



The Comparison of Heavy Metal Level in Surface Water, Sediment and Biota Sampled from the Polluted and Unpolluted Sites in the Northeastern Mediterranean Sea

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Received: 24 February 2020 / Revised: 12 June 2020 / Accepted: 20 July 2020 / Published online: 16 September 2020
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Abstract

The determination of heavy metal level (aluminium, iron, zinc, arsenic strontium) in surface water, sediment, and biota in Iskenderun Bay, polluted site, and Mersin Bay, unpolluted site, in 2018–2019 seasonally were aimed. The muscle and liver (viscera for shrimp) tissue of three fish (*Saurida lessepsianus*, *Nemipterus randalli*, *Mullus barbatus*) and an invertebrate (*Penaeus semisulcatus*) species, which have different habitats and diet, were used to detection of biota metal level. Sediment samples were analyzed in four different particle sizes which were < 63 , $63 \leq - < 125$, $125 \leq - < 250$, $250 \leq - < 500$ μm mesh size. The metal levels in surface water, sediment, and biota samples were analysed with Inductively Coupled Plasma-Mass Spectrometer (ICP-MS). Statistical analysis of the data was carried out using variance analysis and Duncan's procedure was applied. The relationships of the data were compared with Linear Regression Analysis. The heavy metal levels in the surface water, sediment, and biota sampled from the polluted and unpolluted sites in the Northeastern Mediterranean Sea were compared according to metal, species, tissue, season, particle size, and sampling site. Metal levels in seawater detected in polluted and unpolluted sites were $\text{Sr} > \text{Fe} > \text{As} > \text{Al} > \text{Zn}$ and $\text{Sr} > \text{Fe} > \text{As} > \text{Zn} > \text{Al}$ respectively. ($p < 0.05$). The results according to the Multiple Regression Analysis, Sr, Fe, Zn, and As level in seawater were shown strong positive relationships with sediment ($p < 0.0001$). A negative relationship was found between Sr in seawater and seasons ($p < 0.05$). The order of metals determined in sediment was found $\text{Fe} > \text{Al} > \text{Sr} > \text{Zn} > \text{As}$ in both sampling sites. The sediment Zn levels were a negative relationships between biota, particle size, sampling sites and season ($p < 0.001$). There was a strong positive relationship between sediment As level and biota ($p < 0.0001$). The sediment Sr level was shown the negative relationships with biota and season ($p < 0.05$). The metal levels determined in the muscle tissue sampled from polluted and unpolluted sites were $\text{Fe} > \text{As} > \text{Zn} > \text{Sr} > \text{Al}$ and $\text{As} \geq \text{Fe} > \text{Zn} > \text{Sr} > \text{Al}$, while the liver metal level sampled from polluted and unpolluted sites were $\text{Fe} > \text{Zn} > \text{As} > \text{Al} > \text{Sr}$ and $\text{Fe} > \text{Zn} > \text{As} > \text{Sr} > \text{Al}$, respectively. There were significant of metals between species and tissues ($p < 0.05$, 0.001, 0.0001). However, the significant relationships were not found between sampling sites and seasons ($p > 0.05$). Iron accumulated in the liver more than the muscle tissue while arsenic accumulated in the muscle more than the liver tissues due to the functional difference of tissues ($p < 0.05$). The tissue metal level was high abundant in the polluted site. Fe was at the highest level in biota and sediment from both regions may be due to industrial activities in the polluted site whereas agricultural activities or discharged by the Göksu River in the unpolluted site.

Keywords Metals · Seawater · Sediment · Biota · Mersin Bay · Iskenderun Bay

Introduction

The most important feature of heavy metals that differs from other pollutants are transported by physical and biological

routes without structural changes for a long time and are in motion. They are relocated by the hydrologic cycle to the aquatic environments and deposited. Heavy metals sink to the bottom after being suspended in the water column for a certain period (Çoğun et al. 2005). Therefore, results that are more reliably related to the ambient concentration of heavy metals are obtained by research on water, sediment, and the food chain.

Iskenderun and Mersin Bay are important spawning areas for many marine creatures in terms of their geographical

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structure and rich nutrient input thus have an important place in Northeastern Mediterranean fishery (Ayas and Kolonkaya 1996; Yilmaz 2003). An important part of the fishery that is offered for consumption from the Mediterranean coasts of Turkey is met from these gulfs where are under the direct influence of marine pollution caused by industrial and agricultural activities of densely populated cities (Kargin 1996; Kalay et al. 2008). Therefore, human beings need to determine the heavy metal levels in the seafood, which constitutes the healthy protein source and to monitor the current risks.

Previous researches have shown that some essential and toxic metals like Fe, Zn, Cu, Pb, Cd were investigated in various invertebrates and fish species, seawater and sediment sampled from Iskenderun and Mersin Bay (Ayas and Kolonkaya 1996; Ergin et al. 1998; Yilmaz 2003; Çoğun et al. 2005; Türkmen et al. 2005a, b; Dural and Bickici 2010; Ağca and Özdel 2014; Olgunoğlu et al. 2015; Duysak and Uğurlu 2017), however, the number of studies on As, Al, and Sr levels in biota sampled from the NE Mediterranean Sea were very limited (Ayas et al. 2018). Arsenic is a metalloid used as a raw material in various industries (Flora 2015). No biological functions of it in animals were known. It is more abundant in the earth's crust so it is important to determine the level of arsenic in both study areas. Aluminium is a toxic element for animals. It is more abundant in earth crust the fact that aluminum has a value of +3 makes it difficult for animals to be taken (Hydes and Liss 1977). Sr is an alkaline earth element found naturally in the earth's crust. Four different stable isotopes of Sr, especially ^{88}Sr accounts for 83% of natural strontium (Lide 1998). It is transported to the sea through rivers. It is similar to Ca. Its main uptake by aquatic organisms takes place through Ca channels and intestinal system. In nature, radioisotopes released by nuclear waste, particularly ^{90}Sr , cause Sr toxicity.

The determination of heavy metal levels in seawater, sediment, and biota in the polluted site (Iskenderun Gulf) and unpolluted site (Mersin Bay) where industrial fishing is important and to compare both regions' similarities and differences was aimed in the present study. For this purpose, heavy metal levels in muscle and liver tissue (viscera for shrimp) of *S. lessepsianus*, *N. randalli*, *M. barbatus* and *P. semisulcatus* were determined and compared with the results of seawater and sediment metal levels seasonally.

Material and Methods

In this study, Al, Fe, Zn, As, Sr levels were determined in biota, seawater, and sediment sampled from the polluted and unpolluted sites in the NE Mediterranean Sea by trawler seasonally during 2018–2019. The muscle and liver (viscera for shrimp) tissue of three fish (*S. lessepsianus*, *N. randalli*, *M. barbatus*) and an invertebrate (*P. semisulcatus*) species were

used. Sediment sampling was performed by grab from the bottom surface and analyzed in four different particle sizes which were < 63 , $63 \leq - < 125$, $125 \leq - < 250$, $250 \leq - < 500$ μm mesh size. Nansen bottle was used to water sampling from surface water. The sampling area was shown in Fig. 1. Coordinates: $36^{\circ}35'26.0''\text{N}36^{\circ}09'00.5''\text{E}$ (polluted site), $36^{\circ}09'27.1''\text{N}33^{\circ}41'23.5''\text{E}$ (unpolluted site).

