et al., 2015; Linbo et al., 2009; Yılmaz et al., 2010). Therefore, metal accumulation ratios in fish tissues may show fluctuations in different locations, even for the same fish species (Yılmaz, 2003).

Fish are located at the upper part of the aquatic food chain and their normal

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metabolism may tend to accumulate large amounts of certain metals from water, sediment or feed (Authman et al., 2015). Although these metals are required in small amounts for various biochemical and enzymatic reactions, high accumulation values have been shown to have toxic and carcinogenic effects that may cause circulatory system, hepatic, renal, cardiovascular and neurological disorders in humans (Barone et al., 2018; Eisler, 2000; Korkmaz et al., 2017).

The Target Hazard Quotients (THQ) and Total Target Hazard Quotients (TTHQ) values have been used for assessing potential health risks of individual and total effects of heavy metals, respectively. However, TTHQ has been expected much more reliably helpful to assess and compare their combined risks and therefore have been widely employed in recent literature (Copat et al., 2013; Korkmaz et al., 2017; Rajan & Ishak, 2017; Yi et al., 2011). Pollution has been gained attention in the Mediterranean Sea due to intense urbanization near the coastal area, heavy industrial activities, marine traffic issues, and ship accidents (Bosch et al., 2016). In this study, therefore, some selected heavy metals (Cr, Mn, Fe, Cu, and Zn) and fish species [Red Mullet (*Mullus barbatus*), Common Sole (*Solea solea*), and Marbled spinefoot (*Siganus rivulatus*)] from three bays in the North-Eastern Mediterranean (İskenderun, Mersin, and Antalya) were considered to investigate the following issues; (i) to determine the accumulation rates of these metals in the different tissues (muscle, liver, skin, and intestine) of fish, (ii) to compare and to find out any rank pattern of metal accumulation both inter- and intra-species/ tissues/bays, (iii) to find out and to rank metal accumulation stability in different tissues, (iv) to evaluate Target Hazard Quotients (THQ) and Total Target Hazard Quotients (TTHQ) values (both general and fisherman population) of these heavy metals accumulation in muscle tissue as the edible part of fish.

MATERIAL AND METHODS

Fish samples (n=15 specimens for each species) were taken from fishermen in İskenderun, Mersin, and Antalya Bays in April 2016 (Figure 1). Fish were procured as dead and brought to the laboratory on ice immediately and then frozen at -25°C until dissection.
Before dissection, total fish length and weight were measured to the nearest millimeter and gram, respectively. The mean length and weight of *Solea solea*, *Mullus barbatus*, and *Siganus rivulatus* were 25.22± 2.20 cm and 135.90 ± 43.11 g, 19.64 ± 4.07 cm and 158.31 ± 123.61 g and 17.29 ± 2.08 cm and 108.34 ± 72.52 g, respectively. The mean body lengths of each species from three bays were not significantly different ($p>0.05$).

Studied tissue from each fish was transferred to a petri dish after being wet weighed and were added 2 mL nitric acid (HNO_3, % 65, S.W.: 1.40, Merck) and 1 mL perchloric acid ($HClO_4$, % 60, S.W.: 1.53, Merck) mixture in experimental tube and were wet digested on a hotplate at 120 °C for 8 h. They were then transferred to polyethylene tubes and their volumes were made up to 10 mL using deionized water. Samples were passed through a 0.45-μm membrane filter before analysis. All analyses were carried out in triplicate by using Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) (Perkin Elmer Nexion 350 X). The quality of data was verified by the analysis of standard reference material DORM-2 (National Research Council of Canada; dogfish muscle and liver MA-A-2/TM Fish Flesh). The recovery values for Cr, Cu, Fe, Mn, and Zn were measured as 99.98, 91.94, 97.35, 89.83 and 96.59%, respectively. Analytical blanks were run in the same way as the samples, and concentrations were determined using standard solutions prepared in the same acid matrix. Standards for the instrument calibration were prepared based on mono element certified reference solution ICP Standard (Merck). Metal concentrations were calculated in micrograms per gram wet weight (µg metal g^{-1} w.wt.).

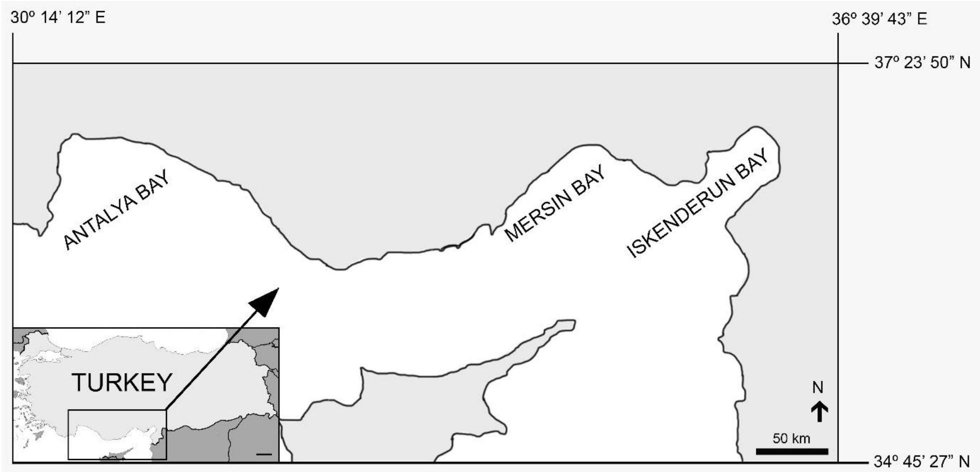

Fig. 1. Map of Studied Areas (İskenderun, Mersin, and Antalya Bay) from North Eastern Mediterranean Sea

To evaluate the potential health risk of *Solea solea*, *Mullus barbatus*, and *Siganus rivulatus* consumption was calculated depending on Target Hazard Quotients (THQ) which is an indication of heavy metal exposure risk. The THQ calculation formula is given below (Chien et al., 2002; Han et al., 1998; Storelli, 2008).

$$THQ = [(E_F x E_D x F_{IR} x C)/(R_{FD} x W_{AB} x T_A)] x 10^{-3}$$

where, E_F is the exposure frequency: 365 days/year, E_D is the exposure duration: the average lifetime is assumed as 70 years according to (Bennett et al., 1999). F_{IR} is the food ingestion rate: 15.06 g/day for Turkish consumers, according to (TUIK, 2018). C is the determined metal concentration in muscle tissue (mg/kg).

