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Assessment of metal element concentrations in mussel (M. Galloprovincialis) in Eastern Black Sea, Turkey

U. Çevik[a,*], N. Damla[a], A.I. Kobya[a], V.N. Bulut[b], C. Duran[c], G. Dalgıc[d], R. Bozacı[a]

[a] Karadeniz Technical University, Department of Physics, 61080 Trabzon, Turkey
[b] Giresun University, Department of Chemistry, 28049 Giresun, Turkey
[c] Karadeniz Technical University, Department of Chemistry, 61080 Trabzon, Turkey
[d] Rize University, Faculty of Fisheries, 53100 Rize, Turkey

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ABSTRACT

The main goal of this work is to determine the effects of pollution of copper, lead and zinc mines on the Eastern Black Sea. Metal and heavy metal concentrations in the Eastern Black Sea mussels were measured using Energy Dispersive X-ray Fluorescence (EDXRF) and Flame Atomic Absorption Spectroscopy (FAAS). The analytical results showed that the tissue of mussel in Eastern Black Sea contains K, Ca, Fe, Cu, Zn, and Sr elements, and the shell of mussel contains Ca, Cu, Sr, and Ba elements. Due to the detection limit of EDXRF, the mussels were analyzed with FAAS for Cr, Mn, Ni, Cd and Pb elements. An ANOVA and Pearson correlation analyses were performed. The results showed although that the mean concentrations of Cu and Zn for the tissue of the mussels were markedly above the permissible levels of the Turkish regulations, Zn concentration is in the limits of the Food and Agriculture Organization (FAO).

1. Introduction

Mussels are widely used as bioindicators of heavy metal pollution in coastal areas because they can accumulate various elements as filter-feeders [1]. They also show the other parameters for being bioindicators such as suitable dimensions, easy identification and collection of organisms, abundance in an ecosystem and accumulation of the elements to a degree suitable to measure [2].

Mussels are well known to accumulate a wide range of contaminants in their soft tissues [3]. Thus, they are often employed to monitor metal pollution in sea. Size has sometimes been shown to be an important variable, but contradictory results have been found. In order to monitor metal content in the mussels, the number of size classes according to shell length as well as the number of samples must be taken into account to obtain a representative sampling [4]. Additionally, the total cost of collection, preparation and analysis of each sample have to be considered in the analysis.

Among the mussels, M. Galloprovincialis is one of the most commonly consumed bivalve mollusks [3]. These bivalves are superior bioaccumulators of many chemical substances, tolerate handling and experimental manipulation, and provide ample tissue and shell material for chemical analysis [5]. The growth of these bivalves is due to their great capacity to filter the water column, which unfortunately, also exposes them to dangerous contaminants, including heavy metals [4]. As known, metals are introduced into the aquatic ecosystems such as lakes, rivers and seas in a number of ways. They may be accumulated by aquatic organism such as fish and mussels and may be a potential risk for ecosystem health and organisms. Industrial wastes and mining of metal create a potential source of heavy metals pollution in the aquatic environment [6–8].

Heavy metal discharges to the marine environment are of great concern all over the world as they have great ecological significances due to their toxicity and accumulative behavior. Metals such as iron, copper, zinc and manganese, are essential metals since they play an important role in biological systems, whereas mercury, lead and cadmium are non-essential metals, as they are toxic, even in traces. However, the essential metals may also produce toxic effects when the metal intake is excessively elevated [9]. Many heavy metals were accumulated in organisms and some were also accumulated in the food chain. The anthropogenic heavy metal outlets may reduce the marine species diversity and acerbate the ecosystem. Further, people consuming seafood would be exposed to the metals with a potential danger to their health.

Eastern Black Sea region has a rich potential in terms of a variety of mines. On the other hand, the industrial and domestic wastes were transported by rivers from Russia and Turkey to Eastern Black Sea. The objective of this study is to determine

* Corresponding author. Tel.: +90 4623773591; fax: +90 4623253195.
E-mail address: berrenazli@yahoo.com (U. Çevik).