Ten individuals were used in each species to determine metal analysis in every season. Analyzes were carried out in triplicates. Samples caught from sampling sites were transported to the laboratory in an icebox. After determining the height and weight of individuals (Table 1), the muscle and liver (viscera for shrimp) tissues were dissected and stored in an Eppendorf tube at -18 °C. At the end of the experimental period, all tissues were dried at 95 °C for 72 h. Dried tissues were weighted and digested in nitric acid (HNO_3 , 65%, Merck) at 120 °C for 4 h. Then digested tissues were transferred to falcon tube and their volumes were made up to 10 ml bidistilled water.

Water samples collected from the sampling sites; 2 ml of HNO_3 was added to 1 L of seawater and stored at -18 °C. Water samples were enriched by 30% volatilization on a hot plate before analysis, then transferred to 9 ml sample polyethylene tubes and 1 ml HNO_3 was added and analyzed by Inductively Coupled Plasma-Mass Spectrometer (ICP-MS).

Sediment samples were frozen in plastic lid sample containers at -18 °C and then thawed samples were dried in Pasteur Oven at 50 °C for 36 h. Dry sediment samples were passed through 63, 125, 250 and 500 μm sieves and 0.1 g samples from each group were burned in 50 ml Teflon beakers by adding 4 ml hydrogen fluoride (HF, 48%, Merck), 2 ml hydrochloric acid (HCL, 37%, Merck) and 2 ml HNO_3 . After the HF was completely evaporated, the sample was transferred to polyethylene tubes, added with 5 ml of bidistilled water, and analyzed by ICP-MS.

Inductively coupled plasma mass spectrometer (ICP-MS, Agilent, 7500ce Model, Japan) was used to determine metals. ICP-MS operating conditions were the following: radio frequency (RF) (W), 1500; plasma gas flow rate (L min^{-1}), 15; auxiliary gas flow rate (L min^{-1}), 1; carrier gas flow rate (L min^{-1}), 1.1; spray chamber T (°C), 2; sample depth (mm), 8.6; sample introduction flow rate (ml min^{-1}), 1; nebuliser pump (rps), 0.1; extract lens (V), 1.5. The levels of Al, Fe, Zn, As, Sr in samples detected as $\mu\text{g metal g}^{-1}$ dry weight. High Purity Multi-Standard (Charleston, SC 29423) was used for the determination of the metal analyses. Standard solutions for calibration curves prepared by dilutions of the trace elements and potentially toxic metals. Solutions have prepared for the toxic metals had a content of Al and Sr in the range of 1–50 ppb (0.001 to 0.050 mg l^{-1}), for the other elements had a content of Fe, Zn, and As in the range of 1–50 ppm (1 to 50 mg l^{-1}). International Atomic Energy Agency (IAEA-436) reference material was used to follow the quality



Fig. 1 The coordinates of the sampling sites (Site 1: unpolluted, Site 2: polluted)

of the analytical process. IAEA-436 was analyzed for all elements. The certified value and observed value of the IAEA-436 reference material were compared. Replicate analysis of this reference material showed good accuracy (Table 2).

Statistical analyses of data were performed using IBM 22 SPSS package program. The metal levels in biota, seawater, and sediment were carried out using variance analysis (ANOVA) and Duncan’s Multiple Range tests were used to compare the distinction between each group. The Linear Regression Analysis was used to compare the relationships between metal levels in seawater, sediment, and biota to species, tissues, sampling sites and seasons. The results were assumed as statistically significant at $p < 0.0001, 0.001, 0.05$.

Table 1 The values of total length and total weight of species

Species	Total length (cm)	Total weight (g)
<i>M. barbatus</i>	11.5–17.5	15.34–56.01
<i>S. lessepsianus</i>	18.5–29.0	65.21–179.64
<i>N. randalli</i>	12.5–19.5	22.76–88.56
<i>P. semisulcatus</i>	12.5–20.5	19.98–57.89

Results

Aluminium, iron, zinc, arsenic, and strontium were determined in seawater, sediment, and biota in polluted and unpolluted sites from NE Mediterranean Sea in 2018–2019 seasonally ($p < 0.05$).

Table 2 The certificated value and observed value of reference material IAEA-436

Analyte	Certified value	95% Confidence interval	Observed value
Al	3.06 ± 0.42	2.68–3.44	3.112 ± 0.132
As	1.98 ± 0.17	1.91–2.06	2.001 ± 0.022
Co	0.042 ± 0.006	0.039–0.045	0.041 ± 0.002
Cr	0.194 ± 0.058	0.168–0.219	0.211 ± 0.009
Cu	1.73 ± 0.19	1.66–1.79	1.731 ± 0.044
Fe	89.3 ± 4.2	87.8–90.9	90.001 ± 0.189
Mn	0.238 ± 0.042	0.218–0.257	0.244 ± 0.007
Ni	0.069 ± 0.041	0.040–0.099	0.065 ± 0.011
Sr	0.564 ± 0.062	0.523–0.606	0.533 ± 0.019
Zn	19.0 ± 1.3	18.6–19.4	18.876 ± 0.066

Table 3 Metal levels ($\mu\text{g l}^{-1}$) in seawater sampled from both sites

Polluted site	Al	Fe	Zn	As	Sr
	$\bar{X} \pm S_x$	$\bar{X} \pm S_x$	$\bar{X} \pm S_x$	$\bar{X} \pm S_x$	$\bar{X} \pm S_x$
Autumn	0.04 ± 0.000^a	0.11 ± 0.009^a	0.01 ± 0.000^a	0.05 ± 0.007^a	15.53 ± 0.44^a
Winter	0.09 ± 0.013^b	0.28 ± 0.059^b	0.01 ± 0.000^a	0.08 ± 0.004^b	9.55 ± 0.307^b
Spring	0.08 ± 0.006^b	0.21 ± 0.006^{ab}	0.05 ± 0.002^b	0.07 ± 0.002^b	4.04 ± 0.096^c
Summer	0.09 ± 0.013^b	1.26 ± 0.54^c	0.01 ± 0.001^a	0.30 ± 0.003^c	3.20 ± 0.225^c
Unpolluted site					
Autumn	0.03 ± 0.005^a	0.03 ± 0.004^a	0.19 ± 0.005^a	0.04 ± 0.003^a	13.83 ± 0.355^a
Winter	0.02 ± 0.001^b	0.10 ± 0.012^a	0.21 ± 0.022^a	0.09 ± 0.010^{ab}	12.47 ± 0.861^{ab}
Spring	0.04 ± 0.01^a	0.20 ± 0.019^a	0.00 ± 0.00^b	0.14 ± 0.012^b	11.40 ± 0.215^b
Summer	0.08 ± 0.01^c	0.60 ± 0.122^b	0.01 ± 0.001^b	0.30 ± 0.048^c	6.21 ± 0.518^c

*Duncan; the letters a, b and c indicate the statistical differences between metal and season. Statistical difference was $p < 0.05$

$\bar{X} \pm S_x$: mean \pm Standard error

The order of metal detected in seawater was shown, $\text{Sr} > \text{Fe} > \text{As} > \text{Al} > \text{Zn}$ in polluted and $\text{Sr} > \text{Fe} > \text{As} > \text{Zn} > \text{Al}$ in unpolluted sites, that strontium had the highest concentration ($p < 0.05$) (Table 3). It was found that strong positive relationships between Sr, Fe, Zn, and As level in seawater and sediment ($p < 0.0001$). A negative relationship was found between Sr in seawater and seasons ($p < 0.05$) (Table 4). No significant relationships were detected between metals in seawater with sampling sites and biota ($p > 0.05$).

