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RESEARCH ARTICLE

Investigation of heavy metal pollution in eastern Aegean Sea coastal waters by using *Cystoseira barbata*, *Patella caerulea*, and *Liza aurata* as biological indicators

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Abstract In order to have an extensive contamination profile of heavy metal levels (Cd, Cu, Fe, Mn, Ni, Pb, and Zn), seawater, sediment, *Patella caerulea*, *Cystoseira barbata*, and *Liza aurata* were investigated by using inductively coupled plasma-atomic emission spectrometry (ICP-AES). Samples were collected from five coastal stations along the eastern Aegean Sea coast (Turkey) on a monthly basis from July 2002 through May 2003. According to the results of this study, heavy metal levels were arranged in the following sequence: Fe > Pb > Zn > Mn > Ni > Cu > Cd for water, Fe > Cu > Mn > Ni > Zn > Pb > Cd for sediment, Fe > Zn > Mn > Pb > Ni > Cd > Cu for *C. barbata*, Fe > Zn > Mn > Ni > Pb > Cu > Cd for *P. caerulea*, and Fe > Zn > Mn > Cu > Ni > Pb > Cd for *L. aurata*. Moreover, positive relationships between Fe in water and Mn in water, Fe in sediment and Mn in sediment, Fe in *C. barbata* and Mn in *C. barbata*, Fe in *P. caerulea* and Mn in *P. caerulea*, and Fe in *L. aurata* and Mn in *L. aurata* may suggest that these metals could be originated from the same anthropogenic source. *C. barbata* represented with higher bioconcentration factor (BCF) values, especially for Fe, Mn, and Zn values. This observation may support that *C. barbata* can be used as an indicator species for the determinations of Fe, Mn, and Zn levels. Regarding Turkish Food Codex Regulation's residue limits, metal values in *L. aurata* were found to be lower than the maximum permissible levels issued by Turkish legislation and also the recommended limits set by FAO/WHO guidelines. The results of the investigation indicated that *P. caerulea*, *L. aurata*, and especially *C. barbata* are quantitative water-quality bioindicators and biomonitoring subjects for biologically available metal accumulation for Aegean Sea coastal waters.

Keywords Heavy metals · Sediment · *Cystoseira barbata* · *Patella caerulea* · *Liza aurata* · Aegean Sea

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Introduction

Among the different types of pollution, heavy metal contamination in the marine environment has become one of the world's major environmental problems and causes serious effects to humans and biota. In the aquatic environment, metals are readily accumulated by aquatic organisms and they transferred through food webs to fish, other piscivorous animals, and humans, and they may affect organisms directly by accumulating in the body or indirectly by transferring to the next trophic level of the food chain (Chen et al. 2000; Prouty et al. 2010). Moreover, they are toxic to aquatic organisms and cause their lethal or sublethal deterioration (Wang et al. 2005). Therefore, monitoring of heavy metal pollutants in the aquatic environment across a variety of sources such as water, sediment, and biota is essential. For this purpose, determination of heavy metal levels by using different indicator organisms, such as algae, mollusks, and fish, which have a greater spatial ability to accumulate some pollutants when compared with water column and bottom sediments can give more reliable information about the status of metal contamination in coastal waters.

In this respect, macroalgae have the ability to accumulate heavy metals at concentrations several orders of magnitude

above those observed in seawater (Conti 2002). Their wide distribution and abundance in the sampling stations could be used in determining their bioaccumulating potential of heavy metals and provide useful information about the levels of metal contamination and environmental quality of an area (Lobban and Harrison 1994). In this study, the brown alga *Cystoseira barbata* was selected as a study organism for obtaining evidence supporting *C. barbata* as an appropriate candidate to be employed in biomonitoring studies for heavy metal pollution in Aegean Sea waters.

Various filter feeding bivalve and gastropod species were abundant in coast sites and play an important role as biomonitors for heavy metal contamination in monitoring programs (Sericano 2000). The gastropod mollusks have the ability to take up metals from different sources either from the aqueous medium or through ingestion from food and inorganic particulate material, and then they heavily concentrate them (Phillips 1977). One of the herbivorous gastropods *Patella caerulea* which fulfills the criterion as being an ideal bioindicator for heavy metal contamination is extensively used in marine monitoring programs (Bu-Olayan and Thomas 2001; Campanella et al. 2001; Storelli and Marcotrigiano 2005). Therefore, in this study, it was used to assess heavy metal pollution levels of the sampling area.

Fish play an important role in human nutrition, and they have the ability to accumulate heavy metals in their muscles and different organs. In order to ensure that high levels of some toxic heavy metals are being transferred to man through fish consumption, they need to be carefully monitored (Olowu et al. 2010). Our study organism *Liza aurata* (*Mugil auratus*) is a good indicator of heavy metal pollution (Filazi et al. 2003; Storelli et al. 2006), and it has also a great impact on human health because it is consumed by the local population.

In this study, for assessing the degree of heavy metal contamination of five stations selected from coast sites along the Aegean Sea, valuable bioindicators such as brown algae *C. barbata*, herbivorous gastropod molluscs *P. caerulea*, and the edible fish species *L. aurata* have been utilized. The results of this study were determined for the first time in this environment. So in order to fully understand and gain valuable information of the study area, heavy metal levels in seawater and sediment samples collected from the same sampling stations were also determined. Heavy metal levels in seawater samples were also used to assess the contamination factors (bioconcentration factors, BCFs) for evaluation of biota's accumulation capability. Moreover, the correlations between metal concentrations in seawater and sediment, between biota and seawater, between biota and sediment, and between marine organisms were statistically examined. Additionally, a comparison of our results with previous studies conducted in other coastal environments is done to assess the degree of metal contamination.

Material and methods

Study area

Seawater, sediment, and biota samples such as *C. barbata*, *P. caerulae*, and *L. aurata* were collected from a total of five stations with different contamination degrees in the coastal area of the eastern Aegean Sea (Turkey) on a monthly basis from July 2002 through May 2003. As shown in Fig. 1, the sampling stations were chosen in order to study spatial trends and to compare different heavy metal concentrations caused by pollution. The sampling stations are exposed to the different sources of pollution including domestic drainage, fishing boat activities, and tourism facilities.

The first sampling station S1 is situated at Sığacık Bay. There are several aquaculture farms in this area where species such as gilthead sea bream and sea bass are cultured. The second sampling station is positioned at Eski Liman which is close to Çayağzı Stream. Moreover, Tekgıda-İş Sendikası recreational facilities and other touristic facilities rank nearby this station. The third sampling station is found at Doğanbey which is a tourist area, and its population increases especially in the summer period. The fourth sampling station S4 is at Ürkmez, where Tahtalı Stream's waters flow in it. This station is also influenced by the domestic sewage from unconnected areas adjoining the shore. The last sampling station S5 is located at Gümüldür; it is subjected to heavy influence from the Şeytan Stream. In this region, DSI rest camps, hotels, and various facilities take place. In addition, during summer, this beach is the site of intense tourism activities (Fig. 1).

Sample collection

Surface water samples were collected with precleaned polyethylene bottles. Sampling bottles were immersed about 10 cm below the water surface. About 1 l seawater was taken at each sampling site, and then they were filtered through acid-cleaned 0.45-μm membrane filters, acidified with 1 ml HCl, and stored at 4 °C for soluble metal analysis. To minimize risks of sample contamination, all the precautions recommended by Kremling et al. (1983) were followed during collection and treatment of samples.

Sediment samples were taken from the same sampling sites. They were collected from the upper layer (0–5 cm) and placed in polyethylene bags, and then the samples were transported to the laboratory.

At each station, 10 samples of *C. barbata* species were handpicked in the subtidal zone and care was taken to choose thalli all at a similar stage of development. The samples were rinsed with seawater and transferred to polyethylene bags. In the laboratory, they were thoroughly cleaned, and any epiphyta and sediments were carefully removed under tap water.



Fig. 1 Map of the study area showing the locations of five sampling stations. *1* Sığacık (S1); *2* Eski Liman (S2); *3* Doğanbey (S3); *4* Ürkmez (S4); *5* Gümüldür (S5) (Altun 2008)

To allow a more precise comparison of metal concentrations among stations and to reduce possible variations in metal concentrations due to size and age, special attention was paid to collecting *P. caerulea* specimens (Nakhlé et al. 2006). About 20 individuals within a very narrow weight and size range were handpicked in the tidal zone at the same depth and distance from the shoreline. And they were transported to the laboratory in polyethylene bags. The soft tissues were separated from the shells with plastic equipment, rinsed with Milli-Q water, and weighed, and the samples were placed under refrigeration at −20 °C pending analysis.

It is important to note that fish were not found at all sites throughout the year. For this reason, *L. aurata* individuals were caught from only three stations (S1, S2, and S5) in July 2002, August 2002, September 2002, and October 2002. A total of five fish were obtained and stored in prewashed polyethylene bags in ice and brought to the laboratory. Edible muscle tissue of fish samples was dissected by using clean equipment, weighed, and then preserved in polyethylene bags, and all studied samples were deep-frozen at −20 °C until treatment.

Analytical procedure

In order to avoid metal contamination, lab treatments were done using non-metal tools. The seawater samples were preconcentrated before analysis with the ammonium 1-pyrrolidinedithiocarbamate (APDC) complexation method (Campanella et al. 2001; Conti and Cecchetti 2003).

Sediment samples were dried (40 °C) to constant weight, homogenized, and reduced to a fine powder by using a sieve (63 μm). For determining total heavy metal levels in sediment, 0.1–0.2 g of finely powdered and dried sample was digested in a microwave digestion system with a mixture of HF/$HClO_4$/HCl (1:1:6) (Gey and Mordoğan 1988).

Algal samples for each station were dried (40 °C) to constant weight, homogenized in a porcelain mortar, and then reduced to a fine powder. In the laboratory, three subsamples of about 1 g dry weight (d.w.) were digested in a microwave digestion system (Milestone 1200) with HNO_3/$HClO_4$(5:1) acid mixture solution (Denton and Burdon-Jones 1986).

P. caerulea specimens were used to prepare a pooled sample to reduce individual variations in heavy metal concentrations. For the determination of heavy metal samples, 5 g of wet sample was placed in flasks and dried to constant weight and then weighed again. Subsequently, 12 ml of concentrated HNO_3/$HClO_4$ (5:1) was added to each flask and the solution was evaporated to dryness on a hot plate. After allowing the flasks to cool, 10 ml of N/10 HCl (per 1 g fresh weight) was added and filtered through Whatman filter paper with a pore size of 0.45 μm (Bernhard 1976).

The muscles of fish were pooled and homogenized and 5 g of tissue was weighed, and then they were dried to constant weight. After then, samples were weighed again and 10 ml of concentrated HNO_3 was added to each flask, and the solution was evaporated to dryness on a hot plate. Then, 10 ml HCl was added to each flask again and the solution was evaporated to dryness on a hot plate again. After allowing the flasks to cool, 10 ml of N/10 HCl (per 1 g fresh weight) was added and filtered through Whatman filter paper with a pore size of 0.45 μm (Bernhard 1976). The obtained data for metal concentrations were expressed on the basis of the dry weight (μg/g dw) for sediment, algae, the soft tissues of *P. caerulea*, and *L. aurata* samples.

All chemicals used in sample treatments were of ultrapure grade (Merck Suprapur). Ultrapure water (Milli-Q System, Millipore) was used for all solutions. All glassware was cleaned prior to use by soaking in 10% *v*/*v* HNO_3 for 24 h and rinsed with Milli-Q water. The standard solutions of metals were prepared from stock standard solutions of ultrapure grade supplied by Merck.

Determination of the elements in all studied samples was carried out by ICP-AES (Varian Terra Model Liberty II). Spectral line emission measurements and limits of detection (LODs) for the ICP-AES are given in Table 1. The accuracy of analytical procedure was checked by analyzing the standard reference materials (water: SRM-143d, National Institute of Standards and Technology; sediment: CRM-277, Community Bureau of Reference; fish: DORM-2, National Research Council). Recovery rates ranged from 79 to 96% for all the elements investigated.

Table 1 Spectral line emission measurements and limits of detection (LODs)[a] for the ICP-AES

	Wavelength (nm)	LOD (μg/l)
Cd	228.8	0.001
Cu	324.8	0.014
Fe	248	0.07
Mn	279	0.05
Ni	232	0.05
Pb	217	0.005
Zn	213.9	0.1

[a] Calculated on the basis of 15 determinations of the blanks as 3 times the standard deviation of the blank

Statistical analysis

Normality and homogeneity of variance tests were carried out by using Shapiro-Wilk, Lilliefors (Kolmogorov-Smirnov), and Levene tests, and transformation of data was conducted when necessary. If data were found to be non-normally distributed, non-parametric tests were preferred for the comparisons of changes in metal concentrations. One-way analysis of variance (ANOVA) (parametric data) and the Kruskal-Wallis test (non-parametric data) were used to find any significant differences in metal concentrations among different stations and seasons. The Tukey (parametric data) and the Mann-Whitney *U* tests (non-parametric data) were used to discriminate significant differences.

Spearman correlation analysis was applied to verify existing relationships (between metal contents in seawater; between metal concentrations in seawater and metal levels in sediment; between metal concentrations in seawater and metal levels in biota; between metal values in sediment and metal contents in biota; and also between heavy metal levels in biota). The data analysis was performed using the STATISTICA v.7.1 (STATSOFT) software package, and statistical significance was defined at the $p < 0.05$ level.

Bioconcentration factors (BCFs) are defined as the ratio of the metal concentrations in biota, expressed in micrograms per gram of dry weight (dw) soft tissue, to the metal concentrations, expressed by micrograms per liter of seawater (Abdallah and Abdallah 2008). The formula is BCFs = C_{org}/C_{water}, where C_{org} is the heavy metal concentration in the organism (μg/g) and C_{water} is the heavy metal level in the water sample (μg/l). BCFs were calculated for each analyte (heavy metal levels in water and *C. barbata*, in water and *P. caerulea*, in water and muscle of fish) for each month using the equation.

Results

In the present study, the mean ± standard deviations (SD) of heavy metal concentrations (Cd, Cu, Fe, Mn, Ni, Pb, and Zn) in coastal water samples, sediments, algae, *C. barbata*, the whole soft tissue of *P. caerulea*, and muscle of *L. aurata* are presented in Tables 2, 3, 4, 5, and 6.

Heavy metals in water

This study showed that water samples were contaminated with different levels of heavy metals. The results clearly indicated that the concentrations of Fe, Pb, and Zn in water were generally higher than the other studied metals (Table 2). Moreover, the overall mean heavy metal levels in water collected from five sampling stations could be arranged in the following sequence: Fe > Pb > Zn > Mn > Ni > Cu > Cd. In this study, the peak concentrations of heavy metals were generally observed during winter, except Cu and Ni. Concerning the stational variation, the highest concentrations were recorded at stations S5 and S4 (Table 2).