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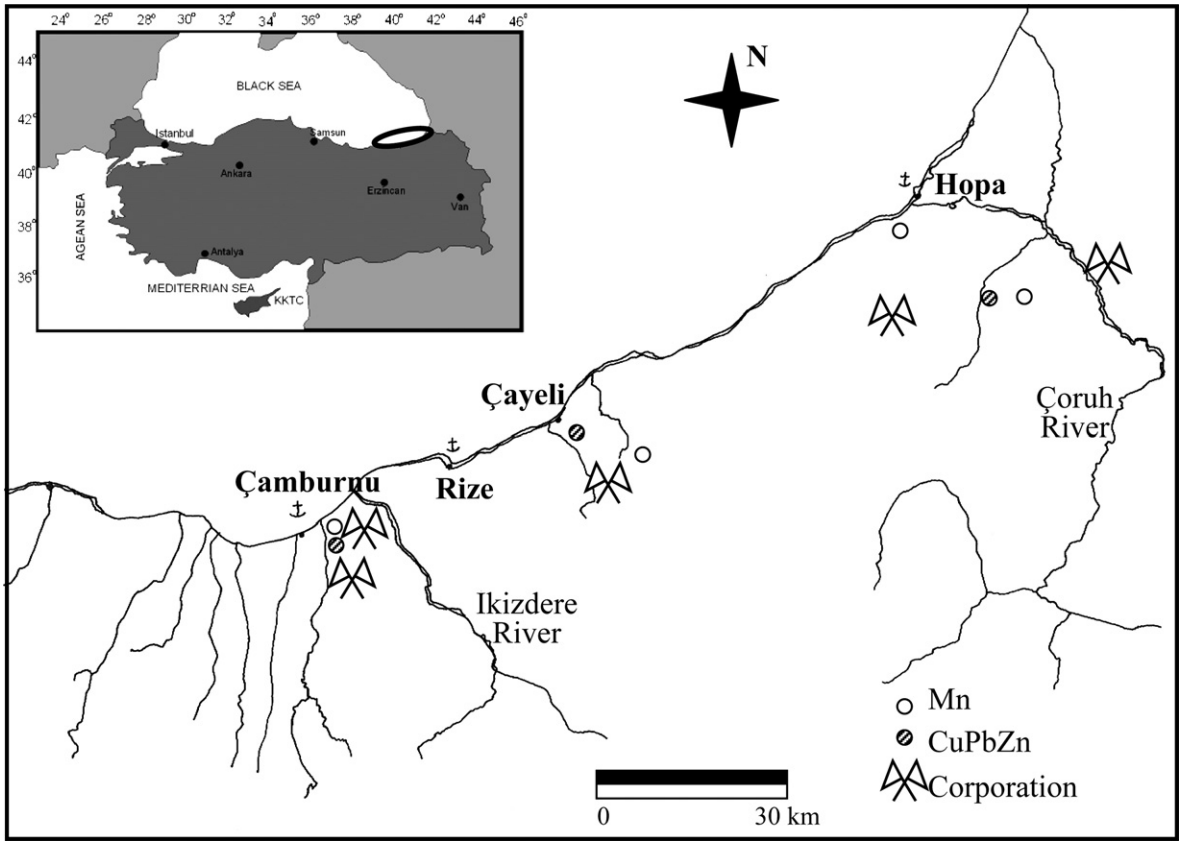

Fig. 1. Sampling sites.

the effects of pollution of mines by measuring essential metal concentrations in the mussels collected from the Eastern Black Sea.

2. Materials and methods

2.1. Study area

The Eastern Black Sea Region is located in the north eastern part of Turkey (Fig. 1) covering deep running water valleys, mountains, steps and broken zones. High water flows such as Ikizdere, Harsit, Firtina, and Coruh rivers originating from this land fall to Black Sea. Moreover, the Eastern Black Sea region has a rich potential in terms of a variety of mines. As shown in the map, the metals wasted due to the industrial activities and carried by means of rivers are drifted and contaminated to the Black Sea. The mine potential of the region is reflected by total reserves; including copper, lead and zinc deposits of 700 million tons and iron reserve of 10 million tons, manganese reserve of 260.000 tons, gold reserve of 36 tons and silver reserve of 1.4 million tons [10].