R_{FD} is the oral reference dose (mg/kg/day): Cr, Mn, Fe, Cu, and Zn have been suggested as 1.5, 0.14, 0.8, 0.5 and 1 respectively (US EPA, 2009).

W_{AB} is the average body weight: 70 kg, according to (Kumar et al., 2013). T_A is the average exposure time for non-carcinogens (365 days/year x E_D, assuming 70 years in this study). In this study, the total THQ (TTHQ) is treated as the arithmetic sum of the individual metal THQ values (Yi et al. 2011) :

TTHQ= THQ (toxicant 1) + THQ (toxicant 2)+ ……. + THQ (toxicant n)

THQ and TTHQ values were estimated for the general population (THQ_{gp}) and fishermen (THQ_f) separately to compare the risk of heavy metals from different consumers. In this study, F_{IR} was assumed for Turkish fishermen to be two times higher than the general population as 30.12 g/day. The THQ and TTHQ $\geq$ 1.0 refers to people may experience significant health risk from the intake of individual metals through fish consumption (Yi et al., 2011).

All data were checked for outliers and then descriptive statistics and box-whisker plots were calculated and drawn, respectively. Both inter- and intra-species/ tissues/bays differences were assessed using by one-way PERMANOVA (Permutational multivariate analysis of variance) test. The coefficient of variation (Cv, %) was used to evaluate metal accumulation stability for different tissues. All computations and statistical analyses were carried out using Microsoft Excel and Past software (V. 3.23) (Hammer et al., 2001).

RESULTS AND DISCUSSION

The mean values with standard deviation (x̄±sd) and coefficient of variation (Cv, %) of measured heavy metals (Cr, Cu, Fe, Mn, and Zn) in the tissues (muscle, intestine, skin, and liver) of *Solea solea, Mullus barbatus, Siganus rivulatus* by studied locations are given in Table 1-5 and Table 6, respectively. Also, Box and Whisker plots with mean and standard deviations of heavy metal concentration in muscle tissues of *Solea solea*, *Mullus barbatus*, and *Siganus rivulatus* are given in Figure 2. The estimated Target Hazard Quotients (THQ) and Total Target Hazard Quotients (TTHQ) were given in Table 7.

Mean Cr concentrations in the tissues of *S. solea* was ordered for İskenderun, Mersin and Antalya Bays as skin (S)> intestine (I)> liver (L)>muscle (M), M>I>S > L and M = S > L > I, respectively. Mean Cr concentration in the *M. barbatus* for İskenderun, Mersin and Antalya Bays ranked as I = L > S > M, M > S > I > L and M > I > S > L, respectively. Cr concentration ranking of *S. rivulatus* for İskenderun Bay was ordered as I > S > L > M; on the other hand, the ranking was same for both Mersin and Antalya Bays as M > I > S > L (Table 1).

One way PERMANOVA results showed that there were significant differences between the tissues of *M. barbatus* and *S. rivulatus* in all locations ($p<0.05$), except *S. solea* ($p>0.05$) (Table 1). When Cr concentration difference among studied species at the same locations was examined, it was found that no significant differences were depending on species ($p>0.05$). The Cr levels in the muscle of these species from Antalya and Mersin Bay were found to be higher than those of other tissues (Table 1, Fig.2a). Considering all species, the Cr accumulation stability was found based on mean Cv % values in descending order as: S> L> I> M (Table 6).

Table 1. Cr concentration (µg metal/g wet weight [w.wt.]) in the tissues of *Solea solea*, *Mullus barbatus*, and *Siganus rivulatus* with respect to studied locations

Heavy Metal	Species	Station		Tissue			
				Muscle	Intestine	Skin	Liver
Cr	*S. solea*	İskenderun	x̄	$0.09^{A}_{a}x$	$0.14^{A}_{a}x$	$0.15^{A}_{a}x$	$0.12^{A}_{a}x$
			±s	0.06	0.08	0.05	0.04
		Mersin	x̄	$0.22^{A}_{a}x$	$0.19a^{A}_{a}x$	$0.13^{A}_{a}x$	$0.11^{A}_{a}x$
			±s	0.19	0.12	0.03	0.04
		Antalya	x̄	$0.14^{A}_{a}x$	$0.08^{A}_{a}x$	$0.14^{A}_{a}x$	$0.11^{A}_{a}x$
			±s	0.13	0.05	0.07	0.06
	M. barbatus	İskenderun	x̄ x̄	$0.07^{A}_{a}x$	$0.18^{A}_{a}xy$	$0.11^{A}_{a}x$	$0.18^{A}_{ab}x$
			±s	0.02	0.11	0.03	0.12
		Mersin	x̄	$0.48^{A}_{a}y$	$0.11^{B}_{a}x$	$0.16^{B}_{a}x$	$0.10^{B}_{a}x$
			±s	0.38	0.03	0.09	0.04
		Antalya	x̄	$0.45^{A}_{a}y$	$0.30^{AB}_{b}y$	$0.18^{B}_{a}x$	$0.11^{AC}_{a}x$
			±s	0.14	0.14	0.07	0.01
	S. rivulatus	İskenderun	x̄	$0.06^{A}_{a}x$	$0.19^{B}_{a}x$	$0.18^{B}_{a}x$	$0.08^{BC}_{b}x$
			±s	0.01	0.12	0.07	0.01
		Mersin	x̄	$0.31^{A}_{a}y$	$0.19^{A}_{a}x$	$0.14^{A}_{a}x$	$0.11^{A}_{a}x$
			±s	0.27	0.11	0.05	0.04
		Antalya	x̄	$0.20^{AB}_{a}y$	$0.16^{B}_{ab}x$	$0.13^{AB}_{a}x$	0.09^{A}_{ax}
			±s	0.15	0.07	0.05	0.03

x̄ Mean values; ±s Standard Deviation
A, B, C denotes differences among tissues in the same species at the same location
a, b, c denotes differences among same tissues of different species at same locations
x, y, z denotes differences among locations in the same tissues of same species

Table 2. Cu concentration (µg metal/g wet weight [w.wt.]) in the tissues of Solea solea, Mullus barbatus, and Siganus rivulatus with respect to studied locations