With the exception of station S4, the higher Cd values were examined during summer months. Moreover, the lowest mean Cd concentration was noted at station S1 (1.00 ± 0.02 μg/l) in November 2002 and at station S3 in January 2003 (1.00 ± 0.03 μg/l) and the highest average Cd level (5.00 ± 0.09 μg/l) was measured at station S4 in December 2002 (Table 2). Regarding Cd levels in water, no statistically significant spatial and temporal differences were found.

The present data indicate that recorded Cu contents in seawater samples during May 2003 exhibited elevated levels at all stations, except station S4. Moreover, the minimum mean Cu level in seawater (1.20 ± 0.06 μg/l) was detected at station S3 in February 2003, whereas the peak mean Cu value (71.0 ± 0.8 μg/l) was recorded at station S4 in July 2002 (Table 2). According to the results, no significant stational changes ($p > 0.05$) were observed in Cu levels in water, while remarkable significant variations ($p < 0.001$) especially for noted values in May 2003 were detected.

As regards Fe in seawater, the recorded values were not always consistent at all stations and the lower average Fe levels were noticed at station S3 in October 2002 (35.0 ± 3.8 μg/l) and February 2003 (35.0 ± 4.7 μg/l), while the highest mean Fe value (372 ± 42 μg/l) was obtained at station S4 in January 2003 (Table 2). Furthermore, statistically significant differences ($p < 0.05$) among stations were exhibited. According to the findings, it can be said that station S3 was different from station S5. However, no statistically significant changes were noted among seasons.

The manganese content of seawater samples at all stations did not vary considerably over the 11-month period, except for the recorded levels at station S5. During all sampling months, measured mean Mn levels in seawater at station S5 were generally represented with considerably higher values (Table 2) and were significantly different from other stations ($p < 0.001$). In addition, the lower mean Mn concentrations in seawater were measured at station S1 in November 2002

Table 2 Dissolved heavy metal concentrations in coastal seawater samples (μg/l) (mean ± SD)

Seawater	Cd					Cu					Fe				
Months	S1	S2	S3	S4	S5	S1	S2	S3	S4	S5	S1	S2	S3	S4	S5
July 2002	4.70±0.01	4.50±0.02	4.50±0.04	3.20±1.01	3.20±0.04	6.30±0.05	5.70±1.12	6.60±0.91	71.0±0.8	4.30±0.05	110±18	359±52	62.0±6.1	106±22	253±42
August 2002	2.00±0.03	3.20±0.04	1.50±0.03	2.40±0.04	4.50±0.03	3.80±0.02	4.50±0.05	2.90±0.08	3.20±0.06	3.20±0.02	62.0±9.3	85.0±9.6	72.0±6.8	110±19	217±36
September 2002	2.90±0.05	2.40±0.03	2.60±0.03	1.70±0.05	4.00±0.03	2.10±0.06	1.60±0.02	1.30±0.06	1.70±0.08	6.20±0.06	215±36	87.0±10.8	53.0±4.7	57.0±4.6	71.0±5.4
October 2002	1.20±0.02	2.30±004	3.60±0.06	1.70±0.06	2.40±0.05	1.70±0.04	3.40±0.03	4.30±0.02	4.50±0.05	4.50±0.04	64.0±8.7	42.0±6.1	35.0±3.8	41.0±3.9	209±30
November 2002	1.00±0.02	1.40±0.03	2.00±0.05	3.40±0.09	3.30±0.04	3.70±0.13	3.30±0.03	5.50±0.21	6.80±0.04	4.80±0.06	46.0±5.4	154±14	177±11	63.0±5.2	232±28
December 2002	3.60±0.07	2.40±0.01	2.60±0.07	5.00±0.09	2.50±0.03	1.80±0.02	7.30±0.18	3.90±0.15	6.60±0.05	4.40±0.04	108±15	232±25	94.0±10.4	72.0±5.5	104±15
January 2003	2.10±0.08	3.10±0.02	1.00±0.03	3.40±0.06	1.50±0.02	5.30±0.09	4.70±0.12	3.20±0.13	3.40±0.05	4.40±0.05	113±14	208±24	82.0±8.5	372±42	178±35
February 2003	1.10±0.03	1.50±0.03	3.30±0.05	1.60±0.05	3.40±0.03	5.40±0.07	2.90±0.09	1.20±0.06	1.40±0.04	6.10±0.07	274±30	123±16	35.0±4.7	212±31	140±21
March 2003	2.50±0.05	2.30±0.05	1.80±0.02	2.30±0.04	4.00±0.04	3.10±0.04	3.20±0.10	1.70±0.05	3.70±0.03	7.40±0.08	53.0±5.2	343±35	91.0±6.5	220±35	124±18
April 2003	2.40±0.03	4.10±0.03	2.20±0.06	1.80±0.06	2.00±0.02	2.60±0.05	3.00±0.14	1.80±0.05	5.80±0.04	7.00±0.07	48.0±4.6	41.0±5.1	81.0±5.8	128±29	127±20
May 2003	3.50±0.04	2.30±0.02	2.20±0.04	1.60±0.06	3.90±0.03	31.4±3.4	32.9±3.6	20.8±2.5	30.0±5.7	52.6±6.5	72.0±8.6	141±14	60.0±6.3	152±14	71.0±8.2

Seawater	Mn					Ni					Pb				
Months	S1	S2	S3	S4	S5	S1	S2	S3	S4	S5	S1	S2	S3	S4	S5
July 2002	6.00±0.34	13.0±0.5	3.00±0.47	7.00±0.35	73.0±0.7	10.0±0.5	15.0±0.3	1.00±0.12	5.00±0.33	3.00±0.2	15.0±2.2	50.0±4.4	79.0±5.1	53.0±5.1	51.0±5.3
August 2002	18.0±0.4	2.00±0.12	3.00±0.37	4.00±0.24	29.0±0.5	9.00±0.61	27.0±0.6	15.0±0.4	9.00±0.52	12.0±0.4	74.0±5.2	47.0±4.1	27.0±3.2	72.0±5.6	65.0±5.4
September 2002	24.0±0.4	5.00±0.24	3.00±0.24	5.00±0.18	54.0±0.6	19.0±1.2	5.00±0.26	11.0±0.3	6.00±0.43	15.0±0.2	12.0±2.3	60.0±5.3	45.0±4.0	29.0±1.5	41.0±4.6
October 2002	9.00±0.15	7.00±0.26	4.00±0.36	5.00±0.14	62.0±0.6	11.0±0.9	2.00±0.15	4.00±0.52	5.00±0.29	21.0±0.3	47.0±3.5	63.0±5.1	58.0±4.8	35.0±3.0	60.0±5.6
November 2002	2.00±0.24	12.0±0.1	7.00±0.19	5.00±0.26	29.0±0.4	2.00±0.12	14.0±0.3	4.00±0.48	6.00±0.35	7.00±0.4	73.0±5.4	34.0±2.8	32.0±3.4	34.0±3.2	40.0±3.6
December 2002	3.00±0.75	6.00±0.24	5.00±0.30	8.00±0.42	35.0±0.3	11.0±0.6	21.0±0.5	21.0±0.4	5.00±0.31	11.0±0.5	10.0±2.2	43.0±3.0	32.0±3.6	38.0±3.6	35.0±2.8
January 2003	6.00±0.24	15.0±0.5	4.00±0.21	5.00±0.33	61.0±0.5	16.0±0.7	16.0±0.4	20.0±0.5	12.0±0.4	11.0±0.6	26.0±4.2	1.00±0.24	37.0±2.9	4.00±0.82	74.0±5.4
February 2003	11.0±0.1	6.00±0.27	2.00±0.14	6.00±0.43	81.0±0.7	6.00±0.66	2.00±0.18	15.0±0.3	5.00±0.25	4.00±0.26	54.0±3.5	64.0±4.6	55.0±4.5	27.0±1.9	30.0±3.5
March 2003	4.00±0.21	16.0±0.3	7.00±0.15	9.00±0.45	74.0±0.6	15.0±0.5	12.0±0.5	20.0±0.6	8.00±0.4	23.0±0.3	55.0±3.7	87.0±5.6	72.0±5.2	19.0±1.3	44.0±4.6
April 2003	3.00±0.16	2.00±0.16	20.0±0.4	12.0±0.3	75.0±0.6	2.00±0.44	19.0±0.6	8.00±0.5	2.00±0.1	21.0±0.4	70.0±5.8	82.0±4.7	62.0±6.1	29.0±1.5	109±8
May 2003	4.00±0.23	8.00±0.23	3.00±0.57	2.00±0.13	16.0±0.3	16.0±0.6	11.0±0.4	14.0±0.3	3.00±0.2	15.0±0.2	8.00±2.18	36.0±2.4	14.0±2.4	39.0±3.5	45.0±4.3

Seawater	Zn				
Months	S1	S2	S3	S4	S5
July 2002	35.0±0.6	31.0±0.4	20.0±0.2	34.0±0.4	39.0±0.4
August 2002	21.0±0.3	64.0±0.5	6.00±0.09	27.0±0.3	3.00±0.30
September 2002	60.0±0.6	83.0±0.7	49.0±0.4	81.0±1.0	53.0±0.4
October 2002	19.0±0.3	10.0±0.2	9.00±0.10	22.0±0.1	60.0±0.5
November 2002	27.0±0.3	38.0±0.4	22.0±0.2	25.0±0.1	12.0±0.2
December 2002	13.0±0.2	19.0±0.3	24.0±0.3	10.0±0.2	17.0±0.2
January 2003	254±36	236±42	233±38	309±54	220±29
February 2003	15.0±0.3	20.0±0.2	27.0±0.3	10.0±0.2	18.0±0.1
March 2003	9.00±0.24	6.00±0.21	12.0±0.2	10.0±0.2	12.0±0.1
April 2003	8.00±0.27	3.00±0.25	8.00±0.26	6.00±0.20	7.00±0.16
May 2003	17.0±0.3	21.0±0.2	5.00±0.05	13.0±0.2	22.0±0.2

Table 3 Heavy metal concentrations in sediment samples (μg/g dry weight)

Sediment	Cd					Cu					Fe				
Months	S1	S2	S3	S4	S5	S1	S2	S3	S4	S5	S1	S2	S3	S4	S5
July 2002	1.90±0.48	1.10±0.49	1.00±0.39	2.90±0.95	3.60±1.20	30.8±8.1	25.9±5.8	22.2±5.7	18.3±4.5	18.1±3.5	25786±45	20563±38	43117±25	21560±24	17566±18
August 2002	0.60±0.19	0.50±0.21	0.20±0.02	0.90±0.35	0.70±0.55	28.0±6.8	19.5±4.1	19.6±4.5	23.6±5.4	14.8±3.8	24857±35	19920±35	34046±24	21907±27	16006±17
September 2002	2.20±0.53	0.70±0.22	0.40±0.03	0.50±0.33	1.80±0.59	12.6±6.3	21.5±3.7	21.8±4.8	19.3±3.8	14.0±3.7	12158±20	20350±36	36655±23	20810±28	17120±18
October 2002	0.30±0.20	0.70±0.25	0.50±0.03	2.00±0.22	3.60±1.10	16.9±5.3	21.5±3.5	20.1±5.0	16.8±3.5	15.9±4.2	13360±22	20603±34	36657±20	20961±24	23140±16
November 2002	1.40±0.40	1.10±0.38	1.10±0.38	0.40±0.31	1.60±0.66	11.5±8.1	16.7±6.6	19.9±3.2	16.6±4.1	9.30±2.61	10573±24	19016±15	36141±27	20090±26	12201±18
December 2002	1.50±0.42	1.60±0.30	0.90±0.25	1.30±0.40	2.30±0.74	9.90±3.21	17.3±5.4	21.8±3.5	14.4±4.3	11.5±2.7	9522±18	15872±18	36219±26	21830±24	14247±15
January 2003	0.30±0.14	0.90±0.26	0.80±0.26	1.00±0.44	1.30±0.57	10.2±2.4	19.3±6.8	16.5±2.8	13.7±3.8	27.1±3.4	9297±18	16732±19	31886±31	17882±23	14301±14
February 2003	1.00±0.48	0.50±0.13	0.80±0.28	0.20±0.11	2.50±0.65	9.90±2.63	37.0±10.2	21.2±2.2	14.1±4.1	16.3±2.8	7748±14	22765±20	36681±29	16280±19	17755±16
March 2003	0.20±0.02	nd	0.60±0.26	1.20±0.35	2.20±0.73	8.20±3.02	19.9±5.4	17.0±5.2	20.2±3.5	16.2±2.6	7974±15	21475±20	34819±24	26574±20	16225±17
April 2003	0.30±0.02	0.40±0.12	1.50±0.28	0.60±0.32	0.80±0.53	11.3±2.9	31.8±5.8	18.6±1.2	16.5±3.8	12.9±1.9	9667±14	23616±21	36526±29	22086±21	15437±13
May 2003	0.70±0.08	0.50±0.03	0.90±0.36	0.50±0.41	1.00±0.42	6.50±4.31	20.4±6.1	17.3±7.5	16.0±4.0	15.6±2.3	4385±10	19333±18	36537±25	17778±19	19525±17

Sediment	Mn					Ni					Pb				
Months	S1	S2	S3	S4	S5	S1	S2	S3	S4	S5	S1	S2	S3	S4	S5
July 2002	425±2	660±5	857±6	496±4	424±4	54.9±6.5	75.5±7.7	262±11	83.9±6.5	56.7±5.6	14.6±2.3	6.20±1.32	25.7±2.6	23.0±2.5	78.3±1.1
August 2002	576±3	333±4	605±5	569±5	385±3	65.9±7.1	102±8	218±10	90.9±7.5	61.9±5.4	20.9±3.4	10.4±1.6	12.0±1.1	23.6±2.5	77.0±1.0
September 2002	255±2	326±4	688±5	395±4	412±4	44.1±5.3	93.6±7.2	271±11	78.6±7.6	58.0±5.7	15.0±2.3	24.0±2.8	31.7±3.5	15.1±2.3	74.0±0.9
October 2002	316±2	326±3	606±6	404±4	483±4	61.9±5.8	86.8±6.3	264±12	79.0±7.7	80.0±5.6	28.8±2.6	21.5±2.0	20.5±2.4	25.2±2.4	61.6±1.0
November 2002	161±3	359±4	597±4	483±3	331±3	18.9±4.6	56.9±5.8	207±11	59.5±6.5	50.7±5.7	32.9±3.8	26.5±2.1	14.1±2.2	22.1±2.3	42.2±3.9
December 2002	263±4	304±3	692±5	624±4	347±4	23.8±8.0	50.6±5.5	223±10	78.1±7.8	39.2±5.4	14.6±2.4	20.2±1.9	24.3±2.3	19.6±2.4	32.2±2.9
January 2003	244±4	299±2	588±5	328±5	415±5	20.7±5.9	71.6±5.7	184±12	59.1±5.6	43.0±4.7	19.2±2.6	19.7±2.1	22.2±2.2	15.3±3.3	59.4±3.8
February 2003	365±3	357±3	775±6	370±4	492±5	21.8±5.7	104±11	241±10	47.4±5.7	60.4±4.9	7.30±1.31	16.7±1.8	26.9±2.5	11.1±2.2	68.9±4.7
March 2003	373±3	348±4	640±4	476±4	492±4	55.6±6.8	110±9	195±9	95.6±6.4	50.8±4.8	17.8±2.5	16.5±1.7	13.2±1.1	16.8±3.2	46.8±4.5
April 2003	264±3	383±4	688±5	421±3	477±5	78.1±8.1	131±10	218±10	85.1±5.7	56.0±3.9	16.4±2.6	22.6±1.9	13.2±1.2	15.8±2.4	55.7±5.6
May 2003	194±2	310±3	671±5	332±4	378±4	38.4±4.1	91.7±5.9	205±10	65.7±5.3	68.2±4.8	9.10±1.45	17.7±1.8	5.80±1.16	29.1±3.5	49.7±4.7

