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Heavy metal concentrations in macroalgae species from Sinop coasts of the Southern Black Sea

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ABSTRACT

Objective: To determine the concentrations of Fe, Zn, Ni, Cu, Mn, Pb, Cd and Co in macroalgae from Sinop coasts of the Sounthern Black Sea.

Methods: Chlorophyta-green algae (*Chaetomorpha* spp., *Cladophora* spp., *Ulva linza*, *Ulva intestinalis*, *Ulva lactuca*, *Ulva rigida*); Ochrophyta-brown alga [*Cystoseira barbata* (*C. barbata*)] and Rhodophyta-red algae (*Ceramium* spp., *Corallina panizzoi*) were collected seasonally in the year 2010 from the upper infralittoral zone of inner harbour, outer harbour, Gerze and Ayancık of Sinop coasts of the Black Sea. The samples were analysed by atomic absorption spectrophotometer in order to determine heavy metal levels in different algal division species along Sinop coasts and to provide information of marine environment quality as marine strategy framework directive is aimed to ensure good environmental status of the seas by 2020 in the European Union.

Results: The results showed that metal concentrations in all studied green algae, brown alga and red algae increase in the order: Cd < Co < Ni < Pb < Cu < Zn < Mn < Fe, Cd < Co < Cu < Ni < Pb < Mn < Zn < Fe and Co < Cd < Ni < Cu < Pb < Mn < Zn < Fe, respectively. In all divisions, among the essential elements the highest concentrations exhibited by Fe, in Gerze station- (2 328 ± 89) µg/g dry weight in *Chaetomorpha* spp.; (2 143 ± 78) µg/g dry weight in *C. barbata* and (968 ± 20) µg/g dry weight in *Ceramium* spp.

Conclusions: The highest accumulation of different metals in the analysed algae species were: Fe and Co in *Chaetomorpha* spp., Zn in *C. barbata*, Ni and Cu in *Ulva linza*, Mn in *Ulva intestinalis*, Pb and Cd in *Corallina panizzoi*.

1. Introduction

Seaweeds play extremely important and vital roles in the marine coastal ecosystems. They are the basis of food web, major food sources for herbivores and also create habitat for many invertebrates and vertebrates of ecological and economic importance. Many Far East and Pacific countries, particularly Japan consume macroalgae as food[1]. Although, the usage of marine algae in human diet is not common in Turkey, excessive levels of heavy metals in sea macroalgae can be potentially toxic to other marine organisms and human through the food chains[2].

Dissimilar to many other chemicals, heavy metals are not biodegradable and therefore heavy metal pollution in coastal areas is one of the severe problems when occurred in high concentrations[3] due to their toxicity and persistence and apt to bioaccumulation[4]. In the marine coastal environment, anthropogenic activities can contribute to heavy metal pollution via industrial, agricultural and domestic wastewaters, atmospheric input, mine runoff and solid waste disposal areas[4,5]. Many living marine organisms directly from surrounding water or through the food chain accumulate heavy metals and this bioaccumulation ability makes them for measuring the bioavailability of the metals[4,6].

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In the European Union, the Marine Strategy Framework Directive [7] includes certain measures to supply or maintain "good environmental status" of the Member States up to 2020. This directive mainly considers ecological status and is based on the assessment and monitoring of biological communities that reflect the quality of habitats. In the Marine Strategy Framework Directive[7] the qualitative Descriptor 8 (Contaminants and Pollution Effects) and Descriptor 9 (Contaminants in Fish and Other Seafood) need to describe toxic substances in biota including macroalgae and if any reduce pollutants by monitoring water bodies with macroalgae. For an organism to be a useful bio-indicator of heavy metal pollution, there should be a relationship between concentrations of metals in organisms and marine coastal environment[8,9]. According to Marine Strategy Framework Directive[7] macroalgae are evaluated as bio-monitors in determining heavy metal concentrations due to their reasonable size, sedentary life, easy collection, uptake and accumulation capacities, relatively easy identification, and considerable abundance[9,10]. The relationship between metal levels in macroalgae and marine environment is linear and illustrate environmental conditions[9].