Heavy Metal	Species	Station		Tissue			
				Muscle	**Intestine**	**Skin**	**Liver**
Cu	*S. solea*	İskenderun	x̄	$0.24_{a}^{A}x$	$1.05_{a}^{B}xy$	$0.39_{a}^{AB}x$	$17.22_{a}^{AB}xy$
			±s	0.03	0.37	0.20	4.96
		Mersin	x̄	$0.31_{a}^{A}y$	$1.00_{a}^{B}x$	$0.31_{a}^{AC}x$	$12.89_{a}^{D}x$
			±s	0.02	0.50	0.19	0.46
		Antalya	x̄	$0.57_{a}^{A}y$	$0.46_{a}^{AB}y$	$3.04_{a}^{C}y$	$42.13_{a}^{D}y$
			±s	0.11	0.16	1.61	26.73
	M. barbatus	İskenderun	x̄ x̄	$0.34_{a}^{A}x$	$2.73_{b}^{B}x$	$0.41_{a}^{A}x$	$1.15_{a}^{B}x$
			±s	0.16	1.83	0.20	0.73
		Mersin	x̄	$0.39_{a}^{A}x$	$0.83_{a}^{AB}y$	$0.66_{a}^{AB}y$	$1.15_{b}^{B}x$
			±s	0.14	0.35	0.42	0.68
		Antalya	x̄	$0.38_{ab}^{A}x$	$1.16_{a}^{B}xy$	$1.50_{ac}^{B}y$	$1.51_{ab}^{AB}x$
			±s	0.19	0.56	0.58	0.04
	S. rivulatus	İskenderun	x̄	$0.23_{a}^{A}x$	$2.73_{b}^{B}x$	$0.48_{a}^{C}x$	$5.17_{a}^{BD}x$
			±s	0.05	1.49	0.16	2.11
		Mersin	x̄	$0.38_{a}^{A}x$	$2.77_{a}^{B}x$	$0.29_{a}^{A}x$	$12.93_{a}^{AC}x$
			±s	0.13	1.59	0.21	8.94
		Antalya	x̄	$0.29_{b}^{A}x$	$2.32_{a}^{B}x$	$0.37_{b}^{A}x$	$5.45_{b}^{C}x$
			±s	0.06	1.44	0.21	1.50

x̄ Mean values; ±s Standard Deviation
A, B, C denotes differences among tissues in the same species at the same location
a, b, c denotes differences among same tissues of different species at same locations
x, y, z denotes differences among locations in the same tissues of same species

Table 3. Fe concentration (µg metal/g wet weight [w.wt.]) in the tissues of *Solea solea*, *Mullus barbatus*, and *Siganus rivulatus* with respect to studied locations

Heavy Metal	Species	Station		Tissue			
				Muscle	**Intestine**	**Skin**	**Liver**
Fe	*S. solea*	İskenderun	x̄	$10.58_{a}^{A}x$	$67.30_{a}^{ABC}x$	$45.85_{a}^{AC}x$	$93.06_{a}^{B}x$
			±s	1.87	30.30	9.60	12.24
		Mersin	x̄	$8.04_{a}^{A}x$	$49.26_{a}^{B}xy$	$15.56_{a}^{C}y$	$67.54_{ab}^{D}xy$
			±s	0.99	10.46	4.97	11.93
		Antalya	x̄	$10.68_{a}^{A}x$	$20.77_{a}^{C}y$	$9.20_{a}^{AB}z$	$182.73_{a}^{D}y$
			±s	2.05	6.81	0.28	68.50
	M. barbatus	İskenderun	x̄	$11.73_{a}^{A}x$	$66.84_{a}^{BC}x$	$34.14_{a}^{B}x$	$65.77_{a}^{C}x$
			±s	2.03	43.58	9.98	0.45
		Mersin	x̄	$9.45_{a}^{A}x$	31.13_{a}^{B}y	18.03_{a}^{C}x	$26.28_{b}^{BC}x$
			±s	3.69	7.42	3.03	8.51
		Antalya	x̄	$11.58_{ab}^{A}x$	$219.58_{a}^{B}xy$	39.62_{b}^{A}x	$108.40_{ab}^{C}x$
			±s	6.72	101.38	11.97	16.18
	S. rivulatus	İskenderun	x̄	9.01_{a}^{A}x	$65.09_{a}^{B}x$	$38.09_{a}^{C}x$	$110.07_{a}^{D}x$
			±s	1.49	16.22	17.39	33.60
		Mersin	x̄	$9.33_{a}^{A}x$	$20.24_{a}^{A}y$	$13.28_{a}^{A}y$	$104.77_{a}^{B}x$
			±s	7.15	13.30	6.43	38.72
		Antalya	x̄	$5.47_{b}^{A}y$	$92.99_{a}^{B}x$	$11.73_{a}^{C}y$	$62.89_{b}^{BD}y$
			±s	2.00	22.88	2.62	28.24

x̄ Mean values; ±s Standard Deviation
A, B, C denotes differences among tissues in the same species at the same location
a, b, c denotes differences among same tissues of different species at same locations
x, y, z denotes differences among locations in the same tissues of same species

In 2004, a ship named "ULLA" sank in the İskenderun Bay which was claimed to be loaded with toxic Cr. After that time, wrong perception occurred in public that there was a risk about Cr level in aquatic organisms (Yılmaz et al., 2017). Contrary to expectations, Cr accumulations in all muscles of three species from İskenderun Bay were found to be the lowest among the three bays.

Mean Cr concentrations we observed in *S. solea* are between those reported in other previous studies carried out in the three bays. For *M. barbatus* and *S. rivulatus*, Cr values obtained in this study were found to be much lower than previous studies (Table 1, Table 8). According to these results, it could be suggested that there is no dense Cr pollution that might have resulted from the ULLA incident in 2004.

While Cu contents in the tissues of *S. solea* in İskenderun, Mersin and Antalya Bays were ranked as L > I > S > M, L > I > M > S and L > S > M > I, also ranking of *M. barbatus* in Mersin, Antalya and İskenderun Bays were L > I > S > M and I > L > S > M, respectively. For *S. rivulatus*, Cu concentration order was similar in İskenderun and Antalya Bays and ranked as L > I > S > M. The same order was applicable for Mersin as L > I > M > S (Table 2). When statistical differences in muscle tissues among the species were examined (Fig 2b, Table 2), only *M. barbatus* and *S. rivulatus* were found to be different in Antalya Bay ($p<0.05$, denoted as a,b).

