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ORIGINAL ARTICLE

Heavy metal concentrations in marine green, brown, and red seaweeds from coastal waters of Yemen, the Gulf of Aden

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Abstract The purpose of this study was to investigate the concentration levels of heavy metals in different species of the main three marine algal divisions from the Gulf of Aden coastal waters, Yemen. The divisions included Chlorophyta—green plants (*Halimeda tuna*, *Rhizoclonium kochiamum*, *Caldophora koiei*, *Enteromorpha compressa*, and *Caulerpa racemosa* species), Phaeophyta—brown seaweeds (*Padina boryana*, *Turbinaria elatensis*, *Sargassum binderi*, *Cystoseira myrica*, and *Sargassum boveanum* species), and Rhodophyta—red seaweeds (*Hypnea cornuta*, *Champia parvula*, *Galaxaura marginate*, *Laurencia paniculata*, *Gracilaria foliifere*, and species). The heavy metals, which included cadmium (Cd), cobalt (Co), copper (Cu), chromium (Cr), Iron (Fe), manganese (Mn), nickel (Ni), lead (Pb), zinc (Zn), and vanadium (V) were measured by Atomic Absorption Spectrophotometer (AAs). The concentrations of heavy metals in all algal species are in the order of Fe >> Cu > Mn > Cr > Zn > Ni > Pb > Cd > V > Co. The results also showed that the uptake of heavy metals by different marine algal divisions was in the order of Chlorophyta > Phaeophyta > Rhodophyta. These heavy metals were several order of magnitude higher than the concentrations of the same metals in seawater. This indicates that marine alga progressively uptake heavy metals from seawater.

Keywords Heavy metals · Chlorophyta · Phaeophyta · Rhodophyta · Gulf of Aden · Yemen

Introduction

The Gulf of Aden extends from the strait of Bab el-Mandab, the southern entrance to the Red Sea, to the northern part of the Arabian Sea (PERSEGA 1981; Crystal 1990). It separates the Arabian Peninsula from the Somali Peninsula with surface area of $\sim 220 \times 10^3$ km^3. The Yemeni shore, which is located along the northern edge of the Gulf, varies in shape according to geological feature of the region. The major features are rocky headlands with series of cuspate beaches between them. The major wadi systems, which are mostly dry river beds in the desert, also influence the coastal shape and environment due to fluvial deposit inputs. There are no major rivers flow into the coastal waters. However, there is seasonal, outflow from the Bana and Hussan wadis (50–70 km east of Aden) that reach the sea from time to time (approximately twice a year) after heavy rain over the mountains (flash flood).

The wind regime and water mass exchange with the Red Sea affect the biogeochemistry of the Gulf of Aden (Morcos 1970; Morcos and Varely 1990; Rushdi et al. 1994). The two seasonal monsoons that occur twice a year affect the hydrographic and ecological structure of the gulf (FAO 1967; Burkill et al. 1993; Cushing 1971, 1973; Pfannkuche and Lochte 2000; van Weering et al. 1997). The southwest monsoon starts late summer (July–September) and generates upwelling system in the Arabian Sea that brings cold nutrient rich water from the deep ocean to the surface (Guraishee 1984; Murty and El-Sabh 1984). This nutrient rich water mass increases the biological productivity in surface water, where chlorophyll *a* level can

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reach 1 mg C m^{-3} h^{-1}, the same level in the Peruvian upwelling region (FAO 1967; Smith 1984). The most obvious coastal ecological impact of the upwelling is the vast growths of sub-tidal and intertidal algal assemblages on the exposed platforms and rocks. The main coastal key habitats include coral reefs, seagrass beds, salt marches, wetlands, mangroves, and nursery areas for fish and shrimps (IUCN 1987b; Rushdi et al. 1994).

There are different types of impacts on the coastal and marine environment of Yemen (Al-Shwafi 2003; Al-Shiwafi et al. 2005; Bawazir and Abu Al-Fatooh 2001; DouAbul and Al-Shiwafi 1998; DouAbul et al. 1997; EPC 1996; Haskoning 1991; IUCN 1987a, b; NAP 2003; Rushdi et al. 1991, 1994). These impacts are mainly caused by human and developmental activities, which introduce pollutants to the marine environment and cause the detraction of some special habitats (Al-Shiwafi et al. 2005; Rushdi et al. 1994). The most widely recognized issue is the oil-related pollution, where considerable attention has been focused (DouAbul and Al-Shiwafi 1998; DouAbul et al. 1997). However, other areas of concern include the impact of growing industrial and domestic effluents, unplanned coastal development as well as various miscellaneous anthropogenic activities such as fishing, hunting, and tourism (EPC 1996; IUCN 1987b; Rushdi et al. 1991, 1994). In fact, the waters off Yemen are now threatened by several kinds of pollution from passing ships and local shore facilities (UNEP 1997).