Bat[4] pointed out that the Black Sea is effected by untreated domestic, industrial and agricultural wastes that drain into via rivers or directly by human activities. Sinop is one of the Black Sea natural harbors and has been subjected mainly domestic wastewater discharges and fisheries activities[11]. There are some available data[12-15] on heavy metal contents of macroalgae collected from the Turkish Black Sea coasts between 1979 and 2001. Also, a number of studies[16-19] have been carried out in Sinop coasts of the Black Sea.

The purpose of this study is to determine heavy metal concentrations in three different algal division species along Sinop coasts and provide information of environmental quality of this area according to Marine Strategy Framework Directive[7] and to evaluate threats of heavy metal in biota as macroalgae.

2. Materials and methods

2.1. Study area

Sinop is situated in the middle part of Turkey and also at the outermost point on the Turkish coastline of the Black Sea and extends between latitude of 42°02′ N and longitude 35°09′ E. Seasonal sampling was carried out at random four stations namely; 1-inner harbor, 2-outer harbor, 3-Ayancık and 4-Gerze) at the upper infralittoral zone along the rocky shores of Sinop coasts of the southern Black Sea (Figure 1).

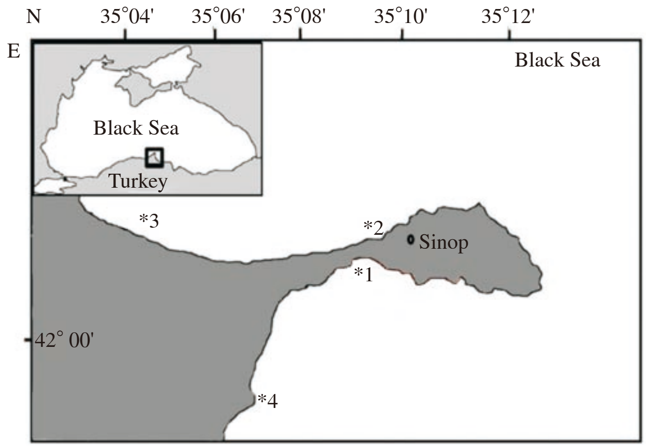

Figure 1. Study area.
1: Inner harbor; 2: Outer harbor; 3: Ayancık; 4: Gerze.

2.2. Sampling

Canopy forming algal samples were hand-picked on rocky substrata in 2010 seasonally at a water depth of 0–0.5 m with different degree of pollution. They were separated from the substrate with a knife and placed into labeled plastic bags. In the laboratory, they were identified, sorted to remove foreign particles, rinsed and dried on blotting papers and then in an oven at 70 °C for 24 h[17,18].

Nine different marine algae species have been analyzed: 6 species belonging to phylum Chlorophyta (*Chaetomorpha* spp., *Cladophora* spp., *Ulva linza*, *Ulva intestinalis*, *Ulva lactuca*, *Ulva rigida*), one species (*Cystoseira barbata*) to phylum Ochrophyta and 2 species (*Ceramium* spp., *Corallina panizzoi*) to phylum Rhodophyta. The main reference used for the identification of major macroalgae groups was Karaçuha and Ersoy Karaçuha[20].

One gram of dried sample was weighed into a 100 mL Erlenmeyer flask, and digested with 10 mL HNO_3:$HClO_4$ (5:1) by heating on a hot-plate at a low temperature (40–50 °C) for 3 days and left to cool to room temperature. Then, the samples were diluted with bi-distilled water to 50 mL. Analysis of Fe, Zn, Ni, Cu, Mn, Pb, Cd and Co was determined by using the Perkin Elmer Model 2280 atomic absorption spectrophotometer with background correction andair acetylene flame, and the data are expressed as μg metal per gram of dry weight (modified from Wahbeh *et al*.[21], and Denton and Burdon-Jones[22].

2.3. Statistical analysis

The concentrations of Fe, Zn, Ni, Cu, Mn, Pb, Cd and Co in macroalgae from each station were compared statistically us One-way ANOVA to determine differences[23].

3. Results

The present study was carried out for assessment of Fe, Zn, Ni, Cu, Mn, Pb, Cd and Co levels in the nine marine algae species collected from Sinop coasts of the Black Sea Turkey, in 2010. The mean ± SE of heavy metal levels of the nine species of algae are presented in Table 1.

