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ORIGINAL PAPER

Heavy metals in *Mytilus galloprovincialis*, suspended particulate matter and sediment from offshore submerged longline system, Black Sea

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Abstract The study was performed between March 2009 and March 2010 at the Sinop Inner Harbor on cultured mussels at the offshore submerged longline system. The samples of mussels, sediment and suspended particulate matter were seasonally taken for the purpose of monitoring differences of metal contents. In addition, burnable organic matter, contamination factors (CF), pollution load index (PLI) in sediment samples, meat yield of mussels were seasonally determined. All metals in suspended particulate matter were found to be above the national limit. Cd concentration in sediment was above the limit according to sediment quality guidelines, and Cd concentration in mussel was above both national and international limit values. According to CF and PLI identified in sediment, it was found that Inner Harbor was under the effect of anthropogenic pollution in terms of Cd accumulation. Provisional tolerable weekly intake calculated in mussel did not exceed the tolerable consumption values. In conclusion, it can be said that this region considered for commercial mussel cultivation is not a suitable area.

Keywords *Mytilus galloprovincialis* · Sediment · Suspended particulate matter · Heavy metals · Submerged longline system · Black Sea

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Introduction

The most commonly cultivated species among invertebrate species is mussel. Bivalve cultivation, which also includes mussels, is of great importance throughout the world and showed a growth of about 6% in the last 30 years. According to 2013 data, the mussel cultivation was about 1,755,694 tons in the world, whereas the mussel cultivation in Turkey was 5 tons in 2011 (Fao/Fishstat Plus 2016). The Black Sea in the north of Turkey was identified to be a potential *Mytilus galloprovincialis* cultivation area due to its suitable water temperature (7–25 °C) and salinity (17–20‰) values (Karayücel et al. 2015; Çelik et al. 2015). However, because mussels feed by filtering water, there are many factors to be considered for commercial mussel cultivation. The most important of these factors are the quality of the living environment, the quality of the water and the quality and reliability of the marketed product. It is necessary to evaluate environmental parameters in the environment where mussel cultivation is carried out in order to ensure a healthy production and safe consumption. As well as nutrients, these organisms intake toxic substances such as heavy metals from water (Nicholson and Szefer 2003; Wang et al. 2005; Funes et al. 2006). Even in very low concentrations, heavy metals may accumulate in excessive amounts in bivalves and consumption of these organisms may be harmful to human health (Belitz et al. 2008). Heavy metal concentrations in surface sediment reveal historical information about their inputs via river, airborne particles and municipal wastewater. Therefore, metal concentrations of sediment are a useful indicator to determine sea water quality, especially in coasts and estuaries (Kılıç and Belivermiş 2013). Suspended particulate matters (SPM), which have a concentration within the 17.3–32.2 mg L^{-1} range in sea water, play an important role in biogeochemical cycles in river mouths and seas. The element composition of SPM is affected by near deep water, entries from surface water and waste water, physicochemical activities occurring in sediment surface and water column, primary productivity, and changes occurring in sediment structure. In addition to above-mentioned factors, human-induced activities greatly contribute to metal entry as well and are seen as the main source of many elements (Pb, Cd, Cu, Cr, Ni, Mn, Fe and Zn) found in rivers and seas (Violintzis et al. 2009).

Several studies were conducted in different countries on heavy metal changes in cultivated mussels (Borchardt et al. 1988; Saavedra et al. 2004; Otero et al. 2005). Studies conducted in Turkish seas are mostly on mussels collected from natural environment (Özden 2008; Özden et al. 2009; Daş et al. 2009; Çolakoğlu et al. 2010, 2011; Türk Çulha et al. 2011; Mol and Üçok-Alakavuk 2011). Türk Çulha et al. (2008) determined Cd, Cu, Pb and Zn concentrations in mussels attempted to be cultivated with the raft system in the Black Sea. However, a comprehensive study on heavy metal concentration in mussels, suspended solids and sediment in the Black Sea is not available. The aims of this study were to (1) determine heavy metal (Cd, Cu, Pb, Zn) levels in cultivated mussels, suspended solids and sediment in submerged longline system installed for the first time in the coast of Sinop Peninsula (Black Sea, Turkey) and identify whether the region is suitable for cultivation and (2) evaluate metal levels according to upper limits set forth by national and international regulations and compare obtained data with other studies. This study is a first in that it assesses health risks caused by heavy metal from cultivated mussel consumption. To this end, we identified provisional tolerable weekly intake (PTWI) from 125 g of consumption (Jović et al. 2012; Stanković and Jović 2012; Jović and Stanković 2014; Stanković et al. 2011). Also, metal accumulation in sediment was evaluated according to sediment quality guidelines (SQG) quality criteria (Yahaya et al. 2012). Contamination factor (CF) and pollution load index (PLI) values in sediment were calculated as well. Biota-sediment accumulation factor (BSAF) was also calculated.

