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RESEARCH ARTICLE

Comparative investigation of heavy metal, trace, and macro element contents in commercially valuable fish species harvested off from the Persian Gulf

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Abstract This study was performed to determine the differences between two commercial species of fish harvested off near the Kharg Island (one of the largest oil terminals in the world) in the Persian Gulf in terms of toxic metals, macro, and trace elements. Samples were analyzed using inductively coupled plasma-optical emission spectrometry (ICP-OES). The results showed that Ca, Li, Mg, P, Se, Sn, Sr, and Zn concentrations were significantly different between the skin and muscle tissues of *Scomberomorus guttatus* but with the exception of P, there was no significant difference between element levels in the skin and muscle tissues of *Otolithes ruber*. The *S. guttatus* contained significantly higher levels

of As, Sn, Se, and P in the muscle tissue and Zn in the skin tissue compared to the muscle and skin tissues of *Otolithes ruber*. The estimated daily intake of the toxic elements including As, Cd, Sb, Pb, and Sn via consumption of these fish were below the established guidelines but due to the potential contamination by oil activities near the island, continuous and permanent monitoring in this region is highly recommended.

Keywords *Scomberomorus guttatus* · *Otolithes ruber* · Heavy metals · Trace elements · Persian Gulf

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Introduction

There is an increasing concern about the quality of foods in some parts of the world. The measurement of toxic elements in different foods has prompted researches to study the toxicological effects of these elements in food. Heavy metals (HMs) are considered as one of the most critical contaminants of the aquatic ecosystem because of their potential to enter water bodies and also their bioaccumulation and biomagnification in the food chain (Olojo et al. 2005). Fish are commonly situated at the top of the food chain and are considered as a susceptible aquatic organism to toxics present in water (Alibabic et al. 2007). Moreover, the increasing importance of aquatic food including fish as protein sources extended the focus toward aquatic ecosystem. So, in recent years, much attention has been focused on the levels of HMs in fish and other food to determine the hazard effects of HMs for human health. HMs can enter into water bodies and aquatic ecosystems through drainage, atmosphere, soil erosion, and all human activities by different ways. Contaminants including HMs enter fish body through several main routes: via gills, non food particles, skin, and oral consumption of

water. These metals concentrate at different contents in organs of fish body. Some metals such as iron, copper, zinc, and manganese are essential for biological systems such as enzymatic activities, whereas, other HMs like arsenic, lead, cadmium, and tin have no known important role in living organs and are toxic even in trace amounts (Fernandes et al. 2008). Essential metals must be taken up from water, food, or sediment by fish for its normal metabolism (Canli and Atli 2003). However, these metals can also have adverse and toxic effects at high concentration (Tüzen 2003). Heavy metal levels of fish body have been broadly studied (Canli and Atli 2003; Fernandes et al. 2008; Mansour and Sidky 2002; Tüzen 2003) and its distribution varies between fish species (Kagi and Schaffer 1988).

Two commercial important species of fish in the Persian Gulf are including *Scomberomorus guttatus* (Indo-Pacific king mackerel) and *Otolithes ruber* (tiger tooth croaker) fish. *S. guttatus* is found in the Indo-West Pacific, Persian Gulf, India, and Sri Lanka to Southeast Asia, north of Hong Kong and Wakasa Bay, Sea of Japan, and Gulf of Thailand. They are a pelagic species and tending to migrate locally around the beach and sometimes entering turbid estuarine areas. Juveniles of *S. guttatus* feed mainly on teleosts, especially anchovies, while adults prey mainly on fish with small quantities of squids and crustaceans. *O. ruber* is found in the West Indian Ocean from the east to Queen’s lands of Australia and Japan except the Red Sea, across the Persian Gulf coastal area, and Oman Sea. They live in the marine coastal area water and mostly on the muddy surfaces and rivers mouths (Collette and Nauen

1983; Fischer and Bianchi 1984). Food of *O. ruber* consists of such crustaceans as shrimp and other invertebrates. Juveniles of *O. ruber* feed mainly on zoobenthos while adults prey mainly on small fish and crustaceans (especially shrimp).

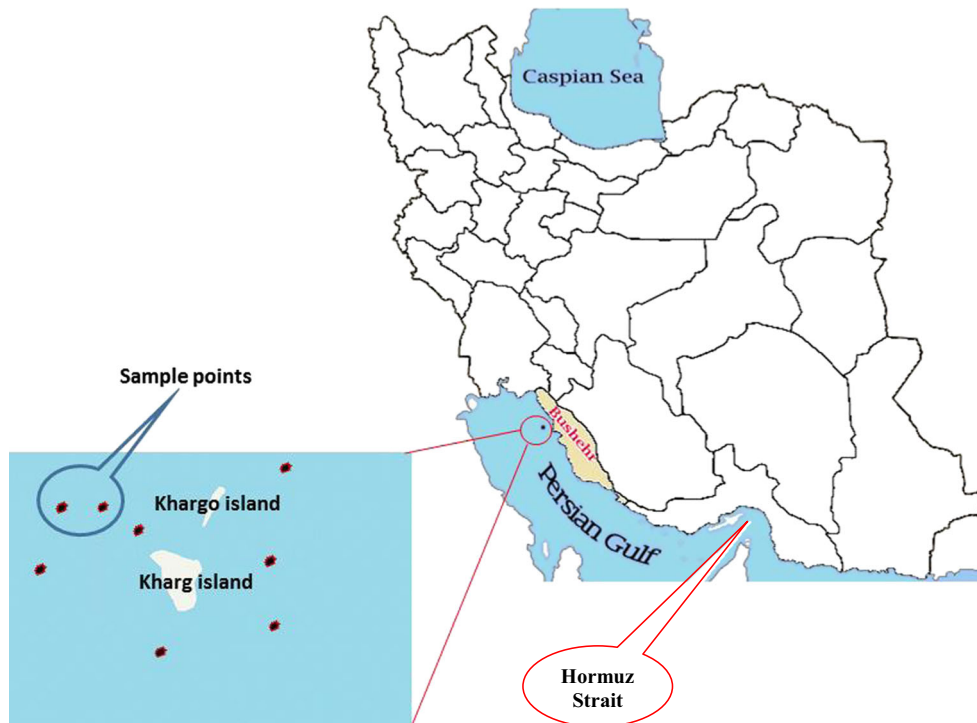
Fish muscle tissue is not always a suitable indicator of the whole fish body contamination and, therefore, it is important to examine other tissues, such as skin. To our knowledge, *S. guttatus* and *O. ruber* fish in the Persian Gulf have not been examined in detail for different content of heavy metals, macro, and trace elements in their skin and muscle tissues. As fish from the Persian Gulf serve as the main source of protein for people living in this region (including Iran in the north and Arabic countries in the south of the Persian Gulf), we measured the heavy metals, macro, and trace elements in the skin, and muscle tissues of these two species of fish that harvested off near the Kharg island, one of the largest oil terminals in the world. In this study we report the Ag, Al, As, B, Ca, Cd, Cu, Fe, Li, Mg, Mn, Na, Ni, P, Pb, Sb, Se, Si, Sn, Sr, and Zn concentrations of the skin and muscle tissues of the mentioned fish to monitoring contaminants accumulation in the edible tissues as well as the nutrition values of these fish.