Sediment samples were separated into four particle sizes as < 63 , $63 \leq - < 125$, $125 \leq - < 250$, $250 \leq - < 500$ μm before metal analyzes. Zn concentration in sediment decreased due to an increase in particle size ($p < 0.05$) (Table 5). The order of metals determined in sediment was found $\text{Fe} > \text{Al} > \text{Sr} > \text{Zn} > \text{As}$ in two sampling sites. The sediment Zn level were negative relationships between biota, particle size, sampling sites and

season ($p < 0.001$). There was a strong positive relationship between sediment As level and biota ($p < 0.0001$). The sediment Sr level was shown the negative relationships with biota and season ($p < 0.05$). However, sediment Fe and Al levels were not significant between biota, particle size, sampling sites and season ($p > 0.05$) (Table 6). Zn was shown positive strong relationships between seawater and sediment ($p < 0.0001$). Sediment Zn level was shown negative relationships between biota ($p < 0.05$), season ($p < 0.001$) and sampling sites ($p < 0.001$).

It was found that the metal levels determined in the muscle tissue sampled from polluted and unpolluted sites were $\text{Fe} > \text{As} > \text{Zn} > \text{Sr} > \text{Al}$ and $\text{As} \geq \text{Fe} > \text{Zn} > \text{Sr} > \text{Al}$, while the liver metal level sampled from polluted and unpolluted sites were $\text{Fe} > \text{Zn} > \text{As} > \text{Al} > \text{Sr}$ and $\text{Fe} > \text{Zn} > \text{As} > \text{Sr} > \text{Al}$, respectively (Tables 7 and 8). Fe was the highest level in biota except to the

Table 4 Factors predicting metal accumulation in seawater

Seawater	<i>F</i>	df	β	<i>t</i>	<i>R</i> ²
Sr	7.819**	3			0.662
sediment			0.687	4.090***	
seasons			-0.398	-2.369*	
Sampling sites and biota			NS ^a	NS ^a	
Fe	14.606	3			0.785
sediment			0.841	6.280***	
Seasons, sampling sites and biota			NS ^a	NS ^a	
As	71.385***	3			0.934
sediment			0.967	14.547***	
Seasons, sampling sites and biota			NS ^a	NS ^a	
Zn	112.911***	3			0.966
sediment			0.979	18.340***	
Seasons, sampling sites and biota			NS ^a	NS ^a	

* $p < 0.05$, ** $p < 0.001$, *** $p < 0.0001$, NS not significant, ^a $p > 0.05$

Table 5 Metal levels ($\mu\text{g g}^{-1}$ dw) in sediment sampled from both sites

Polluted site					
Autumn	Al	Fe	Zn	As	Sr
	$\bar{X} \pm S_x$	$\bar{X} \pm S_x$	$\bar{X} \pm S_x$	$\bar{X} \pm S_x$	$\bar{X} \pm S_x$
<63	3872.7 ± 47.69 ^a	19,313.0 ± 557.32 ^a	69.0860 ± 0.16 ^a	8.1056 ± 0.32 ^a	405.48 ± 3.47 ^a
63 ≤ - < 125	2167.0 ± 329.60 ^b	30,661.0 ± 4064.87 ^a	57.1991 ± 0.87 ^b	14.4369 ± 0.26 ^b	37.6010 ± 0.24 ^b
125 ≤ - < 250	3317.4 ± 68.60 ^a	28,400.0 ± 1155.39 ^a	52.3744 ± 1.30 ^c	14.8738 ± 1.10 ^b	44.7222 ± 0.07 ^b
250 ≤ - < 500	5855.8 ± 305.80 ^c	24,199.0 ± 2602.08 ^a	52.9122 ± 0.001 ^c	10.7927 ± 0.84 ^c	35.6942 ± 4.91 ^b
Winter					
<63	4034.3 ± 52.27 ^a	51,951.0 ± 1683.31 ^a	139.80 ± 2.20 ^a	20.8134 ± 0.48 ^a	160.75 ± 1.75 ^a
63 ≤ - < 125	1063.6 ± 142.85 ^b	16,340.0 ± 402.07 ^b	36.5555 ± 1.96 ^b	9.7016 ± 1.31 ^b	39.4287 ± 2.24 ^b
125 ≤ - < 250	4841.4 ± 114.49 ^c	15,655.0 ± 1105.07 ^b	34.1861 ± 0.64 ^{bc}	15.3544 ± 0.09 ^c	68.1764 ± 2.16 ^c
250 ≤ - < 500	2391.9 ± 224.45 ^d	10,867.0 ± 354.96 ^c	30.2901 ± 0.31 ^c	10.8136 ± 0.34 ^b	41.6713 ± 1.29 ^b
Spring					
<63	5546.2 ± 500.5 ^a	14,012.0 ± 878.49 ^a	70.6588 ± 0.22 ^a	9.7910 ± 0.77 ^a	100.39 ± 7.92 ^a
63 ≤ - < 125	554.60 ± 45.58 ^b	8332.2 ± 245.5 ^b	34.3644 ± 0.66 ^b	11.7224 ± 0.08 ^b	23.6210 ± 1.35 ^b
125 ≤ - < 250	2419.5 ± 85.2 ^c	8829.9 ± 77.16 ^{bc}	32.0252 ± 0.13 ^b	16.4181 ± 0.24 ^c	36.7932 ± 2.59 ^b
250 ≤ - < 500	3261.0 ± 124.68 ^c	10,248.0 ± 264.20 ^c	30.2053 ± 1.05 ^c	11.8293 ± 0.40 ^b	53.9618 ± 0.06 ^c
Summer					
<63	5038.9 ± 273.97 ^a	16,709.0 ± 1902.98 ^a	80.7728 ± 1.21 ^a	10.2542 ± 0.63 ^a	106.52 ± 15.97 ^a
63 ≤ - < 125	635.16 ± 50.69 ^b	10,492.0 ± 21.54 ^b	36.4131 ± 1.22 ^b	7.2523 ± 0.03 ^b	28.4040 ± 1.43 ^b
125 ≤ - < 250	2951.6 ± 134.02 ^c	8439.0 ± 272.23 ^b	34.1572 ± 0.61 ^b	17.3207 ± 0.20 ^c	33.9570 ± 0.69 ^b
250 ≤ - < 500	2203.8 ± 13.55 ^d	9483.3 ± 52.22 ^b	35.4936 ± 0.73 ^b	14.8873 ± 0.03 ^d	42.9228 ± 1.16 ^b
Unpolluted site					
Autumn					
<63	7536.4 ± 183.55 ^a	40,701.0 ± 3981.81 ^a	74.0162 ± 15.61 ^a	15.0955 ± 1.74 ^a	467.50 ± 42.78 ^a
63 ≤ - < 125	3298.3 ± 97.21 ^b	4569.2 ± 355.87 ^b	29.2191 ± 2.91 ^b	12.2717 ± 0.83 ^{ab}	220.60 ± 27.83 ^b
125 ≤ - < 250	3296.1 ± 83.77 ^b	3747.6 ± 21.65 ^b	33.2646 ± 0.83 ^b	10.3666 ± 1.04 ^b	198.33 ± 25.25 ^b
250 ≤ - < 500	3047.6 ± 22.04 ^b	2853.4 ± 450.29 ^b	37.2157 ± 1.94 ^b	9.7580 ± 0.78 ^b	160.18 ± 20.09 ^b
Winter					
<63	361.08 ± 28.95 ^a	18,644.0 ± 777.41 ^a	97.0754 ± 0.56 ^a	10.6292 ± 0.92 ^a	51.7583 ± 7.57 ^a
63 ≤ - < 125	456.14 ± 43.47 ^a	2707.6 ± 894.75 ^b	26.5060 ± 3.72 ^b	10.2397 ± 0.47 ^{ab}	97.6336 ± 6.69 ^b
125 ≤ - < 250	230.29 ± 3.43 ^b	748.52 ± 77.64 ^b	16.1470 ± 0.71 ^c	9.0052 ± 0.04 ^b	99.7877 ± 3.46 ^b
250 ≤ - < 500	237.86 ± 17.43 ^b	1479.6 ± 247.78 ^b	19.2441 ± 0.78 ^c	9.1726 ± 0.05 ^b	35.3747 ± 4.09 ^a
Spring					
<63	25,105.0 ± 420.41 ^a	23,998.0 ± 403.6 ^a	90.5760 ± 0.42 ^a	23.6292 ± 0.37 ^a	407.28 ± 4.27 ^a
63 ≤ - < 125	7839.4 ± 252.01 ^b	16,898.0 ± 260.5 ^b	35.0964 ± 1.07 ^b	21.3562 ± 0.94 ^a	32.6146 ± 0.58 ^b
125 ≤ - < 250	12,101.0 ± 1777.6 ^c	10,158.0 ± 935.40 ^c	44.6890 ± 0.34 ^c	13.8369 ± 1.05 ^b	29.7454 ± 2.17 ^b
250 ≤ - < 500	7441.6 ± 276.68 ^b	8496.5 ± 546.07 ^c	33.2556 ± 0.71 ^b	11.0568 ± 0.20 ^c	19.6653 ± 1.28 ^c
Summer					
<63	27,151.0 ± 1288.6 ^a	16,341.0 ± 42.01 ^a	61.3805 ± 0.53 ^a	19.4772 ± 1.12 ^a	56.8275 ± 2.02 ^a
63 ≤ - < 125	8678.4 ± 294.44 ^b	8578.5 ± 71.82 ^b	29.2765 ± 0.40 ^b	16.1865 ± 0.36 ^b	34.7856 ± 4.12 ^b
125 ≤ - < 250	13,901.0 ± 260.51 ^c	11,785.0 ± 157.67 ^c	41.4047 ± 1.18 ^c	16.3435 ± 0.74 ^b	27.9142 ± 1.30 ^b
250 ≤ - < 500	18,932.0 ± 11.98 ^d	17,306.0 ± 380.16 ^d	46.3187 ± 1.08 ^d	15.2886 ± 0.81 ^b	28.7992 ± 0.94 ^b