Sediment	Zn				
Months	S1	S2	S3	S4	S5
July 2002	87.1±25.7	92.4±35.3	77.0±13.6	78.7±13.5	258±82
August 2002	80.2±20.2	69.7±26.6	65.3±12.7	60.4±14.1	206±75
September 2002	35.8±23.3	51.4±18.2	69.9±11.1	53.7±20.7	309±89
October 2002	48.8±20.6	63.3±13.3	90.1±19.8	73.7±11.6	445±102
November 2002	31.1±11.1	45.7±15.4	58.7±14.1	45.9±14.3	194±56
December 2002	31.0±11.2	49.9±17.2	73.5±13.5	54.2±18.4	154±50
January 2003	39.4±15.4	68.6±16.3	55.4±11.0	52.8±16.3	184±50
February 2003	49.7±20.5	73.8±12.2	70.7±10.5	65.6±19.5	293±76
March 2003	57.1±20.3	67.4±17.1	56.3±12.1	57.9±16.4	257±76
April 2003	38.3±15.1	78.4±14.3	65.9±13.4	53.5±14.3	154±45
May 2003	21.8±10.2	54.0±12.0	59.7±14.7	47.1±17.6	194±50

nd not detectable

Table 4 Heavy metal concentrations in *Cystoseira barbata* samples (μg/g dry weight) (mean ± SD)

Cystoseira barbata	Cd					Cu					Fe				
Months	**S1**	**S2**	**S3**	**S4**	**S5**	**S1**	**S2**	**S3**	**S4**	**S5**	**S1**	**S2**	**S3**	**S4**	**S5**
July 2002	4.78±0.99	0.03±0.00	3.69±0.55	3.01±0.44	2.53±0.56	0.95±0.21	nd	3.04±0.54	1.97±0.42	1.42±0.10	1218±100	2330±200	2124±185	1636±131	755±78
August 2002	4.56±0.87	3.13±0.68	2.52±0.44	1.21±0.24	4.40±0.86	1.54±0.56	5.06±1.03	0.26±0.08	1.98±0.35	1.50±0.19	1729±112	3670±175	3006±231	3287±201	5976±286
September 2002	3.93±0.75	3.83±0.75	2.23±0.42	2.53±0.46	3.08±0.72	4.22±0.75	5.26±0.89	5.87±1.15	3.04±0.46	5.45±1.10	3977±245	2729±210	4383±200	2786±175	1323±119
October 2002	3.47±0.72	3.30±0.63	2.26±0.52	5.84±0.71	3.70±0.68	1.57±0.33	1.46±0.56	2.52±0.72	2.67±0.25	1.04±0.14	3719±263	1888±203	2778±186	1100±90	1715±124
November 2002	4.24±0.88	2.42±0.65	3.24±0.60	4.06±0.72	1.69±0.25	2.37±0.63	4.22±0.16	2.29±0.65	0.24±0.09	0.33±0.12	1526±111	1044±101	2218±197	717±85	903±81
December 2002	3.40±0.69	2.12±0.71	5.01±0.76	4.78±0.70	1.32±0.36	3.48±0.54	4.87±0.32	1.00±0.30	0.48±0.07	3.82±0.35	4092±245	1577±115	2061±165	1759±146	1382±118
January 2003	2.45±0.88	2.56±0.69	5.92±0.73	3.21±0.68	3.32±0.56	0.86±0.13	2.36±0.44	0.93±0.25	0.21±0.06	1.45±0.43	4947±236	2226±214	2013±145	821±83	1116±106
February 2003	4.13±0.96	0.93±0.12	3.01±0.68	7.23±0.78	2.44±0.45	1.47±0.31	4.40±0.86	1.68±0.41	3.64±0.56	1.21±0.10	5093±278	6058±301	4083±215	1439±104	570±45
March 2003	2.94±0.78	3.58±0.89	7.10±0.85	3.78±0.86	4.94±0.76	nd	0.78±0.13	2.92±0.35	3.46±0.37	1.68±0.14	5257±289	3657±185	499±217	1257±100	2053±174
April 2003	1.99±0.33	4.75±0.96	3.23±0.62	0.93±0.35	1.28±0.30	0.50±0.12	1.26±0.32	0.90±0.18	2.65±0.35	1.24±0.16	4846±255	2944±213	2373±158	3546±153	4725±198
May 2003	0.65±0.20	3.17±0.69	3.46±0.59	3.91±0.63	2.68±0.50	4.42±0.96	2.02±0.50	0.46±0.08	0.42±0.10	6.64±1.13	6288±301	3247±163	2400±142	1280±100	1545±110

Cystoseira barbata	Mn					Ni					Pb				
Months	**S1**	**S2**	**S3**	**S4**	**S5**	**S1**	**S2**	**S3**	**S4**	**S5**	**S1**	**S2**	**S3**	**S4**	**S5**
July 2002	134±15	149±16	84.2±4.7	80.7±4.2	57.2±2.9	5.00±0.45	2.80±0.44	18.0±1.1	9.20±0.85	4.60±0.42	2.30±0.69	28.9±2.3	29.1±2.7	37.2±3.5	23.8±2.4
August 2002	125±11	234±14	152±13	114±12	245±16	28.5±2.1	21.2±0.52	15.1±1.8	20.6±2.4	24.2±2.1	29.6±2.1	22.6±2.4	34.6±3.4	11.2±1.5	36.0±3.7
September 2002	267±16	414±15	140±14	107±17	70.4±41	25.9±2.4	12.7±0.84	30.8±2.3	25.1±2.6	22.7±2.3	25.0±2.4	41.1±3.6	10.1±1.8	62.1±5.4	20.3±2.6
October 2002	391±18	358±13	122±16	30.6±2.8	101±16	5.00±0.64	8.00±0.67	15.8±1.5	3.90±0.42	7.50±0.73	6.50±1.3	25.3±2.9	4.20±0.83	20.0±2.6	23.8±2.4
November 2002	138±11	95.9±4.5	127±17	34.7±2.5	103±14	17.2±1.6	3.70±0.31	22.2±1.9	13.5±0.79	1.80±0.46	25.6±2.7	33.7±3.2	11.7±1.3	4.50±0.78	44.0±4.2
December 2002	339±14	155±13	96.5±4.9	31.2±	117±12	5.70±0.87	2.80±0.61	9.50±0.89	4.90±0.68	10.9±0.39	14.7±1.6	0.90±0.13	34.2±3.4	8.3±1.5	22.7±2.9
January 2003	382±16	97.7±4.8	107±11	32.8±2.9	91.5±4.5	33.0±2.7	2.70±0.82	2.90±0.30	9.40±0.75	1.10±0.33	20.3±2.4	8.5±1.3	30.1±3.8	12.5±1.4	24.9±2.7
February 2003	278±14	321±21	82.6±4.6	22.4±2.1	42.1±2.5	21.4±2.6	1.90±0.4	0.90±0.21	6.60±0.54	1.50±0.35	14.7±1.3	39.0±0.96	27.6±2.7	22.5±2.8	34.2±3.6
March 2003	204±12	87.9±4.5	105±10	28.5±2.8	87.6±2.4	4.20±0.75	3.70±0.10	11.9±0.18	8.00±0.61	3.90±0.49	28.3±2.1	1.50±0.45	4.60±0.75	15.1±1.5	50.6±4.9
April 2003	186±10	88.6±4.3	65.1±3.5	48.0±3.1	350±21	14.3±1.1	20.6±1.12	5.40±0.52	3.30±0.24	27.7±2.5	4.30±0.36	18.9±1.8	24.3±2.4	23.1±2.4	15.8±1.9
May 2003	236±13	188±14	29.4±2.8	15.9±1.5	46.8±3.2	5.60±0.86	12.4±0.78	8.70±0.35	5.90±0.46	22.8±2.9	25.4±2.7	7.90±1.1	32.3±3.9	20.2±2.6	46.7±4.8

Cystoseira barbata	Zn				
Months	**S1**	**S2**	**S3**	**S4**	**S5**
July 2002	109±22	120±17	98.3±12.1	108±20	132±16
August 2002	110±19	130±16	117±16	107±16	167±18
September 2002	228±21	165±14	112±15	151±16	136±14
October 2002	149±23	242±19	114±17	112±17	159±14
November 2002	116±18	124±18	115±19	84.1±10.6	150±12
December 2002	165±16	134±14	95.8±11.7	106±15	104±10
January 2003	110±15	153±16	128±17	80.3±10.4	163±13
February 2003	238±22	295±21	148±21	117±16	100±11
March 2003	198±20	139±17	145.1±23	143±14	129±17
April 2003	190±19	139±18	269±18	160±18	234±19
May 2003	117±14	141±20	118±15	124±14	132±21

nd not detectable

Table 5 Heavy metal concentrations in *Patella caerulea* samples (μg/g dry weight)

Patella caerulea	Cd					Cu				
Months	**S1**	**S2**	**S3**	**S4**	**S5**	**S1**	**S2**	**S3**	**S4**	**S5**
July 2002	0.048±0.012	0.041±0.008	0.037±0.012	0.037±0.014	0.011±0.004	0.249±0.051	0.180±0.020	0.192±0.018	0.137±0.036	0.209±0.021
August 2002	0.015±0.003	0.009±0.002	0.004±0.001	0.024±0.010	0.005±0.001	0.050±0.012	0.050±0.011	0.056±0.010	0.067±0.012	0.058±0.011
September 2002	0.009±0.002	0.010±0.002	0.030±0.011	0.013±0.005	0.012±0.002	0.036±0.008	0.044±0.010	0.067±0.012	0.038±0.009	0.050±0.009
October 2002	0.021±0.003	0.011±0.003	0.038±0.015	0.027±0.003	0.016±0.006	0.074±0.016	0.040±0.009	0.057±0.011	0.053±0.008	0.042±0.006
November 2002	0.018±0.002	0.017±0.003	0.032±0.014	0.028±0.006	0.002±0.000	nd	0.015±0.003	0.041±0.036	0.039±0.001	0.044±0.006
December 2002	0.021±0.003	0.010±0.002	0.021±0.009	0.020±0.002	0.018±0.003	nd	nd	nd	nd	nd
January 2003	0.024±0.003	0.009±0.001	0.015±0.006	0.023±0.006	0.007±0.001	nd	0.025±0.008	nd	nd	nd
February 2003	0.057±0.013	0.028±0.009	0.054±0.010	0.060±0.012	0.025±0.006	0.024±0.006	nd	0.045±0.009	0.031±0.009	nd
March 2003	0.017±0.006	0.014±0.003	0.041±0.013	0.024±0.007	0.043±0.010	nd	nd	nd	0.022±0.004	0.034±0.003
April 2003	0.042±0.012	0.016±0.004	0.065±0.019	0.025±0.009	0.031±0.009	nd	nd	0.035±0.007	nd	0.016±0.002
May 2003	0.046±0.014	0.015±0.006	0.056±0.016	0.039±0.009	0.044±0.011	0.031±0.009	nd	nd	0.024±0.008	0.009±0.001

Patella caerulea	Fe					Mn				
Months	**S1**	**S2**	**S3**	**S4**	**S5**	**S1**	**S2**	**S3**	**S4**	**S5**
July 2002	25.2±2.1	20.5±2.5	13.4±1.5	21.5±2.1	21.4±2.3	0.095±0.024	0.109±0.035	0.049±0.016	0.245±0.048	0.115±0.024
August 2002	9.61±1.8	5.70±1.5	18.3±1.3	1.85±0.89	13.1±1.2	0.048±0.016	0.054±0.036	0.105±0.025	0.141±0.033	0.110±0.023
September 2002	16.13±2.4	18.3±2.4	21.2±2.4	18.0±2.4	20.6±2.7	0.064±0.021	0.070±0.035	0.070±0.024	0.092±0.021	0.095±0.024
October 2002	34.5±3.5	19.8±2.6	35.6±2.8	50.0±5.5	29.7±3.2	0.140±0.036	0.109±0.033	0.156±0.031	0.281±0.035	0.117±0.021
November 2002	58.8±4.2	54.5±3.5	32.5±2.9	53.4±5.3	24.9±3.4	0.186±0.052	0.194±0.032	0.140±0.029	0.284±0.038	0.096±0.018
December 2002	9.48±1.7	44.3±3.2	40.7±3.4	49.0±5.4	41.3±4.5	0.098±0.026	0.320±0.098	0.260±0.052	0.207±0.042	0.174±0.018
January 2003	18.6±2.6	9.5±1.1	12.3±2.2	17.8±2.7	7.3±1.1	0.113±0.033	0.166±0.042	0.180±0.043	0.188±0.021	0.061±0.020
February 2003	69.8±4.7	25.7±2.4	41.2±3.6	59.1±4.7	35.8±2.8	0.308±0.099	0.171±0.043	0.167±0.036	0.247±0.037	0.148±0.041
March 2003	21.7±2.3	15.8±2.1	27.1±2.7	76.0±5.7	53.1±4.7	0.130±0.045	0.089±0.026	0.143±0.040	0.370±0.043	0.216±0.036
April 2003	25.4±2.4	57.6±4.2	62.4±5.2	48.3±3.5	75.7±5.6	0.110±0.043	0.268±0.072	0.495±0.078	0.213±0.038	0.231±0.033
May 2003	24.7±2.6	6.61±1.1	61.3±5.3	12.8±2.5	19.5±2.1	0.135±0.042	0.157±0.052	0.263±0.043	0.222±0.020	0.144±0.024