Materials and methods

Sampling and analytical methods

The study was performed at the Sinop Peninsula Inner Harbor (İçliman) of Turkish Black Sea coasts between March 2009 and March 2010. The region's economy is largely based on agriculture and fishing. The system used for Mediterranean mussel (*Mytilus galloprovincialis*) cultivation is found at the Sinop Peninsula Inner Harbor (420 01′30″ north–350 12′85″ east), partly shows offshore property, has a depth about 25 m depth, located under 10 m of the water surface (Fig. 1). *Mytilus galloprovincialis* samples were collected from longline system, whereas sediment samples were collected by underwater

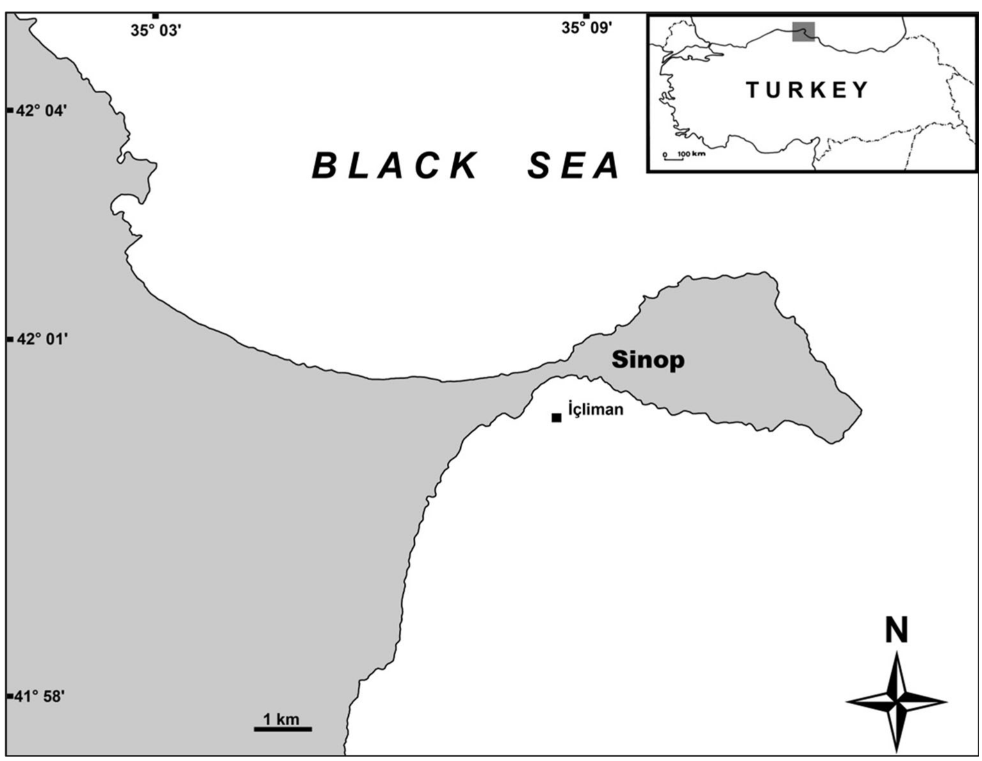

Fig. 1 Sampling sites from the Sinop Peninsula, Inner Harbor (İçliman), Black Sea

Table 1 Certified and measured values of heavy metal concentrations in reference materials ERM-CE278 mussel sediment RTC-CRMO16 (mean ± SE; dw)

Trace metals	ERM-CE278 mussel tissue	Measured (mg kg^{-1}; $n = 5$)	RTC-CRM016 sediment	Measured (mg kg^{-1}; $n = 5$)
Cd	0.348	0.301 ± 0.063	0.6	0.542 ± 0.076
Cu	9.45	9.821 ± 1.026	16.3	14.926 ± 1.072
Pb	2.00	2.280 ± 0.639	15.5	14.655 ± 7.271
Zn	83.1	74.858 ± 2.183	74.0	68.141 ± 4.237

divers on a monthly basis from the area located under longlines that mussels hang on, from 3- to 5-cm surface section of the sediment. Water samples were collected from 13 m depth where mussels hang on ropes in the system using 5-L Niskin water sampler. The samples that were provided to laboratory with a cold chain were stocked in cold storage at −21 °C until analyses were carried out. In addition, temperature, salinity, dissolved oxygen (DO) and pH values of the surface water were measured in situ using a handheld portable WTW Multi 340 i Set Model Multiparameter device during sampling from the stations.

Mytilus galloprovincialis samples were washed with tap water firstly and then bidistilled water before the analyses. The samples were dissected using a stainless steel scissor and a lancet after filtering water on blotting papers. The edible muscle tissue was homogenized for use in the analysis and dried in an oven at 105 °C for 24 h until it had a fixed weight. 10 mL of HNO_3 was added per dry weight (dw) of 1 g, and the mixture was left at room temperature overnight with a watch glass thereon. Then, these erlenmeyer flasks were heated firstly at a low temperature (40 °C) for 1 h and then at a slowly increasing temperature (140 °C) for 3 h in a fume hood on a heat-adjustable hot plate until colored vapor of the samples disappeared. 1 mL of HNO_3 was added to samples upon completion of organic degradation, and the volume was brought to 25 mL with bidistilled water (Bernhard 1976; Yap et al. 2004). The meat yield of mussels was calculated according to Lutz (1980). The sediments sampled from the stations were classified according to the particle size (Buchanan 1984), and the sediment samples smaller than 125–63 µm were used in order to determine the heavy metals in sediment. The sediment samples of 0.5 g were added from 4:1 HNO_3/$HClO_4$ Aqua regia mixture and thereafter burned firstly at a low temperature (40 °C) for 1 h and then at a slowly increasing temperature (140 °C) for 3 h in a fume hood on a heat-adjustable hot plate until colored vapor of the samples disappeared. The samples were brought to 25 mL with bidistilled water upon completion of organic degradation (Bernhard 1976; Yap et al. 2004). In addition, burnable substance amounts (%) in sediments sampled were calculated (Buchanan 1984). Metal analysis for suspended particulate matter was performed according to UNEP (1984).