Table 2
Instrumental conditions for the measurements of the working elements by FAAS

Element	Wavelength (nm)	Slit width (nm)	Lamp current (mA)	Flow rate of acetylene ($L\,min^{-1}$)
Cd	228.8	0.5	5.1	1.2
Ni	232.0	0.2	11.5	0.9
Pb	217.0	0.5	6.0	1.1
Mn	279.5	0.2	9.0	1.2
Cr	357.9	0.5	9.0	4.2

2.2. Sample treatment

In this work, the different sized mussel samples were collected at five stations in the Eastern Black Sea. As seen in Fig. 1, the stations are scattered throughout the area marked on the map. Some information related to the sampling points is given in Table 1. The specimens were sorted with respect to their sizes given as follows: Group A: <1.5 cm; Group B: 1.6–2.5 cm; Group C: 2.6–4.0 cm; Group D: 4.1–5.0 cm; Group E: >5.0 cm. Around twenty mussels were selected from each group. After the samples were sorted out, they were dried in ovens at 100 °C and then their tissues and shells were

Table 1
Information related to collected samples

Location	Coordinate	Sample depth (m)	Maximum depth (m)	Salinity (%)	pH (20 °C)	Water temperature (°C)
Çamburnu	40°55′N 40°13′E	2	5	19.1	8.3	9.1
Rize (inner of harbor)	41°01′N 40°31′E	2	12	18.9	8.3	8.0
Rize (out of harbor)	41°01′N 40°31′E	2	28	17.8	8.2	23.6
Çayeli	41°05′N 40°43′E	0.5	1	13.0	8.3	19.6
Hopa	41°24′N 41°25′E	2	16	18.9	7.8	24.0

Table 3
Concentration of metals found in mussel samples ($\mu g\,g^{-1}$)

Samples		Çamburnu		Rize (inner of harbor)		Rize (out of harbor)		Çayeli		Hopa	
Elements		Tissue	Shell	Tissue	Shell	Tissue	Shell	Tissue	Shell	Tissue	Shell
EDXRF	K	4230 ± 126	< DL	6110 ± 306	<DL	7870 ± 236	<DL	6930 ± 208	<DL	7960 ± 239	<DL
	Ca	20,400 ± 612	6,23,640 ± 1870	11,000 ± 330	4,89,760 ± 1469	14,270 ± 428	6,46,680 ± 19,400	10,370 ± 311	4,99,820 ± 14,995	6960 ± 209	6,01,880 ± 18,056
	Fe	3340 ± 165	<DL	2390 ± 72	<DL	1400 ± 42	<DL	4030 ± 121	<DL	1150 ± 35	<DL
	Cu	190 ± 6	120 ± 4	260 ± 8	110 ± 3	90 ± 3	170 ± 5	130 ± 4	110 ± 3	130 ± 4	180 ± 5
	Zn	630 ± 32	<DL	600 ± 30	<DL	340 ± 10	<DL	230 ± 7	<DL	180 ± 5	<DL
	Sr	160 ± 8	920 ± 28	100 ± 3	950 ± 29	190 ± 6	870 ± 26	100 ± 3	800 ± 24	60 ± 2	840 ± 25
	Ba	<DL	50 ± 2	<DL	110 ± 4	<DL	70 ± 2	<DL	<DL	<DL	110 ± 3
AAS	Cr	3.0 ± 0.2	<DL	2.0 ± 0.1	<DL	1.0 ± 0.1	<DL	1.0 ± 0.1	<DL	2.0 ± 0.1	<DL
	Mn	59 ± 3	39 ± 1	54 ± 3	21 ± 1	41 ± 2	11 ± 1	46 ± 2	23 ± 1	47 ± 2	21 ± 1
	Ni	6.0 ± 0.3	<DL	1.0 ± 0.1	<DL	3.0 ± 0.2	<DL	3.0 ± 0.2	<DL	2.0 ± 0.1	<DL
	Cd	4.0 ± 0.2	1.0 ± 0.1	3.0 ± 0.2	1.0 ± 0.1	3.0 ± 0.2	2.0 ± 0.1	2.0 ± 0.1	1.0 ± 0.1	3.0 ± 0.2	1.0 ± 0.1
	Pb	21.0 ± 1.0	<DL	5.0 ± 0.3	<DL	9.0 ± 0.5	<DL	5.0 ± 0.2	<DL	3.0 ± 0.1	<DL

The results are reported as average of five subsamples (in a sample of 20 mussels) with the relative standard error, DL detection limit

separated. After that they were put into powder by using a Spex mill during 20 min of time. To reduce the size effects of particles, the powder was sieved by using a 400 mesh sieve. Forty milligrams of this powder was pressed in 13 mm diameter.