*Duncan; the letters a, b, c and d indicate the statistical differences between metal and particle size. Statistical difference was $p < 0.05$

$\bar{X} \pm Sx$: mean ± Standard error

muscle tissues sampled from unpolluted sites. Arsenic was more accumulate in muscle than liver tissue ($p < 0.05$).

Results of the Multiple Regression Analysis, there were significant relationships of metals between species and tissues,

Table 6 Factors predicting metal accumulation in sediment

Sediment	<i>F</i>	df	β	<i>t</i>	R ²
Biota	9.508***	7			0.486
Zn			−0.304	−2.792*	
As			0.383	3.989***	
Sr			−0.233	−2.042*	
Fe, Al			NS ^a	NS ^a	
Particle size	5.357**	7			0.413
Zn			−0.574	−3.442**	
Fe, Al, As, Sr			NS ^a	NS ^a	
Season	4.649**	6			0.414
Sr			−0.340	−2.004*	
Zn			−0.576	−3.459**	
Fe, Al, As			NS ^a	NS ^a	
Sampling sites	4.292**	6			0.389
Zn			−0.574	−3.282**	
Fe, Al, As, Sr			NS ^a	NS ^a	

* $p < 0.05$, ** $p < 0.001$, *** $p < 0.0001$, NS not significant, ^a $p > 0.05$

however, not significant between sampling sites and seasons were found (Table 9). In generally, the muscle arsenic level was found to be higher in *M. barbatus* sampled from the polluted site while it was found high in the other fish and invertebrate sampled from the unpolluted site. The level of metals examined in shrimp sampled from the unpolluted site was higher than the polluted site ($p < 0.05$) (Tables 7 and 8). The muscle Fe level was determined at high levels in *M. barbatus* and *N. randalli* sampled from the polluted site while it was found to be high in *S. lessepsianus* sampled from the unpolluted site (Table 10) ($p < 0.05$). Zn was found positive relationship with species ($p < 0.05$) and tissues ($p < 0.0001$).

Discussion

Heavy metals are bioaccumulative at the increasing amounts and cause toxic effects on a given concentration in biological systems. It is important for the environment and human health so environmental monitoring activities should be carried out continuously to reflect the current state of pollution. Numerous studies on metal levels in various invertebrates and fish species, seawater and sediment sampled from Iskenderun and Mersin Bay (Ayas and Kolonkaya 1996; Ergin et al. 1998; Yilmaz 2003; Çoğun et al. 2005; Türkmen et al. 2005a, b; Dural and Bickici 2010; Ağca and Özdel 2014; Olgunoğlu et al. 2015; Duysak and Uğurlu 2017; Ayas et al. 2018). However, the number of studies on As, Al, and Sr levels in biota sampled from the NE Mediterranean Sea were very limited.

In the present study, As was found in high concentration in biota in addition to Fe. Iron is a trace element that participates in the structural composition of hemoglobin and has a biological function in animals. The liver Fe concentration was higher than muscle concentration may be because the liver is metabolically active tissue. Fe concentration in all tissues was higher in the polluted site than in the unpolluted site (Tables 7 and 8). This may be due to the impact of industrial waste in the contaminated area. These were the expected results. Our findings were compared with metal levels of some fish species sampled from the Mediterranean Sea (Table 10). It was reported that Fe had the highest concentration in six different ray species sampled from Iskenderun Bay, followed by Zn. It was stated that the highest metal levels were found in the intestine and liver tissue than the muscle tissue (Türkmen et al. 2013). In the present study, Fe was more accumulated in the liver of all fish and shrimp, although, the highest muscle level was found in *M. barbatus* and *N. randalli* sampled from the polluted site. The muscle Fe level was detected at a high level in *S. lessepsianus* sampled from the unpolluted site (Tables 7, 8, 10). In a similar study, it was reported that Fe accumulation was high in different fish species sampled from Paradeniz lagoon (Türkmen et al. 2011). This finding may be related to the feeding habits of the species studied.