The concentrations of metals were found to vary from 98.0 to 2328.0 μg/g dry weight for Fe, 1.6 to 76.0 μg/g dry weight for Zn, 0.18 to 18.40 μg/g dry weight for Ni, 1.10 to 14.00 μg/g dry weight for Cu, 1.78 to 67.00 μg/g dry weight for Mn, 0.40 to 28.00 μg/g dry weight for Pb, 0.04 to 3.10 μg/g dry weight for Cd and 0.09 to 3.10 μg/g dry weight for Co at four sites.

In the present study, the results showed that metal concentrations are Fe > Mn > Zn > Cu > Pb > Ni > Co > Cd for green algae, Fe > Zn > Mn > Pb > Ni > Cu > Co > Cd for brown alga and Fe > Zn > Mn > Pb > Cu > Ni > Cd > Co for red algae, respectively. In all divisions, the highest levels were found for

Table 1

The mean ±SE of heavy metal concentrations in macroalgae samples collected from Sinop coasts of the southern Black Sea in 2010 (μg/g dry weight).

Algae		Sites	Fe	Zn	Ni	Cu	Mn	Pb	Cd	Co
Chlorophyta	*Chaetomorpha* spp.	SIH	2 145.0 ± 78.0	22.0 ± 5.0	1.25 ± 0.10	3.00 ± 0.40	42.00 ± 5.00	5.00 ± 0.50	1.50 ± 0.10	2.80 ± 0.70
		SOH	982.0 ± 34.0	18.0 ± 2.3	–	2.20 ± 0.40	33.00 ± 3.40	2.00 ± 0.20	0.90 ± 0.10	1.20 ± 0.40
		Gerze	2 328.0 ± 89.0	25.7 ± 6.0	1.30 ± 0.10	3.50 ± 0.50	57.00 ± 9.00	6.00 ± 0.70	2.10 ± 0.20	3.30 ± 0.80
		Ayancık	751.0 ± 27.0	12.0 ± 2.0	–	4.80 ± 0.70	21.00 ± 8.00	1.40 ± 0.10	0.60 ± 0.10	1.00 ± 0.20
	Cladophora spp.	SIH	328.0 ± 34.0	37.0 ± 6.0	2.20 ± 0.15	3.40 ± 0.50	8.00 ± 2.00	0.90 ± 0.10	0.90 ± 0.10	1.80 ± 0.70
		SOH	98.0 ± 11.0	21.0 ± 4.0	–	2.10 ± 0.30	6.00 ± 1.00	0.60 ± 0.10	0.70 ± 0.06	1.10 ± 0.05
		Gerze	232.0 ± 28.0	25.0 ± 7.0	–	2.40 ± 0.50	1.78 ± 0.20	0.80 ± 0.10	0.09 ± 0.01	0.90 ± 0.10
		Ayancık	111.0 ± 14.0	8.0 ± 2.0	–	1.10 ± 0.20	8.00 ± 2.00	0.40 ± 0.05	1.00 ± 0.04	1.50 ± 0.30
	U. linza	SIH	1 342.0 ± 53.0	4.1 ± 0.2	18.40 ± 3.20	1.70 ± 0.10	31.00 ± 4.00	6.00 ± 0.50	0.04 ± 0.01	0.32 ± 0.07
		SOH	738.0 ± 28.0	11.0 ± 1.5	6.00 ± 0.90	2.20 ± 0.10	7.00 ± 1.00	–	–	1.40 ± 0.30
		Gerze	1 130.0 ± 44.0	19.0 ± 3.0	1.20 ± 0.05	14.00 ± 2.00	3.00 ± 0.40	5.20 ± 0.20	0.06 ± 0.01	0.70 ± 0.09
		Ayancık	681.0 ± 19.0	9.0 ± 1.0	4.00 ± 0.30	1.50 ± 0.07	5.00 ± 0.90	–	–	1.20 ± 0.10
	Ulva intestinalis	SIH	1 512.0 ± 51.0	32.0 ± 5.0	2.66 ± 0.40	4.70 ± 0.50	53.00 ± 7.00	8.00 ± 1.00	2.00 ± 0.30	0.90 ± 0.10