Table 4. Mn concentration (µg metal/g wet weight [w.wt.]) in the tissues of *Solea solea*, *Mullus barbatus*, and *Siganus rivulatus* with respect to studied locations

Heavy Metal	Species	Station		Tissue			
				Muscle	Intestine	Skin	Liver
Mn	*S. solea*	İskenderun	x̄	$0.29^{A}_{ab}\,x$	$1.02^{AB}_{a}\,x$	$1.29^{B}_{a}\,x$	$1.08^{AB}_{ab}\,x$
			±s	0.25	0.17	0.54	0.59
		Mersin	x̄	$0.31^{A}_{a}\,x$	$1.21^{AB}_{a}\,x$	$0.97^{B}_{a}\,x$	$0.77^{AB}_{a}\,x$
			±s	0.12	0.60	0.28	0.44
		Antalya	x̄	$0.31^{A}_{a}\,x$	$0.70^{A}_{a}\,x$	$1.71^{A}_{a}\,x$	$0.96^{A}_{a}\,x$
			±s	0.17	0.42	1.49	0.68
	M. barbatus	İskenderun	x̄	$0.52^{A}_{a}\,x$	$2.46^{B}_{a}\,xy$	$0.78^{AB}_{a}\,xy$	$4.20^{B}_{a}\,x$
			±s	0.29	1.38	0.39	3.57
		Mersin	x̄	$0.25^{A}_{a}\,y$	$1.57^{B}_{a}\,x$	$0.34^{AD}_{b}\,x$	$1.13^{C}_{a}\,x$
			±s	0.08	0.82	0.23	0.40
		Antalya	x̄	$0.20^{A}_{a}\,y$	$7.82^{B}_{a}\,y$	$0.94^{C}_{a}\,y$	$0.81^{ABC}_{a}\,x$
			±s	0.12	3.75	0.54	0.24
	S. rivulatus	İskenderun	x̄	$0.28^{A}_{b}\,x$	$2.86^{B}_{a}\,x$	$2.83^{B}_{b}\,x$	$0.80^{C}_{b}\,x$
			±s	0.07	1.78	0.87	0.17
		Mersin	x̄	$0.13^{A}_{a}\,y$	$0.83^{A}_{a}\,x$	$1.74^{B}_{c}\,y$	$0.76^{C}_{a}\,xy$
			±s	0.04	0.32	0.50	0.30
		Antalya	x̄	$0.11^{A}_{a}\,y$	$0.89^{C}_{a}\,x$	$1.04^{C}_{a}\,y$	$0.48^{B}_{a}\,y$
			±s	0.03	0.42	0.32	0.15

x̄ Mean values; ±s Standard Deviation
A, B, C denotes differences among tissues in the same species at the same location
a, b, c denotes differences among same tissues of different species at same locations
x, y, z denotes differences among locations in the same tissues of same species

Table 5. Zn (µg metal/g wet weight [w.wt.]) in the tissues of *Solea solea*, *Mullus barbatus*, and *Siganus rivulatus* with respect to studied locations

Heavy Metal	Species	Station		Tissue			
				Muscle	Intestine	Skin	Liver
Zn	*S. solea*	İskenderun	x̄	$3.11_{a}^{A}x$	$7.90_{a}^{B}x$	$9.14_{a}^{BC}x$	$12.94_{a}^{C}x$
			±s	0.51	2.31	2.04	2.52
		Mersin	x̄	$3.31_{a}^{A}x$	$9.83_{a}^{B}x$	$8.25_{a}^{B}x$	$12.50_{a}^{A}x$
			±s	0.39	2.79	2.11	2.90
		Antalya	x̄	$3.22_{a}^{A}x$	$36.97_{a}^{B}y$	$5.37_{a}^{A}x$	$9.00_{a}^{A}x$
			±s	1.01	7.37	0.73	1.99
	M. barbatus	İskenderun	x̄	$2.81_{a}^{A}x$	$14.61_{b}^{B}x$	$28.43_{b}^{B}x$	$20.40_{ab}^{B}x$
			±s	1.16	3.40	19.12	12.37
		Mersin	x̄	$2.84_{a}^{A}x$	$11.72_{a}^{B}x$	$5.67_{a}^{C}y$	$10.79_{a}^{B}x$
			±s	0.64	5.03	2.30	2.29
		Antalya	x̄	$2.89_{a}^{A}x$	$11.56_{a}^{B}x$	$19.88_{a}^{B}x$	$14.91_{ab}^{B}x$
			±s	0.99	3.06	4.75	1.95
	S. rivulatus	İskenderun	x̄	$3.51_{a}^{A}x$	$12.14_{b}^{B}x$	$51.43_{c}^{C}x$	$68.63_{b}^{B}x$
			±s	0.94	3.77	7.39	9.97
		Mersin	x̄	$4.15_{a}^{A}x$	$23.46_{b}^{B}x$	$33.00_{b}^{B}y$	$51.12_{b}^{B}xy$
			±s	1.49	15.06	14.62	29.63
		Antalya	x̄	$3.34_{a}^{A}x$	$14.15_{a}^{B}x$	$35.70_{b}^{C}y$	$21.76_{b}^{B}y$
			±s	0.65	5.96	8.80	7.21

x̄ Mean values; ±s Standard Deviation
A, B, C denotes differences among tissues in the same species at the same location
a, b, c denotes differences among same tissues of different species at same locations
x, y, z denotes differences among locations in the same tissues of same species

Table 6. Coefficient of variation (Cv, %) of heavy metals in the tissues of *Solea solea, Mullus barbatus, Siganus rivulatus* with respect to studied locations

Metal	Species	Station	Muscle	Intestine	Skin	Liver
Cr	*S. solea*	İskenderun	59.95	56.51	32.28	33.23
		Mersin	87.86	65.40	20.56	33.63
		Antalya	88.36	66.21	49.58	54.66
	M. barbatus	İskenderun	23.82	65.53	28.63	68.12
		Mersin	79.30	29.31	57.92	44.32
		Antalya	30.16	46.39	37.42	9.12
	S. rivulatus	İskenderun	12.59	64.22	35.49	16.33
		Mersin	84.62	56.60	36.89	37.13
		Antalya	77.66	41.08	37.44	28.26
Cu	*S. solea*	İskenderun	11.44	35.40	52.89	28.79
		Mersin	7.11	50.00	62.46	3.55
		Antalya	18.69	35.14	52.85	63.45
	M. barbatus	İskenderun	47.60	67.10	47.82	63.65
		Mersin	36.81	41.89	64.15	58.93
		Antalya	48.84	48.52	38.85	2.72
	S. rivulatus	İskenderun	20.12	54.59	33.10	40.89
		Mersin	34.98	57.31	71.78	69.12
		Antalya	19.26	62.05	56.11	27.48
Fe	*S. solea*	İskenderun	17.69	45.03	20.94	13.16
		Mersin	12.29	21.24	31.94	17.67
		Antalya	17.33	65.20	29.25	0.69
	M. barbatus	İskenderun	39.09	23.83	16.80	32.40
		Mersin	58.03	46.17	30.22	14.93
		Antalya	58.03	46.17	30.22	14.93
	S. rivulatus	İskenderun	16.56	24.91	45.66	30.52
		Mersin	76.67	65.69	48.41	36.96
		Antalya	36.65	24.60	22.30	44.91
Mn	*S. solea*	İskenderun	83.88	16.66	41.49	54.91
		Mersin	54.61	56.08	49.38	84.93
		Antalya	32.06	51.92	67.53	35.50