2.3. Instrumentation

Metal elements were analyzed using Energy Dispersive X-Ray Fluorescence method (EDXRF) by employing the standard addition method in sample preparation. The samples have been stimulated by ^{55}Fe and ^{241}Am radioactive sources. To detect the radiation scattered from the sample, Si(Li) detector manufactured by PGT having 150 eV full width at half maximum (FWHM) for 5.96 keV photons was used. Two thousand and forty-eight channels of a multi-channel analyzer were employed in data acquisition. In quantitative analysis, characteristic X-rays emitted by excited atoms of the sample were registered for a time interval of 5000 s [11–14].

In order to determine the detection limit of EDXRF for Cd, Cr, Ni and Pb we used the reference material which was prepared by International Atomic Energy Agency. Characteristic X-ray peak intensities for Cd, Cr, Ni and Pb were obtained through spectrum analysis of the reference material. Sensitivity of this element was calculated by the following equation [15]:

$$C_i = \frac{I_i}{S_i} \tag{1}$$

where C_i is the concentration ($mg\,kg^{-1}$), I_i the characteristic X-ray intensity (cps), and S_i is the elemental sensitivity ($cps\,mg^{-1}\,kg$) for the element i. The detection limit was calculated by

$$DL_i = \frac{3}{S_i}\sqrt{\frac{I_i(BG)}{t}} \tag{2}$$

where DL_i is the detection limit ($mg\,kg^{-1}$), I_i (BG) the background intensity (cps) under element I peak, and t is the counting time (s). Detection limit for Cd, Cr, Ni and Pb were calculated to be 7.2, 14.5, 9.8 and 5.4 $mg\,kg^{-1}$, respectively.

Due to the detection limit of EDXRF, the samples were analyzed with Flame Atomic Absorption Spectroscopy (FAAS) for Cr, Mn, Ni, Cd and Pb elements. The measurements of metal ions were performed with a Unicam Model AA-929 flame atomic absorption spectrometer equipped with a single element hollow-cathode lamps and 5.0 cm of an air/acetylene burner head. Instrumental conditions for working elements are summarized in Table 2 [16].

One gram of fine powdered and the dried mussel samples were weight into Teflon vessel and 8 mL of HNO_3 (65%), 1 mL of H_2O_2 (30%) and 0.5 mL of concentrated HF (39%) were added. Then, the content of the vessel was digested by microwave irradiation. The digestion conditions of a microwave system for the samples were applied as (45 bar) 1 min for 250 W, 1 min for 0 W, 10 min for 650 W, 5 min for 250 W, vents: 3 min, respectively. The residue diluted to 25.0 mL with deionized water. A blank digest was carried out in the same way [16].

3. Results and discussion

The metal and heavy metals concentration in the mussels can provide the measure for the effects of the mixtures of potential pollutants in the marine environment. However, when no data are available on the pollutant content of the mussels in the study areas, the research about the contaminant concentrations in organisms through the accumulation of contaminants in their tissue remains to be necessarily worked out.

The concentrations of elements ($\mu g\,g^{-1}$) in the mussel samples in five different sampling stations are shown in Table 3. In order to show the general distribution of elemental concentrations in the mussels for all over the studied area, the results were expressed as

Table 4
Metal concentrations ($\mu g\,g^{-1}$) in sediment collected from the sampling sites

	K	Ca	Cr	Mn	Fe	Ni	Cu	Zn	Cd	Pb
Çamburnu	5,700	2,076	12.0	504	2,55,000	ND	4,436	1,836	3.8	134.8
Rize (inner of harbor)	10,700	283	9.0	985	53,000	ND	122	305	ND	41.7
Rize (out of harbor)	7,800	314	52.9	903	72,000	21.4	161	125	ND	43.7
Çayeli	9,000	324	5.6	1,089	53,000	ND	414	359	ND	50.6
Hopa	7,300	434	56.9	1,674	88,000	16.1	6,259	2,344	5.9	355.1

Table 5
Metal concentrations ($\mu g\,L^{-1}$) in sea water collected from the sampling sites