The metal concentrations of the samples were measured using ICP-AES Varian Liberty Series 2. The wavelengths for metal reading were 324.754 nm for Cd, 220.353 nm for Pb, 226.205 nm for Cu and 213.856 nm for Zn, respectively. The concentration values for the *M. galloprovincialis*, sediment and suspended particulate matter (SPM) were expressed in mg kg^{-1} dry weight. The standard reference material (SRM) used to test accuracy and precision of the device and the calibration curve was analyzed initially and after analysis of each sample in triplicate. In this study, "ERM-CE278" supplied for determining heavy metals in *M. galloprovincialis* and "RTC-CRM016" supplied for the sediment samples are certified reference materials (Table 1).

Statistical analysis

The data obtained from mussel, sediment and suspended particular matters were recorded and analyzed using the SPSS statistical software version 21 to obtain the seasonal differences. In order to analyze the statistical differences among four seasons, the ANOVA test was applied. The one-way ANOVA analysis was performed at 5% of significance level using the SPSS software package in this study. However, ANOVA never gives which season(s) differ(s) from the others. The homogeneity of variances of seasons was prerequisite for applying the post hoc tests to obtain differences. The Levene statistic is probably the most commonly used test for this purpose. After applying this test, the value shows that the hypothesis on the homogeneity of variances is accepted for mussel Cd, mussel Pb, sediment Cd, sediment Pb, sediment Zn, SPM-Cu, SPM-Pb. In order to see the differences, Tukey's honestly significant difference (HSD) test for homogenous variables and Dunnet's T3 test for inhomogeneous variables are applied multiple comparison tests in statistics. Spearman correlation analysis was applied to verify existing relationships.

Biota-sediment accumulation factor (BSAF) was also calculated. BSAF is the ratio between the metal concentration in biota and that found in sediment (Abdallah and Abdallah 2008). The formula is BSAF = Corg/Csed, where Corg is the heavy metal concentration in the organism (mg kg^{-1} dw) and Csed is the heavy metal level in sediment (mg kg^{-1} dw). BSAF was calculated for concentrations recorded for *M. galloprovincialis*, suspended particulate matter and sediment. Also, the provisional tolerable weekly intake (PTWI) value defines the amount of mussel one can consume each week without any health risks and has been determined by FAO/WHO (Jović et al. 2012; Stanković and Jović 2012; Jović and Stanković 2014; Stanković et al. 2011). FAO/WHO (2007) values were used for PTWI*Zn* and PTWI*Cu*, WHO (1989) values were used for PTWI*Cd*, and FAO/WHO (2004) values were used for PTWI*Pb*.

Contamination factor (CF) was calculated according to Pekey et al. (2004). The formula is CF: Ca/Cb, where Ca is the heavy metal concentration in the sample and Cb is the shale average concentrations of the calculated metal according to Krauskopf (1979). Pollution load index (PLI) was calculated according to the following (Tomlinson et al. 1980). The formula is $PLI = (CF_1 \times CF_2 \times CF_3 \times \cdots \times CF_n)^{1/n}$. CF is metals' contamination factor and n is the number of metals analyzed in the study.

Results and discussion

Table 2 shows physicochemical parameter measurement values of water sampled from the Inner Harbor area and analysis results related to SPM, combustible organic matter in sediment and meat yield of mussels. The lowest sea water temperature was measured in spring (9.53 ± 0.92 °C) and the highest in summer (20.29 ± 3.86 °C), the lowest salinity was measured in autumn (17.37 ± 0.09‰) and the highest in summer (17.70 ± 0.05‰), the lowest pH values were measured in autumn (8.47 ± 0.07) and the highest in winter (8.74 ± 0.08), and the lowest dissolved oxygen value was measured in autumn (5.51 ± 1.25 mg L^{-1}) and the highest in spring (8.55 ± 0.16 mg L^{-1}). The annual average SPM value at the Inner Harbor was found to be 11.99 ± 0.48 mg L^{-1}. The lowest value was measured in winter (11.23 ± 0.20 mg L^{-1}) and the highest in autumn (12.63 ± 0.60 mg L^{-1}). The lowest BS value in sediment samples from the study area was seen in winter 6.42 ± 0.15% and the highest value was seen in summer 9.53 ± 0.52%. The annual average meat yield (MY) from mussels sampled from the cultivation system was 23.54 ± 1.05%. The seasonal MY variation was the lowest in winter (21.28 ± 1.30%) and the highest in summer (26.56 ± 0.63%). As a result of the statistical analysis, it was found that the variation in SPM value and BS % value in sediment between seasons was not statistically significant ($p > 0.05$). Physicochemical parameters found in the study were determined to be within limits necessary for areas where mussel cultivation is carried out

Table 2 Physicochemical, suspended particulate matter (SPM), burnable substance (BS), meat yield (MY), variables at the sampling station