Materials and methods

Sample collection and preparation

Fish samples including *S. guttatus* and *O. ruber* were harvested and collected near the Kharg island, one of the largest oil terminals in the world, in the Bushehr province. Kharg Island

Fig. 1 Location of sample points near the Kharg Island in the Persian Gulf



is located 25 km off the coast in the Bushehr province of Iran and 483 km in the northwest of the Hormuz Strait (Fig. 1). About 20 % of the world's petroleum, and nearly 35 % of the petroleum traded by sea, passes through the Hormuz Strait, making it a highly important strategic location for international oil trade. This island provides a sea port for the export of the Iranian oil. The mean weight and length (mean±SD) of fish were 905±25.4 g and 39.3±4.3 cm for *S. guttatus* and 965±31.8 g and 42.1±5.1 cm for *O. ruber*. The fish were brought to the laboratory and washed with distilled water. Muscle and skin were dried, homogenized, and ground first with a meat grinder and later, with a mortar and pestle to a fine powder.

Reagents

HNO₃ (65 %) and H₂O₂ (30 %) were of supra pure quality (Merck, Darmstadt, Germany). All the plastic and glassware was cleaned by soaking in dilute nitric acid and rinsed with distilled water before use. All solutions were prepared using ultrapure water (resistivity 18.2 MΩ cm).

Analytical procedures

A weighed sample (1 g) of dry skin and muscle powder was mixed with 4 ml 30 % of H₂O₂ and 8 ml of 65 % HNO₃ in a Teflon container and the samples were digested by boiling until all the contents dissolved. After cooling to room laboratory temperature, the solution was filtered through filter (a 0.45-μm nitrocellulose membrane filter) and transferred to an acid-washed volumetric flask (25 ml) and ultrapure water was added to bring the final sample volume up to 25 ml. Blank digest was also performed in the same way.

Analysis of the elements including Ag, Al, As, B, Ca, Cd, Cu, Fe, Li, Mg, Mn, Na, Ni, P, Pb, Sb, Se, Si, Sn, Sr, and Zn was carried out by inductively coupled plasma optical spectrometry (ICP-OES). In Table 1, details of the instrumental operating conditions are presented. All heavy metal, macro, and trace element concentrations were expressed as micrograms per gram wet weight (*ww*). Statistical analysis of data was carried out with the statistical package for the social sciences (SPSS) and independent sample *t* test and Pearson's correlation coefficient were performed for statistical significant differences. Differences in mean values were accepted as being significant if *P*<0.05.

Results and discussion

The heavy metal, macro, and trace element concentrations in the muscle and skin tissue samples of the *S. guttatus* and the *O. ruber* are presented in Table 2.

Table 1 ICP-OES instrumental operating details

Parameter	
Company, model	SPECTRO (Germany), Spectro arcos
RF generator power (W)	1400
Frequency of RF generator (MHz)	27.12 MHz
Type of detector	Charge coupled devices (CCD)
Torch type	Flared-end EOP torch 2.5 mm
Plasma, auxiliary, and nebulizer gas	High purity (99.99 %) argon
Plasma gas flow rate (l/min)	14.5
Auxiliary gas flow rate (l/min)	0.9
Nebulizer gas flow rate (l/min)	0.85
Sample uptake time (s)	240 total
Delay time of (s)	–
Rinse time of (s)	45
Initial stabilization time (s)	Preflush:45
Time between replicate analysis (s)	–
Measurement replicate	3
Pump rate	30 RPM
Element (λ/nm)	Ag 328.068; Al 396.152 As 189.042; B 249.773 Ca 422.673; Cd 228.802 Cu 324.754; Fe 259.941 Li 670.780; Mg 279.553 Mn 257.611; Na 589.592 Ni 231.604; P 177.495 Pb 283.305; Sb 206.833 Se 196.090; Si 251.611 Sr 346.446; Sn 189.991 Zn 268.416

As shown in Fig. 2a, b, the orders of heavy metal, macro, and trace element concentrations of *O. ruber* were P > Na > Ca > Mg > Zn > Sr > Al > Fe > Si > Cu > Se > Ag > B > As > Ni > Cd > Pb > Mn > Sb > Li > Sn in the muscle and P > Ca > Na > Mg > Zn > Sr > Fe > Al > Si > Mn > Se > Cu > As > Ni > B > Ag > Pb > Sb > Li > Sn in the skin.

In the case of *S. guttatus*, the orders of heavy metal, macro, and trace element levels of fish were P > Mg > Na > Ca > Zn > Si > Fe > Sr > As > Se > Al > Cu > Mn > B > Pb > Ni > Sn > Cd > Sb > Ag > Li in the muscle and Ca > P > Na > Mg > Zn > Sr > Al > Fe > Si > Cu > As > Mn > B > Ag > Se > Ni > Cd > Sb > Li > Sn in the skin (Fig. 2c, d). Recently, considerable studies have been done to determine the distribution of heavy metals, macro, and trace elements in different aquatic ecosystems and different parts of fish body (Dobaradaran et al. 2010, 2011; Dural et al. 2007; Fallah et al. 2011; Mansour and Sidky 2002; Mendil et al. 2010; Saei-Dehkordi and Fallah 2011; Santos et al. 2013; Tüzen 2003; Tuzen et al. 2009; Yilmaz