		SOH	956.0 ± 32.0	13.0 ± 2.0	1.34 ± 0.20	2.80 ± 0.20	23.00 ± 4.00	4.00 ± 0.50	1.20 ± 0.10	1.00 ± 0.20
		Gerze	655.0 ± 23.0	17.0 ± 3.0	3.20 ± 0.50	6.20 ± 1.30	28.00 ± 6.00	3.60 ± 0.20	0.07 ± 0.01	1.20 ± 0.25
		Ayancık	1 382.0 ± 42.0	36.0 ± 6.0	1.96 ± 0.50	5.80 ± 0.70	67.00 ± 8.00	6.40 ± 0.70	1.00 ± 0.10	0.70 ± 0.10
	U. lactuca	SIH	158.0 ± 48.0	14.0 ± 3.0	2.40 ± 0.12	6.60 ± 0.09	15.40 ± 2.00	5.60 ± 0.50	1.30 ± 0.50	1.40 ± 0.20
		SOH	117.0 ± 13.0	12.0 ± 2.0	2.70 ± 0.41	6.80 ± 0.10	13.00 ± 1.40	–	–	–
		Gerze	1 375.0 ± 42.0	30.0 ± 7.0	2.11 ± 0.14	10.00 ± 1.00	8.20 ± 0.60	–	0.11 ± 0.10	0.20 ± 0.01
		Ayancık	271.0 ± 11.0	39.0 ± 9.0	–	5.50 ± 0.09	11.00 ± 1.00	4.00 ± 0.20	–	–
	Ulva rigida	SIH	1 112.0 ± 47.0	3.0 ± 0.6	2.40 ± 0.15	3.80 ± 0.70	18.00 ± 3.00	5.90 ± 0.60	1.50 ± 0.30	1.10 ± 0.10
		SOH	1 382.0 ± 51.0	13.0 ± 3.0	2.30 ± 0.11	2.60 ± 0.30	21.00 ± 3.80	4.00 ± 0.30	2.30 ± 0.40	2.60 ± 0.40
		Gerze	2 002.0 ± 0.4	32.0 ± 0.2	1.80 ± 0.10	9.00 ± 0.60	19.00 ± 3.00	–	–	1.10 ± 0.10
		Ayancık	2 002.0 ± 0.4	32.0 ± 0.2	1.20 ± 0.04	7.00 ± 0.40	10.00 ± 1.00	–	–	1.00 ± 0.09
Phaeophyta	*C. barbata*	SIH	748.0 ± 29.0	65.0 ± 6.0	3.80 ± 0.60	4.80 ± 0.90	33.00 ± 4.00	8.00 ± 2.00	1.20 ± 0.10	2.40 ± 0.10
		SOH	308.0 ± 21.0	13.0 ± 2.0	1.74 ± 0.10	3.10 ± 0.30	2.40 ± 0.20	–	–	–
		Gerze	2 143.0 ± 78.0	76.0 ± 11.0	15.00 ± 2.00	7.00 ± 1.00	64.00 ± 9.00	10.00 ± 1.00	1.30 ± 0.10	0.30 ± 0.01
		Ayancık	261.0 ± 22.0	5.0 ± 1.0	2.30 ± 0.50	1.30 ± 0.06	19.00 ± 3.00	1.50 ± 0.10	0.09 ± 0.01	1.65 ± 0.07
Rhodophyta	*Ceramium* spp.	SIH	881.0 ± 32.0	50.0 ± 8.0	2.90 ± 0.30	3.70 ± 0.30	29.00 ± 5.00	2.00 ± 0.10	2.20 ± 0.10	0.09 ± 0.01
		SOH	561.0 ± 18.0	1.6 ± 0.3	0.18 ± 0.02	1.55 ± 0.40	22.50 ± 4.00	–	–	–
		Gerze	968.0 ± 20.0	38.0 ± 9.0	2.00 ± 0.30	8.00 ± 1.00	12.00 ± 2.00	–	0.82 ± 0.04	–
		Ayancık	311.0 ± 28.0	9.0 ± 2.0	0.30 ± 0.01	1.22 ± 0.04	15.00 ± 2.00	0.50 ± 0.01	0.08 ± 0.01	–
	C. panizzoi	SIH	402.0 ± 22.0	26.0 ± 5.0	3.00 ± 0.20	2.00 ± 0.10	29.00 ± 3.00	16.00 ± 1.00	1.90 ± 0.05	0.40 ± 0.01
		SOH	540.0 ± 32.0	29.0 ± 5.0	5.00 ± 0.40	3.00 ± 0.30	32.00 ± 8.00	28.00 ± 5.00	3.10 ± 0.02	1.00 ± 0.10
		Gerze	805.0 ± 55.0	33.0 ± 6.0	3.10 ± 0.40	2.10 ± 0.30	38.00 ± 7.00	14.00 ± 2.00	3.10 ± 0.30	1.10 ± 0.01
		Ayancık	465.0 ± 43.0	31.0 ± 5.0	4.20 ± 0.30	1.78 ± 0.10	7.00 ± 1.00	11.00 ± 2.00	1.10 ± 0.02	0.10 ± 0.01