High levels of heavy metals (e.g., cadmium, cobalt, mercury, copper lead, vanadium, and zinc) in aquatic ecosystems are regarded as serious pollutants, because they can be toxic and incorporated into the food chain (Kishe and Machiwa 2003). EPC (1996), studied heavy metal concentrations in sediments and mollusks and recommended continuous monitoring programs for the Gulf of Aden and Arabian Sea coastal zones to insure that the concentrations of heavy metals remain within the baseline levels recommended by the survey. Thus, the aim of the present study is to investigate the levels of heavy metals (cadmium, cobalt, chromium, copper, iron, manganese, nickel, lead, vanadium, and zinc) in different marine algal groups.

Methodology

Sample collection

Fifteen species of three marine algal plants were collected from the Yemeni coastal waters of the Gulf of Aden during the winter of 2005. They were obtained from about 20 miles west of the Aden Harbor (Fig. 1) at depths ranging from half to 1 m. The algal groups included Chlorophyta—green plants (*Halimeda tuna*, *Rhizoclonium kochiamum*, *Caldophora koiei*, *Enteromorpha compressa*, and *Caulerpa racemosa* species), Phaeophyta—brown seaweeds (*Padina boryana*, *Turbinaria elatensis*, *Sargassum binderi*, *Cystoseira myrica*, and *Sargassum boveanum* species), and Rhodophyta—red seaweeds (*Hypnea cornuta*, *Champia parvula*, *Galaxaura marginate*, *Laurencia paniculata*, and *Gracilaria foliifere* species). The samples were taken from the algal flesh for the determination of heavy metal concentrations. The samples were washed with sea water at the sampling site and transferred to the laboratory in polyethylene boxes under refrigeration (4°C).

Chemical analysis

After the arrival of the samples to the laboratory, they were rinsed and washed carefully with di-ionized water. Then, they were dried at 60°C (to constant weights), homogenized by grinding each sample in a porcelain pestle and mortar and kept away from metallic materials and dusty conditions to avoid contamination.

Sub-samples ($\sim$1.5 g of each sample) were submitted to acid digestions in a microwave oven by using concentrated 5 ml HNO_3 in Teflon-lined vessel. Each digested sample was diluted to 25 ml with double distilled water and analyzed for metals contents (Ho 1987). The analysis of cadmium (Cd), cobalt (Co), copper (Cu), chromium (Cr), Iron (Fe), manganese (Mn), nickel (Ni), lead (Pb), zinc (Zn), and vanadium (V) were performed with a flame atomic absorption spectrophotometer (Perkin-Elmer, Foster City, CA, USA, Model 2380). Procedural blanks were performed between each run. All glasswares, plastic devices and Teflon devices were thoroughly acid washed (Moody and Lindstrom 1977). Reagents of analytical grade were used for blanks and calibration curves.

Results

The analysis results of the three algal group are summarized in Table 1 and are illustrated in Fig. 2. The reported results are mean values of triplicate determinations and are expressed as μg g^{-1} dry weight (DW). The results showed that Chlorophyta division have the highest concentrations of heavy metals followed by Phaeophyta division and then by Rhodophyta division.

Chlorophyta

The concentrations of various heavy metals in different Chlorophyta species vary (Table 1, Fig. 2a). The concentration of Cd ranged from 0.38 to 2.3 μg g^{-1} DW with

Table 1 Concentrations (μg g^{-1} DW) of heavy metals in marine green plant (Chlorophyta), brown seaweed (Phaeophyta), and red seaweed (Rhodophyta) algal species