Table 2 Mean and range concentration of the elements in the muscle and skin of the fish ($\mu\text{g/g}$ /wet weight; values are mean \pm SD, sample number for each parameter=7)

Elements	<i>O. ruber</i>		<i>S. guttatus</i>	
	Muscle	Skin	Muscle	Skin
Ag	0.43 \pm 0.40 (0.05–0.93)	0.36 \pm 0.46 (0.01–0.97)	0.09 \pm 0.08 (0.01–0.24)	0.54 \pm 0.66 (0.03–1.67)
Al	4.01 \pm 3.49 (0.47–7.52)	3.41 \pm 1.58 (0.28–4.48)	0.99 \pm 0.26 (0.74–1.25)	7.14 \pm 5.94 (0.79–17.85)
As	0.36 \pm 0.08 (0.27–0.46)	0.46 \pm 0.18 (0.31–0.76)	1.17 \pm 0.34 (0.83–1.63)	0.80 \pm 0.35 (0.43–1.30)
Fe	2.34 \pm 0.46 (1.79–2.84)	3.67 \pm 1.87 (1.75–6.44)	2.93 \pm 1.43 (1.37–5.29)	4.80 \pm 2.35 (2.54–8.82)
Mn	0.18 \pm 0.16 (0.03–0.40)	0.74 \pm 1.00 (0.20–2.77)	0.55 \pm 0.43 (0.05–1.09)	0.57 \pm 0.21 (0.43–0.95)
Sb	0.11 \pm 0.02 (0.09–0.13)	0.14 \pm 0.03 (0.11–0.19)	0.13 \pm 0.05 (0.08–0.21)	0.10 \pm 0.02 (0.07–0.14)
Sn	0.04 \pm 0.00 (0.03–0.04)	0.04 \pm 0.01 (0.02–0.04)	0.23 \pm 0.21 (0.04–0.50)	0.04 \pm 0.01 (0.02–0.04)
Zn	7.04 \pm 3.15 (3.25–10.15)	14.92 \pm 10.78 (5.54–33.20)	18.37 \pm 23.08 (4.27–64.09)	53.15 \pm 21.68 (27.91–78.81)
B	0.38 \pm 0.17 (0.18–0.63)	0.40 \pm 0.23 (0.23–0.86)	0.46 \pm 0.21 (0.22–0.75)	0.55 \pm 0.17 (0.43–0.88)
Ca	262.51 \pm 153.22 (99.26–510.72)	406.40 \pm 233.58 (150.49–613.5)	237.23 \pm 138.42 (110.57–411.4)	631.28 \pm 173.56 (357.5–791.7)
Li	0.06 \pm 0.02 (0.04–0.09)	0.08 \pm 0.03 (0.05–0.12)	0.06 \pm 0.02 (0.03–0.10)	0.10 \pm 0.01 (0.09–0.12)
Mg	184.16 \pm 29.80 (146.89–229.82)	132.35 \pm 53.20 (84.3–198.84)	265.50 \pm 96.97 (152.6–440.8)	121.57 \pm 26.97 (74–147.31)
Na	329.61 \pm 102.00 (224.71–490.38)	279.87 \pm 88.52 (148.68–424.0)	261.26 \pm 154.76 (109.96–521.3)	212.05 \pm 82.15 (116.82–307.5)
P	847.52 \pm 80.13 (765.77–1000.8)	473.75 \pm 302.18 (174.92–863.7)	1339.68 \pm 446.60 (745–2121.5)	478.75 \pm 123.66 (261.30–606.57)
Se	0.67 \pm 0.09 (0.60–0.85)	0.67 \pm 0.13 (0.46–0.82)	1.04 \pm 0.39 (0.73–1.60)	0.50 \pm 0.15 (0.32–0.67)
Si	2.32 \pm 0.92 (1.18–3.28)	3.00 \pm 1.32 (1.98–5.35)	3.41 \pm 3.08 (0.92–9.10)	3.41 \pm 1.51 (1.21–5.28)
Sr	4.47 \pm 3.51 (0.74–8.66)	6.11 \pm 4.06 (1.92–12.17)	2.04 \pm 1.23 (0.85–4.22)	8.84 \pm 2.16 (5.74–12.45)
Cd	0.33 \pm 0.13 (0.13–0.43)	–	0.21 \pm 0.11 (0.07–0.33)	0.19 \pm 0.2 (0.01–0.45)
Cu	0.98 \pm 0.86 (0.33–2.1)	0.59 \pm 0.19 (0.38–0.9)	0.62 \pm 0.22 (0.44–0.93)	0.82 \pm 0.63 (0.52–2.1)
Ni	0.34 \pm 0.12 (0.2–0.53)	0.44 \pm 0.14 (0.3–0.66)	0.35 \pm 0.15 (0.25–0.64)	0.49 \pm 0.15 (0.38–0.73)
Pb	0.19 \pm 0.87 (0.09–0.3)	0.28 \pm 0.1 (0.18–0.41)	0.41 \pm 0.325 (0.06–1.00)	–

et al. 2007). However, there is no consensus among the published reports. This may be due to different factors including location of capture and habitats, species, water concentration of elements, growth rate, metal accumulation, and detoxification mechanism (Marin-Guirao et al. 2008). In a recent

study, the accumulation of trace elements in different organs of fish including muscle, skin, and gills of four edible fish species from the Rawal lake Reservoir, Pakistan, in a different season was described. The accumulation of trace metals in different organs of fish in pre-monsoon was lower than in