In this study, arsenic was found the highest level in muscle tissues in the unpolluted site while second high level after Fe in the polluted site. However, it is still unknown whether arsenic has any biological function in animals such as Fe. Arsenic is a metalloid of earth origin; it is known to exist in different chemical forms in nature and to form organoarsenic by forming complexes with organic substances. This may be the reason why arsenic muscle concentration was higher than liver tissue concentration in the present study. In a similar study conducted by Storelli et al. (2005) the muscle arsenic levels (2.61 and 3.68 $\mu\text{g g}^{-1}$ ww respectively) in *Thunnus thynnus* and *Xiphias gladius* sampled from the Mediterranean Sea were found to be lower than the liver levels (7.07 and 6.23 $\mu\text{g g}^{-1}$ ww). The arsenic levels were lower than those found in this study.

Arsenic accumulation varies depending on the feeding preference of the species. Arsenic enters the aquatic ecosystem, is taken up by the primary producers and is released into the ecosystem after it has been transformed into harmless forms at the end of the methylation process. This may be the reason for high concentrations in the lower trophic levels of the food chain. Therefore, it is also present in high concentrations in primary consumers. In this study, the arsenic level in *M. barbatus* and *N. randalli* fed with benthic invertebrates was found to be higher than *S. lessepsianus* fed with fish can support this information. It was reported that muscle arsenic level (69.44 $\mu\text{g g}^{-1}$ dw) in *N. randalli* sampled from the NE Mediterranean, was higher than *S. lessepsianus* (25.58 $\mu\text{g g}^{-1}$ dw) (Karaytuğ

Table 7 Metal levels ($\mu\text{g g}^{-1}$ dw) in biota sampled from the polluted site (M: muscle, L: Liver, V: Viscera)

Autumn		Al	Fe	Zn	As	Sr
		$\bar{X} \pm S_x$	$\bar{X} \pm S_x$	$\bar{X} \pm S_x$	$\bar{X} \pm S_x$	$\bar{X} \pm S_x$
<i>S. lessepsianus</i>	(M)	2.5574 ± 0.41 ^a	26.4800 ± 2.22 ^a	17.3838 ± 0.87 ^a	19.4982 ± 1.89 ^{ab}	3.5462 ± 0.66 ^{ab}
	(L)	6.0642 ± 0.83 ^a	357.18 ± 6.84 ^b	80.6457 ± 2.11 ^b	11.8480 ± 0.65 ^c	7.8779 ± 0.67 ^c
<i>N. randalli</i>	(M)	2.2450 ± 0.24 ^a	49.6451 ± 8.31 ^a	16.9999 ± 0.89 ^a	10.8533 ± 0.79 ^c	1.4575 ± 0.08 ^a
	(L)	22.0283 ± 2.68 ^b	1717.3 ± 69.92 ^c	146.66 ± 7.03 ^c	26.0304 ± 1.76 ^{bd}	11.1352 ± 0.55 ^d
<i>M. barbatus</i>	(M)	5.2036 ± 0.95 ^a	271.11 ± 7.76 ^d	17.5931 ± 0.71 ^a	118.86 ± 4.95 ^e	3.0434 ± 0.24 ^{ab}
	(L)	3.0607 ± 0.44 ^a	435.66 ± 5.16 ^b	48.6473 ± 1.82 ^d	30.6562 ± 2.32 ^d	4.1154 ± 0.48 ^b
<i>P. semiculcatus</i>	(M)	2.3651 ± 0.28 ^a	47.7555 ± 2.61 ^a	65.3001 ± 0.44 ^c	24.8998 ± 0.25 ^{bd}	11.9254 ± 1.15 ^d
	(V)	32.7294 ± 3.11 ^c	252.80 ± 3.16 ^d	184.15 ± 12.28 ^f	16.0032 ± 1.09 ^{ac}	36.2115 ± 0.46 ^c
Winter						
<i>S. lessepsianus</i>	(M)	4.2758 ± 0.18 ^{ab}	21.9547 ± 2.08 ^a	17.1645 ± 0.61 ^a	52.2628 ± 2.13 ^a	5.1724 ± 0.67 ^{ab}
	(L)	7.6659 ± 1.88 ^{ab}	443.42 ± 13.38 ^b	78.7909 ± 6.14 ^b	30.2698 ± 1.78 ^b	28.3815 ± 3.02 ^c
<i>N. randalli</i>	(M)	1.9328 ± 0.44 ^a	122.22 ± 6.26 ^c	15.1510 ± 0.24 ^a	18.5057 ± 0.12 ^c	1.4406 ± 0.13 ^a
	(L)	8.8511 ± 0.70 ^{ab}	371.64 ± 1.17 ^d	107.34 ± 6.42 ^c	15.8888 ± 1.12 ^c	8.9851 ± 0.04 ^b
<i>M. barbatus</i>	(M)	4.5405 ± 0.74 ^{ab}	190.21 ± 5.18 ^c	15.3712 ± 0.62 ^a	152.00 ± 5.74 ^d	2.1374 ± 0.61 ^a
	(L)	18.2338 ± 8.36 ^b	250.36 ± 0.48 ^f	94.4621 ± 2.16 ^d	72.1184 ± 1.84 ^e	5.0484 ± 0.07 ^{ab}
<i>P. semiculcatus</i>	(M)	6.2139 ± 1.23 ^{ab}	61.2304 ± 3.31 ^e	48.1944 ± 1.58 ^c	30.3574 ± 1.40 ^b	7.6667 ± 1.30 ^b
	(V)	109.45 ± 4.27 ^c	274.43 ± 19.22 ^f	616.67 ± 13.07 ^f	33.7904 ± 1.73 ^b	25.8728 ± 2.43 ^c
Spring						
<i>S. lessepsianus</i>	(M)	4.8020 ± 0.37 ^a	196.02 ± 3.39 ^a	11.1194 ± 0.75 ^a	11.5456 ± 0.93 ^{ab}	3.4293 ± 0.38 ^a
	(L)	16.7275 ± 2.68 ^b	533.46 ± 9.80 ^b	150.01 ± 16.41 ^b	6.3957 ± 0.63 ^a	3.6344 ± 0.25 ^a
<i>N. randalli</i>	(M)	8.3362 ± 0.76 ^c	240.26 ± 14.70 ^c	10.3670 ± 0.53 ^a	67.63 ± 0.47 ^c	16.1592 ± 1.74 ^b
	(L)	8.5600 ± 0.16 ^c	548.07 ± 13.70 ^b	187.23 ± 14.51 ^c	45.9386 ± 2.21 ^d	14.0552 ± 0.88 ^b
<i>M. barbatus</i>	(M)	16.6078 ± 0.19 ^b	62.9188 ± 7.011 ^d	10.6218 ± 0.13 ^a	208.43 ± 3.88 ^e	3.5059 ± 0.34 ^a
	(L)	69.2500 ± 0.75 ^d	129.25 ± 2.75 ^c	6.3966 ± 1.18 ^a	18.4967 ± 1.50 ^b	18.8966 ± 0.10 ^c
<i>P. semiculcatus</i>	(M)	2.0554 ± 0.30 ^a	71.0540 ± 1.05 ^d	43.7847 ± 1.65 ^d	16.7192 ± 2.82 ^b	22.0442 ± 0.37 ^d
	(V)	7.9883 ± 0.33 ^c	64.0670 ± 2.07 ^d	138.74 ± 6.26 ^b	40.1460 ± 1.85 ^d	91.0239 ± 1.02 ^c
Summer						
<i>S. lessepsianus</i>	(M)	4.7675 ± 0.79 ^a	2.5014 ± 0.50 ^a	11.5399 ± 1.15 ^a	34.3480 ± 2.44 ^a	2.5108 ± 0.89 ^a
	(L)	6.7729 ± 0.69 ^a	365.63 ± 20.79 ^b	183.34 ± 17.41 ^b	34.3081 ± 4.14 ^a	10.5844 ± 0.34 ^c
<i>N. randalli</i>	(M)	3.7688 ± 0.44 ^a	11.5665 ± 0.43 ^{ac}	11.8601 ± 0.79 ^a	37.3768 ± 2.70 ^a	2.9008 ± 0.22 ^a
	(L)	4.9182 ± 1.28 ^a	442.51 ± 9.03 ^d	136.71 ± 3.72 ^c	60.3584 ± 6.37 ^b	3.0629 ± 0.92 ^a
<i>M. barbatus</i>	(M)	6.3405 ± 0.73 ^a	34.1378 ± 0.42 ^c	12.7200 ± 0.83 ^a	13.2382 ± 1.37 ^c	5.8249 ± 0.49 ^b
	(L)	12.446 ± 3.45 ^a	804.17 ± 11.80 ^c	108.14 ± 7.77 ^d	44.5834 ± 1.42 ^a	9.4108 ± 1.26 ^c
<i>P. semiculcatus</i>	(M)	6.2970 ± 1.43 ^a	16.9072 ± 0.87 ^{ac}	42.4571 ± 1.04 ^c	90.9980 ± 9.49 ^d	10.4702 ± 0.04 ^c
	(V)	192.15 ± 63.84 ^b	411.17 ± 1.038 ^f	101.90 ± 3.90 ^d	58.9808 ± 0.49 ^b	84.0631 ± 1.30 ^d