Periods	Temperature (°C)	Salinity (‰)	pH	Dissolved oxygen (mg L^{-1})	SPM (mg L^{-1})	BS (%)	MY (%)
Spring	7.84–12.00 (9.53 ± 0.92)	17.03–17.77 (17.56 ± 0.18)	8.31–8.59 (8.59 ± 0.14)	8.19–8.96 (8.55 ± 0.16)	9.43–104.75 (11.90 ± 1.36)	7.02–12.72 (9.22 ± 1.77)	14.57–28.31 (23.25 ± 3.09)
Summer	12.63–24.90 (20.29 ± 3.86)	17.61–17.77 (17.70 ± 0.05)	8.60–8.69 (8.57 ± 0.08)	4.77–7.89 (6.20 ± 0.91)	11.20–13.03 (12.63 ± 0.60)	8.65–10.44 (9.53 ± 0.52)	25.31–27.33 (26.56 ± 0.63)
Autumn	15.20–22.50 (19.33 ± 2.16)	17.20–17.40 (17.37 ± 0.09)	8.35–8.60 (8.47 ± 0.07)	3.41–7.74 (5.51 ± 1.25)	12.93–13.93 (13.34 ± 0.30)	6.89–11.01 (9.21 ± 1.22)	21.94–23.82 (23.19 ± 0.63)
Winter	8.07–12.45 (10.40 ± 1.27)	17.30–17.85 (17.61 ± 0.16)	8.60–8.86 (8.74 ± 0.08)	4.66–11.4 (7.24 ± 2.10)	9.90–11.43 (11.23 ± 0.20)	6.21–6.71 (6.42 ± 0.15)	19.87–23.87 (21.28 ± 1.30)

(temperature, 12–30 °C; salinity 10–37‰; DO $\geq$ 5 mg L^{-1}; pH 6.5–8.5; SPM, 5–80 mg L^{-1}) (SÜY 2006). Temperature of the sea water is one of the most important factors affecting mussel cultivation (Okumuş and Stirling 1998; Rajagopal et al. 1998).

The correlation analysis between heavy metal concentrations found in mussel, sediment and SPM is shown in Table 3. There was a positive correlation between Cd and Cu in mussel ($r = -0.000$, $p < 0.01$) and a negative correlation between Cd in mussel and Pb in sediment ($r = 0.033$, $p < 0.05$). There was a negative correlation between Cu in mussel and Pb in sediment ($r = 0.017$, $p < 0.05$), a positive correlation between Cu in mussel and Pb in SPM ($r = 0.009$, $p < 0.01$), and a positive correlation between Zn in mussel and Pb in SPM ($r = 0.042$, $p < 0.05$). There was a positive correlation between Cd and Zn in sediment ($r = 0.045$, $p < 0.05$), a positive correlation between Cd in sediment and Zn in SPM ($r = 0.003$, $p < 0.01$), a negative correlation between Pb and Zn in sediment ($r = 0.031$, $p < 0.05$), a negative correlation between Pb in sediment and Zn in SPM ($r = 0.020$, $p < 0.05$), a negative correlation between Zn in sediment and Cd in SPM ($r = 0.007$, $p < 0.01$), and a positive correlation between Zn in sediment and Zn in SPM ($r = 0.039$, $p < 0.05$). The identified relationship among the metals shows that they come from same origin or synergic interaction amongst themselves.

Heavy metal concentration in SPM

Heavy metals concentrations in SPM sampled from Sinop Peninsula Inner Harbor were as follows: Zn (119.997 mg kg^{-1}) > Cu (0.737 mg kg^{-1}) > Pb (0.638 mg kg^{-1}) > Cd (0.052 mg kg^{-1}). The difference in Cd, Cu and Pb values between seasons was found to be statistically insignificant ($p < 0.05$), whereas the difference in Zn value was found to be significant ($p > 0.05$) (Fig. 2). Metal concentrations are reported to increase in autumn and winter due to terrestrial inputs to sea with rainfall (Niencheski and Baumgarten 2000; Kar et al. 2008). In these periods where rainfall is intense, the amount of suspended particulate matter in water increases and since it is easier for heavy metal ions to hang on these particulate matters, an increase in metal levels is observed (Dural and Göksu 2006). It was reported in several studies that heavy metal concentrations in SPM vary seasonally (Akçay et al. 2003; Turgut 2003; Türkmen and Türkmen 2004). Balkıs et al. (2010) reported that natural and anthropogenic pollution was carried from land to sea with increased river input especially in periods with high rainfall in Gulf of Gökova, which may have caused to an increase in Pb and Cd concentrations. Similarly, Dural and Göksu (2006) stated that the highest Cd concentration was observed in winter and metal accumulation usually occurred in winter and rainy spring months. In another study, the authors reported high Cd, Pb, Cu and Zn concentrations in Gulf of İskenderun in winter (Türkmen and Türkmen 2004). In our study, Zn concentration was found to be high in winter as well. Heavy metal values found by Balkis et al. (2007) for the coast of Sinop (Cd: 0.13–0.50 mg kg^{-1}, Cu: 0.26–0.40 mg kg^{-1}, Pb: 0.01–1.5 mg kg^{-1}) are considerably higher compared to the data from Sinop Inner Harbor. When heavy metal concentrations in SPM measured for different seas are considered, concentrations determined by Violintzis et al. (2009) in Gulf of Thermaikos (Cd: 0.1–66 mg kg^{-1}, Cu: 32–81 mg kg^{-1}, Pb: 38–223 mg kg^{-1}; Zn: 60–244 mg kg^{-1}), by Niencheski and Baumgarten (2000) in Patos Lagoon (Cu:

Table 3 Correlation coefficient (r) of mussel, sediment and SPM metals

	M-Cd	M-Cu	M-Pb	M-Zn	S-Cd	S-Cu	S-Pb	S-Zn	SPM-Cd	SPM-Cu	SPM-Pb	SPM-Zn
M-Cd	1.00	0.86**	0.37	0.46	0.18	−0.17	−0.62	0.13	0.08	−0.30	0.61*	0.25
M-Cu		1.00	0.19	0.53	0.36	−0.06	−0.67*	0.11	0.12	−0.33	0.72**	0.49
M-Pb			1.00	−0.04	−0.09	−0.01	−0.11	−0.35	0.10	−0.42	0.17	0.13
M-Zn				1.00	0.47	0.46	−0.18	0.06	0.23	0.04	0.59*	0.12
S-Cd					1.00	0.15	−0.43	0.59*	−0.41	0.09	0.15	0.77**
S-Cu						1.00	0.49	−0.29	0.32	0.09	−0.11	−0.04
S-Pb							1.00	−0.62*	0.46	0.17	−0.49	−0.66*
S-Zn								1.00	−0.73**	0.11	−0.35	0.60*
SPM-Cd									1.00	−0.29	0.34	−0.43
SPM-Cu										1.00	−0.51	−0.26
SPM-Pb											1.00	0.19
SPM-Zn												1.00

** Correlation significant at the 0.01 level (two-tailed)

* Correlation significant at the 0.05 level (two-tailed)

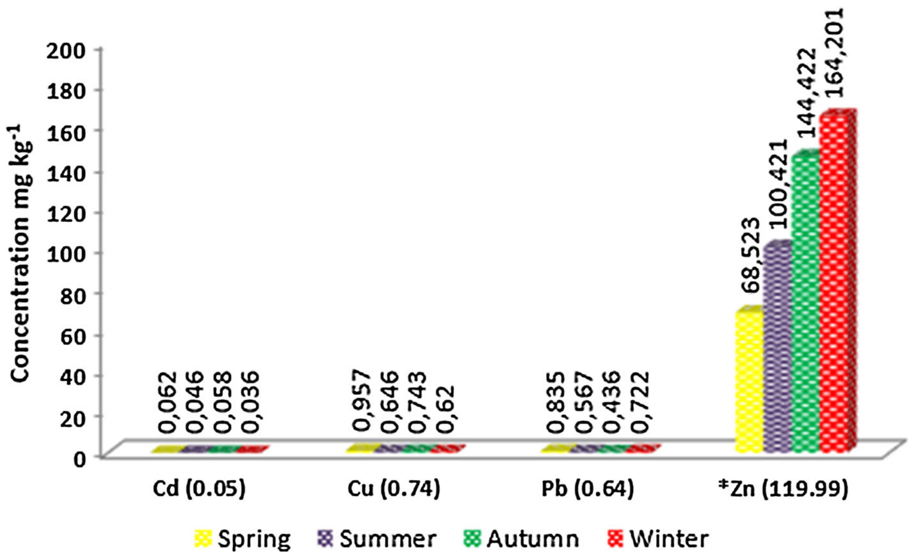

Fig. 2 Heavy metal concentrations (mg kg^{-1} dw; mean ± SE) values in SPM during samples among periods (*asterisk* shows statistical differences among periods with $p < 0.05$)

0.40–216.7 mg kg^{-1}, Pb: 1.3–65.7 mg kg^{-1}; Zn: 2.7–489.7 mg kg^{-1}) and by Kamidis et al. (2004) in Gulf of Kavala (Cd: 4.4 mg kg^{-1}, Cu: 24 mg kg^{-1}, Pb: 37.6 mg kg^{-1}; Zn: 173 mg kg^{-1}) are considerably higher from the data of this study. The reason of determining different values for heavy metal concentrations in a number of previous studies is associated with varying pollutant resources (industrial, agricultural and urban activities) of territorial environments surrounding the study fields (Altuğ and Güler 2004; Çevik et al. 2008; Balkıs et al. 2010; Ayas et al. 2009). It was found that heavy metal concentrations in SPM were considerably higher than national standard values (Cd: 0.01 mg L^{-1}, Cu: 0.01 mg L^{-1}, Pb: 0.1 mg L^{-1}, Zn: 0.1 mg L^{-1}) (SKKY 2004).

Heavy metal concentration in sediment

Heavy metal concentrations in sediment sampled from the Inner Harbor station were as follows: Zn (41.876 mg kg^{-1}) > Cu (19.454 mg kg^{-1}) > Pb (7.881 mg kg^{-1}) > Cd (5.949 mg kg^{-1}). The seasonal distribution was winter > autumn > spring > summer. The difference in Cd, Cu and Pb values between seasons was found to be statistically significant ($p < 0.05$). No significant difference was found in Zn concentration ($p > 0.05$) (Fig. 3). Considering the seasonal distribution of heavy metal concentrations in sediment samples in the present study, they were generally higher in winter and autumn and lower in summer and spring months. Sewage and domestic wastes discharged from the coastal area of Sinop are directly canalized to the sea from three different points in autumn and winter, which is believed to be one of the most important human-induced pollution factors. Metal concentration values found by Topçuoğlu et al. (2002) at the coast of Sinop were Zn 91.5 mg kg^{-1}, Cd 0.89 mg kg^{-1}, Cu 37.3 mg kg^{-1} and Pb 15.1 mg kg^{-1}. Only the Cd concentration is lower than the data in the present study. Concentrations found by Bat et al. (2015) were Zn 13.8–19.3 mg kg^{-1}, Cd 0.04–0.06 mg kg^{-1}, Cu 5.09–7.24 mg kg^{-1} and Pb 6.32–7.12 mg kg^{-1}. Compared to these values, concentrations found by Topçuoğlu et al. (2002) seem to be much higher except for Cd. In a study conducted at the coastal area of the Aegean Sea, high heavy metal contamination was reported at sewage discharge points (Aloupi and Angelidis 2001). One of the reasons behind high Zn concentration at the Sinop Inner Harbor is the increase in organic matter content of sediment. Increased population in summer (Türk Çulha et al. 2008; Bat et al. 2009, 2015), sewage discharge at the coastal area and terrestrial inputs (Bat et al. 2012; Bat and Gökkurt-Baki 2014) are believed to be effective in this increase. The high amount of BS found in summer supports this idea as well. The annual average BS value in sediment sampled from the Inner Harbor is 8.60 ± 0.61%. These results are higher than BS values (0.4–2.0%) found by Bat et al. (2001), but lower than BS values (16.8%) found for the Southern Aegean Sea (Aydın and Sunlu 2004). The study area (Sinop) is highly affected by winds and currents. It can be said that terrestrial organic matters affect the marine environment. Also, BS values are found to be quite high, since the region has high human activity and vessel traffic. Similar results were obtained by Aydın and Sunlu (2004) as well.