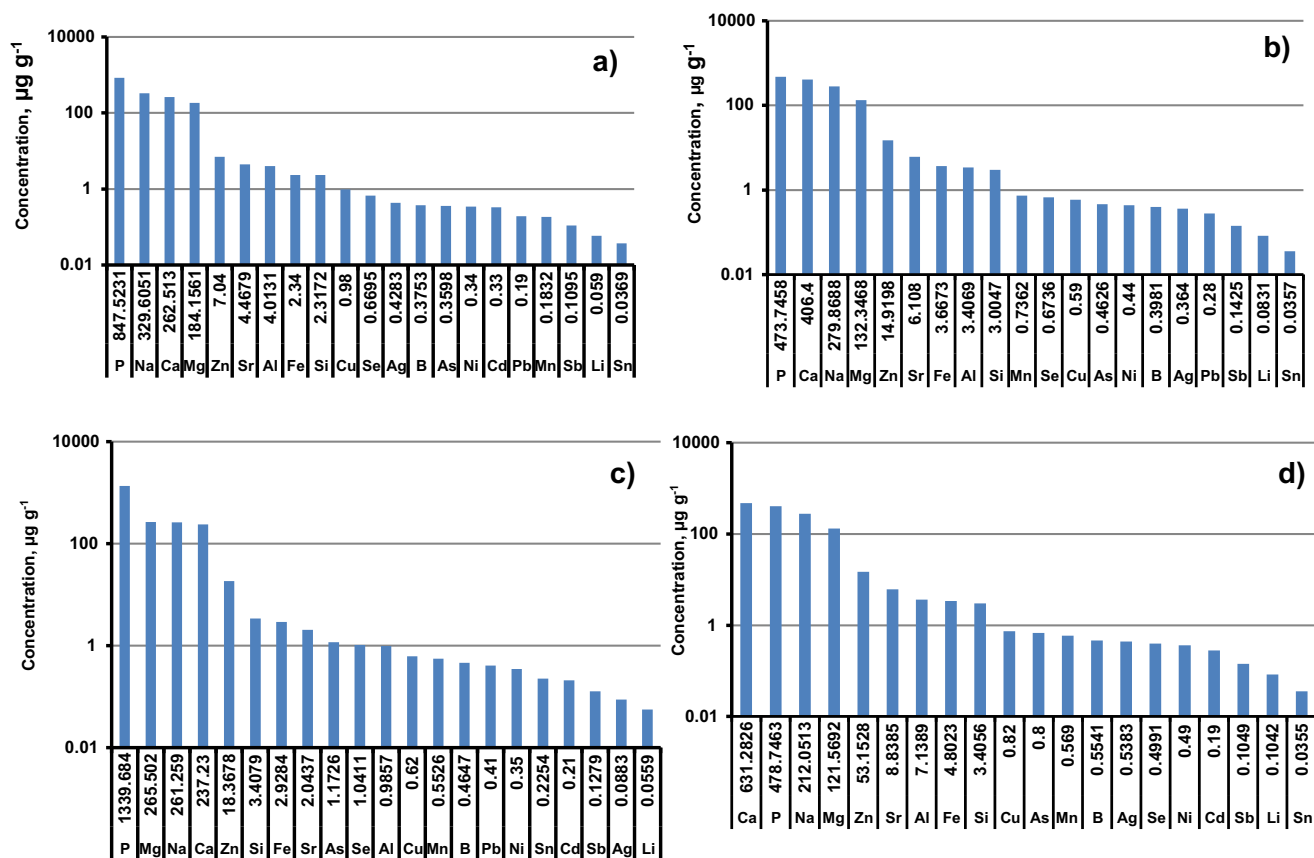


Fig. 2 Order of heavy metal, nutrient, and trace element concentrations in the muscle (a) and skin (b) tissues of *O. ruber* and muscle (c) and skin (d) tissues of *S. guttatus*

post-monsoon. In pre-monsoon season, the trace metals followed the order Zn > Pb > Fe > Cr > Ni > Mn > Co > Cu > Cd > Li, while in the post-monsoon, the order was Fe > Pb > Cr > Ni > Zn > Cu > Co > Mn > Cd > Li (Malik et al. 2014).

The results of the present study showed that with the exception of P (with higher level in the muscle tissue of fish ($P < 0.05$, Fig. 2a–d) there were no significant difference ($P > 0.05$) between heavy metal, macro, and trace element levels in the skin and muscle tissues of the *O. ruber* (Table 3) while Ca, Li, Mg, P, Se, Sn, Sr, and Zn concentrations were significantly different ($P < 0.05$) in the skin and muscle tissues of *S. guttatus* (Table 3) (Ca, Li, Sr, and Zn concentrations were higher in the skin tissue of fish, Fig. 2a–d). Our study showed *S. guttatus* contained significantly higher concentrations of As, Se, Sn, and P in the muscle tissue and Zn in skin tissue ($P < 0.05$) compared to muscle and skin tissues of *O. ruber* (Table 3).

The elements such as iron, copper, zinc, and manganese are essential for biological systems such as enzymatic activities and functional and structural roles, whereas, elements like lead, cadmium, and arsenic are known as potentially toxic (Fernandes et al. 2008). The skin tissue of *S. guttatus* contained significantly higher concentrations ($P < 0.05$) of Zn and Ca compared to the muscle tissue of *S. guttatus* but the muscle tissue of *S. guttatus* contained significantly higher

levels ($P < 0.05$) of Mg, Se, and P compared to the skin tissue of *S. guttatus*. There were no significant differences ($P > 0.05$) between Mn, Fe, Cu, and Na concentrations in the skin and muscle tissues of *S. guttatus* (Table 3). In the case of *O. ruber*, with the exception of P (with higher concentration in the muscle tissue of fish) ($P < 0.05$), there were no significant differences between Se, Zn, Mn, Ca, Fe, Cu, Na, and Mg concentrations in the skin and muscle tissues of *O. ruber* (Table 3). The concentrations of Se and P in the muscle tissue of *S. guttatus* compared to *O. ruber* were higher and significantly different ($P < 0.05$) but no statistically significant differences ($P > 0.05$) were observed in the Mn, Ca, Fe, Cu, Na, and Mg levels between *O. ruber* and *S. guttatus* tissues (both skin and muscle tissues); while the skin tissue of *S. guttatus* contained significantly higher concentration ($P < 0.05$) of Zn (Table 3). The difference in accumulation potential between skin and muscle of fish can be due to greater tendency of the element to react with the proteins that are present in the skin and muscle tissues of these fish (Uysal et al. 2008; Visnjic-Jeftic et al. 2010).