*Duncan; the letters a, b, c, d, e, f and g indicate the statistical differences between metal and species. Statistical difference was $p < 0.05$

$\bar{X} \pm S_x$: mean ± Standard error

et al. 2018). The muscle arsenic concentration of *P. semisulcatus* (29.254 mg kg⁻¹ ww) sampled from Iskenderun Bay was reported to be higher than fish tissue (*Solea solea* 4.397 mg kg⁻¹ ww; *Merlangius merlangus* 10.320 mg kg⁻¹ ww; *Sillago sihama* 1.741 mg kg⁻¹ ww) (Kaya and Turkoglu 2017). The arsenic levels in muscle tissues in previous studies were lower than those found in this study.

Metal accumulation in aquatic organisms varies depending on species, tissue, ambient concentration, sampling region and season. Al level in viscera of *P. semisulcatus* increased in summer ($p < 0.05$) (Tables 7 and 8). The positive relationships between Al level in biota was shown in Table 9 ($p < 0.001$). Al has a very high concentration in the earth's crust is known (Brown et al. 2010). It is economically important because

Table 8 Metal levels ($\mu\text{g g}^{-1}$ dw) in biota sampled from the unpolluted site (M: muscle, L: Liver, V: Viscera)

Autumn		Al	Fe	Zn	As	Sr
		$\bar{X} \pm S_x$	$\bar{X} \pm S_x$	$\bar{X} \pm S_x$	$\bar{X} \pm S_x$	$\bar{X} \pm S_x$
<i>S. lessepsianus</i>	(M)	3.9500 \pm 0.82 ^{ab}	112.96 \pm 2.61 ^{ab}	14.0619 \pm 0.82 ^a	24.0721 \pm 0.83 ^{ab}	3.1791 \pm 0.32 ^{ab}
	(L)	6.8436 \pm 2.35 ^b	521.01 \pm 2.94 ^c	52.6998 \pm 1.61 ^b	22.2582 \pm 1.84 ^a	11.7801 \pm 0.22 ^c
<i>N. randalli</i>	(M)	4.3370 \pm 0.69 ^{ab}	39.081 \pm 3.63 ^d	13.7428 \pm 0.35 ^a	41.0841 \pm 2.21 ^c	19.2872 \pm 1.99 ^d
	(L)	15.9298 \pm 3.22 ^c	359.96 \pm 37.92 ^c	86.2372 \pm 1.20 ^c	63.3340 \pm 2.49 ^d	5.7441 \pm 0.01 ^b
<i>M. barbatus</i>	(M)	2.0909 \pm 0.23 ^a	35.3122 \pm 0.40 ^d	12.6791 \pm 1.10 ^a	61.0727 \pm 1.90 ^d	1.6002 \pm 0.22 ^a
	(L)	13.9811 \pm 2.44 ^c	122.97 \pm 1.87 ^b	74.9354 \pm 0.80 ^d	38.9888 \pm 5.24 ^b	3.9600 \pm 0.16 ^{ab}
<i>P. semiculcatus</i>	(M)	3.4396 \pm 0.58 ^{ab}	73.2766 \pm 0.19 ^d	68.5894 \pm 2.52 ^e	117.45 \pm 10.90 ^e	11.1278 \pm 0.14 ^c
	(V)	40.8516 \pm 0.38 ^d	374.08 \pm 1.96 ^e	204.74 \pm 1.70 ^f	53.6843 \pm 0.22 ^{cd}	38.5723 \pm 3.16 ^d
Winter						
<i>S. lessepsianus</i>	(M)	3.0686 \pm 0.72 ^a	58.2594 \pm 2.19 ^{ab}	17.0627 \pm 1.72 ^a	76.7097 \pm 2.88 ^a	2.0481 \pm 0.16 ^a
	(L)	15.0106 \pm 1.93 ^b	420.05 \pm 26.07 ^c	100.19 \pm 9.73 ^b	40.5655 \pm 3.18 ^b	10.8244 \pm 0.71 ^b
<i>N. randalli</i>	(M)	2.4061 \pm 0.28 ^a	38.1806 \pm 7.70 ^a	16.1615 \pm 0.28 ^a	45.62 \pm 3.62 ^{bc}	4.5762 \pm 0.41 ^{ac}
	(L)	18.8362 \pm 0.94 ^c	143.37 \pm 12.90 ^d	169.45 \pm 10.03 ^c	61.9680 \pm 3.70 ^d	17.1522 \pm 3.54 ^d
<i>M. barbatus</i>	(M)	2.6735 \pm 0.43 ^a	47.0218 \pm 4.71 ^{ab}	16.8070 \pm 0.85 ^a	42.1526 \pm 0.99 ^{bc}	2.5456 \pm 0.51 ^a
	(L)	7.3006 \pm 0.54 ^d	86.9332 \pm 10.70 ^b	108.39 \pm 11.64 ^b	40.7242 \pm 2.23 ^b	7.2175 \pm 0.44 ^{bc}
<i>P. semiculcatus</i>	(M)	6.0440 \pm 1.16 ^d	44.5599 \pm 0.30 ^{ab}	49.7179 \pm 1.61 ^d	52.6671 \pm 3.05 ^{cd}	16.8740 \pm 2.30 ^d
	(V)	47.2540 \pm 0.90 ^c	233.00 \pm 13.50 ^c	298.71 \pm 9.41 ^e	46.1262 \pm 5.39 ^{bc}	42.7122 \pm 0.84 ^e
Spring						
<i>S. lessepsianus</i>	(M)	4.5686 \pm 0.49 ^a	20.2009 \pm 3.37 ^a	17.0747 \pm 0.55 ^a	27.9735 \pm 1.12 ^a	2.7412 \pm 0.42 ^a
	(L)	3.4216 \pm 0.70 ^{ab}	1148.7 \pm 51.26 ^b	81.5296 \pm 1.29 ^b	10.4459 \pm 0.74 ^b	6.0722 \pm 0.40 ^b
<i>N. randalli</i>	(M)	1.8556 \pm 0.04 ^b	44.7602 \pm 2.56 ^a	10.8596 \pm 0.75 ^a	32.2319 \pm 1.46 ^a	4.4853 \pm 0.28 ^c
	(L)	11.0720 \pm 0.20 ^c	648.03 \pm 30.58 ^c	133.15 \pm 2.49 ^e	49.3586 \pm 0.60 ^c	9.3606 \pm 0.35 ^d
<i>M. barbatus</i>	(M)	3.3151 \pm 0.13 ^{ab}	56.6190 \pm 3.29 ^a	17.5230 \pm 0.93 ^a	56.7853 \pm 0.41 ^c	11.3019 \pm 0.05 ^e
	(L)	17.8643 \pm 0.36 ^d	753.65 \pm 1.94 ^d	89.5628 \pm 9.41 ^b	29.5714 \pm 0.92 ^a	8.6625 \pm 0.51 ^d
<i>P. semiculcatus</i>	(M)	3.4030 \pm 1.03 ^{ab}	19.0761 \pm 0.46 ^a	42.3727 \pm 1.33 ^d	115.24 \pm 2.74 ^d	13.1282 \pm 0.40 ^f
	(V)	9.1659 \pm 0.49 ^e	47.6716 \pm 1.04 ^a	73.672 \pm 1.28 ^e	87.2204 \pm 11.32 ^e	26.4860 \pm 0.84 ^e
Summer						
<i>S. lessepsianus</i>	(M)	3.5244 \pm 0.55 ^a	101.14 \pm 0.86 ^a	15.2861 \pm 0.90 ^a	9.4648 \pm 2.09 ^a	5.6116 \pm 0.01 ^a
	(L)	4.0307 \pm 0.18 ^a	542.13 \pm 23.46 ^b	90.0552 \pm 13.75 ^b	12.5340 \pm 1.48 ^a	5.3612 \pm 1.93 ^a
<i>N. randalli</i>	(M)	3.7485 \pm 0.34 ^a	57.1600 \pm 0.84 ^{ac}	13.0854 \pm 0.61 ^a	116.21 \pm 6.24 ^b	3.7339 \pm 0.92 ^a
	(L)	11.5572 \pm 1.35 ^b	454.96 \pm 17.32 ^d	74.0522 \pm 16.57 ^b	31.4316 \pm 6.21 ^{ce}	12.4824 \pm 0.5 ^a
<i>M. barbatus</i>	(M)	3.3928 \pm 0.36 ^a	37.6664 \pm 1.33 ^c	13.0848 \pm 0.61 ^a	48.6142 \pm 3.46 ^d	2.1893 \pm 0.26 ^a
	(L)	4.5905 \pm 0.69 ^a	145.83 \pm 5.70 ^c	43.9418 \pm 0.35 ^c	24.3519 \pm 1.75 ^c	5.9048 \pm 0.89 ^a
<i>P. semiculcatus</i>	(M)	3.6643 \pm 0.62 ^a	59.2587 \pm 0.56 ^{ac}	37.1292 \pm 0.90 ^c	37.5475 \pm 0.54 ^c	8.5246 \pm 2.36 ^a
	(V)	72.1712 \pm 1.59 ^c	530.31 \pm 22.72 ^b	111.09 \pm 13.64 ^d	36.0081 \pm 0.23 ^e	128.55 \pm 14.5 ^b