	K	Ca	Cr	Mn	Fe	Ni	Cu	Zn	Cd	Pb
Çamburnu	3,08,000	5,37,000	ND	ND	680	ND	19.5	6.5	3.0	ND
Rize (inner of harbor)	2,86,000	5,10,000	ND	ND	210	ND	7.5	207.5	ND	29.0
Rize (out of harbor)	2,80,000	4,64,000	ND	ND	130	ND	ND	12.0	ND	ND
Çayeli	1,71,000	2,70,000	ND	ND	290	ND	9.0	6.0	ND	17.5
Hopa	3,00,000	4,40,000	ND	ND	340	ND	20.5	81.5	ND	39.0

ND: not detected.

an average value of five different sizes. The obtained results from EDXRF show that the tissue of mussel samples contains K, Ca, Fe, Cu, Zn, and Sr elements and the shell of mussel contains Ca, Cu, Sr, and Ba elements.

The maximum value of Cu for the tissue of the mussels was found in the inner harbor of Rize ($260\,\mu g\,g^{-1}$) while the minimum value was measured in the outer harbor of Rize ($90\,\mu g\,g^{-1}$). Furthermore, the maximum concentration of Zn for the tissue of the mussels was measured in Çamburnu coast as $600\,\mu g\,g^{-1}$ while the minimum value as $180\,\mu g\,g^{-1}$ was measured in Hopa coast. The obtained results show that the average concentrations of Cu and Zn were approximately eight times higher than the maximum value specified by the Turkish Fisheries Law and Regulations (TKB) [17]. The most important reason is that the Cu and Zn reserve of Turkey, like Pb reserve, mostly takes place in the Eastern Black Sea region.

Since this study aims to determine the effects of pollution of mines on the mussels collected from the Eastern Black Sea, one would expect to detect the other metals such as Cr, Mn, Ni, Cd and Pb. However, Cr, Mn, Ni, Cd and Pb were not observed with EDXRF because of the lower detection limit of the system. As such, we analyzed the samples with FAAS for Cr, Mn, Ni, Cd and Pb elements. The obtained results with FAAS were also shown in Table 3. The maximum values of Cr, Ni, Cd and Pb for tissue of mussels were found in the Çamburnu coast (3, 6, 4 and $21\,\mu g\,g^{-1}$). However, these elements except Cd were not detected in the shells of mussel. Mn concentrations range between $46\,\mu g\,g^{-1}$ in Çayeli coast and $59\,\mu g\,g^{-1}$ in the Çamburnu coast for the tissue of the mussels. The maximum value of Mn for shell of mussel was detected in the Çamburnu coast ($39\,\mu g\,g^{-1}$) while that of value was found minimum in the outer harbor of Rize ($11\,\mu g\,g^{-1}$).

Tables 4 and 5 indicate the metal concentrations in the sediment and the water in the samples in studied area. These measurements are important to show the elemental concentrations in the water and the sediments and to make a correlation with those in the mussels. Considering the sea water density, it has been observed that the concentrations in the mussels are relatively higher than that

Table 6
Concentrations of metals in various size of mussel tissue

Station		K	Ca	Cr	Mn	Fe	Ni	Cu	Zn	Sr	Cd	Pb
Çamburnu	A	3,860	33,540	4.8	56	3,290	5.4	210	400	230	3.5	9.7
	B	3,720	20,490	3.8	11	2,840	ND	169	590	150	5.4	31.9
	C	5,050	15,510	1.5	106	4,600	17.9	190	440	120	2.3	ND
	D	4,910	15,420	ND	39	3,580	ND	180	518	150	1.8	45.9
	E	3,610	17,040	5.1	24	2,390	5.7	203	1,204	150	6.2	17.6
Rize (inner of harbor)	A	5,740	14,510	7.2	36	2,830	3.2	362	660	130	4.6	10.2
	B	6,450	14,200	1.8	26	3,090	ND	323	710	110	5.1	16.1
	C	6,740	13,910	1.7	149	2,370	ND	110	520	120	2.1	ND
	D	5,970	7,150	ND	26	1,830	ND	325	480	71	2.6	ND
	E	5,650	5,230	ND	36	1,830	3.6	180	630	71	2.8	ND
Rize (out of harbor)	A	8,720	17,820	ND	27	1,620	ND	60	310	330	2.4	ND
	B	7,570	13,910	1.1	25	1,140	ND	130	370	210	4.2	15.9
	C	6,250	14,770	1.5	93	1,170	ND	90	330	220	1.4	ND
	D	8,360	13,050	ND	18	1,630	ND	110	370	50	2.6	ND
	E	8,900	12,340	1.8	41	1,440	14.5	60	320	140	6.0	26.9
Çayeli	A	7,350	9,180	4.2	43	3,890	9.4	80	240	95	3.3	5.9
	B	6,870	9,420	1.2	32	4,700	3.6	140	260	100	3.6	16.9
	C	7,570	15,850	1.4	117	3,810	3.3	110	250	110	2.2	ND
	D	6,260	8,100	ND	18	3,880	ND	120	190	97	1.0	ND
	E	6,600	9,300	ND	20	3,870	ND	200	210	97	0.6	ND
Hopa	A	7,670	14,940	8.8	48	1,630	6.2	200	120	100	6.2	12.9
	B	8,020	6,350	ND	50	870	ND	90	210	55	3.0	ND
	C	8,760	4,400	ND	80	660	3.9	60	210	44	1.7	ND
	D	7,550	5,000	ND	21	1,440	ND	170	220	55	2.1	ND
	E	7,800	4,110	ND	36	1,150	ND	130	140	44	2.8	ND