The similar pattern has been extensively reported in several studies for different species (Brucka-Jastrzëbska et al. 2009; Jarić et al. 2011; Verep et al. 2007; Visnjic-Jeftic et al. 2010). In a report, the accumulation of heavy metals in the muscle

Table 3 Comparison between the means of the element levels among the samples

Comparison	Skin and muscle of <i>O. ruber</i>		Skin and muscle of <i>S. guttatus</i>		Skin of <i>O. ruber</i> and skin of <i>S. guttatus</i>		Muscle of <i>O. ruber</i> and muscle of <i>S. guttatus</i>	
	Mean difference (Std. Error)	Sig*	Mean difference (Std. Error)	Sig*	Mean difference Std. Error	Sig*	Mean difference Std. Error	Sig*
Ag	0.064 (0.259)	0	-0.450 (0.271)	0	-0.174 (0.351)	0	0.340 (0.166)	0
Al	0.606 (1.578)	0	-6.153 (3.554)	0	-3.732 (2.511)	0	3.027 (2.082)	0
As	-0.102 (0.081)	0	0.372 (0.197)	0	-0.337 (0.159)	0	-0.812 (0.142)	1
B	-0.022 (0.118)	0	-0.089 (0.109)	0	-0.155 (0.117)	0	-0.089 (0.110)	0
Ca	-143.8 (116.9)	0	-394.05 (93.9)	1	-224.9 (130.1)	0	25.28 (84.29)	0
Cd	-	0	0.026 (0.112)	0	-	0	0.115 (0.086)	0
Cu	0.387 (0.359)	0	-0.148 (0.271)	0	-0.228 (0.267)	0	0.307 (0.362)	0
Fe	-1.32731 (0.78820)	0	-1.873 (1.123)	0	-1.135 (1.228)	0	-0.588 (0.612)	0
Li	-0.024 (0.014)	0	-0.048 (0.010)	1	-0.021 (0.012)	0	0.003 (0.012)	0
Mg	51.81 (24.894)	0	143.9 (41.09)	1	10.77 (24.35)	0	-81.34 (41.41)	0
Mn	-0.553 (0.413)	0	-0.016 (0.196)	0	0.167 (0.416)	0	-0.369 (0.189)	0
Na	49.74 (55.13)	0	49.20 (71.53)	0	67.81 (49.30)	0	68.34 (75.67)	0
Ni	-0.099 (0.076)	0	-0.140 (0.087)	0	-0.053 (0.085)	0	-0.011 (0.078)	0
P	373.7 (127.63)	1	860.9 (189.2)	1	-5.00 (133.29)	0	-492.16 (185.23)	1
Pb	-0.092 (0.054)	0	-	0	-	0	-0.218 (0.137)	0
Sb	-0.033 (0.015)	0	0.023 (0.021)	0	0.037 (0.017)	0	-0.018 (0.020)	0
Se	-0.004 (0.066)	0	0.542 (0.170)	1	0.174 (0.081)	0	-0.371 (0.164)	1
Si	-0.687 (0.657)	0	0.002 (1.400)	0	-0.400 (0.817)	0	-1.090 (1.313)	0
Sn	0.001 (0.004)	0	0.189 (0.084)	1	0.0002 (0.004)	0	-0.188 (0.084)	1
Sr	-1.64 (2.191)	0	-6.794 (1.015)	1	-2.730 (1.878)	0	2.424 (1.518)	0
Zn	-7.88 (4.58)	0	-34.78 (12.92)	1	-38.23 (9.88)	1	-11.327 (9.509)	0

* Significance of 1 indicates that the difference of averages is significant at 0.05; Significance of 0 indicates that the difference of averages is not significant at 0.05

and bone of four fish species from the central Adriatic Sea were measured. Perugini et al. found no significant variations of heavy metal concentrations including As, Cd, Cu, Pb, Zn, Hg, and Se in the muscle of the examined species, but a significant difference was found for Cd, Pb, Se, and As levels in the bones of the examined fish species (Perugini et al. 2014). In another study in the Pagasitikos Gulf in Greece, heavy metal concentrations in two fish species were determined. Significant differences were detected between two examined fish species' tissues concerning Cu and Zn levels (Giannakopoulou and Neofitou 2014). The essential trace element levels of Fe, Mn, Cu, Zn, and Se in the skin and muscle tissues of fish samples were found as 2.34–4.8 µg/g for Fe, 0.18–0.74 µg/g for Mn, 0.59–0.98 µg/g for Cu, 7.04–53.15 µg/g for Zn, and 0.5–1.04 µg/g for Se. Various previous studies have reported the essential element contents of different fish species samples. For example, in a study, the levels of Cu, Zn, and Se have been reported as 0.65–2.75, 38.8–93.4, and 0.19–0.85 µg/g, respectively (Tuzen 2009). In another report, the contents of these elements were found 1.10–2.50 µg/g for Cu, 7.57–34.4 µg/g for Zn, and 0.96–3.64 µg/g for Se (Tuzen and Soylak 2007).

Se is an essential element and acts as a regulator of thyroid hormone metabolism, antagonistic role to the toxicological effects of mercury, antioxidant, and anticarcinogenic (Khan et al. 1987). The Se concentration in our study was below the recommended toxicity threshold for aquatic biota (i.e., 10 µg/g of dry weight) (Pyle et al. 2005). Ni is not an essential element but in a few trace levels, it may be useful to activate some enzyme systems. Chronic intake of Ni can be associated with increased risk of lung cancer (Expert Group on Vitamins and Minerals (EGVM) 2003). The level of Ni in the skin and muscle tissues of *S. guttatus* compared to the skin and muscle tissues of *O. ruber* were not significantly different ($P>0.05$); also, there was no significant difference ($P>0.05$) between the skin tissue and muscle tissue of every fish (Table 3). Because of the unknown metabolic function in organism, Sr is not an essential element, but it can be used as a marker to differentiate between meat and fish due to the fact that its level is noticeably higher in fish than in meat (Carvalho et al. 2005). The concentration of Sr was significantly higher ($P<0.05$) in the skin tissue compared to the muscle tissue of *S. guttatus* but no significant difference ($P>0.05$) was found in the Sr concentration between the skin and muscle tissues of *O. ruber*.

Also, no statistically significant differences ($P>0.05$) were observed in the Sr concentration in the muscle tissues of fish as well as their skin tissues (Table 3). The concentrations of Al, Si, B, and Li in the skin and muscle tissues of *S. guttatus* compared to the skin and muscle tissues of *O. ruber* were not significantly different ($P>0.05$). Also, there were no significant differences ($P>0.05$) between the Al, Si, B, and Li concentrations in the skin and muscle tissues of *O. ruber*, but the skin tissue of *S. guttatus* contained significantly a higher level of Li compared to its muscle tissue.