Table 6. Coefficient of variation (Cv, %) of heavy metals in the tissues of *Solea solea, Mullus barbatus, Siganus rivulatus* with respect to studied locations

Metal	Species	Station	Muscle	Intestine	Skin	Liver
Mn	*M. barbatus*	İskenderun	57.37	48.02	57.96	29.73
		Mersin	25.53	62.36	30.70	21.07
		Antalya	31.13	38.91	28.55	39.93
	S. rivulatus	İskenderun	25.53	62.36	30.70	21.07
		Mersin	31.13	38.91	28.55	39.93
		Antalya	22.66	47.07	30.45	30.29
Zn	*S. solea*	İskenderun	16.36	29.22	22.30	19.48
		Mersin	11.80	28.44	25.57	23.20
		Antalya	31.36	19.93	13.58	22.06
	M. barbatus	İskenderun	41.29	23.25	67.26	60.63
		Mersin	22.32	42.96	40.58	21.20
		Antalya	34.42	26.43	23.87	13.09
	S rivulatus	İskenderun	26.88	31.09	14.37	14.52
		Mersin	35.96	64.19	44.31	57.95
		Antalya	19.40	42.13	24.65	33.13

Table 7. THQ values in the tissues of *Solea solea*, *Mullus barbatus,* and *Siganus rivulatus* with respect to studied locations

Station	Heavy Metal		*S. solea*		*M. barbatus*		*S. rivulatus*	
			THQ_{gp}	THQ_f	THQ_{gp}	THQ_f	THQ_{gp}	THQ_f
İskenderun		Mean	0	0	0	0	0	0
	Cr	Min	0		0	0	0	0
		Max	0		0	0	0.0001	0.0002
		Mean	0.0013	0.0025	0.0018	0.0032	0.0012	0.006
	Cu	Min	0.0011	0.0022	0.0011	0.0022	0.0010	0.0020
		Max	0.0014	0.0028	0.0034	0.0068	0.0016	0.0032
		Mean	0.0031	0.062	0.0035	0.007	0.0027	0.0054
	Fe	Min	0.0025	0.0050	0.0028	0.0056	0.0024	0.0048
		Max	0.0036	0.0072	0.0041	0.0082	0.0032	0.0064
		Mean	0.0043	0.0086	0.0077	0.0154	0.0041	0.0082
	Mn	Min	0.0021	0.0042	0.0047	0.0094	0.0031	0.0062
		Max	0.0098	0.0130	0.0151	0.0302	0.0055	0.011
		Mean	0.0113	0.0226	0.0285	0.057	0.0185	0.037
	Zn	Min	0.0018	0.0036	0.0020	0.0040	0.0015	0.0030
		Max	0.0026	0.0052	0.0046	0.0092	0.0026	0.0052
	Total*	Mean	0.02	0.04	0.0415	0.083	0.0265	0.053
Mersin		Min	0	0	0.0001	0.0001	0	0
	Cr	Max	0	0	0	0	0	0
		Mean	0	0	0.0001	0.002	0.0001	0.002
		Min	0.0016	0.0035	0.0020	0.0040	0.0020	0,0039
	Cu	Max	0.0014	0.0028	0.0012	0.0024	0.0012	0.0024
		Mean	0.0017	0.0034	0.0031	0.0062	0.0024	0.0048
		Min	0.0024	0.0048	0.0028	0.0056	0.0028	0.0056
	Fe	Max	0.0022	0.0044	0.0018	0.0036	0.0009	0.0018
		Mean	0.0028	0.0056	0.0041	0.0082	0.0057	0.0104
		Min	0.0045	0.0090	0.0037	0.0074	0.0019	0.0038
	Mn	Max	0.0025	0.0050	0.0025	0.0050	0.0015	0.0030
		Mean	0.0064	0.0128	0.0056	0.0102	0.0027	0.0054
		Min	0.0081	0.0162	0.0154	0.0308	0.0248	0.496
	Zn	Max	0.0020	0.0040	0.0020	0.0040	0.0015	0.0030
		Mean	0.0025	0.0050	0.0046	0.0092	0.0026	0.0052
	Total*	Min	0.0166	0.0332	0.024	0.048	0.0315	0.063
Antalya		Max	0	0	0.0001	0.0001	0	0
	Cr	Mean	0	0	0	0	0	0
		Min	0	0	0.0001	0.0002	0.0001	0.0002
		Max	0.0030	0.0024	0.002	0.0039	0.0015	0.0030
	Cu	Mean	0.0023	0.0046	0.001	0.002	0.0011	0.0022
		Min	0.0034	0.0068	0.0031	0.0062	0.0018	0.0036
		Max	0.0032	0.0064	0.0034	0.0068	0.0016	0.0032
	Fe	Mean	0.0025	0.0050	0.0017	0.0034	0.0009	0.0018
		Min	0.0037	0.0074	0.0056	0.0102	0.0023	0.0046
		Max	0.0046	0.0091	0.0030	0.006	0.0017	0.0034
	Mn	Mean	0.0021	0.0042	0.0013	0.0026	0.0012	0.0024
		Min	0.0071	0.0142	0.0052	0.0104	0.0021	0.0042
		Max	0.0216	0.0432	0.0237	0.0474	0.0134	0.0268
	Zn	Mean	0.0014	0.0028	0.0019	0.0038	0.0011	0.0022
		Min	0.0028	0.0056	0.0028	0.0056	0.0027	0.0054
	Total*	Max	0.0324	0.0648	0.0322	0.0644	0.0182	0.0364

*Total THQ was estimated based on average values.