ND: not detected, mussel size—A: <1.5 cm; B: 1.6–2.5 cm; C: 2.6–4.0 cm; D: 4.1–5.0; E: >5.0 cm.

Table 7
Pearson correlation coefficients between metal levels in tissue of mussels in Eastern Black Sea

	K	Ca	Cr	Mn	Fe	Ni	Cu	Zn	Sr	Cd	Pb
K	1										
Ca	**−0.517**[a]	1									
Cr	−0.336	*0.499*[b]	1								
Mn	0.065	0.130	0.017	1							
Fe	*−0.467*[b]	0.284	0.137	0.073	1						
Ni	−0.051	0.157	0.331	0.268	0.287	1					
Cu	**−0.535**[a]	0.162	0.373	−0.267	0.254	−0.087	1				
Zn	**−0.662**[a]	0.321	0.279	−0.104	0.103	0.041	*0.492*[b]	1			
Sr	−0.197	**0.698**[a]	0.182	0.031	0.035	0.000	−0.108	0.184	1		
Cd	−0.163	0.321	**0.690**[a]	−0.282	−0.122	0.323	0.276	*0.444*[b]	0.119	1	
Pb	−0.368	0.361	0.276	−0.300	0.198	0.091	0.152	0.352	0.236	**0.523**[a]	1

[a] Correlation is significant at the 0.01 level (2-tailed).
[b] Correlation is significant at the 0.05 level (2-tailed).

Table 8
Comparison of the metal concentrations ($\mu g\,g^{-1}$) in tissue of mussels with the other studies and guidelines

Elements	Mean	Range	FAO [18]	TKB [17]	Morocco [19]	Korean [20]	Spanish [21]	USA [22]
K	6,620	4,230–7,960						
Ca	12,600	6,960–20,400						
Cr	2	1–3	5		2–29			17
Mn	49	41–59			7–28	7–227		
Fe	2,462	1,150–4,030						
Ni	3	1–6			12–32			11
Cu	160	90–260	50–150	20	4–43	4–14	5–7	6
Zn	396	180–630	200–500	50	107–366	70–157	176–316	75
Sr	122	60–190						
Cd	3	2–4	10	0.1	2–35	<1–2	<1–1.4	1
Pb	9	5–21	5–30	1	1–26	4–53	1–3	1

of the water. As seen from tables, while Cr, Mn, Ni and Cd are not detected in the water samples, they are detected in the sediments and the mussels. Another important point is that the detected Pb concentrations in sediment samples are quite high. This can be attributed to the unsolved lead in the sea water adsorbed by the sediment.

Table 6 shows the metal concentrations with respect to the tissue sizes in each sampling site. According to results of one-way ANOVA there is no statistical difference between the mean values of concentrations of metals in tissues. As a result of Pearson correlation, metal concentrations in the tissues did not seem to depend on the mussel length and sampling sites. Table 7 summarizes the correlation coefficients among the metal levels of the tissue of mussels in Eastern Black Sea. Significant correlations coefficients ($p < 0.05$) and ($p < 0.01$) are in bold and italics, respectively. As clearly seen from the table, the most noticeable correlations were found between the pairs K–Ca ($r = -0.517$), K–Fe ($r = -0.467$), K–Cu ($r = -0.535$), K–Zn ($r = -0.662$), Ca–Cr ($r = 0.499$), Ca–Sr ($r = 0.698$), Cr–Cd ($r = 0.690$), Cu–Zn ($r = 0.492$), Zn–Cd ($r = 0.444$), and Cd–Pb ($r = 0.523$). These results suggest that the correlated metals share a common accumulation process in tissue of mussels.