Some elements such as Ag, Cd, Pb, Sn, Sb, and As, classify as toxic elements and have no metabolic function but can be harmful for human health, even at low levels, if ingested over a long time period (Somers 1974). The toxic element levels of

Ag, As, Cd, Sb, Pb, and Sn in the skin and muscle tissues of fish samples were found as 0.09–0.54 $\mu\text{g/g}$ for Ag, 0.36–1.17 $\mu\text{g/g}$ for As, 0.19–0.33 $\mu\text{g/g}$ for Cd, 0.1–0.14 $\mu\text{g/g}$ for Sb, 0.19–0.41 $\mu\text{g/g}$ for Pb, and 0.04–0.23 $\mu\text{g/g}$ for Sn. In a study, the toxic element levels of Pb and Cd in fish species harvested off from the Black and Aegean Seas were found in the ranges of 0.33–0.93 and 0.45–0.9 $\mu\text{g/g}$, respectively (Uluozlu et al. 2007). Fallah et al. also have reported the toxic levels of Pb, Cd, and As in edible tissues of farmed and wild rainbow trout in the ranges of 0.99–5.32, 0.00–1.21, and 0.00–3.56 $\mu\text{g/g}$, respectively (Fallah et al. 2011). Another study has reported the levels of Pb and Cd in fish species harvested off from the Tokat Lake in the range of 0.7–2.4 and 0.1–1.2 $\mu\text{g/g}$, respectively (Mendil et al. 2005).

Table 4 The estimated daily intakes of elements for the fish sample and comparison with recommended safety guidelines

Analytes	EDI (Estimated daily intake)				EWI (Estimated weekly intake)				PTWI ^c or PMTDI ^{d#} $\mu\text{g/kg BW/day}$
	<i>O. ruber</i>		<i>S. guttatus</i>		<i>O. ruber</i>		<i>S. guttatus</i>		
	$\mu\text{g/kg}^a$	$\mu\text{g/kg BW/day}^b$	$\mu\text{g/kg}^a$	$\mu\text{g/kg BW/day}^b$	$\mu\text{g/kg}^a$	$\mu\text{g/kg BW/day}^b$	$\mu\text{g/kg}^a$	$\mu\text{g/kg BW/day}^b$	
Toxic elements									
Ag	9.03	0.150	1.89	0.031	63.21	1.053	13.23	0.220	
AS	7.56	0.126	24.57	0.409	52.92	0.882	171.99	2.866	15
Cd	6.93	0.115	4.41	0.073	48.51	0.808	30.87	0.514	1
Sb	2.31	0.038	2.73	0.045	16.17	0.269	19.11	0.318	6*
Pb	3.99	0.066	8.61	0.143	27.93	0.465	60.27	1.004	3.6
Sn	0.84	0.014	4.83	0.080	5.88	0.098	33.81	0.563	220 [‡]
Essential elements									
Ca	5512.7	91.87	4981.8	83.03	38,588.9	643.2	34,872.8	581.2	
Cu	20.58	0.343	13.02	0.217	144.06	2.401	91.14	1.519	50–500;160 [†]
Fe	49.14	0.819	61.53	1.025	343.98	5.733	430.71	7.178	
Mg	3867.3	64.45	5575.5	92.92	27,071.5	451.19	39,028.5	650.47	
Mn	3.78	0.063	11.55	0.192	26.46	0.441	80.85	1.347	200 or 150 [‡]
Na	6921.81	115.36	5486.46	91.441	48,452.67	807.54	38,405.2	640.08	
P	17,797.9	296.63	28,133.3	468.9	124,585.4	2076.4	196,933	3282.2	
Zn	147.84	2.464	385.77	6.429	1034.88	17.248	2700.39	45.006	300–1000
Se	14.07	0.234	21.84	0.364	98.49	1.641	152.88	2.548	5 [†]
Nonessential									
Ni	7.14	0.119	7.35	0.122	49.98	0.833	51.45	0.857	4.3 [‡]
Sr	93.87	1.564	42.84	0.714	657.09	10.955	299.88	4.998	
Al	84.21	1.403	20.79	0.346	589.47	9.824	145.53	2.425	143

^a Estimated daily intake ($\mu\text{g/day}$)

^b Estimated daily intake ($\mu\text{g/day}/60$ kg body weight)

^c Provisional tolerable weekly intake ($\mu\text{g/week/kg}$ body weight)

^d Provisional maximum tolerable daily intake ($\mu\text{g/day/kg}$ body weight)

[#] The numerical values shown are the tolerable daily intake for a 60 kg person derived from PTWIs or PMTDIs recommended by JECFA (WHO 2006) unless mentioned otherwise

* TDI derived by WHO (WHO 2003)

[‡] Expert group on vitamins and minerals—guidance level (EVM 2003)

[†] Expert group on vitamins and minerals—safe upper level (EVM 2003)

The concentrations of Sb and Ag in the muscle and skin tissues of *S. guttatus* were not significantly different ($P>0.05$) compared to the muscle and skin tissues of *O. ruber*. Also, the concentrations of Cd and Pb in the muscle tissues of *S. guttatus* and *O. ruber* were not significantly different ($P>0.05$). There were no significant difference ($P>0.05$) between the concentrations of Ag, Sb, and As in the skin and muscle tissues of every examined fish while the muscle tissue of *S. guttatus* contained significantly higher concentrations ($P<0.05$) of As and Sn compared to the muscle tissue of *O. ruber*. Also, results showed that the muscle tissue of *S. guttatus* had significantly higher levels ($P<0.05$) of Sn compared to its skin tissue. In a research, Mendil et al. measured heavy metal contents in four commercially fish species from the Black Sea. The concentrations of all examined trace metals such as Fe, Zn, Pb, Cr, Mn, Cu, Cd, and Co depended on the fish species. The metals followed the trend $Fe > Zn > Mn > Cu > Cr > Pb > Co > Cd$ (Mendil et al. 2010). In another study, significant difference was observed between two examined fish species tissues in term of Cd concentration (Giannakopoulou and Neofitou 2014).