Patella caerulea	Ni					Pb				
Months	**S1**	**S2**	**S3**	**S4**	**S5**	**S1**	**S2**	**S3**	**S4**	**S5**
July 2002	0.309±0.065	0.238±0.045	0.090±0.031	0.289±0.045	0.130±0.013	0.166±0.019	0.121±0.016	0.061±0.015	0.124±0.014	0.112±0.015
August 2002	0.054±0.012	0.047±0.017	0.067±0.015	0.065±0.011	0.031±0.004	0.191±0.028	0.030±0.008	0.022±0.008	0.029±0.007	0.022±0.006
September 2002	0.031±0.011	0.042±0.016	0.038±0.007	0.048±0.005	0.042±0.006	0.031±0.004	0.016±0.002	0.025±0.006	0.044±0.005	0.030±0.005
October 2002	0.023±0.008	0.159±0.035	0.021±0.006	0.031±0.006	0.019±0.002	0.036±0.008	0.024±0.003	0.032±0.005	0.034±0.003	0.021±0.003
November 2002	0.055±0.010	0.115±0.036	0.025±0.009	0.136±0.011	0.021±0.002	0.044±0.010	0.051±0.002	0.019±0.003	0.037±0.005	0.026±0.004
December 2002	0.047±0.016	0.052±0.013	0.115±0.021	0.060±0.012	0.068±0.006	0.015±0.003	0.107±0.010	0.101±0.010	0.020±0.004	0.010±0.003
January 2003	0.132±0.055	0.090±0.015	0.128±0.035	0.083±0.013	0.046±0.005	0.052±0.013	0.025±0.003	0.081±0.009	0.025±0.004	0.048±0.006
February 2003	0.168±0.053	0.131±0.010	0.101±0.025	0.091±0.013	0.115±0.018	0.083±0.016	0.023±0.004	0.026±0.005	0.031±0.003	0.148±0.014
March 2003	0.073±0.025	0.190±0.016	0.041±0.006	0.143±0.012	0.235±0.033	0.045±0.011	0.068±0.006	0.076±0.013	0.123±0.010	0.053±0.009
April 2003	0.339±0.067	0.155±0.012	0.173±0.023	0.131±0.028	0.227±0.035	0.042±0.010	0.123±0.010	0.088±0.018	0.051±0.003	0.037±0.006
May 2003	0.166±0.054	0.190±0.014	0.295±0.037	0.256±0.036	0.176±0.021	0.081±0.015	0.063±0.015	0.032±0.009	0.075±0.005	0.074±0.005

Table 5 (continued)

Patella caerulea	Zn				
Months	**S1**	**S2**	**S3**	**S4**	**S5**
July 2002	0.84±0.16	0.61±0.12	0.85±0.16	0.39±0.10	0.82±0.14
August 2002	0.52±0.14	0.38±0.09	0.36±0.08	0.42±0.15	0.37±0.06
September 2002	0.35±0.06	0.28±0.07	0.31±0.08	0.31±0.12	0.29±0.04
October 2002	0.42±0.04	0.27±0.06	0.28±0.05	0.41±0.16	0.26±0.06
November 2002	0.72±0.05	0.71±0.15	0.29±0.06	1.19±0.24	0.32±0.08
December 2002	0.63±0.05	0.79±0.19	0.67±0.10	0.68±0.12	1.00±0.21
January 2003	1.11±0.10	0.80±0.20	0.64±0.12	1.03±0.26	0.56±0.12
February 2003	1.23±0.13	1.05±0.35	0.85±0.16	1.39±0.25	0.93±0.15
March 2003	0.94±0.08	0.94±0.32	1.05±0.21	1.17±0.14	1.64±0.31
April 2003	1.15±0.16	1.21±0.42	1.66±0.23	0.91±0.10	0.96±0.25
May 2003	1.24±0.18	1.17±0.21	1.02±0.23	1.38±0.31	1.41±0.32

nd not detectable

(2.00 ± 0.24 μg/l), at station S2 in August 2002 (2.00 ± 0.12 μg/l) and April 2003 (2.00 ± 0.16 μg/l), at station S3 in February 2003 (2.00 ± 0.14 μg/l), and at station S4 in May 2003 (2.00 ± 0.13 μg/l), while the highest average Mn value (81.0 ± 0.7 μg/l) was found at station S5 in February 2003 (Table 2). Concerning Mn levels in water samples, no statistically significant temporal changes were seen.

In the present study, the minimum mean Ni value in seawater (1.00 ± 0.12 μg/l) was detected at station S3 in July 2002, where the maximum Ni level (27.0 ± 0.6 μg/l) was noted at station S2 in August 2002 (Table 2). However, no statistically significant stational and seasonal differences in Ni contents in water samples were recorded.

Among the different locations, it can be concluded that samples measured at station S2 in January 2003 showed the lowest mean concentration of Pb in seawater (1.00 ± 0.24 μg/l), whereas the highest mean content of Pb (109 ± 8 μg/l) was observed at station S5 in April 2003 (Table 2). Moreover, ANOVA results did not show statistically significant spatial and temporal changes in the observed levels of Pb in water samples.

Recorded Zn values in seawater in January 2003 were higher compared to the measured contents in other sampling months. Results from the analysis of seawater registered the lowest average Zn value (3.00 ± 0.25 μg/l) at station S2 in April 2003 and at station S5 in August 2002 (3.00 ± 0.30 μg/l), while the elevated mean level of Zn in seawater (309 ± 54 μg/l) was observed at station S4 in January 2003 (Table 2). As for water samples, the findings of ANOVA performed with Zn data showed significant variations among sampling months ($p < 0.001$). However, no significant stational differences were found for Zn contents in seawater samples.

Heavy metals in sediment

In general, the analyzed heavy metals in sediment samples showed higher concentrations at stations S3 and S5. The results showed that the pattern of overall average metal levels in sediment samples decreased in the following order: Fe > Cu > Mn > Ni > Zn > Pb > Cd. The highest concentrations in sediment samples were measured in July 2002, except Cu and Ni (Table 3).

In this study, with the exception of stations S3 and S2, higher mean Cd levels in sediment were obtained in July 2002. The higher average Cd levels were measured at station S5 in July 2002 (3.60 ± 1.20 μg/g) and October 2002 (3.60 ± 1.10 μg/g) (Table 3). The ANOVA test concluded that there were statistically significant differences among stations for Cd values in sediment ($p < 0.05$), while no differences among seasons were detected.

Stations S3 and S2 generally represented a bit higher mean Cu level in sediment during all months, and recorded concentrations ranged between 6.50 ± 4.31 and 37.0 ± 10.2 μg/g. The

Table 6 Heavy metal concentrations in *Liza aurata* samples (μg/g dry weight)

Mugil auratus	Cd			Cu			Fe			Mn	
Months	S1	S2	S5	S1	S2	S5	S1	S2	S5	S1	S2
July 2002	0.004 ± 0.001	0.003 ± 0.001	0.001 ± 0.001	0.051 ± 0.002	0.052 ± 0.002	0.058 ± 0.002	31.9 ± 12.4	64.9 ± 16.4	44.9 ± 18.7	0.284 ± 0.075	0.302 ± 0.098
August 2002	0.001 ± 0.001	0.003 ± 0.001	0.002 ± 0.001	nd	0.028 ± 0.001	0.027 ± 0.002	16.4 ± 5.4	25.2 ± 10.3	16.0 ± 8.1	0.072 ± 0.019	0.134 ± 0.450
September 2002	0.002 ± 0.001	0.003 ± 0.001	0.001 ± 0.001	0.021 ± 0.001	0.023 ± 0.001	0.019 ± 0.001	18.4 ± 6.7	37.1 ± 12.4	11.6 ± 6.5	0.078 ± 0.020	0.131 ± 0.046
October 2002	0.001 ± 0.001	0.003 ± 0.001	0.001 ± 0.001	0.026 ± 0.001	0.022 ± 0.001	0.024 ± 0.001	13.1 ± 3.2	10.7 ± 4.1	8.06 ± 3.21	0.090 ± 0.027	0.075 ± 0.018

Mugil auratus	Mn	Ni			Pb			Zn			
Months	S5	S1	S2	S5	S1	S2	S5	S1	S2	S5	
July 2002	0.272 ± 0.090	0.028 ± 0.009	0.042 ± 0.010	0.037 ± 0.013	0.039 ± 0.001	0.036 ± 0.002	0.026 ± 0.001	0.549 ± 0.123	0.437 ± 0.142	0.555 ± 0.111	
August 2002	0.122 ± 0.041	0.005 ± 0.001	0.013 ± 0.005	0.030 ± 0.008	nd	0.005 ± 0.001	nd	0.187 ± 0.060	0.334 ± 0.098	0.304 ± 0.086	
September 2002	0.057 ± 0.018	0.009 ± 0.002	0.017 ± 0.007	0.025 ± 0.007	nd	nd	nd	0.147 ± 0.042	0.179 ± 0.064	0.142 ± 0.042	
October 2002	0.065 ± 0.020	nd	0.022 ± 0.004	0.022 ± 0.005	nd	0.006 ± 0.001	0.007 ± 0.002	0.189 ± 0.053	0.229 ± 0.068	0.138 ± 0.043	

nd not detectable

lowest average Cu value in sediment was found at station S1 in May 2003, while the peak mean Cu content was recorded at station S2 in February 2003 (Table 3). Statistical analysis findings showed that Cu levels in sediment exhibited significant spatial variations ($p < 0.001$), while no significant temporal differences were found for Cu levels in sediment.

The monthly detected Fe concentrations in sediments over the sampling periods were generally very high at station S3. In addition, the lowest mean concentration of iron (4385 ± 10 μg/g) was measured at station S1 in May 2003 and the highest average Fe level (43,117 ± 25 μg/g) was found at station S3 in July 2002 (Table 3). The sampling sites have a wide range of Fe values in sediment samples. In addition, station S3 was the only one differing significantly from the other stations ($p < 0.001$); however, no statistical variations among seasons were observed.

Generally, a higher sedimentary Mn content was noted at station 3. Moreover, the lowest average Mn level in sediment (161 ± 3 μg/g) was detected at station S1 in November 2002, while the highest mean Mn value (857 ± 6 μg/g) was recorded at station S3 in July 2002 (Table 3). As for Mn values in sediment, statistically significant variations were highlighted among stations ($p < 0.001$). Station S3 was different from other stations, and also station S1 was different from station S4. However, no statistical differences in the concentrations of manganese in water among the sampling seasons were detected.

An overall increase in Ni concentrations was detected at station S3 during all months. Furthermore, the lowest mean Ni level in sediment (18.9 ± 4.6 μg/g) was measured at station S1 in November 2002, while the highest average Ni value (271 ± 11 μg/g) was recorded at station S3 in September 2002 (Table 3). ANOVA analysis obscured statistically significant changes among stations ($p < 0.001$); however, no differences among seasons were found.

Higher Pb values in sediments were recorded at station S5 in all sampling months. The lowest mean Pb level in sediment (5.80 ± 1.16 μg/g) was measured at station S3 in May 2003, while the highest mean value (78.3 ± 1.1 μg/g) was noted at station S5 in July 2002 (Table 3). Furthermore, statistically significant changes among stations were detected for Pb contents in sediments ($p < 0.001$) and station S5 differed significantly from other sampling stations; however, no significant changes in Pb levels in sediment were defined among seasons.

In this study, except station S5, higher Zn levels in sediment were found at all sampling stations during July 2002. Concerning Zn in sediment, the lowest average concentration (21.8 ± 10.2 μg/g) was measured at station S1 in May 2003, while the highest mean value (445 ± 102 μg/g) was found at station S5 in October 2002 (Table 3). According to the findings of this study, station S5 exhibited significant stational differences ($p < 0.001$) in Zn values in sediment. However,

no significant variations among seasons were detected for accumulated Zn concentration in sediment.

Heavy metals in *Cystoseira barbata*

According to the overall means, the order of heavy metal level accumulation was as follows: Fe > Zn > Mn > Pb > Ni > Cd > Cu. Among the different locations, it can be concluded that *C. barbata* measured at station S2 in July 2002 showed the lowest mean level of Cd (0.03 ± 0.00 μg/g), whereas the highest average concentration of Cd (7.23 ± 0.78 μg/g) was observed at station S4 in February 2003 (Table 4). In terms of the five sampling stations and also sampling months, Cd levels in *C. barbata* did not present statistically significant spatial and temporal differences.

Mean Cu levels in *C. barbata* were below the detection limit in March 2003 at station S1 and in July 2002 at station S2. Furthermore, the highest mean Cu concentration (6.64 ± 1.13 μg/g) was found in May 2003 at station S5 (Table 4). Concerning copper, statistical analyses showed significant changes among sampling months ($p < 0.05$). In addition, September 2002 differed significantly from July 2002, January 2003, and April 2003. In contrast, no significant variations among stations were observed.

Regarding Fe level in *C. barbata*, the minimum average value (570 ± 45 μg/g) was noted in February 2003 at station S5 and the maximum mean Fe content (6288 ± 301 μg/g) was detected at station S1in May 2003 (Table 4). Although the detected concentrations at station S1 differed from stations S4 and S5 ($p < 0.05$), no differences among seasons were found.

Manganese concentration in *C. barbata* showed a relatively wider variation, and the lowest mean Mn content (15.9 ± 1.5 μg/g) was measured at station S4 in May 2003, while the highest average Mn value (414 ± 15 μg/g) was detected at station S2 in September 2002 (Table 4). Moreover, Mn values recorded in all sampling months were generally higher at stations S2 and S1. As for Mn in *C. barbata*, stational changes ($p < 0.001$) were detected. On the other hand, no significant differences were found among seasons.

Compared to other stations, detected Ni concentrations in autumn months were higher at station S3. In addition, the mean Ni levels in *C. barbata* ranged between 0.90 ± 0.21 and 33.0 ± 2.7 μg/g, with the lowest average content observed at station S3 in February 2003 (Table 4). On the other hand, the highest mean Ni level in *C. barbata* was recorded at the same station in September 2002. It can be noticed that Ni levels in *C. barbata* exhibited temporal variations ($p < 0.01$). Measured values in September 2002 were significantly different ($p < 0.05$) from detected levels in December 2002, February 2003, and March 2003; however, no remarkable spatial differences were exhibited.

The results obtained for Pb in *C. barbata* showed that the lowest average value of Pb (0.90 ± 0.13 μg/g) was noted in December 2002 (station S2) and the peak mean Pb concentration of 62.1 ± 5.4 μg/g was determined in September 2002 (station S4) (Table 4). Concerning Pb in *C. barbata*, measured values at all stations exhibited great seasonal fluctuations. In contrast, no significant spatial and temporal changes in Pb levels in *C. barbata* occurred.

Zinc values were higher at stations S5 and S3 in April 2003, at station S2 in February 2003, and at station S1 in September 2002 and February 2003. The lowest mean Zn level in *C. barbata* (80.3 ± 10.4 μg/g) was measured at station S4 in January 2003, while the highest average Zn value (295 ± 21 μg/g) was recorded at station S2 in February 2003 (Table 4). There were significant differences in Zn concentrations in *C. barbata* among all the sampling months ($p < 0.05$). However, Zn contents in *C. barbata* revealed no significant variations among stations.