In Table 8, the elemental concentration levels in the tissue of mussels are compared with the national and international standards for metals in mollusks compiled by the TKB [17] and Food and Agriculture Organization (FAO) [18]. Also, some experimental values presented for the other countries were given in Table 8 for comparison. Although the mean concentrations of Cu and Zn in the tissues of mussels are higher than those of given in the literature, it is consistent with the values of FAO. For the mean concentrations of Cr, the observed value is lower than the literature value. The mean concentration value of Cd ($3\,\mu g\,g^{-1}$) and Pb ($9\,\mu g\,g^{-1}$) are generally lower than the limits of Morocco [19] and FAO [18], but are higher than those of Spanish [21], USA [22] and TKB [17]. A comparison in the concentrations values measured in the Eastern Black Sea mussels shows that the levels of Cr, Cd and Pb are well below the converted FAO limits. The concentrations of Cu and Zn in the mussel samples rank within the different standards defined by the FAO limits, but over the highest value.

From the obtained data, the average metal concentrations decrease in following order: Zn > Cu > Mn > Pb > Ni > Cd > Cr. This heavy metal sequence is for the soft tissues of the mussel or environmental complex. Accumulation of metals in the shell also shows the exposure periods and the degree of contamination. These heavy metal concentrations are generally higher than the limits of TKB [17] and FAO [18]. This is because of the discharge of untreated domestic wastes into the sea, the various harbor activities, the dumping of ship wastes into the sea, and the oil spills. Also, the results of our study showed that the Eastern Black Sea was contaminated by the waste over metals because of the nearby industrial activities drifted to the region by means of rivers.

4. Conclusion

Pollution measurement in marine environment includes the analysis of mussels. Thus, biological characteristics of mussels may provide critical information about water pollution. The soft tissues of mussels are generally more efficient accumulators of metal elements than shells. Nevertheless, studies on metal accumulation in shells are also useful since they can be used as a record of environmental metal levels [23]. Cu and Zn are the critical elements to be determined in the pollution analysis. The obtained results show that the mean concentrations of Cu and Zn for the tissue of mussels were markedly above the permissible levels according to Turkish permissible concentrations. Zn concentration in the mussels samples rank within the limits of different standards defined by the FAO. The concentration of Cu is above the permissible level of FAO limits. In coastal waters, the fluctuations in metal and heavy metal concentrations are similar depending on the natural factors or the anthropogenic pollution. The people living along the coastal areas

consume the mussels in considerable amount. Therefore, the metal and heavy metal concentrations measured in the present study may help to determine the dose contribution of the seafood consumed by these people.