Sediment quality guidelines (SQG) values and obtained results were compared in order to determine whether heavy metal found in sediment samples from the Inner Harbor had any adverse effects (Yahaya et al. 2012). It was found that the Cd concentration was considerably high (Table 4).

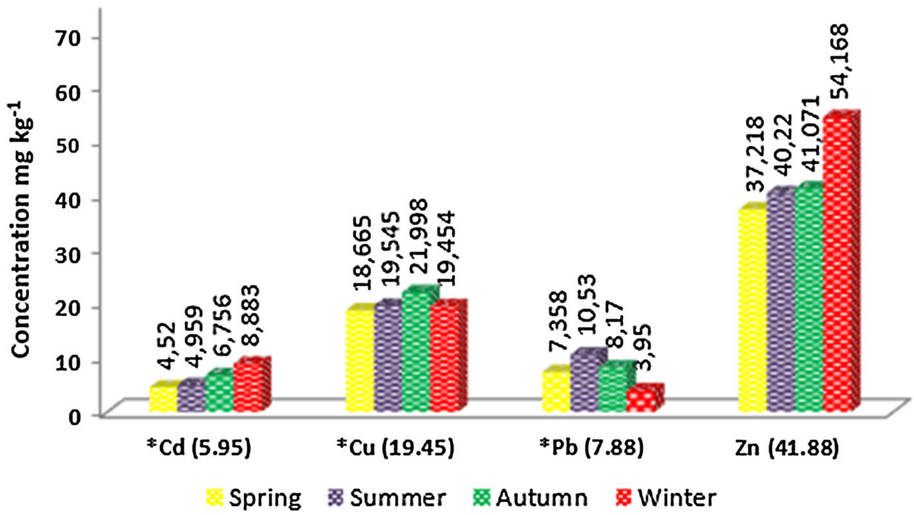

Fig. 3 Heavy metal concentrations (mg kg^{-1} dw; mean ± SE) values in sediment during samples among periods (*asterisk* shows statistical differences among periods with $p < 0.05$)

Table 4 Comparison of results obtained with world shale value and sediment quality guideline values (mg kg^{-1}; dw)

Metal	Cd	Cu	Pb	Zn
Earth's background values				
WS	0.3	45	20	95
US	0.3	33	19	95
Guideline value				
ISQG	0.6	35	35	123
PEL	3.5	197	91.3	315
LEL	0.6	16	31	120
SEL	10	110	250	820
Present study	5.949	19.454	7.881	41.876

WS world shale value, *US* unpolluted sediments, *ISQG* interim sediment quality guideline, *PEL* probable effect level, *LEL* lowest effect level, *SEL* severe effect level (Yahaya et al. 2012)

The CF value was found to be 19.8 for Cd in sediment, and the PLI value calculated using CF was found to be 1.09. Hakanson (1980) notes that if CF ≥6 value is higher in sediment, there is a significant increase in pollution in the environment. It can be said that the amount of Cd heavy metal in the environment increased at the Sinop Peninsula Inner Harbor due to anthropogenic factors. It was reported in other studies conducted at the coastal area of Sinop that the pollution in the area was caused by sewage and domestic waste discharged without treatment (Topçuoğlu et al. 2002; Bat et al. 2009, 2012, 2015; Türk Çulha et al. 2008; Bat and Gökkurt-Baki 2014).

Heavy metal concentration in *M. galloprovincialis*

Seafood is an important source of minerals for people. Mussels, rich in nutrients, feed by filtering the water and thus accumulate higher amounts of heavy metal compared to their environment (Oliver et al. 2001). With this property of mussels, they provide important data about pollutants in the marine life and adverse effects of these pollutants on human health can be determined by evaluating measurement results. The annual average Cd, Cu, Pb and Zn heavy metal concentrations in mussels cultivated in this study were, respectively, as follows: 1.182 ± 0.112, 4.774 ± 0.297, 1.081 ± 0.138 and 84.730 ± 4.044 mg kg^{-1}. It was also found that metals showed seasonal variations except for Zn ($p > 0.05$) (Fig. 4). Türk Çulha et al. (2008) found the highest metal accumulation in mussels in the same region in autumn. In another study, it was reported that many elements except for Cd were in high concentrations in spring, autumn and summer (Vlahogianni et al. 2007). Seasonal variations in metal concentrations may occur due to physiological changes in the organism rather than exposure to metals (Mubiana et al. 2005). It is also affected by the reproductive cycle involving the maturation of gametes and changes in the amount of available nutrients (Bryan 1976; Usero et al. 1997; Adami et al. 2002; Ansari et al. 2004; Cardellicchio et al. 2008). Meat weight, which depends on gonadal development and nutrition conditions in mussels, is a value used for determination of metal concentration. Amount of nutrients, filtration rate, seasonal meat weight changes due to gonadal development affect metal concentration rate and may cause metal concentration in mussel to seem increased. Borchardt et al. (1988) note that low condition value in mussels causes higher metal concentrations, which is observed in seasons when ovulation occurs. For this reason, it is important to evaluate metal concentrations in mussels taking organisms' physiology into account.