All examined heavy metals and toxic elements were found to be lower than the maximum level allowed in food that is recommended by WHO (WHO 1999). Similar acceptable concentrations of heavy metal contents in different fish species have been reported (Dobaradaran et al. 2010; Perugini et al. 2014; Saei-Dehkordi and Fallah 2011; Uysal et al. 2008). Though Mendil et al. have reported that the levels of lead and cadmium in the examined fish from the Black Seas were higher than the recommended limits while the concentrations of other examined metals were acceptable (Mendil et al. 2010). In another study, the concentrations of Ni, Cr, and Pb in the muscle of all examined fish species were higher than the WHO guideline values (Malik et al. 2014).

Dietary exposure assessment for the toxic elements and other elements through fish consumption was carried out by evaluating daily and weekly intakes. According to a report of the Fishery Statistics of Iran (Annual Fishery Statistics of Iran 2010), the fish consumption per day and week in Iran has been considered as 21 and 147 g, respectively. The estimated daily/weekly intakes (EDI/EWI) due to consumption of the muscle of *O. ruber* and *S. guttatus* for an adult with 60-kg body weight are demonstrated in Table 4 and have been compared with the guidelines. All calculated EDI and EWI values revealed that the dietary exposure to toxic elements due to the consumption of these fish could be considered safe. EDI/EWI values for arsenic were calculated due to the total arsenic not inorganic arsenic. It is well known that most arsenic in fish is organic arsenic which is the less toxic form of arsenic. It has been reported that the percentage of inorganic arsenic in fish is between 0.02 and 11 % (Muñoz et al. 2000) whereas the WHO set a 3000 µg for a subject of 60 kg as the maximum acceptable daily load for arsenic (FAO/WHO 1989). Results

indicated that EDI/EWI values of examined toxic elements in the present study could be considered safe in comparison to the guidelines limits, so there is no health threatening concern due to consumption of *S. guttatus* and *O. ruber* in Iran. In another study in Iran, the estimated daily and weekly intakes of zinc, lead, and copper via fish consumption flesh from the Iranian market were below the established values by WHO (Saei-Dehkordi and Fallah 2011).

Conclusions

The results of the present study provides important information on the toxic, macro, and trace element accumulation in the skin and muscles tissues of *O. ruber* and *S. guttatus* harvested commercially off the Persian Gulf. Concentrations of some elements (Ca, Li, Mg, P, Se, Sn, Sr, and Zn) were significantly different between the skin and muscle tissues of the *S. guttatus* but with the exception of P, there were no significant difference between the element concentrations in the skin and muscle tissues of *O. ruber*. *S. guttatus* contained significantly higher levels of As, Sn, Se, and P in the muscle tissue and Zn in the skin tissue compared to the muscle and skin tissues of *O. ruber*. The estimated daily and weekly intakes of toxic elements such as As, Cd, Sb, Pb, and Sn showed that the safety of dietary intakes of these elements via consumption of these two important commercial species should be considered acceptable. Finally, in view of the potential contamination by the oil activities near the fishing points, continuous and permanent monitoring in this region is highly recommended.

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References

- Alibabic V, Vahcic N, Bajramovic M (2007) Bioaccumulation of metals in fish of Salmonidae family and the impact on fish meat quality. *Environ Monit Assess* 131:349–364. doi:10.1007/s10661-006-9480-6
- Annual Fishery Statistics of Iran (2010) Consumption of fish in Iran. Ministry of Agriculture: pp 36–40.
- Brucka-Jastrzëbska E, Kawczuga D, Rajkowska M, Protasowicki M (2009) Levels of microelements [Cu, Zn, Fe] and macroelements [Mg, Ca] in freshwater fish. *J Elem* 14:437–447
- Canli M, Atli G (2003) The relationships between heavy metal (Cd, Cr, Cu, Fe, Pb, Zn) levels and the size of six Mediterranean fish species. *Environ Pollut* 121:129–136. doi:10.1016/S0269-7491(02)00194-X
- Carvalho ML, Santiago S, Nunes ML (2005) Assessment of the essential element and heavy metal content of edible fish muscle. *Anal Bioanal Chem* 382:426–432. doi:10.1007/s00216-004-3005-3
- Collette BB, Nauen CE (1983) Scombrids of the world: an annotated and illustrated catalogue of tunas, mackerels, bonitos and related species known to date. *FAO Species Catalog* 2