Table 8. Heavy metal accumulation (µg/g w.wt.) and THQ values of muscle tissue of fish from previous studies evaluated in Iskenderun, Mersin and Antalya Bays

Species	Metal	Bay	w.wt*	THQ	Reference
M. barbatus	Cr	İskenderun	0.482	<0.0001	(Bıçkıcı, 2010)
M. barbatus	Cr	İskenderun	0.096	<0.0001	(Dural et al., 2010)
M. barbatus	Cr	Mersin	1.54	<0.0001	(Korkmaz et al., 2017)
M. barbatus	Cr	Mersin	1.54	<0.0001	(Korkmaz et al., 2017)
M. barbatus	Cr	İskenderun	0.3786	<0.0001	(Turan et al., 2009)
M. barbatus	Cr	Antalya	0.55	<0.0001	(Türkmen & Pınar, 2018)
M. barbatus	Cr	İskenderun	0.6578	<0.0001	(Türkmen et al., 2005)
S. rivulatus	Cr	Antalya	0.36	<0.0001	(Ateş et al., 2015)
S. rivulatus	Cr	İskenderun	1.24	<0.0001	(Ateş et al., 2015)
S. solea	Cr	İskenderun	0.05	<0.0001	(Ersoy & Çelik, 2010)
S. solea	Cr	Samandağ	0.146	<0.0001	(Kaya & Turkoglu, 2017)
S. solea	Cr	Mersin	0.001	<0.0001	(Korkmaz et al., 2017)
S. solea	Cr	Mersin	0.001	<0.0001	(Korkmaz et al., 2017)
S. solea	Cr	İskenderun	0.29	<0.0001	(Türkmen, 2011)
S. solea	Cr	Mersin	1.78	<0.0001	(Türkmen, 2011)
M. barbatus	Cu	İskenderun	3.5	0.0002	(Çoğun et al., 2006)
M. barbatus	Cu	İskenderun	1.176	0.0181	(Kalay et al., 1999)
M. barbatus	Cu	Mersin	0.72	0.0061	(Korkmaz et al., 2017)
M. barbatus	Cu	Mersin	0.72	0.0037	(Korkmaz et al., 2017)
M. barbatus	Cu	Mersin	0.234	0.0037	(Külcü et al., 2014)
M. barbatus	Cu	İskenderun	1.22	0.0012	(Tepe et al., 2008)
M. barbatus	Cu	Antalya	0.26	0.0063	(Türkmen & Pınar, 2018)
M. barbatus	Cu	İskenderun	0.5952	0.0013	(Türkmen et al., 2005)
M. barbatus	Cu	Antalya	0.9562	0.0031	(Yipel & Yarsan, 2014)
S. rivulatus	Cu	Antalya	0.44	0.0049	(Ateş et al., 2015)
S. rivulatus	Cu	İskenderun	0.35	0.0023	(Ateş et al., 2015)
S. solea	Cu	İskenderun	1.66	0.0018	(Çoğun et al., 2005)
S. solea	Cu	İskenderun	1.06	0.0086	(Ersoy & Çelik, 2010)
S. solea	Cu	İskenderun	1.54	0.0055	(Ersoy & Çelik, 2010)
S. solea	Cu	Mersin	0.49	0.0080	(Korkmaz et al., 2017)
S. solea	Cu	Mersin	0.49	0.0025	(Korkmaz et al., 2017)
S. solea	Cu	Mersin	1.336	0.0025	(Külcü et al., 2014)
S. solea	Cu	İskenderun	1.82	0.0069	(Türkmen, 2011)
S. solea	Cu	Mersin	0.38	0.0094	(Türkmen, 2011)
M. barbatus	Fe	İskenderun	3.314	0.0020	(Bıçkıcı, 2010)
M. barbatus	Fe	İskenderun	217.08	0.0010	(Çiçek et al., 2008)
M. barbatus	Fe	İskenderun	0.662	0.0642	(Dural et al., 2010)
M. barbatus	Fe	Mersin	10.4	0.0002	(Korkmaz et al., 2017)
M. barbatus	Fe	Mersin	0.4	0.0031	(Korkmaz et al., 2017)
M. barbatus	Fe	Mersin	13.97	0.0001	(Külcü et al., 2014)
M. barbatus	Fe	İskenderun	15.132	0.0041	(Manaşırlı et al., 2015)
M. barbatus	Fe	İskenderun	34.8	0.0045	(Tepe et al., 2008)
M. barbatus	Fe	İskenderun	4.3802	0.0103	(Turan et al., 2009)
M. barbatus	Fe	İskenderun	2.686	0.0013	(Türkmen et al., 2005)
S. rivulatus	Fe	Antalya	39.1	0.0008	(Ateş et al., 2015)
S. rivulatus	Fe	İskenderun	25.2	0.0116	(Ateş et al., 2015)
S. solea	Fe	İskenderun	3.18	0.0074	(Çoğun et al., 2005)
S. solea	Fe	İskenderun	1.11	0.0009	(Ersoy & Çelik, 2010)
S. solea	Fe	Mersin	13.52	0.0003	(Korkmaz et al., 2017)
S. solea	Fe	Mersin	13.52	0.0040	(Korkmaz et al., 2017)
S. solea	Fe	Mersin	18.382	0.0040	(Külcü et al., 2014)
S. solea	Fe	İskenderun	59.7	0.0054	(Türkmen, 2011)
S. solea	Fe	Mersin	15.5	0.0176	(Türkmen, 2011)
M. barbatus	Mn	Mersin	0.33	0.0046	(Korkmaz et al., 2017)
M. barbatus	Mn	Mersin	0.33	0.0005	(Korkmaz et al., 2017)
M. barbatus	Mn	İskenderun	0.88	0.0005	(Tepe et al., 2008)
M. barbatus	Mn	İskenderun	0.0206	0.0013	(Turan et al., 2009)
M. barbatus	Mn	Antalya	0.14	0.0000	(Türkmen & Pınar, 2018)
M. barbatus	Mn	İskenderun	0.5274	0.0002	(Türkmen et al., 2005)
S. rivulatus	Mn	Antalya	0.33	0.0008	(Ateş et al., 2015)
S. rivulatus	Mn	İskenderun	0.86	0.0005	(Ateş et al., 2015)
S. solea	Mn	İskenderun	0.41	0.0013	(Ersoy & Çelik, 2010)
S. solea	Mn	Mersin	1.13	0.0006	(Korkmaz et al., 2017)
S. solea	Mn	Mersin	1.135	0.0017	(Korkmaz et al., 2017)
S. solea	Mn	İskenderun	1.11	0.0017	(Türkmen, 2011)
S. solea	Mn	Mersin	0.9	0.0016	(Türkmen, 2011)
M. barbatus	Zn	İskenderun	0.111	0.0013	(Bıçkıcı, 2010)
M. barbatus	Zn	İskenderun	39.41	0.0001	(Çiçek et al., 2008)