It was found in this study that the meat yield, which was 28.31% in April 2009, dropped to 14.57% in May. Similarly, Çelik et al. (2012) found the minimum mussel meat yield in the same period in the same region. This period

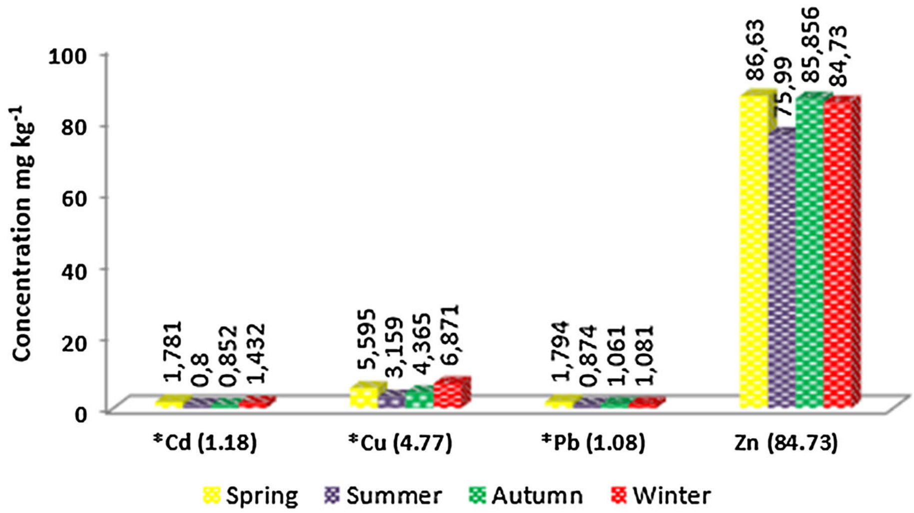

Fig. 4 Heavy metal concentrations (mg kg^{-1} dw; mean ± SE) values in mussel during samples among periods (*asterisk* shows statistical differences among periods with $p < 0.05$)

Table 5 A comparison of reported concentrations (mg kg^{-1}, dw) of Cd, Cu, Pb and Zn in *M. galloprovincialis* from Sinop coasts (Black Sea) and different sea coasts studies with the present results

Areas	Cd	Cu	Pb	Zn
Sinop coast (Bat et al. 1999)	0.03–0.27	0.10–1.89	0.11–1.18	1.58–7.28
Sinop coast (Topçuoğlu et al. 2002)	1.79	8.01	0.31	256.4
Sinop/İçliman (Türk Çulha et al. 2008)	2.53	6.98	–	228
Sinop coast (Bat et al. 2012)	0.27–0.98	2.41–4.82	2.10–4.10	79–163
Sinop coast (Belivermis et al. 2016)	0.55	25.5	0.51	70
Dardanelles Strait/North Aegean (Çayır et al. 2012)	0.20–1.59	4.2–12.0	1.8–6.0	139–319
Boka Kotorska Bay/Montenegro (Jović et al. 2012)	0.33–0.48	1.05–1.24	0.64–0.73	27.9–34.5
Oran Harbor/Algeria (Rouane-Hacene et al. 2012)	0.67	3.63	10.67	89.29
Sinop/İçliman (present study)	1.182 (0.089–3.638)	4.774 (0.334–1.827)	1.081 (0.002–3.872)	84.730 (9.385–182.616)

corresponds to the ovulation period. It was reported by researchers that meat yield in mussels is closely related to nutrient reserve, maturation of gonads and ovulation and the highest meat yield is obtained just before the ovulation period (Çelik et al. 2012; Karayücel et al. 2015). It is noted that meat yield declines in mussels after ovulation (Okumuş and Stirling 1998). In our study, Zn and Cu values found in mussel tissues were found to be high in winter. This increase is believed to be associated with the decline in meat weight. Cd and Pb heavy metal values were measured to be high in spring. Mussels want to refill nutrient reserves emptied due to ovulation during spring when nutrients are abundant in the water by increasing their filtration rate (Bayne et al. 1993). It is believed that higher amount of Cd and Pb in the environment compared to other seasons accumulates more in the structure of the organism as a result.

It was noted in another study conduct by Bat et al. (1999) at the coastal area of Sinop that Zn concentration was higher in mussels collected in early spring and early summer compared to other metals, Cd accumulated at peak levels, and this accumulation was at its highest in mussels sampled from the Inner Harbor. Similar to this finding, Cd concentration was found to be above national and international limit values in our study as well. It is seen that heavy metal concentrations in mussels sampled from the longline system are higher than results of Bat et al. (1999) and Daş et al. (2009) for all metals, higher than results of Topçuoğlu et al. (2002) for Zn and Cu concentrations and lower than results for Pb and Cd. However, our results are considerably lower than metal concentrations (2.53 mg kg^{-1} Cd; 6.98 mg kg^{-1} Cu and 228 mg kg^{-1} Zn) in mussels sampled from the raft system installed in the coastal area of Inner Harbor by Türk Çulha et al. (2008). The reason behind this difference is that the study was conducted in the coastal area, exposed to human-induced pollution due to discharge of unrefined domestic waste in this area, harbor activities, waste discharge by ships and coastal activities (Bat et al. 1999; Türk Çulha et al. 2008). This leads to a higher metal accumulation in mussels.