*Duncan; the letters a, b, c, d, e, f and g indicate the statistical differences between metal and species. Statistical difference was $p < 0.05$

$\bar{X} \pm S_x$: mean \pm Standard error

it does not contain iron, has low density, is resistant to corrosion and does not have magnetic properties and is widely used especially in transportation and aviation fields. It is possible to participate in nature because of a natural cycle or anthropogenic activities. Al is insoluble in neutral pH, therefore its toxicity occurs in extreme pH ranges. It is found in nature with a value of +3. It is absorbed by aquatic organisms through gills and by

means of nutrients. Therefore, Al is more limited to be taken than metals with a +2 value. The toxicity mechanism is not clear enough. No toxicity record was found in the seas. The concentration in ocean waters is less than $1 \mu\text{g l}^{-1}$ (Brown et al. 2010). Its main participation in the seas is through rivers. However, Al mixed with brackish waters forms a complex with clay minerals in sediment and collapses (Hydes and Liss 1977; Walker et al. 1988).

Table 9 Factors predicting metal accumulation in biota

Biota	<i>F</i>	df	β	<i>t</i>	R ²
Fe	10.776***	4			0.422
species			−0.204	−2.062*	
tissue			0.612	6.184***	
sampling sites, seasons			<i>NS</i> ^a	<i>NS</i> ^a	
As	2.896*	4			0.107
species			0.303	2.545*	
tissue			−0.265	−2.230	
sampling sites, seasons			<i>NS</i> ^a	<i>NS</i> ^a	
Zn	10.396***	4			0.413
species			0.238	2.382*	
tissue			0.584	5.858***	
sampling sites, seasons			<i>NS</i> ^a	<i>NS</i> ^a	
Sr	7.340***	4			0.332
species			0.434	4.079***	
tissue			0.351	3.300**	
sampling sites, seasons			<i>NS</i> ^a	<i>NS</i> ^a	
Al	5.839***	4			0.284
species			0.338	3.071**	
tissue			0.372	3.374**	
sampling sites, seasons			<i>NS</i> ^a	<i>NS</i> ^a	

* $p < 0.05$, ** $p < 0.001$, *** $p < 0.0001$, *NS* not significant, ^a $p > 0.05$

High temperature decreases Al solubility and causes uptake and accumulation through the gills. In this study, it was determined that liver Al level in fish and shrimp sampled from both sampling site in summer increased compared to other seasons ($p < 0.05$) (Tables 7 and 8). However, no statistical significance was found between muscle Al level and season ($p > 0.05$). In some fish species sampled from Iskenderun Bay, the Al level was reported between 0.02–5.41 $\mu\text{g g}^{-1}$ dw (Türkmen et al. 2005a, b). This result correlates with the results of the present study (Table 7).

Al level in sediment was found a second-high concentration after Fe. The sediment Al concentration in the unpolluted site (8725.81 $\mu\text{g g}^{-1}$ dw) was higher than the polluted site (3134.68 $\mu\text{g g}^{-1}$ dw) (Table 5). It was reported that Al and Fe were high level among examined metals in sediment sampled from Kızıkalesi in Mersin Bay. The Al concentration in the region was reported to be 8267.17 mg kg^{-1} and Fe concentration was 18,803.63 mg kg^{-1} (Yalçın and İlhan 2008). While sediment iron level in previous study was higher than the present study, Al levels were similar in both studies. There was no statistical significance was found between sediment Al level and biota ($p > 0.05$). This can be explained by the fact that Al intake and transport to the body is slower than other metals or due to the complexes with the clay minerals in

the sediment. Zn levels in sediment decreased due to an increase in particle size ($p < 0.05$) (Table 5). It was found that a negative correlation between Zn levels and particle size ($p < 0.001$). The particle size reduction causes expansion in the surface area, the rate of binding of the metal at the lower particle size increases.