Table 8. Heavy metal accumulation (µg/g w.wt.) and THQ values of muscle tissue of fish from previous studies evaluated in Iskenderun, Mersin and Antalya Bays

Species	Metal	Bay	w.wt*	THQ	Reference
M. barbatus	Zn	Mersin	17.12	0.0272	(Korkmaz et al., 2017)
M. barbatus	Zn	Mersin	17.12	0.0118	(Korkmaz et al., 2017)
M. barbatus	Zn	Mersin	5.024	0.0118	(Külcü et al., 2014)
M. barbatus	Zn	İskenderun	6.634	0.0035	(Manaşırlı et al., 2015)
M. barbatus	Zn	İskenderun	10.2	0.0046	(Tepe et al., 2008)
M. barbatus	Zn	İskenderun	1.174	0.0070	(Turan et al., 2009)
M. barbatus	Zn	Antalya	4.38	0.0008	(Türkmen & Pınar, 2018)
M. barbatus	Zn	İskenderun	1.163	0.0030	(Türkmen et al., 2005)
S. rivulatus	Zn	Antalya	5.36	0.0008	(Ateş et al., 2015)
S. rivulatus	Zn	İskenderun	5.64	0.0037	(Ateş et al., 2015)
S. solea	Zn	İskenderun	6.76	0.0039	(Çoğun et al., 2005)
S. solea	Zn	İskenderun	2.76	0.0047	(Ersoy & Çelik, 2010)
S. solea	Zn	Mersin	23.58	0.0019	(Korkmaz et al., 2017)
S. solea	Zn	Mersin	4.568	0.0163	(Külcü et al., 2014)
S. solea	Zn	İskenderun	5.66	0.0031	(Türkmen, 2011)
S. solea	Zn	Mersin	6.8	0.0039	(Türkmen, 2011)

* Dry weight values were converted to wet wt. dividing by 5 (According to Yılmaz 2010)

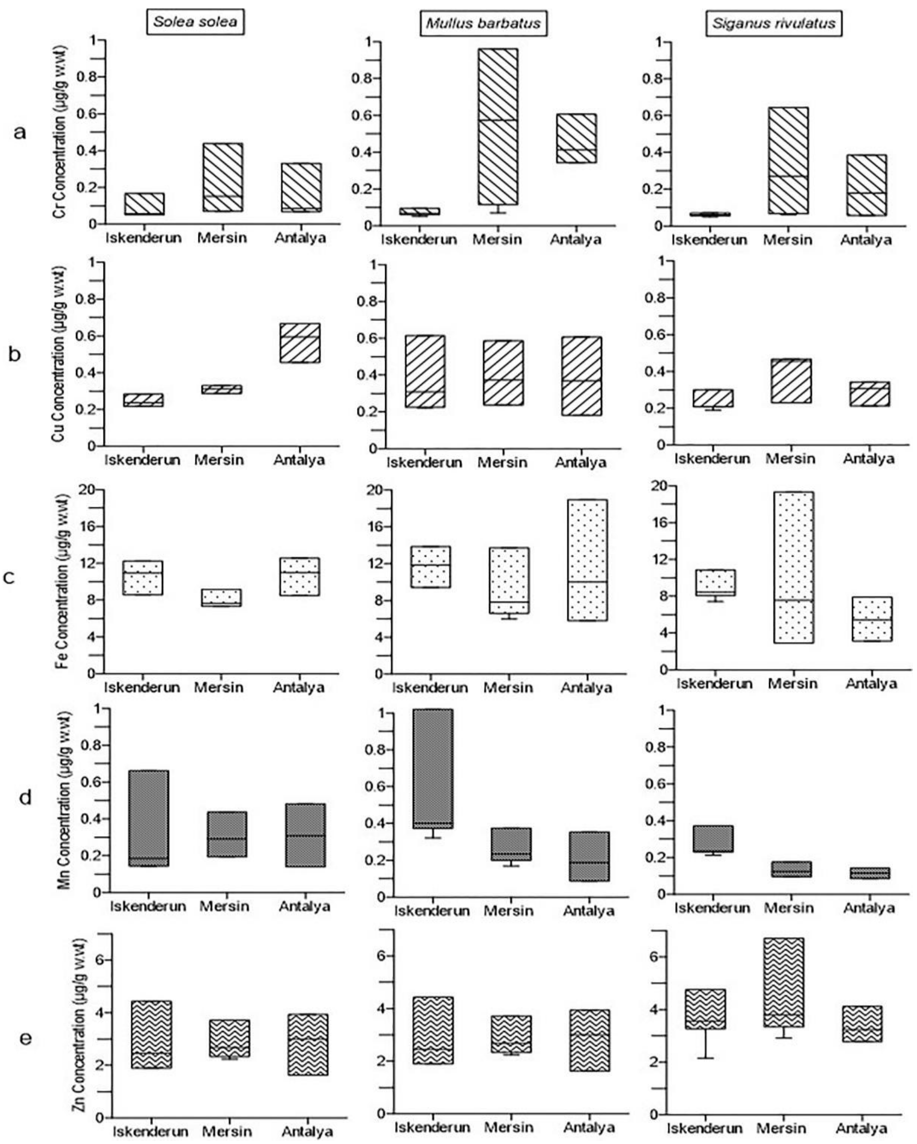

Fig. 2. Box and whisker plots indication heavy metal concentration in muscle tissues of *Solea solea, Mullus barbatus, Siganus rivulatus*

Results showed that Cu levels in the muscle tissue of *S. solea* and *M. barbatus* in İskenderun, Mersin and Antalya Bays were significantly different from the other tissues ($p<0.05$). For *S. rivulatus*, Cu accumulation in the muscle tissue was significantly different from all other tissues in İskenderun Bay, and also it was only significantly different intestine in Mersin Bay and intestine and liver in Antalya Bay ($p<0.05$) (Table 2). Considering all species, the Cu accumulation stability was found based on mean Cv% values in descending order as: M > L > I > S (Table 6).

In general, our results for mean Cu concentrations (µg metal g^{-1} w.wt.) in muscle tissue of *S. solea* (0.24-0.57) , *M. barbatus* (0.34-0.38) and, *S. rivulatus* (0.23-0.38) were consistent with the previous studies as 0.38- 1.66, 0.23-3.5, 0.35-0.44, respectively (Table 2, Table 8).