- Dobaradaran S, Naddafi K, Nazmara S, Ghaedi H (2010) Heavy metals (Cd, Cu, Ni and Pb) content in two fish species of Persian Gulf in Bushehr Port. *Iran Afr J Biotechnol* 9:6191–6193
- Dobaradaran S, Abadi DRV, Mahvi AH, Javid A (2011) Fluoride in skin and muscle of two commercial species of fish harvested off the Bushehr Shores of the Persian Gulf fluoride 44:143–146
- Dural M, Goksu MZL, Ozak AA (2007) Investigation of heavy metal levels in economically important fish species captured from the Tuzla lagoon. *Food Chem* 102:415–421. doi:10.1016/j.foodchem.2006.03.001
- EGVM (Expert Group on Vitamins and Minerals) (2003) Risk assessment: nickel. http://archive.food.gov.uk/dept_health/pdf/evmpdf/evm9924.pdf
- Fallah AA, Saei-Dehkordi SS, Nematollahi A, Jafari T (2011) Comparative study of heavy metal and trace element accumulation in edible tissues of farmed and wild rainbow trout (*Oncorhynchus mykiss*) using ICP-OES technique. *Microchem J* 98:275–279. doi:10.1016/j.microc.2011.02.007
- FAO/WHO (1989) Evaluation of certain food Additives and contaminants; technical report series 759. World Health Organization, Geneva
- Fernandes C, Fontainhas-Fernandes A, Cabral D, Salgado MA (2008) Heavy metals in water, sediment and tissues of *Liza saliens* from Esmoriz-Paramos Lagoon. *Port Environ Monit Assess* 136:267–275. doi:10.1007/s10661-007-9682-6
- Fischer W, Bianchi G (1984) FAO species identification sheets for fishery purposes. Western Indian Ocean (Fishing Area 51) 4
- Giannakopoulou L, Neofitou C (2014) Heavy metal concentrations in *Mullus barbatus* and *Pagellus erythrinus* in relation to body size, gender, and seasonality. *Environ Sci Pollut Res* 21:7140–7153
- Jarić I, Višnjić-Jeftić Ž, Cvijanović G, Gačić Z, Jovanović L, Skorić S, Lenhardt M (2011) Determination of differential heavy metal and trace element accumulation in liver, gills, intestine and muscle of sterlet (*Acipenser ruthenus*) from the Danube River in Serbia by ICP-OES. *Microchem J* 98:77–78
- Kagi JH, Schaffer A (1988) Biochemistry of metallothionein. *Biochem* 27:8509–8515
- Khan AH, Ali M, Biaswas SK, Hadi DA (1987) Trace-elements in marine fish from the Bay of Bengal. *Sci Total Environ* 61:121–130. doi:10.1016/0048-9697(87)90362-7
- Malik RN, Hashmi MZ, Huma Y (2014) Heavy metal accumulation in edible fish species from Rawal Lake Reservoir. *Pak Environ Sci Pollut Res* 21:1188–1196
- Mansour SA, Sidky MM (2002) Ecotoxicological studies. 3. Heavy metals contaminating water and fish from fayoum governorate. *Egypt Food Chem* 78:15–22. doi:10.1016/S0308-8146(01)00197-2
- Marin-Guirao L, Lloret J, Marin A (2008) Carbon and nitrogen stable isotopes and metal concentration in food webs from a mining-impacted coastal lagoon. *Sci Total Environ* 393:118–130. doi:10.1016/j.scitotenv.2007.12.023
- Mendil D, Uluözülü ÖD, Hasdemir E, Tüzen M, Sarı H, Suiçmez M (2005) Determination of trace metal levels in seven fish species in lakes in Tokat. *Turk Food Chem* 90:175–179. doi:10.1016/j.foodchem.2004.03.039
- Mendil D, Demirci Z, Tuzen M, Soylyak M (2010) Seasonal investigation of trace element contents in commercially valuable fish species from the Black sea. *Turk Food Chem Toxicol* 48:865–870. doi:10.1016/j.fct.2009.12.023
- Muñoz O et al (2000) Total and inorganic arsenic in fresh and processed fish products. *J Agric Food Chem* 48:4369–4376
- Olojo EAA, Olurin KB, Mbaka G, Oluwemimo AD (2005) Histopathology of the gill and liver tissues of the African catfish *Clarias gariepinus* exposed to lead. *Afr J Biotechnol* 4:117–122
- Perugini M, Visciano P, Manera M, Zaccaroni A, Olivieri V, Amorena M (2014) Heavy metal (As, Cd, Hg, Pb, Cu, Zn, Se) concentrations in muscle and bone of four commercial fish caught in the central Adriatic Sea. *Italy Environ Monit Assess* 186:2205–2213
- Pyle GG, Rajotte JW, Couture P (2005) Effects of industrial metals on wild fish populations along a metal contamination gradient. *Ecotox Environ Safe* 61:287–312. doi:10.1016/j.econenv.2004.09.003
- Saei-Dehkordi SS, Fallah AA (2011) Determination of copper, lead, cadmium and zinc content in commercially valuable fish species from the Persian Gulf using derivative potentiometric stripping analysis. *Microchem J* 98:156–162. doi:10.1016/j.microc.2011.01.001
- Santos LFP, Trigueiro INS, Lemos VA, Furtunato DMD, Cardoso RDV (2013) Assessment of cadmium and lead in commercially important seafood from Sao Francisco do Conde, Bahia, Brazil. *Brazil Food Control* 33:193–199. doi:10.1016/j.foodcont.2013.02.024
- Somers E (1974) Toxic potential of trace-metals in foods. *Rev J Food Sci* 39:215–217. doi:10.1111/j.1365-2621.1974.tb02860.x
- Tüzen M (2003) Determination of heavy metals in fish samples of the middle Black Sea (Turkey) by graphite furnace atomic absorption spectrometry. *Food Chem* 80:119–123
- Tuzen M (2009) Toxic and essential trace elemental contents in fish species from the Black Sea. *Turk Food Chem Toxicol* 47:1785–1790. doi:10.1016/j.fct.2009.04.029
- Tuzen M, Soylyak M (2007) Determination of trace metals in canned fish marketed in Turkey. *Food Chem* 101:1378–1382. doi:10.1016/j.foodchem.2006.03.044
- Tuzen M, Karaman I, Cıtak D, Soylyak M (2009) Mercury (II) and methyl mercury determinations in water and fish samples by using solid phase extraction and cold vapour atomic absorption spectrometry combination. *Food Chem Toxicol* 47:1648–1652. doi:10.1016/j.fct.2009.04.024
- Uluozlu OD, Tuzen M, Mendil D, Soylyak M (2007) Trace metal content in nine species of fish from the Black and Aegean Seas. *Turk Food Chem* 104:835–840. doi:10.1016/j.foodchem.2007.01.003
- Uysal K, Emre Y, Köse E (2008) The determination of heavy metal accumulation ratios in muscle, skin and gills of some migratory fish species by inductively coupled plasma-optical emission spectrometry (ICP-OES) in Beymelek Lagoon (Antalya/Turkey). *Microchem J* 90:67–70. doi:10.1016/j.microc.2008.03.005
- Verep B, Akin S, Mutlu C, Ertugral B, Apaydin G, Cevik U (2007) Assessment of trace elements in rainbow trout (*Oncorhynchus mykiss*) cultured in the marine aquaculture cages on the black sea coast. *Fresenius Environ Bull* 16:1005
- Visnjic-Jeftic Z, Jarić I, Jovanovic L, Skoric S, Smederevac-Lalic M, Nikcevic M, Lenhardt M (2010) Heavy metal and trace element accumulation in muscle, liver and gills of the Pontic shad (*Alosa immaculata* Bennet 1835) from the Danube River (Serbia). *Microchem J* 95:341–344. doi:10.1016/j.microc.2010.02.004
- WHO (1999) Food safety issues associated with product from aquaculture. WHO Technical Report Series 883
- WHO (2003) Chemical hazards in drinking water WHO Guidelines for Drinking-water Quality
- WHO (2006) Sixty-seventh meeting of the joint FAO/WHO Expert Committee on Food Additives; 20–29 June 2006, Rome. <http://www.who.int/ipcs/food/jecfa/summaries/summary67pdf>
- Yilmaz F, Ozdemir N, Demirak A, Tuna AL (2007) Heavy metal levels in two fish species *Leuciscus cephalus* and *Lepomis gibbosus*. *Food Chem* 100:830–835. doi:10.1016/j.foodchem.2005.09.020