Heavy metals in *Patella caerulea*

The results showed that the pattern of the overall average heavy metal values in *P. caerulea* could be ranked from highest to lowest as follows: Fe > Zn > Mn > Ni > Pb > Cu > Cd. Throughout the study period in the investigated area, higher Cd levels in *P. caerulea* were measured at station S3 during sampling months, except the detected Cd value in August 2002. The lowest mean Cd concentration in *P. caerulea* (0.004 ± 0.001 μg/g) was found at station S3 in August 2002, while the highest average Cd value (0.065 ± 0.019 μg/g) was recorded at station S3 in April 2003 (Table 5). Regarding cadmium, the statistical analysis highlighted significant spatial ($p < 0.05$) and temporal differences ($p < 0.01$) in the tissues of *P. caerulea*. Furthermore, measured Cd levels at station S2 differed significantly from recorded values at station S3 and the noted Cd levels in August 2002 were different from the obtained contents in April 2003.

The highest mean copper content (0.249 ± 0.051 μg/g) in *P. caerulea* was found at station S1 in July 2002 (Table 5). Furthermore, it was also noted that Cu levels in the studied limpet samples were not detectable at some stations. Cu values in *P. caerulea* exhibited statistically significant changes ($p < 0.001$) among sampling months. On the other hand, samples collected from sampling sites did not show any consistent significant variations.

The peak mean Fe concentration (76.0 ± 5.7 μg/g) was recorded in the whole soft tissue of *P. caerulea* at station S4 in March 2003. Moreover, the lowest average Fe level in *P. caerulea* (1.85 ± 0.89 μg/g) was measured at station S4 in August 2002 (Table 5). Although no overall significant stational variations in Fe level in *P. caerulea* occurred,

statistically significant differences among seasons were detected ($p < 0.001$).

A slight monthly variation in the mean Mn levels in *P. caerulea* samples was observed. The minimum Mn content was determined at station S1 in August 2002 (0.048 ± 0.016 μg/g), and the maximum Mn level was found at station S3 in April 2003 (0.495 ± 0.078 μg/g) (Table 5). Based on the results of the one-way ANOVA, statistically significant differences ($p < 0.05$) were exhibited among sampling months for Mn levels in *P. caerulea*.

Nickel levels in *P. caerulea* displayed seasonal variations ($p < 0.01$) throughout the sampling periods with the lower average values (0.021 ± 0.002 μg/g) measured at station S3 in October 2002 and at station S5 in November 2002, and the highest level (0.339 ± 0.067 μg/g) was found at station S1 in April 2003 (Table 5). On the other hand, statistically significant spatial differences in Ni levels in *P. caerulea* were not found.

The levels of Pb in *P. caerulea* showed very narrow changes in all stations as well as seasons. The lowest Pb level in *P. caerulea* (0.010 ± 0.003 μg/g) was measured at station S5 in December 2002, while the highest value (0.191 ± 0.028 μg/g) was noted at station S1 in August 2002 (Table 5). Concerning sampling seasons, *P. caerulea* samples showed statistically significant differences in the observed levels of Pb. In addition, results showed that recorded Pb contents in September 2002 differed significantly from observed Pb values in October 2002.

For Zn levels in *P. caerulea*, higher levels were generally recorded in spring months. Furthermore, the lowest Zn content in *P. caerulea* (0.26 ± 0.06 μg/g) was detected at station S5 in November 2002, while the highest Zn value (1.66 ± 0.23 μg/g) was observed at station S3 in April 2003 (Table 5). Concerning ANOVA results, significant differences in zinc contents in limpet samples were observed among seasons ($p < 0.001$). Besides, no spatial variations were detected.

Heavy metals in *Liza aurata*

Regarding all studied metals, the overall mean heavy metal levels in *L. aurata* were arranged in the following sequence: Fe > Zn > Mn > Cu > Ni > Pb > Cd. Furthermore, the highest levels were generally recorded at station S2.

With respect to Cd level in *L. aurata*, the lower mean values appeared at station S1 both in August 2002 and in September 2002 (0.001 ± 0.0001 μg/g) and in July 2002, September 2002, and October 2002 (0.001 ± 0.0001 μg/g) at station S5, while the highest average content (0.004 ± 0.0001 μg/g) was noted at station S1 in July 2002 (Table 6). Concerning the ANOVA results, statistically significant spatial and temporal differences in Cd levels in *L. aurata* were not observed.

Measured Cu values in *L. aurata* were generally very low and showed very small variations in concentrations over the sampling periods. The mean Cu level in *L. aurata* was not detectable at station S1 in August 2002, and the highest average Cu value (0.058 ± 0.0002 μg/g) was recorded at station S5 in July 2002 (Table 6). The statistical analysis highlighted that the contents of Cu in *L. aurata* did not show statistically significant differences among stations. However, the monthly variations for the concentrations of copper in *L. aurata* revealed significant changes ($p < 0.001$).

The mean levels of Fe in *L. aurata* displayed remarkable variations and ranged from 8.06 ± 3.21 to 64.9 ± 16.4 μg/g (Table 6). In the present study, detected Fe levels in *L. aurata* in July 2002 revealed statistically significant differences than the measured concentrations in October 2002 ($p < 0.05$). On the other hand, no stational variations were detected.

A great variability was not observed in Mn concentrations in *L. aurata*, and the lowest (0.057 ± 0.018 μg/g) and the higher average Mn values (0.302 ± 0.098 μg/g) were recorded at station S5 in September 2002 and at station S2 in July 2002, respectively (Table 6). In this study, statistically significant spatial variations were not detected. In addition, ANOVA applied with manganese data showed that recorded values in July 2002 exhibited significant differences ($p < 0.001$) compared to the other sampling months.

The variations in Ni level in *L. aurata* were approximately similar. Moreover, the mean Ni level in *L. aurata* was not detected at station S1 in October 2002, while the highest average Ni value (0.042 ± 0.010 μg/g) was recorded at station S2 in July 2002 (Table 6). Statistically, no significant spatio-temporal variations ($p > 0.05$) were observed for Ni levels in *L. aurata*.

Lead level in *L. aurata* reached its maximum mean value (0.039 ± 0.001 μg/g) at station S1 and station S2 (0.036 ± 0.002 μg/g) in July 2002. Moreover, Pb contents in fish samples were not detected at some stations (Table 6). Furthermore, the results showed that Pb concentrations in *L. aurata* did not exhibit significant changes among sampling stations and months.

Zinc values in *L. aurata* recorded in July 2002 were threefold to fourfold higher as compared to the minimum levels measured in September 2002. Regarding Zn concentration in *L. aurata*, station S5 in October 2002 exhibited the lowest mean Zn level (0.138 ± 0.043 μg/g) (Table 6). Moreover, the peak average Zn value (0.555 ± 0.111 μg/g) was noted at station S5 in July 2002. Statistical analysis indicated that levels of Zn in *L. aurata* from all sampling stations did not change significantly ($p > 0.05$). The statistical comparison of Zn contents in *L. aurata* revealed that measured concentrations in July 2002 showed significant differences ($p < 0.001$) from other sampling months.

Discussion

Heavy metals are among the most common chemical pollutants in the marine environment. It is necessary to monitor the

levels of these contaminants in waters, in sediments, and in marine organisms because of their toxic effects on aquatic organisms, their long residence time within food chains, and their potential to have serious adverse effects on human health (Ferreira et al. 2005).

Compared to several marine biomonitors, seawater and sediments are not enough to predict entirely the bioavailability of heavy metals (Roberts et al. 2008). For this reason, in this study, several biomonitors such as *C. barbata*, *P. caerulea*, and *L. aurata* were also used with the aim of assessing their utility as the potential biomonitors indicative of the pollution levels of heavy metals at coast sites along the Aegean Sea. And they were also used to achieve a better estimate of biologically available heavy metal accumulation.

According to some authors, a number of processes, namely, biological uptake, scavenging by particulate matter, release from bottom sediments, advection and mixing of water masses, and aeolian transport of terrestrial materials, can affect the concentrations of dissolved heavy metals in seawater (Leal et al. 1997).

The results clearly indicated that the mean levels of Fe, Pb, and Zn in water were generally higher than the other studied metals. Concerning the stational variation, the highest average concentrations were found at stations S5 and S4. Generally, the highest mean heavy metal values were observed during the winter period, except for the recorded average Cu and Ni contents (Table 2).

Regarding Cd and Cu levels in seawater, higher mean levels were measured in July 2002 and May 2003, respectively. Furthermore, the concentrations of dissolved metals in seawater varied markedly during the sampling periods and exhibited seasonality. In a study, Leal et al. (1997) investigated the levels of dissolved Cd, Cu, Hg, and Pb in seawater and algae (*Enteromorpha* spp. and *Porphyra* spp.) collected from three beaches located in the Oporto coast (Portugal). They found that concentrations of Pb in seawater were low in winter and both Cu and Pb values in seawater peaked in spring months.

According to some authors, Cd, Cu, and Pb contents in seawater are typically related to the nutrient levels and show marked seasonal variations, with peak periods of primary productivity in spring-summer associated with a depletion of heavy metals from surface waters due to sorption by phytoplankton cells. Moreover, inputs from rivers and estuaries as well as from the atmosphere are very significant and can be dominant, masking and counteracting biological removal associated with phytoplanktonic blooms (Leal et al. 1997).

Our recorded Cu and Pb concentrations in seawater were similar to those reported findings of the study conducted by Leal et al. (1997). However, in our study, only Cu levels in seawater showed marked seasonality. Although phytoplanktonic populations were not studied in the present work, it can be said that temporal changes in sampling months for Cu levels in seawater ($p < 0.001$), at least partially, can be due to the biological activity.

Generally, the highest mean values of measured heavy metals in seawater were observed during winter (Cd, Fe, Mn, Pb, and Zn) and summer (Cu and Ni). The higher metal concentrations in winter may result from land-based runoff by relatively high seasonal rain input, and thus, metal pollution may be carried through the rivers into the bay. These results were supported by the meteorological rainfall data. According to the results, there were heavy rains especially in December 2002, January 2003, and February 2003 in the study area (http://www.mgm.gov.tr/veridegerlendirme/il-ve-ilceler-istatistik.aspx?m=IZMIR).

The higher metal concentrations in summer could be attributed to an increase in the rate of metal accumulation due to the higher temperature and high evaporation rate. Furthermore, summer could increase the amount of toxicants flushed into the water, in particular sewage and high fishing activities. But the lower salinity in autumn could reduce the rate of metal accumulation due to sedimentation (Coulibaly et al. 2012).

Regarding Fe ($p < 0.05$) and Mn levels in water ($p < 0.001$), results recorded exhibited stational variations. The mean Fe levels in seawater at station S3 were generally a little bit lower compared to other sampling stations, and also detected average Mn levels in water were higher at station S5. Ni and Pb concentrations in seawater did not show significant changes among sampling stations and months. Moreover, Mn in water displayed a significant positive correlation with Fe in water ($r = 0.545$, $p < 0.001$).

The higher levels of Zn in seawater were recorded at all stations during January 2003, and the low values of Zn were measured during summer and spring months and also showed temporal differences ($p < 0.001$); this may be due to its consumption by phytoplankton, which increases in spring and summer months as stated by El-Samra et al. (1995).

The data available on heavy metal levels in the water of this coastal area are quite scarce. In this study, generally measured metal concentrations in seawaters from the sampling stations showed increased values when compared to other areas of the world. Conti and Cecchetti (2003) investigated the Cd, Cr, Cu, Pb, and Zn concentrations in seawater, *Ulva lactuca* L., *Padina pavonica* (L.) Thivy, *Mytilus galloprovincialis* Lamarck, *Monodonta turbinata* Born, and *P. caerulea* L. This study clearly indicated that our measured levels were much higher than those measured Cd, Cu, Pb, and Zn values in seawater taken from six coastal stations in the area of the Gulf of Gaeta (Tyrrhenian Sea, central Italy).

Conti et al. (2006) studied the heavy metal levels (Cd, Cr, Cu, Pb, and Zn) in seawater, *M. turbinata* B., and *P. caerulea* L. taken from selected stations located in Linosa Island (Sicily, Italy). Comparing the findings of this study, it can be said that our recorded heavy metals in seawater samples were higher than the reported values by Conti et al. (2006).

Hamed and Emera (2006) determined Cu, Zn, Pb, Cd, Cr, Ni, Fe, and Mn levels in coastal water samples, sediments, and soft tissues of the gastropod limpet, *P. caerulea*, and the bivalve, *Barbatus barbatus*, collected from seven different stations in the western coast of the Gulf of Suez. Comparing the results, it can be seen that our findings were higher than the determined heavy metal levels by Hamed and Emera (2006) (Table 7).

Nakhlé et al. (2006) monitored the cadmium, lead, and mercury concentrations in water, mussel, *Brachidontes variabilis*, and the limpet, *Patella* sp., along the Lebanese coasts (Eastern Mediterranean Sea) over a 2-year period. They were measured Cd (0.062 μg/l) and Pb levels in seawater (0.221 μg/l) (Table 7). According to the findings, it can be said that these values were very low compared with the data obtained in the present study.

Our detected Cd and Pb values in seawater were markedly higher (Table 2) than those reported Cd levels (0.06–0.73 μg/l) and Pb concentrations in water (0.21–15.79 μg/l) taken from stations in Kazanlı, Karaduvar, Mersin Port, and the Mezitli region in Mersin Bay which were mainly affected by pollution (Table 7) (Ayas et al. 2009).

Concentrations of Cd, Cr, Cu, Pb, and Zn were measured in seawater, *Posidonia oceanica* L. Delile tissues, *P. pavonica* (L.) Thivy, *Cystoseira* sp., *M. turbinata* Born, and *P. caerulea* L. collected at five sampling stations along the coastal areas of Linosa Island, Sicily (southern Tyrrhenian Sea, Italy) (Conti et al. 2010). It can be seen that our findings were higher than the given mean results of Cd, Cu, Pb, and Zn levels in seawater by Conti et al. (2010) (Table 7).

In aquatic environments, metal concentrations in solution are often near analytical detection limits and may be highly variable over time (Luoma et al. 1982), so that sediments can act as an adsorptive sink and a scavenger agent for heavy metals (Tessier and Campbell 2005). Moreover, heavy metal concentrations in surface sediments can provide historical information on heavy metal inputs at those locations (Förstner and Salamons 1980).

According to the results of the present study, heavy metal levels in sediment samples were found to be many times greater than the measured metal values in the water column. All of the studied metals exhibited stational variations [Cd ($p < 0.05$), Cu ($p < 0.001$), Fe ($p < 0.001$), Mn ($p < 0.001$), Ni ($p < 0.001$), Pb ($p < 0.001$), and Zn ($p < 0.001$)]. The recorded metal contents in sediment samples were generally higher at the southern part of the sampling area (S3, S4, and S5) (Table 3). This result concluded that anthropogenic pollution of the heavy metal levels at the southern part of the area was clearly noticed in sediments.