Mean iron (Fe) concentrations of fish tissues from three bays were shown in Table 3 (µg g^{-1} w.wt.). The accumulation rates in the tissues of *S. solea* were the same for all stations and ranked as L > I > S > M. There were two different patterns on ranking of heavy metals in tissues of *M. barbatus and S. rivulatus as* I>L>S>M *(S. rivulatus* from Antalya, *M.barbatus* from all Bays) and L>I>S>M *(S. rivulatus* from Mersin and İskenderun).

Statistical differences in muscle tissues among the species were examined (Table 3, Fig 2 c) and only *S. rivulatus* was found to be different in Antalya Bay ($p<0.05$, denoted as b). Fe concentrations in the muscle tissue (µg g^{-1} w.wt.) of *S. solea*, *M. barbatus*, and *S. rivulatus* changed as 8.55-7.39, 12.25-10.85, 9.4-13.85 in İskenderun Bay, 7.32-9.17, 2.91-9.31, 5.98-13.73 in Mersin Bay and 0.46-0.67, 3.12-7.89, 5.79-18.95 in Antalya Bay, respectively (Fig. 2c). The Fe accumulation stability was found in descending order as: L > S >M > I (Table 6).

Our findings on mean Fe concentrations (µg metal g^{-1} w.wt.) in the muscle tissues of *S. solea* (8.04-10.68) and *M. barbatus* (9.45-11.73) were found to be in the range of previous studies as 1.11-59.7 and 0.4-217.08, respectively. But, for *S. rivulatus* our results (5.47- 9.33) were far below reported studies (25.2- 39.1) from the same bays (Table 3, Table 8).

Mean Mn concentration in different tissues of *S. solea* for İskenderun and Antalya Bays was ranked as S>L>I>M. On the other hand, this ranking was found as I>S>L>M in Mersin Bay. There was no regular accumulation order for *M. barbatus* and rankings were found as L>I>S>M, I>L>S>M and I>S>L>M for İskenderun, Mersin and Antalya Bays, respectively. Mn accumulation orders for *S. rivulatus* in Mersin and Antalya Bays were the same and as follows S>I>L>M and, I>S>L>M for İskenderun Bay. It should be noted that muscle was the least Mn accumulating tissue regardless of species and location (Table 4).

No significant variations in the muscle tissues of *S. solea* were observed depending on bays ($p>0.05$). On the other hand, there were significant differences in muscle tissues between İskenderun-Mersin Bays and İskenderun-Antalya Bays for both *M. barbatus* and *S. rivulatus* in terms of Mn accumulation ($p<0.05$) (Table 4).

Results showed that the highest Mn accumulation level was observed in *M. barbatus* at İskenderun Bay (Fig. 2d). Mn accumulation stability among the tissues was found in descending order as: L > M >S > I (Table 4).

The range of mean Mn accumulation (µg metal g^{-1} w.wt.) in muscle tissues of *M. barbatus* (0.20-0.52) in our study was consistent with previous reports (0.14-0.88). However, our findings on Mn in *S. rivulatus* (0.11-0.28) and *S. solea* (0.29-0.31) were lower than that of reported studies from the same bays as 0.33-0.86 and 0.90-1.13, respectively (Table 4, Table 8).

Mean Zn concentrations in the tissues of *S. solea* were ranked as L > S > I > M, L > I > S > M and I > L > S > M for İskenderun, Mersin and Antalya Bays, respectively. Ranking of mean Zn levels in the *M. barbatus* for İskenderun, Mersin and Antalya Bays were same (S > L > I > M), and for *S. rivulatus* in all three bays it was ranked as L > S > I > M. It was seeing that muscle was found to be the least Zn accumulating tissue regardless of species and location (Table 5).

Statistical differences among the tissues of *S. solea* were examined and results showed that Zn concentration in the muscle tissue was different from other tissues in İskenderun Bay, from intestine and skin in Mersin Bay and intestine in Antalya Bay. When *M. barbatus* and *S. rivulatus* were examined, it was found that the Zn level in the muscle tissue was significantly different from all other tissues in all stations ($p<0.05$). Also, considering muscle tissue, there were no significant differences in mean Zn concentrations depending on either location or species ($p>0.05$).

As seen in Table 6, the Zn accumulation stability among the tissues in descending order was found as: M >L >S > I.

The mean Zn concentrations in the muscle tissues of (µg metal g^{-1} w.wt.) *S. solea* (3.11-3.31) and *M. barbatus* (2.81-2.89) were consistent with previous studies as 2.76-23.58 and 0.11-25.12, respectively. However, the mean range of Zn values for *S. rivulatus* obtained in this study (3.34-4.15) was found to be lower than that of previous studies (5.36-5.64) (Table 5, Table 8).

The highest levels of THQ_{gp} and THQ_{f} for *S. solea*, *M. barbatus and S. rivulatus* in İskenderun, Mersin and Antalya Bays were calculated for Zn (Table 7). The $TTHQ_{gp}$ and $TTHQ_{f}$ for *S. solea*, *M. barbatus and S. rivulatus* in İskenderun, Mersin, and Antalya Bays were calculated as (0.020 and 0.096), (0.042 and 0.083), and (0.027 and 0.053); (0.017 and 0.033), (0.024 and 0.048), (0.032 and 0.063); (0.032 and 0.065), (0.032 and 0.064), and (0.018 and 0.036), respectively.

In this study, calculated THQ and TTHQ values were not exceeded the threshold point (<1.00) for any cases and these results were also valid for fishermen who were assumed to consume two times more fish than that of the general population. In general, therefore, our findings were consistent with previous studies (Table 8). Results show that there is no evidence and acceptable non-cancer risk for both the general population and fishermen eating the considered fish in this study from three bays.

CONCLUSION

This study was carried out to provide some information on heavy metal accumulation and related health risk assessment of both general and fishermen populations for different fish species from İskenderun, Mersin and, Antalya Bays. There were some significant inter- and intra- species/ tissues/bays differences. The stability in heavy metal accumulation in fish tissues varied and the most stable tissue by Cr, Cu, Fe, Mn and Zn were determined as skin, muscle, liver, liver and, muscle, respectively. In general, the lowest heavy metals value was observed in fish muscle tissue. THQ and TTHQ values were not exceeded by 1.00. Therefore, these results suggest that both general and fishermen populations are not subjected to the significant potential health risk from these bays, yet.

Although our findings on heavy metal accumulation in fish are mostly consistent with previous studies conducted in the same areas, monitoring programs should be continued for protecting the environment and human health.

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CONFLICT OF INTEREST

The authors declare that there is not any conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/ or falsification, double publication and/or submission, and redundancy has been completely observed by the authors.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research.

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