Table 5 shows the results of studies on heavy metal concentrations in mussels sampled from the Black Sea and seas in different countries. It is seen that different metal concentration values are found depending on the region. These differences result from properties of terrestrial environments surrounding regions, mussels sampled from different stations, different nutrient content due to different sediment structures in stations, physiological attitude of mussels, concentrations of active metals in sampling regions, inputs due to land and sea traffic and anthropogenic factors.

Bioaccumulation of metals by organism occurs if the BSAF is >1 (Aydin-Önen et al. 2015). The efficiency of metal bioaccumulation in *M. galloprovincialis* was evaluated by calculating the biosediment accumulation factor (BSAF), which is defined as the ratio between the metal concentration in the organism and that in the sediment (Lau et al. 1998). BSAF values found in the study were 0.20 for Cd, 0.25 for Cu, 0.14 for Pb and 2.02 for Zn. The highest BSAF value was detected in Zn concentration with 2.02. *M. galloprovincialis* is a good indicator species to determine Zn heavy metal in the environment (Besada et al. 2002; Rainbow 2002; Topçuoğlu et al. 2002; Saavedra et al. 2004).

Provisional tolerable weekly intake in *M. galloprovincialis*

Cd is a non-base toxic metal found in low concentrations in nature. High Cd levels are usually a result of anthropogenic factors (Stanković et al. 2011). For Cd, the PTWI value was established as 0.007 mg kg^{-1} body weight $week^{-1}$ (0.49 mg $week^{-1}$ for a 70-kg adult) (WHO 1989). The highest value measured for Cd was 1.781 mg kg^{-1}. The value calculated for weekly 125-g mussel consumption was 0.22 mg (44.9%). Considering the highest Cd concentration in mussels, PTWI was 0.28 kg of mussel for Cd. The PTWI value for Pb is 25 µg kg^{-1} body weight $week^{-1}$ (1.75 mg $week^{-1}$ for a 70-kg adult) (FAO/WHO 2004). The value calculated for weekly 125-g mussel consumption was 0.22 mg (12.6%) for Pb (1.794 mg kg^{-1}). Considering the highest Pb concentration in mussels, PTWI value was 0.98 kg of mussel for Pb. Zn is abundant in bivalves and although it is an important heavy metal for nutrition, it may be harmful for human health when a certain limit value is exceeded (Plum et al. 2010; Jović et al. 2012). The PTWI was established as 7 mg kg^{-1} body weight $week^{-1}$ for Zn, which is equivalent to 490 mg $week^{-1}$ for a 70-kg adult (FAO/WHO 2007). The highest Zn concentration found in the study was 93.439 mg kg^{-1}. The value calculated for weekly 125-g mussel consumption was 85.62 mg $person^{-1}$ $week^{-1}$, which represents 2.16% of the Zn PTWI value. Considering the highest Pb concentration in mussels, the amount of mussel needed to exceed PTWI Zn value was 5.2 kg. Cu is an element found in large quantities in shellfish. Although it is a basic element for human health, high accumulation may be highly toxic similar to Zn (Gorell et al. 1997). For Cu, the PTWI value was established as 3.5 mg kg^{-1} body weight $week^{-1}$ (245 mg $week^{-1}$ for a 70-kg adult) (FAO/WHO 2007). The highest value measured for Cu was 6.871 mg kg^{-1}. A weekly 125-g mussel consumption corresponds to weekly 0.86 mg of Cu (max 6.871 mg kg^{-1}), which is 24.6% of the PTWI Cu value. Considering the highest Cu concentration in mussels, the amount of mussel needed to exceed PTWI Cu value was 35.7 kg. These results show that none of the elements exceeded the tolerable limit value.

Conclusion

When concentration values found in the study are compared to consumable limit values determined for mollusk species, Cd concentration seems to be high according to Turkish Food Codex (2008) Cd: 1.0 mg kg^{-1}, Pb: 1.5 mg kg^{-1}, EU (2008) Cd: 0.05–1.0 mg kg^{-1}, Pb: 1.5 mg kg^{-1} and FDA (2001) Cd: 0.2 mg kg^{-1}, Pb: 1.5 mg kg^{-1}, Cu: 100 mg kg^{-1} and Zn: 150 mg kg^{-1}. Relations found between metals are believed to arise from metals' coming from similar sources or the same source or synergistic interactions between metals. It was found that SPM level in mussel and sediment had a significant correlation. It was determined as a result of positive correlations between metals that metal pollution in the water arose from the same source (Rubio et al. 2000). At the end of the study, Cd concentrations in mussels were found to be higher than limit values determined by TFC (2008), EU (2008) and FDA (2001). It was identified that metal concentrations in SPM exceeded standard values and Cd concentration in sediment was considerably high according to the CF value. The fact that Sinop Inner Harbor was under the effect of anthropogenic pollution in terms of Cd accumulation is also supported by PLI results. In conclusion, it can be said that this region considered for commercial mussel cultivation is not a suitable area.

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