Iron, which had the highest concentration in biota and sediment in the present study, was found quite low in the seawater ($p < 0.05$). It can be shown that Fe collapsed into the sediment. Sr was found the highest concentration in surface water sampled from both sampling sites. Sr is an alkaline earth element found naturally in the earth's crust. It is found in nature as celestite (SrSO_4) and strontianite (SrCO_3). These consist of a mixture of four different stable isotopes of Sr, ⁸⁴Sr, ⁸⁶Sr, ⁸⁷Sr, ⁸⁸Sr. ⁸⁸Sr accounts for 83% of natural strontium (Lide 1998). It is transported to the seas through rivers. Sr is similar to Ca. Its main uptake by aquatic organisms takes place through Ca channels and intestinal system. Hanlon et al. (1989) reported that Sr is necessary for shell growing and the embryonic development of statolith in the aquatic invertebrates. In nature, radioisotopes released by nuclear waste, particularly ⁹⁰Sr, cause Sr toxicity. The concentration of Sr in seawater was reported as 20 and 92 μM (Chowdhury and Blust 2002). In this study, its concentration was found between 3.20 and 15.53 $\mu\text{g l}^{-1}$ in the polluted site, and between 6.21 and 13.83 $\mu\text{g l}^{-1}$ in the unpolluted site. In both sampling sites, the highest concentration was found in the autumn and decreased towards the summer season ($p < 0.05$) (Table 3). The highest level of Sr in the viscera of *P. semisulcatus* (128.55 $\mu\text{g g}^{-1}$ dw) sampled from the unpolluted site was determined in the summer ($p < 0.05$) (Table 8). The reason for this may be due to the change in the physicochemical parameters of the environment, as well as the fact that *P. semisulcatus* is located at the lower trophic level of the food chain and shows detritivore nutrition feature different from fish. On the other hand, the increase of metabolic rate due to the high temperature in the summer will raise energy requirement, therefore, the enhancement amount of respiration and nutrition will elevate intake and accumulation. Ecotoxicological records of strontium could not be reached in the Mediterranean Sea. This research carried out in Iskenderun and Mersin Bay in the Northeastern Mediterranean may be the first study to reflect the current status of Sr. Limited study was found Sr level in biota. Cravo et al. (2002) reported that Sr level in shell was 1339 $\mu\text{g g}^{-1}$ dw in *Patella aspera* sampled from the contaminated and uncontaminated region in Portugal. It is known that the current accumulation of Sr are caused by the natural resources. In this research, water, sediment, and biota analysis sampled from the Northeastern Mediterranean constitute the first toxicological records of Sr for the Mediterranean Sea.

Table 10 Comparison of metal level in some fish sampled from Northeastern Mediterranean Sea

Species	Metals	Iskenderun Bay		Mersin Bay		References
		Muscle	Liver	Muscle	Liver	
<i>M. barbatus</i>	Fe	71.7	151	103.1	258	Kalay et al. 1999
	Zn	16.1	55.3	25.8	55.3	
<i>Mugil cephalus</i>	Fe	73.4	310	129	262	
	Zn	23.5	52.2	30.9	57.3	
<i>Sparus aurata</i>	Fe	14.5	–	–	–	Türkmen et al. 2008
	Zn	6.2	–	–	–	
<i>Diplodus vulgaris</i>	Fe	34.9	–	–	–	
	Zn	11.2	–	–	–	
<i>M. barbatus</i>	Fe	56.7*	385.2*	–	–	Çoğun et al. 2006
	Zn	34.5*	130.2*	–	–	
<i>Mugil cephalus</i>	Fe	41.2*	284.5*	–	–	
	Zn	29.7*	86.5*	–	–	
<i>M. barbatus</i>	Fe	34.8	135	11.5	889	Tepe et al. 2008
	Zn	5.09	26	10.2	70.9	
<i>Upeneus mollucensis</i>	Fe	3.5	211.7	–	–	Dural and Bickici 2010
	Zn	0.2	66.3	–	–	
<i>Mugil cephalus</i>	Fe	–	–	34.8	112	Türkmen et al. 2011
	Zn	–	–	6.6	17.8	
<i>Mugil cephalus</i>	Fe	70.3	–	–	–	Yilmaz 2003
	Zn	38.2	–	–	–	
<i>Trachurus mediterraneus</i>	Fe	41.8	–	–	–	
	Zn	19.6	–	–	–	
<i>Mugil cephalus</i>	Fe	38.7	370.4	–	–	Canlı and Atlı 2003
	Zn	37.4	110	–	–	
<i>Sardina pilchardus</i>	Fe	39.6	225.5	–	–	
	Zn	34.6	73.2	–	–	
<i>M. barbatus</i>	Fe	–	–	69.9	–	Külcü et al. 2014
	Zn	–	–	25.1	–	
<i>Scomber japonicas</i>	Fe	–	–	86.1	–	
	Zn	–	–	26.3	–	
<i>Solea solea</i>	Fe	–	–	91.9	–	
	Zn	–	–	22.8	–	
<i>M. barbatus</i>	Fe	139.6	404.9	44.2	277.3	Present study
	Zn	14.1	64.4	15	80	
<i>S. lessepsianus</i>	Fe	61.8	424.9	73.1	710.5	
	Zn	14.3	123.0	15.9	81.0	
<i>N. randalli</i>	Fe	105.9	769.9	44.8	401.6	
	Zn	13.6	144.5	13.5	115.7	

*indicates that the findings belong to both sampling sites

Conclusion

It is important to compare metal concentrations in biota, water, and sediment in Iskenderun and Mersin Bay, are the most important fishing areas, and are directly affected by industrial, domestic and agricultural wastes. Certain parts of heavy metals discharged in the aquatic systems are suspended while a major part of these is precipitated in the sediment. Food and

water constitute the main uptake way of metals in the environment. The metal accumulation varies depending on the ambient concentration, species, tissues, sampling region, particle size, and season.

In this study, metal concentrations in fish and shrimp tissues sampled from the polluted and unpolluted sites have changed depending on the species. Fe showed the highest concentration in biota and sediment in both sampling sites.

Arsenic was the second-highest element in biota. The muscle level of arsenic was higher than the liver. Al was the second-highest accumulate in sediment. Zn concentration in sediment decreased due to an increase in particle size.

In the surface water, Sr was found the highest level. Its concentration was decreased from autumn to summer. The Sr data in water, sediment, and biota sampled from the Iskenderun and Mersin Bay are the first toxicological records for the Mediterranean Sea.

Acknowledgements This study was supported by the Research Fund of Mersin University in Turkey with Project Number: 2018-2-AP3-3014.

Compliance with Ethical Standards

Conflict of Interest The authors, Nuray Çiftçi, Deniz Ayas and Misra Bakan declare that they have no conflict of interest.

Ethical Approval There is no ethical issue concerning this article.

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