Generally, measured heavy metals in sediment samples were higher in summer months (Table 3). During summer, the analyzed heavy metal levels increased markedly. Fishing boats, shipping, tourism activities, and antifouling paints may be associated with increased heavy metal levels in sediment during this period.

The results showed that the accumulation rates of the overall mean metal levels in sediment samples decreased as follows: Fe > Cu > Mn > Ni > Zn > Pb > Cd. All measured heavy metals showed spatial variations. In contrast, no clear temporal patterns were observed.

Higher mean Cd values were assessed at station S5. Moreover, the average Cu contents were represented with a slightly higher level at stations S2 and S3 (Table 3). Compared to the other sampling stations, the determined Fe, Mn, and Ni levels in sediment samples during all seasons were generally higher at station S3. The high metal concentrations are most likely due to the local mineralogy and the natural origin of this station, rather than pollution.

The local distribution of the mean Pb and Zn contents in sediment at station 5 showed higher values compared to the other stations (Table 3). The high sedimentary Pb and Zn levels were recorded during summer and winter. This can be due to the precipitation of decomposed organic matter and antifouling paints (Bazzi 2014).

As for sediment samples, significant correlations existed between Mn and Fe ($r = 0.776$, $p < 0.001$), Mn and Ni ($r = 0.615$, $p < 0.001$), Zn and Pb ($r = 0.502$, $p < 0.001$), Cu and Fe ($r = 0.705$, $p < 0.001$), and Cu and Ni ($r = 0.642$, $p < 0.001$). Moreover, Ni and Fe levels in sediment samples revealed a high-degree relation ($r = 0.882$, $p < 0.001$). These correlations may suggest a common pattern of the sediments at the sampling stations.

A comparison of our results with the findings of Güven et al. (1998) showed that Zn levels in sediment samples collected from Sinop Stations (Şile, Riva, and İğneada) were lower than those recorded values in our study (Table 7).

In another study, Topcuoğlu et al. (2004) studied the biota and sediment samples collected from the Marmara Sea in Turkey. They found that the measured Cd, Cu, Fe, Mn, Ni, Pb, and Zn values in sediment samples taken from Şarköy, Ereğli, and Menekşe stations were higher than our reported findings (Table 7).

Hamed and Emera (2006) reported that levels of Cd, Cu, Fe, Mn, Ni, Pb, and Zn in sediment collected from the Gulf of Suez were 1.29–3.23, 2.79–8.65, 1.24–3.28, 12.9–64.2, 16.8–34.3, 12.1–36.5, and 27.4–51.8 μg/g, respectively (Table 7). By comparing the present data with that of Hamed and Emera (2006), it can be seen that our findings were higher than the given results.

It has been reported that the high levels of some heavy metals in the algae reflect firstly the high bioavailability of the metals in the study area and secondly the capacity of the alga to accumulate them. Many factors may influence the bioavailability of metals in algae including pH, salinity, temperature, light, oxygen, nutrient concentrations, complexing agents, particulate matters, and organic matters (Garnharm et al. 1992; Favero et al. 1996; Jothinayagi and Anbazhagan 2009).

Table 7 Comparisons of recorded heavy metal levels in seawater (μg/l) and in sediment samples (μg/g) during the present study with literature reported from different areas

	Sites	Cd	Cu	Fe	Mn	Ni	Pb	Zn	Reference
Seawater	Gulf of Gaeta (central Italy)	0.115 ± 0.006–0.142 ± 0.004	0.79 ± 0.03–1.67 ± 0.03				1.05 ± 0.02–2.83 ± 0.01	4.75 ± 0.11–6.37 ± 0.21	Conti and Cecchetti (2003)
	Linosa Island (Sicily Italy)	0.16 ± 0.04	0.96 ± 0.08				0.68 ± 0.15	10.66 ± 1.11	Conti et al. (2006)
	Gulf of Suez, Red Sea	0.04–0.27	1.15–4.78	6.33–32.78	1.28–4.56	1.36–3.65	0.56–3.17	6.79–25.19	Hamed and Emara (2006)
	Lebanese Coasts (Eastern Mediterranean Sea)	0.062					0.221		Nakhle et al. (2006)
	Mersin Bay (Turkey)	0.06–0.73					0.21–15.79		Ayas et al. (2009)
	Linosa Island, Sicily (southern Tyrrhenian Sea, Italy)	0.33 ± 0.03	2.34 ± 0.26				0.97 ± 0.15	13.0 ± 1.0	Conti et al. (2010)
	Aegean Sea (Turkey)	1.00 ± 0.02–5.00 ± 0.09	1.20 ± 0.06–71.0 ± 0.8	35.0 ± 3.8–372 ± 42	2.00 ± 0.12–81.0 ± 0.7	1.00 ± 0.12–27.0 ± 0.6	1.00 ± 0.24–109 ± 8	3.00 ± 0.25–309 ± 54	This study
Sediment	Sinop, Black Sea (Turkey)							29.4 ± 3.4	Güven et al. (1998)
	Şile,Black Sea (Turkey)							16.8 ± 3.1	Güven et al. (1998)
	Riva, Black Sea (Turkey)							11.7 ± 2.7	Güven et al. (1998)
	Şarköy, Marmara Sea (Turkey)	<0.02	12.7 ± 0.4	14896 ± 45	372.9 ± 1.9	53.88 ± 0.70	22.7 ± 2.1	43.6 ± 0.1	Topcuoglu et al. (2004)
	Ereğli, Marmara Sea (Turkey)	<0.02	30.6 ± 0.1	5956 ± 36	273.6 ± 0.5	20.53 ± 0.47	31.9 ± 1.7	34.1 ± 0.1	Topcuoglu et al. (2004)
	Menekşe, Marmara Sea (Turkey)	0.50 ± 0.12	16.8 ± 0.2	11875 ± 36	384.2 ± 1.9	41.30 ± 1.59	21.6 ± 3.6	50.9 ± 0.1	Topcuoglu et al. (2004)
	Gulf of Suez, Red Sea	1.29–3.98	2.79–12.16	1.24–4.56	12.90–70.22	16.84–49.63	12.10–42.15	25.48–58.06	Hamed and Emara (2006)
	Aegean Sea (Turkey)	nd −3.60 ± 1.20	6.50 ± 4.31–37.0 ± 10.2	4385 ± 10–43117 ± 25	161 ± 3–857 ± 6	18.9 ± 4.6–271 ± 11	5.80 ± 1.16–78.3 ± 1.1	21.8 ± 10.2–445 ± 102	This study

nd not detectable

In addition to variations in the available metal concentrations in the ambient water, other factors, such as water conditions, and structural differences among the algae species such as the stage of development and variation in growth and chemical composition of the algae may influence the pattern of accumulation (Wai-Yin and Wen-Xiong 2001; Jothinayagi and Anbazhagan 2009).

As for *C. barbata*, the accumulation order of overall mean metal levels was as follows: Fe > Zn > Mn > Pb > Ni > Cd > Cu. According to the results, it can be seen that essential metals in cell metabolism such as Fe and Zn were presented in relatively high amounts in algal tissue (Malea and Haritonidis 1999).

Fe values in *C. barbata* showed stational variations ($p < 0.05$), and especially station S1 generally showed higher Fe values. Furthermore, Mn contents in *C. barbata* ($p < 0.001$) showed stational variations, while Cu ($p < 0.05$), Ni ($p < 0.01$), and Zn levels ($p < 0.05$) exhibited seasonal differences. The high Fe levels found in seaweed as compared to the other heavy metals, e.g., Zn and Cu, are probably due to several factors such as the established need for Fe for normal growth of marine plants (Goldberg 1952) and the ability of most algal species to biomagnify Fe from the surrounding environment (Eisler 1981).

Algae, in general, accumulate Zn and Cu readily from seawater (Ho 1988). According to Moore and Ramamurti (1987), in benthic macrophytes, Zn levels not exceeding 100 μg/g are suggested as background for non-polluted areas. In this study, mean Zn levels in *C. barbata* were mostly recorded to be higher than both 100 μg/g and measured seawater values. It can be concluded that higher Zn concentrations in *C. barbata* are probably partly related to some of the characteristics of the sampling stations subjected to anthropogenic contamination (Table 4).

In literature, Cu levels of 200–300 μg/g have been recorded in species from polluted areas (Haug et al. 1974). Recorded average Cu levels in *C. barbata* changed between not determined and 6.64 ± 1.13 μg/g and exhibited temporal variations ($p < 0.05$). Measured Cu levels in *C. barbata* in September 2002 represented higher values than in other sampling seasons. The relatively low mean concentrations of Cu determined in the present study may indicate the lack of anthropogenic sources of this metal in the investigated areas (Table 4).

In the present study, Cu ($p < 0.05$), Ni ($p < 0.01$), and Zn levels in *C. barbata* ($p < 0.05$) exhibited statistical differences among sampling months. The bioaccumulation of metals by a plant depends on time of year, as there are seasonal changes in growth and chemical composition influencing the pattern of accumulation, in addition to differences in activity concentrations in the environment (Carlson and Erlandsson 1991).

Manganese in *C. barbata* showed stational variations ($p < 0.001$). Moreover, stations (stations S1 and S2) located in the northern part of the sampling area showed higher Mn levels in *C. barbata*. Seaweeds have the ability to concentrate metals from solution, accumulate only those metals that are biologically available, and integrate short-term temporal fluctuations in concentrations, so that seaweeds have commonly been employed as indicators of biologically available heavy metals in aqueous environments (Alahverdi and Savabieasfahani 2012; Søndergaard et al. 2014) Although measured mean Mn levels in seawater were low, it can be seen that *C. barbata* accumulated Fe, Mn, and Zn in a higher degree than seawater. This observation may support the use of *C. barbata* as an indicator species for determination of Fe, Mn, and Zn levels.

A positive correlation between Fe concentrations in *C. barbata* and Mn in *C. barbata* ($r = 0.638$, $p < 0.001$) indicated that these metals enter from the same sources and/or that, among these metals, there are synergistic interactions for the binding sites of the plant (Villares et al. 2002).

A comparison of our results with the findings of Güven et al. (1998) who have worked with *C. barbata* collected from Sinop, Şile, Riva, and İğneada stations showed that the highest concentrations of Cd, Fe, Pb, and Zn recorded herein are lower than those noted values in our study, except Cu levels in sediment (Table 8).

Similarly, our recorded Cd, Cu, Mn, Ni, Pb, and Zn levels in *C. barbata* were generally higher than the mean heavy metal concentrations in *C. barbata* species collected from different sites in N. Fokaea, in Kalandra, in A Trias, in Alykes, in Lehonia, in Monolithos, in Perivolos, in Heraklion, and in Chania (Sawidis et al. 2001) (Table 8).

Caliceti et al. (2002) investigated the accumulation levels of heavy metals (Fe, Zn, Cu, Cd, Ni, Pb, Cr, and As) in seven seaweeds collected from four sampling sites in the lagoon of Venice, in the 1999 spring and autumn periods. Our findings showed that the obtained Cd, Fe, Ni, Pb, and Zn levels in *C. barbata* were considerably higher and Cu values were lower than those recorded values in the Venice lagoon (Table 8).

This study shows that our recorded mean metal levels in *C. barbata* were higher than those noted values in Pazar Station and in Rize Station (Topcuoğlu et al. 2003a); also, our measured average Cu contents in *C. barbata* were generally similar, and Cd, Fe, Pb, Mn, Ni, and the mean Zn values were higher than the detected values in *C. barbata* at Şile and Sinop stations (Topcuoğlu et al. 2003b (Table 8).

A comparison of the findings showed that our recorded heavy metal results in *C. barbata* were generally higher than the reported values for Cd, Cu, Fe, Mn, Ni, Pb, and Zn in Şarköy Station, except for the recorded Cu level in *C. barbata* from M. Ereğli station; the obtained findings [Cd (<0.02 μg/g), Fe (114 ± 0.1 μg/g), Mn (23.8 ± 0.1 μg/g), Ni (<0.1 μg/g), Pb (<0.1 μg/g), and Zn (41.8 ± 0.1 μg/g)] were generally higher than the present study results (Topcuoğlu et al. 2004) (Table 8).

Al-Masri et al. (2003) studied three alga species along the Syrian coast and analyzed for radioactivity and heavy metals.

Table 8 Comparisons of recorded heavy metals levels in *Cystoseira barbata* (μg/g dry weight) during the present study with literature reported from other locations in the world