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References

[1] F. Regoli, Metals and antioxidant enzymes in gills and digestive gland of the Mediterranean mussel M. Galloprovincialis, Arch. Environ. Contam. Toxicol. 34 (1998) 48–63.
[2] A. Wagner, J. Boman, Biomonitoring of trace elements in Vietnamese freshwater fish, Spectrochim. Acta B 59 (2004) 1125–1132.
[3] E.D. Goldberg, V.T. Bowen, J.W. Farrington, G. Harvey, J.H. Martin, P.L. Parker, R.W. Risebrough, W. Robertson, E. Schneider, E. Gamble, The mussel watch, Environ. Conserv. 5 (1978) 101–125.
[4] Y. Saavedra, A. Gonzalez, P. Fernandez, J. Blanco, The effect of size on trace metal levels in raft cultivated mussels (M. Galloprovincialis), Sci. Total Environ. 318 (2004) 115–124.
[5] R.T. Angelo, M.S. Cringan, D.L. Chamberlain, A.J. Stahl, S.G. Haslouer, C.A. Goodrich, Residual effects of lead and zinc mining on freshwater mussels in the Spring River Basin (Kansas, Missouri, and Oklahoma, USA), Sci. Total Environ. 384 (2007) 467–496.
[6] M. Türkmen, C. Ciminli, Determination of metals in fish and mussel species by inductively coupled plasma-atomic emission spectrometry, Food Chem. 103 (2007) 670–675.
[7] B. Gümgüm, E. Ünlü, Z. Tez, Z. Gülsün, Heavy metal pollution in water, sediment and fish from the Tigris River in Turkey, Chemosphere 29 (1994) 111–116.
[8] Y.H. Lee, R.B. Stuebing, Heavy metal contamination in the River Toad, *Bufo juxtasper* (Inger), near a copper mine in East Malaysia, Bull. Environ. Contam. Toxicol. 45 (1990) 272–279.
[9] J. Matta, M. Milad, R. Manger, T. Tosteson, Heavy metals, lipid prexidation, and cigateratoxicity in liver of the Caribben barracuda (*Sphyraena barracuda*), Biol. Trace Elem. Res. 70 (1999) 69–79.
[10] MTA, Mine and mineral sources of Turkey, MTA General Directorate, No: 185 Ankara (1989) (in Turkish).
[11] A. Aslan, G. Budak, E. Tirasoglu, A. Karabulut, Y. Karagoz, G. Apaydin, B. Ertugral, U. Çevik, Analysis of elements in some lichens by radioisotope X-ray fluorescence spectrometry, Fres. Environ. Bull. 13 (2004) 740–747.
[12] U. Çevik, A.I. Kopya, H. Karal, Y. Sahin, Quantitative analysis of sea-bed sediments from Eastern Black Sea by EDXRF spectrometry, J. Radional. Nucl. Chem. Lett. 201 (1995) 241–249.
[13] M. Kobya, M. Ertugrul, O. Dogan, Ö. Şimşek, Analysis of titanium and zirconium in red mud with energy dispersive X-ray spectrometry, Instrum. Sci. Technol. 24 (1996) 272–282.
[14] U. Çevik, E. Ergen, G. Budak, A. Karabulut, E. Tirasoglu, G. Apaydin, A.I. Kopya, Elemental analysis of Akçaabat tobacco and its ash by EDXRF spectrometry, J. Quant. Spectrosc. RA. 78 (2002) 409–415.
[15] B. Koz, U. Cevik, T. Ozdemir, C. Duran, S. Kaya, A. Gundogdu, N. Celik, Analysis of mosses along Sarp-Samsun highway in Turkey, J. Hazard. Mater. 153 (2008) 646–654.
[16] V.N. Bulut, A. Gundogdu, C. Duran, H.B. Senturk, M. Soylak, L. Elci, M. Tufekci, A multi-element solid-phase extraction method for trace metals determination in environmental samples on Amberlite XAD-2000, J. Hazard. Mater. 146 (2007) 155–163.
[17] TKB, Turkish Fisheries Laws and Regulations, Ministry of Agriculture and Rural Affairs, Conservation and Control General Management, Ankara, 2002.
[18] FAO (Food and Agriculture Organization), Compilation of legal limits for hazardous substances in fish and fishery products, FAO fishery circular, No. 464, pp. 5–100, 1983.
[19] M. Maanan, Biomonitoring of heavy metals using M. Galloprovincialis in Safi coastal waters, Morocco, Environ. Toxicol. 22 (2007) 525–531.
[20] P. Szefer, B.S. Kim, C.K. Kim, E.H. Kim, C.B. Lee, Distribution and coassociations of trace elements in soft tissue and byssus of M. Galloprovincialis relative to the surrounding seawater and suspend matter of the southern part of the Korean Peninsula, Environ. Pollut. 129 (2004) 209–228.
[21] V. Besada, J. Fumega, A. Vaamonde, Temporal trends of Cd, Cu, Hg, Pb and Zn in mussel (M. Galloprovincialis) from the Spanish North-Atlantic coast 1991–1999, Sci. Total Environ. 288 (2002) 239–253.
[22] T. Cohen, S.S. Que Hee, R.F. Ambrose, Trace metals in fish and invertebrates of Three California coastal Wetlands, Mar. Pollut. Bull. 42 (2001) 224–232.
[23] M. Protasowicki, M. Dural, J. Jaremek, Trace metals in the shells of blue mussels (Mytilus edulis) from the Poland coast of Baltic Sea, Environ. Monit. Assess. 141 (2008) 329–337.