SIH: Sinop inner harbour; SOH: Sinop outer harbour

Fe in Gerze station 2328±89 μg/g dry weight in *Chaetomorpha* spp.; 2143±78 μg g^{-1} dry wt. in *C. barbata* and 968±20 μg/g dry weight in *Ceramium* spp. The highest accumulation of different metals in the analysed algae species were: Fe and Co in *Chaetomorpha* spp, Zn in *C. barbata*, Ni and Cu in *U. linza*, Mn in *U. intestinalis*, Pb and Cd in *C. panizzoi*.

4. Discussion

Various factors such as shipping and fishing activities, industrial and urban effluents and usage of agricultural fertilizer effect heavy metal levels in biota[4]. Inner harbor and Gerze sites are relatively intensive areas with fishing activities. Moreover, there is a fish flour factory that leaves raw materials into the sea in Gerze. Thus, these stations have higher heavy metal concentrations compared to other stations.

Heavy metals may be toxic to marine biota through the food web, including human as being top predator[2,4,24,25]. The non-essential metals are extremely harmful even at low levels[24,26-29]. Pb and Cd levels are associated with generally highest human activities and contaminated areas with sewage[30]. Bat *et al*.[31] investigated the changes in macrofauna community structure along an organic enrichment gradient in outer harbor caused by sewer outfall in Sinop, focusing on the infra-littoral zoneand showed increases in the growth of opportunistic green algae including *U. lactuca* inresponse to increased nutrient supply in the coastal ecosystems of Sinop.

In the present study, *C. panizzoi* species shows the highest accumulation of Pb and Cd in Gerze. Actually, phylum Rhodophyta may reside in clean and contaminated regions[32] and the results of *C. panizzoi* and *Ceramium* spp. confirm it.

The essential metal, Fe needs for normal growth of marine plants[33], and several factors such as industrial activities and abilities of algal species to biomagnifying Fe from the surrounding environmentlead to emergence of high Fe level. Caliceti *et al*.[34]showed that Fe levels were always higher than those in Zn. In the present study results showed that metal levels in all studied algae species decrease in the order Fe > Zn. It is indicated that all algae species had the potential as biomonitors for essential metals such as Fe, Zn and Mn in Sinop coasts of the southern Black Sea.

Although in Turkey, macroalgae arenot used as a food source, seaweeds are one of the most important vegetable sources of calcium[35]. Macroalgae are the main primary producers in marine food web providing habitat, food and nursery grounds for many other species. The coastal waters have been affected by industrial and agricultural pollutants such as heavy metals that can accumulate within aquatic organisms at much higher levels than those in water column and sediment[4,36,37]. In order to reverse the deterioration of the European seas, European Union has adopted the Marine Strategy Framework Directive, aiming at improved status of the coastal waters. Monitoring programmes of the Marine Strategy Framework Directive includes studies of toxic substances of macroalgae. Therefore, it may be preliminary study to ensure “good environmental status” for implementation Marine Strategy Framework Directive in the Black Sea coastal waters.

In conclusion the presented data on heavy metals in macroalgaewill be used as a baseline for further investigations of contaminationof marine ecosystems along the southern Black Sea coast.

Conflict of interest statement

We declare that we have no conflict of interest.

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