	Site	Cd	Cu	Fe	Mn	Ni	Pb	Zn	Reference
C. barbata	Sinop (Turkey)	1.30 ± 0.02	4.15 ± 0.03	446 ± 88			5.3 ± 2.4	12.1 ± 5.1	Güven et al. (1998)
	Şile (Turkey)			231 ± 89				18.6 ± 7.7	Güven et al. (1998)
	Riva (Turkey)	1.08 ± 0.02	12.33 ± 0.18	604 ± 101			8.3 ± 1.2	7.0 ± 2.8	Güven et al. (1998)
	İğneada (Turkey)			<100				<7	Güven et al. (1998)
	N. Fokaea (Turkey)	0.47	6.1		70.5	8.7	2.1	22.3	Sawidis et al. (2001)
	Kalandra (Turkey)	0.83	8.8		40.5	14.2	2.5	16.4	Sawidis et al. (2001)
	A Trias (Turkey)	0.06	1.7		22.2	9.7	0.02	29.8	Sawidis et al. (2001)
	Alykes (Turkey)	0.4	2.1		30.9	4.3	0.02	18.7	Sawidis et al. (2001)
	Lehonia (Turkey)	2.6	3.2		181.3	28.5	0.02	58.1	Sawidis et al. (2001)
	Monolithos (Turkey)	0.4	2.7		47.0	14.6	0.02	17.2	Sawidis et al. (2001)
	Perivolos (Turkey)	1.4	0.7		7.9	6.8	0.02	8.8	Sawidis et al. (2001)
	Heraklion (Turkey)	1.1	3.8		5.6	4.3	0.02	14.2	Sawidis et al. (2001)
	Chania (Turkey)	2.7	1.7		34.0	16.0	0.02	24.5	Sawidis et al. (2001)
	Venice Lagoon (Italy)	<0.1–0.2 ± 0.0	2 ± 0–21 ± 0	199 ± 7–609 ± 100		1.4 ± 0.1–2.7 ± 0.6	1.4 ± 0.0–5.6 ± 0.7	15 ± 4–88 ± 11	Caliceti et al. (2002)
	Pazar (Turkey)	0.64 ± 0.06	4.12 ± 2.18		5.4 ± 2.1		6.7 ± 1.87	12.0 ± 2.6	Topcuoglu et al. (2003a)
	Rize (Turkey)	0.66 ± 0.12	6.30 ± 0.04		22.2 ± 1.3		14.1 ± 1.21	61.2 ± 1.1	Topcuoglu et al. (2003a)
	Şile (Turkey)	<0.02–0.78 ± 0.1	2.2 ± 0.08–5.7 ± 0.12	130 ± 1–427 ± 3	6.7 ± 0.1–32.1 ± 0.2	<0.1–9.1 ± 0.1	<0.1–1.4 ± 0.2	13.9 ± 0.3–35.1 ± 0.3	Topcuoglu et al. (2003b)
	Sinop (Turkey)	<0.02–<0.2	1.7 ± 0.02–6.0–0.01	463 ± 2–593 ± 4	22.7 ± 0.1–33.5 ± 3.0	4.7 ± 0.5–7.2 ± 0.1	<0.1–3.5 ± 0.4	6.5 ± 0.9–191.5 ± 0.4	Topcuoglu et al. (2003b)
	Şarköy (Turkey)	<0.02	5.2 ± 0.1	1511 ± 1.5	143.6 ± 0.6	12.36 ± 0.14	3.7 ± 0.2	113.5 ± 0.2	Topcuoglu et al. (2004)
	M. Ereğli (Turkey)	<0.02	164.3 ± 0.8	114 ± 0.1	23.8 ± 0.1	<0.1	<0.1	41.8 ± 0.1	Topcuoglu et al. (2004)
	Black Sea	0.55 ± 0.04	2.47 ± 0.18	242 ± 15	14.9 ± 1.3	2.05 ± 0.14	4.62 ± 0.32	6.62 ± 0.26	Tuzen et al. (2009)
Cystoseira sp.	Syrian Coast	<0.1–<0.5	7.21 (2.22)				1.31 (0.70)	14.37 (2.87)	Al-Masri et al. (2003)
	Aegean Coast	0.005 ± 0.00–0.19 ± 0.001	2.25 ± 0.17–11.28 ± 0.48	97.62 ± 3.01–526.38 ± 2.15	8.43 ± 0.15–131.37 ± 1.40		0.002 ± 0.00–0.008 ± 0.00	10.81 ± 0.29–62.48 ± 1.03	Akçali and Kücüksezgin (2009)
	Linosa Island (Sicily, Italy)	0.32 ± 0.04–2.58 ± 0.88	4.44 ± 0.16–8.24 ± 1.47				1.33 ± 0.16–14.57 ± 1.70	22.5 ± 2.5–28.2 ± 2.8	Conti et al. (2010)
	South-western Sardinia (Italy)	1.72	1.80				10.3	52.4	Schintu et al. (2010)
	Marsa-Matrouh Beaches (Egypt)	0.54	8.66	204.28			18.31	27.90	Khaled et al. (2014)
	Aegean Sea (Turkey)	0.03 ± 0.00–7.23 ± 0.78	nd–6.64 ± 1.13	570 ± 45–6288 ± 301	15.9 ± 1.5–414 ± 15	0.90 ± 0.21–33.0 ± 2.7	0.90 ± 0.13–62.1 ± 5.4	80.3 ± 10.4–295 ± 21	This study

nd not detectable

The findings showed that our measured Cd, Cu, Pb, and Zn levels were higher than the given results by Al-Masri et al. (2003).

Akçalı and Küçüksezgin (2009) analyzed the heavy metal levels in *Cystoseira* sp. and also in seawater samples collected from six sampling stations along the Aegean coast. Results clearly showed that our study findings were consistently higher than the reported values for Cd, Fe, Mn, Pb, and Zn values in *C. barbata*, except Cu levels in *C. barbata* (Table 8).

Tuzen et al. (2009) investigated the contents of Cd, Cu, Fe, Mn, Ni, Pb, and Zn of different marine algae species collected from the Black Sea coasts. Comparing the results showed that the detected Cu, Cd, Fe, Mn, Ni, Pb, and Zn levels in this study were higher than the obtained findings by Tuzen et al. (2009) (Table 8).

The highest mean Cd, Pb, and Zn values of *C. barbata* reported in this study were, however, higher than those noted mean Cd, Pb, and Zn levels in *C. barbata* collected from five sampling stations along the coastal areas of Linosa Island, Sicily (southern Tyrrhenian Sea, Italy), by Conti et al. (2010). Moreover, the highest average Cu content in *C. barbata* measured in our study was lower than the detected Cu levels in *C. barbata* by Conti et al. (2010) (Table 8).

Schintu et al. (2010) investigated Cd, Cu, Pb, and Zn in green, red, and brown algae species collected from five coastal sites in south-western Sardinia (Italy). Our results in *C. barbata* were higher than the detected values reported by Schintu et al. (2010).

Khaled et al. (2014) studied the accumulation of some heavy metals in *Cystoseira* sp. collected from ten locations along the Marsa-Matrouh beaches (Egypt). They found 0.54 μg/g for Cd, 8.66 μg/g for Cu, 204.28 μg/g for Fe, 18.31 μg/g for Pb, and 27.90 μg/g for Zn (Table 8). Comparing our results with those of Khaled et al. (2014), it can be seen that our Cd levels in *C. barbata* were higher, Fe and Zn levels markedly higher, and Pb values considerably higher, while Cu contents were lower than the reported findings.

In the present research, the mean metal levels in *P. caerulea* from all stations were in the following order: Fe > Zn > Mn > Ni > Pb > Cu > Cd. According to the results, it can be seen that Fe, Mn, and Zn were the dominant metals among all the seven studied metals. Furthermore, similar Mn and Zn concentrations and also Cu and Pb levels were determined in the tissue of limpets from the investigation (Table 5).

The maximum heavy metal levels in limpet samples were generally noted in winter and spring periods. This high metal concentration during these seasons can be related to rain and runoff. Similar findings were reported by Fowler and Oregioni (1976) and Bordbar et al. (2015).

In the present study, concentrations of Cd ($p < 0.05$) in *P. caerulea* showed location-dependent variation. This situation described above could arise as a result of both environmental and physiological factors. So the significant differences in metal levels in *P. caerulea* at the sampling stations may be related to the availability of different metals to the animal with varying stations and also their ability for different uptake and retention mechanisms for the same metal at different stations (Bazzi 2014).

The present study has highlighted seasonal variations for Cd ($p < 0.01$), Cu ($p < 0.001$), Fe ($p < 0.001$), Mn ($p < 0.05$), Ni ($p < 0.01$), Pb (0.05), and Zn ($p < 0.001$) levels in *P. caerulea*. Generally higher concentrations of heavy metals occurred during the winter and spring months (Table 5). So, seasonal variations in metal content are influenced by a number of intrinsic factors such as size, age, sex, feeding behavior, and growth rate and extrinsic factors such as metal concentrations and speciation in surrounding waters, salinity, hardness, and temperature (Powell and White 1990; Hernandez et al. 1992; Pastor et al. 1994).

It is now a well-established fact that the metal content in mollusk is a function of inherent (biological) and external (including the contaminant bioavailability in the environment) factors, which combine to produce a quasi steady state between the metal concentration within the organism and its surrounding environment. For contamination monitoring purposes, it is important to maximize the significance of external factors by minimizing the biological ones such as metal retention in soft tissues of mollusks in their growth and reproduction. In fact, the growth rate and reproduction cycling may change the metal level of the organisms by diluting or concentrating the metal concentration of the organisms (Phillips and Rainbow 1993).

Cadmium has the ability to bound to metallothioneins, and those proteins cannot be found in detectable amounts in limpets caught from unpolluted waters (Noël-Lambot et al. 1978). This may explain how limpets can tolerate such high Cd levels in their tissues.

From a statistical point of view, there were significant relations ($p < 0.05$) for concentrations of Cd in *P. caerulea* and Fe in *P. caerulea* ($r = 0.473$, $p < 0.001$); Cd in *P. caerulea* and Mn in *P. caerulea* ($r = 0.423$, $p < 0.01$); Cd in *P. caerulea* and Ni in *P. caerulea* ($r = 0.487$, $p < 0.001$); Cd in *P. caerulea* and Pb in *P. caerulea* ($r = 0.268$, $p < 0.05$); Cd in *P. caerulea* and Zn in *P. caerulea* ($r = 0.883$, $p < 0.001$); Fe in *P. caerulea* and Mn in *P. caerulea* ($r = 0.713$, $p < 0.001$); Fe in *P. caerulea* and Zn in *P. caerulea* ($r = 0.486$, $p < 0.001$); Mn in *P. caerulea* and Ni in *P. caerulea* ($r = 0.365$, $p < 0.05$); Mn in *P. caerulea* and Zn in *P. caerulea* ($r = 0.539$, $p < 0.001$); Ni in *P. caerulea* and Pb in *P. caerulea* ($r = 0.530$, $p < 0.001$); Ni in *P. caerulea* and Zn in *P. caerulea* ($r = 0.686$, $p < 0.001$); and Pb in *P. caerulea* and Zn in *P. caerulea* ($r = 0.415$, $p < 0.01$). The results indicate that these metals probably originated from some common sources. On the other hand, Cu in *P. caerulea* showed negative weak correlations with Mn in *P. caerulea* ($r = -0.358$, $p < 0.05$) and also Zn in *P. caerulea* ($r = -0.299$, $p < 0.05$).

Compared with other studies, it was observed that our Cd levels in *P. caerulea* were lower than the findings detected from stations situated at Kazanlı, Karaduvar, Mersin Port, and the Mezitli region (0.11–2.04 μg/g dry weight) (Ayas et al. 2009). In addition, our measured cadmium concentrations were markedly lower than those reported values in *P. caerulea* from five stations in Favignana Island (Sicily, Italy) by Campanella et al. (2001) (4.41 μg/g) and Cubadda et al. (2001) (1.07–9.12 μg/g) (Table 9).

Mollusks accumulate the bioavailable fraction of the contaminants present in the water, and the metal content of their tissues reflect generally the profile of the pollution (Storelli and Marcotrigiano 2001). According to Nicolaidou and Nott (1989), Campanella et al. (2001), and Cubadda et al. (2001), the species of the genus *Patella* was the strongest accumulator of Cd. On this basis, the relatively low cadmium levels observed in this study may indicate the lack of anthropogenic sources of this metal in the investigated areas.

In the present study, the measured iron values in *P. caerulea* were much greater than those of the other metals in each station. Similar results have been found in a study carried out by Miguel et al. (1999) and Cravo et al. (2004), and they reported that Fe is the predominant metal in marine mollusks from the Mediterranean Sea.

Iron and generally zinc in the tissues of *P. caerulea* showed higher levels in every season compared to the rest of the elements. According to Pourang et al. (2005), Zn, Fe, and Cu are essential elements and play important roles in growth, cell metabolism, and survival of most animals including mollusks. Hence, the relatively high levels of these metals can be attributed to their essentiality. Similar results were also obtained by Yüzereroğlu et al. (2010) in *P. caerulea* samples collected from polluted and non-polluted areas from the Iskenderun Gulf (Mediterranean Turkey).

In particular, snails of the genus *Patella* take up large amounts of Zn which is an essential element ensuring the proper functioning of many enzymes and other compounds' crucial significance in metabolism. Furthermore, snails also have high content of iron (Jakimska et al. 2011), a constituent of goethite (FeOOH). Zn and Cu are constituents of haemocyanin (Cravo and Bebianno 2005). The correlation between these two elements can be explained by the fact that they are bound metallothionein (MT), a compound of great significance in homeostasis and in the binding and release of the metals (Krone et al. 1999).

Compared with other studies, the recorded concentrations in this study were slightly lower than those reported values in Cu, Ni, Pb, and Zn concentrations in *P. caerulea* collected along the Mediterranean Sea of Israel by Ramelow (1985) and also lower than Cu, Mn, Ni, and Zn levels in *P. caerulea* species taken from 3 clean stations at Central Aegean Sea, Milos Island, Greece, by Catsiki et al. (1991); Cd, Cu, Pb, and Zn contents in specimens collected from 5 stations located along the coast of Favignana Island, Sicily, Italy, by Cubadda et al. (2001); Cd, Cu, Pb, and Zn levels in samples obtained from 6 coastal stations in the area of the Gulf of Gaeta (Tyrrhenian Sea, central Italy) by Conti and Cecchetti (2003); Cu, Ni, and Zn contents in limpet samples taken from 11 stations located at the coastlines of Saronic Gulf from Sounia to Poros Island, Greece, including areas affected by naval and industrial operations, and a naturally enriched heavy metal site by Kontopoulos et al. (2003); Cu, Pb, and Zn concentrations in studied samples collected from Ustica Island, Sicily (Italy), by Conti et al. (2004) and also except Fe values; Cd, Cu, Mn, Ni, Pb, and Zn contents in samples obtained from İskenderun Bay (northern East Mediterranean Sea) by Turkmen et al. (2005); Cd, Cu, Pb, and Zn levels in samples taken from stations positioned in Linosa Island, Sicily (Italy), by Conti et al. (2006) and except Fe concentrations; Cd, Cu, Mn, Ni, Pb, and Zn values in *P. caerulea* samples obtained from the western coast of the Gulf of Suez by Hamed and Emera (2006); Cd, Cu, Pb, and Zn levels in samples taken from Ustica Island, Sicily (Italy), by Conti et al. (2007); Cd and Pb levels in samples collected from Mersin Bay by Ayas et al. (2009); Cd, Cu, Mn, Ni, Pb, and Zn, except Fe values in samples obtained from 3 sites (Bizerta Channel, La Goulette, and Sidi Rais) along the north coasts of Tunisia by Belkhodja et al. (2010); Cd, Cu, Pb, and Zn contents in samples collected at 5 stations placed along the coasts of Linosa Island by Conti et al. (2010); Cd and Pb levels in samples collected from 12 coastal stations in the Gulf of Gabes by Rabaoui et al. (2013); Cu, Fe, Mn, and Zn concentrations in specimens taken from stations along the coastline of Larymna Bay (Greece) by Bordbar et al. (2015); and Cu, Mn, Ni, Pb, and Zn values in the samples obtained from stations at Stratoni and Artemida coastal waters (North East Greece) by Kelepertzis (2013) (Table 9).

The findings of the present study for limpet samples were slightly lower than the recorded Cd (4.41 μg/g), Cu (1.7 μg/g), and Zn contents (5 μg/g), and also these study findings were also similar to the Pb levels (0.20 μg/g) than that reported by Campanella et al. (2001) from the Favignana Island, Sicily (Italy) (Table 9).

Meanwhile, Yüzereroğlu et al. (2010) studied heavy metal levels in *P. caerulea* specimens collected from two coastal sites in Iskenderun Gulf. They reported higher concentrations for Cd, Cu, Fe, Ni, Pb, and Zn than that recorded in the present study (Table 9).

The relative sequence of the overall mean metal levels evaluated in the musell of *L. aurata* could be arranged in the following order: Fe > Zn > Mn > Cu > Ni > Pb > Cd. In relation to the impact of metals on human health, the muscle tissue of fish has been investigated more than the other organs, because it is the main fish part consumed by humans.

Iron and zinc were most abundant in all fish tissues under investigation. Golden grey mullet are a kind of filter feeders,

Table 9 Comparisons of recorded heavy metal levels in *Patella caerulea* (μg/g dry weight) during the present study with literature reported from different areas

	Site	Cd	Cu	Fe	Mn	Ni	Pb	Zn	Reference
P. caerulea	Mediterranean Sea of Israel		3.5–13.7			2.1–83.7	0.3–3.2	44–96	Ramelow (1985)
	Central Aegean Sea, Milos Island (Greece)	7.6–12.0			11.3–32.1	19.1–28.6		31.9–56.5	Catsiki et al. (1991)
	Favignana Island (Sicily, Italy)	3.30 ± 0.37–6.30 ± 2.08	1.21 ± 0.30–2.35 ± 0.67				0.14 ± 0.05–1.52 ± 0.34	3.5 ± 1.6–14.6 ± 3.8	Campanellaet al. (2001)
	Favignana Island (Sicily, Italy)	1.7–11.8	0.47–3.79				0.06–2.18	2.2–19.1	Cubadda et al. (2001)
	Gulf of Gaeta (Tyrrhenian Sea, central Italy)	2.89 ± 0.57–4.06 ± 0.84	10.2 ± 1.4–19.2 ± 3.6				0.51 ± 0.15–1.50 ± 0.29	87.4 ± 11.8–117.1 ± 25.0	Conti and Cecchetti (2003)
	Saronic Gulf from Sounia to Poros Island (Greece)		5.0–77.4	96–3045		6.0–31.6		43–367	Kontopoulos et al. (2003)
	Ustica Island (Sicily, Italy)		4.87 ± 2.19–9.50 ± 5.63				0.85 ± 0.57–1.30 ± 0.66	44.20 ± 11.09–56.62 ± 12.80	Conti et al. (2004)
	Iskenderun Bay, northern East Mediterranean Sea	2.39 ± 0.92–4.97 ± 1.04	1.58 ± 0.59–4.02 ± 0.67	15.34 ± 1.76–41.20 ± 7.82	1.39 ± 0.48–4.31 ± 0.94	3.60 ± 1.02–12.21 ± 1.83	4.28 ± 0.86–14.53 ± 2.01	23.13 ± 3.94–46.59 ± 6.71	Turkmen et al. (2005)
	Linosa Island(Sicily, Italy)	2.69 ± 0.63–4.97 ± 1.31	2.78 ± 0.88–9.71 ± 2.25				0.40 ± 0.11–2.41 ± 0.65	38.40 ± 6.26–51.47 ± 9.62	Conti et al. (2006)
	Gulf of Suez, Red Sea	0.63–2.13	1.61–12.17	1.24–2.94	24.91–76.30	3.06–9.88	6.23–70.91	56.47–191.42	Hamed and Emara (2006)
	Ustica Island(Sicily, Italy)	4.7	6.3				1.02	51	Conti et al. (2007)
	Mersin Bay (Turkey)	0.11–2.04					0.00–3.74		Ayas et al. (2009)
	North Coasts of Tunisia	0.43–1.50	5.59–9.29	1.86–2.59	5.4–10.76	3–4.14	2.73–3.61		Belkhodja et al. (2010)
	Linosa Island(Sicily, Italy)	2.69 ± 0.63–4.97 ± 1.31	2.78 ± 0.88–9.71 ± 2.25				0.40 ± 0.11–2.41 ± 0.65	38.4 ± 6.3–51.5 ± 9.6	Conti et al. (2010)
	İskenderun Bay (Turkey)	0.24 ± 0.03–0.68 ± 0.03	1.09 ± 0.01–5.58 ± 0.18	36.56 ± 1.25–212.9 ± 6.60		0.39 ± 0.03–1.60 ± 0.05	0.05 ± 0.01–0.70 ± 0.03	3.70 ± 0.20–13.71 ± 0.15	Yüzeroğlu et al. (2010)
	Gulf of Gabes	0.800 ± 0.0288–4.300 ± 0.3398					0.823 ± 0.0575–2.123 ± 07075		Rabaoui et al. (2013)
	Larymna Bay (Greece)		7.2–95.8	843–16232	1.9–33.6			38.1–126.0	Bordbar et al. (2015)
Patella sp.	Stratoni and Artemida (North East Greece)		6 ± 1–28 ± 4		10 ± 5–36 ± 17	9 ± 3–9 ± 4	8 ± 5–96 ± 19	48 ± 4–196 ± 31	Kelepertzis (2013)
P. caerulea	Aegean Sea (Turkey)	0.004 ± 0.001–0.065 ± 0.019	nd–0.249 ± 0.051	1.85 ± 0.89–76.0 ± 5.7	0.048 ± 0.016–0.495 ± 0.078	0.021 ± 0.002–0.339 ± 0.067	0.010 ± 0.003–0.191 ± 0.028	0.26 ± 0.06–1.66 ± 0.23	This study

nd not detectable

and they are benthofagous and omnivorous species. They filter plankton and fine suspended particles in water and consume organic matter, such as bacteria, bottom diatom, and algae encrusting on mineral grains. Moreover, they are known to uptake and utilize sand granules to grind the cell wall of microalgae in digestion. Mullets are particularly exposed to possibly sediment-associated contamination. Furthermore, these organisms seem to be appropriate for monitoring contaminants in estuarine environments and in coastal waters (Licata et al. 2003; Marcovecchio 2004).

Seasonal changes in metal Cu ($p < 0.001$), Fe ($p < 0.05$), Mn ($p < 0.001$), Pb ($p < 0.001$), and Zn concentrations ($p < 0.001$) in the tissues of *L. aurata* were observed with slightly higher concentrations during July 2002. In contrast, no statistically significant stational differences were found. Seasonal variations of metal concentrations in fish may result from intrinsic factors such as growth and reproductive cycle and from changes in water temperature. Additionally, the differences noted in the metal concentrations in different tissues among seasons could have been the result of local pollution. A number of studies have shown that various factors, such as season (Kargın 1996), length and weight (Uysal and Tuncer 1982), and physical and chemical status of water (Johnson 1988), can play a major role in the tissue accumulation of heavy metals. Temperature is known to be a significant factor affecting metal uptake by aquatic organisms through increased metabolism (Voutsino-Taliadouri 1982). Larson et al. (1985) indicated that many physiological parameters in fish are subject to seasonal variations.

The results obtained from samples collected in summer show comparatively high metal concentrations (Table 6). This high metal accumulation could be due to heavy rainfall during spring, which increases the metal burden of the water by washing down the metal contained in soil.

With regard to correlation results, Cu in *L. aurata* showed positive relations with Fe in *L. aurata* ($r = 0.587$, $p < 0.05$), with Mn in *L. aurata* ($r = 0.853$, $p < 0.001$), with Ni in *L. aurata* ($r = 0.601$, $p < 0.05$), with Pb in *L. aurata* ($r = 0.620$, $p < 0.05$), and with Zn in *L. aurata* ($r = 0.811$, $p < 0.01$). Moreover, Fe in *L. aurata* exhibited a positive correlation with Mn in *L. aurata* ($r = 0.860$, $p < 0.001$) and Fe in *L. aurata* ($r = 0.587$, $p < 0.05$) revealed a positive relationship with Zn in *L. aurata* ($r = 0.643$, $p < 0.05$). Moreover, Mn in *L. aurata* ($r = 0.846$, $p < 0.01$) showed a positive relation with Zn in *L. aurata* ($r = 0.846$, $p < 0.01$). The results exhibited that these metals might originate mainly from common sources.

Filazi et al. (2003) investigated the heavy metal levels in muscle tissues of fish (*M. auratus*) collected from the stations located in Black Sea coasts (Sinop-İçliman). Comparison of Cd, Cu, and Pb levels in the present study revealed that our detected concentrations in this study were lower than the reported Cd, Cu, and Pb values by Filazi et al. (2003) (Table 10).

Amini Ranjbar and Sotoudebnia (2005) investigated the concentrations of heavy metals (Cd, Cu, Pb, and Zn) in the muscle tissue of *M. auratus* caught from important parts of the Caspian Sea in Mazandaran Province. Regarding the results, our findings were slightly lower than the reported levels by Amini Ranjbar and Sotoudebnia (2005) (Table 10).

Krishna et al. (2014) studied the heavy metal levels in muscle of *M. cephalus* collected from the Machilipatnam coast. According to the findings of the present study, the results were lower than the average concentrations of Cu, Mn, Pb, and Zn noted by Krishna et al. (2014) (Table 10).

Results of the present investigation were lower than the measured Cd, Cu, Mn, Ni, Pb, and Zn contents in *M. cephalus* collected from Iskenderun Bay, Northern East Mediterranean (Turkey), but the detected highest Fe value was lower than that found in the present study (Türkmen et al. 2006) (Table 10).

Analysis of tissue metal concentrations, besides from the purpose of extending our knowledge about the biological role of the various organs and tissues examined, can also provide reliable information on the contamination degree of the marine environment, as well as constitute a tool to check the quality of marine foodstuffs concerning human health. According to the Turkish Food Codex (2011), the maximum levels of the metals for human consumption of fish may not exceed 0.05 and/or 0.1 mg/kg for Cd, 20 mg/kg for Cu, 0.3 mg/kg fish for Pb, and 50 mg/kg fish for Zn. In addition, heavy metal limits for fish samples set by FAO/WHO guidelines were 1.0 μg/g for Cd, 30 μg/g for Cu, 2 μg/g for Pb, 10 μg/g for Ni, and 100 μg/g for Zn (FAO 1983; WHO 1989). In this study, the measured heavy metal values in *L. aurata* were found to be lower than the maximum permissible levels issued by the Turkish legislation. Also, the studied metal concentrations in the muscle of *L. aurata* were not higher than the permissible limits proposed by FAO (1983) and WHO (1989).

The bioconcentration factor (BCF) may be used to evaluate the bioaccumulation ability of a species in an ecosystem (Chong and Wang 2001). BCF in the present study ranged from 0.01 to 5.92 (Cd), 0.01 to 4.52 (Cu), 2.98 to 116.65 (Fe), 0.52 to 117 (Mn), 0.06 to 18.00 (Ni), 0.01 to 8.50 (Pb), and 0.43 to 55.73 (Zn) in *C. barbata*; 0.001 to 0.052 (Cd), 0.00 to 9.71 (Cu), 0.02 to 1.40 (Fe), 0.00 to 0.09 (Mn), 0.00 to 0.17 (Ni), 0.00 to 0.03 (Pb), and 0.00 to 0.40 (Zn) in *P. caerulea*; and 0.0001 to 0.0012 (Cd), 0.0031 to 0.0155 (Cu), 0.0386 to 0.4269 (Fe), 0.0010 to 0.0669 (Mn), 0.0004 to 0.0139 (Ni), 0.0000 to 0.0026 (Pb), and 0.0022 to 0.1014 (Zn) in *L. aurata*. According to the findings of this study, it can be seen that *C. barbata* represented with higher BCF values especially for Fe, Mn, and Zn than *P. caerulea* and *L. aurata*. The results indicated that the accumulation potential of *C. barbata* for Fe, Mn, and Zn was higher than the other studied biota samples.

Table 10 Comparisons of recorded heavy metal levels in *Liza aurata* (*Mugil auratus*) (μg/g dry weight) during the present study with literature reported from different areas

	Site	Cd	Cu	Fe	Mn	Ni	Pb	Zn	Reference
M. auratus	Sinop-Icliman (Turkey)	0.11 ± 0.00–0.30 ± 0.05	0.51 ± 0.07–0.75 ± 0.09				0.67 ± 0.03–0.88 ± 0.01		Filazi et al. (2003)
M. auratus	Caspian Sea in Mazandaran Province	0.321	0.996				2.337	14.327	Amini Ranjbar and Sotoudebnia (2005)
M. auratus	Machilipatham Coast		5.5		6.3		8.4	25.2	Krishna et al. (2014)
M. cephalus	Iskenderun Bay, Northern East Mediterranean (Turkey)	0.195–0.551	0.738–2.243	8.869–18.75	1.296–3.092	0.661–1.587	1.263–2.088	3.202–8.032	Türkmen et al. (2006)
M. auratus	Aegean Sea (Turkey)	0.001 ± 0.0001–0.004 ± 0.0001	nd–0.058 ± 0.0002	8.06 ± 3.21–64.9 ± 16.4	0.057 ± 0.018–0.302 ± 0.098	nd–0.042 ± 0.010	nd–0.039 ± 0.001	0.138 ± 0.043–0.555 ± 0.111	This study

nd not detectable

Conclusion

The findings of this study suggest that the use of different bioindicator organisms for biomonitoring of heavy metals in the marine environment provides quantitative information on the quality of the studied area. Concerning stational variations, both stations S4 and S5 represented with higher heavy metal levels in seawater and sediment samples. According to the results, anthropogenic pollution of the heavy metal values at the southern part of the area was clearly noticed both in seawater and sediment. Furthermore, elevated concentrations of Fe, Zn, and Mn were noted in the studied biota samples.

Although measured Mn levels in seawater were low, it can be seen that *C. barbata* accumulated Fe, Mn, and Zn concentrations in a higher degree than seawater. This observation may support the use of *C. barbata* as an indicator species for Fe, Mn, and Zn contents. It can be concluded that higher Zn concentrations in *C. barbata* can be related to the characteristics of the sampling stations subjected to anthropogenic contamination.

According to the findings, positive correlations were found between Fe in water and Mn in water, Fe in sediment and Mn in sediment, Fe in *C. barbata* and Mn in *C. barbata*, Fe in *P. caerulea* and Mn in *P. caerulea*, and Fe in *L. aurata* and Mn in *L. aurata*. The results indicated that positive relations between these metals indicate that they might likely originate from common sources.

The highest mean Cu content in *C. barbata* measured in our study was lower than the reported Cu levels in previous studies. The relatively low concentrations of Cu in *C. barbata* in the present study may indicate the lack of anthropogenic sources of this metal in the investigated areas.

As for the species of the genus *Patella*, being a stronger accumulator of Cd, the relatively low cadmium levels observed in this study may indicate the lack of anthropogenic sources of this metal in the sampling stations.

A comparison of the findings with the literature showed that our recorded heavy metal results in seawater, sediment, and *C. barbata* were generally higher than the reported values. Compared with previous studies, it can be said that our recorded concentrations in *P. caerulea* and *L. aurata* were slightly lower than those reported levels from different parts of the world.

Based on the results, detected heavy metal values in *L. aurata* were found to be lower than the maximum permissible levels issued by the Turkish legislation. Also, the measured heavy metal levels in the muscle of *L. aurata* did not exceed the permissible limits issued by FAO (1983) and WHO (1989).

According to the findings of this study, it can be seen that *C. barbata* represented with high BCF values, especially for Fe, Mn, and Zn levels, compared to the recorded BCF values in *P. caerulea* and *L. aurata*. Therefore, further research works

are necessary to fully clarify the actual accumulation patterns of these organisms.

In conclusion, this study confirms that *P. caerulea*, *L. aurata*, and especially *C. barbata* are promising water-quality bioindicators and biomonitoring subjects for biologically available metal accumulation for Aegean Sea coastal waters.

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