	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	V	Zn
Chlorophyta										
Halimeda tuna	0.38	0.3	3.4	8.31	60.2	3.5	4.7	3.2	0.3	2.53
Rhizoclonium kochiamum	0.75	0.75	7.3	9.93	15.7	4.3	1.2	0.8	0.7	3.4
Caldophora koiei	2.3	0.9	10	15.4	40.2	5.4	3.9	0.73	1.2	2.35
Enteromorpha compressa	1.9	1	15.3	17.53	35.7	12.9	13.3	6	0.6	8.1
Caulerpa racemosa	0.93	0.8	13.2	20.5	90.1	13.7	5.3	2.5	0.3	7.9
Range	0.38–2.30	0.30–1.00	3.40–15.30	8.31–20.50	15.70–90.10	3.50–13.70	1.20–13.30	0.73–6.00	0.30–1.20	2.35–8.10
Phaeophyta										
Padina boryana	0.65	0.73	2.2	5.14	50.3	2.2	3.3	0.9	0.6	3.5
Turbinaria elatensis	0.35	0.35	4.4	6.3	13.81	7.3	0.9	0.7	0.4	3.3
Sargassum binderi	0.76	0.6	3.7	10.2	30.6	3.55	2.8	2.4	0.9	2.8
Cystoseira myrica	0.53	0.53	9.8	14.1	15.1	7.9	4.73	5.3	1	9.2
Sargassum boveamum	0.45	0.95	11	17.2	60.2	5.4	9.57	3.2	0.6	7.4
Range	0.35–0.76	0.35–0.95	2.20–11.00	5.14–17.20	13.81–60.2	2.20–7.90	0.90–9.57	0.70–5.30	0.40–1.00	2.80–9.20
Rhodophyta										
Hypnea cornuta	0.93	0.14	1.13	2.31	30.73	6.73	2.35	2.8	0.5	4.53
Champia parvula	0.26	0.35	0.98	4.5	13.55	2.55	1.36	1.75	0.64	3.66
Galaxaura marginate	0.46	0.3	2.35	3.66	10.75	7.37	0.8	0.35	0.44	5.3
Laurencia paniculata	0.17	0.63	1.11	17.01	25.3	5.56	0.73	0.91	0.87	2.71
Gracilaria foliifere	0.55	0.44	0.7	9.83	41.2	8	7.3	0.93	0.73	6.35
Range	0.17–0.93	0.14–0.63	0.70–2.35	2.31–17.01	10.75–41.2	2.55–8.00	0.73–7.30	0.35–2.80	0.44–0.87	2.71–6.35
Seawater[a]	0.11–1.12[c]	0.24–17.68[c]	0.16–0.26	0.03–0.08	0.00–0.17	0.03–0.16		1.49–15.96	1.17–1.63	0.007–0.013
Sediment[b]	0.9–3.3	18–42	120–390	10–120	40–100	0.5–2.6	18–30	50–800		50–350
Mulluscs[b]	0.5–3.3	0.2–1.1	3.2–6.2	7.5–10.5	0.2–2.2	150–180	1.0–5.6	2.5–5.9		60–320

[a] Donat and Dryden (2001)

[b] Szefer et al. (1999)

[c] Concentration = ng l^{-1}

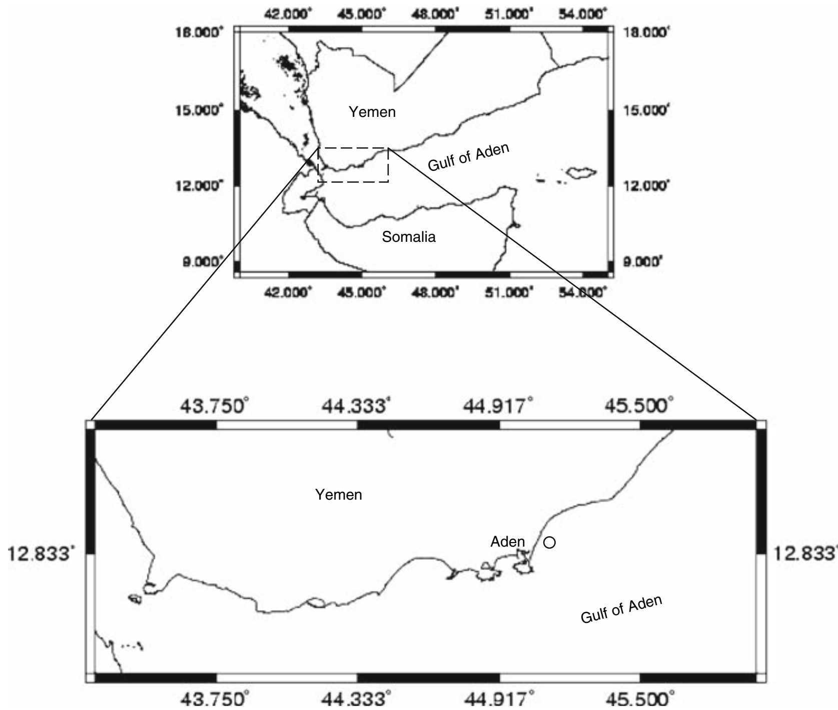

Fig. 1 Map showing **a** the location of study area and **b** the site of sample collection indicated by the symbol ○

minimum and maximum concentrations in the *H. tuna* and *C. koiei* species, respectively. The Co concentration, which ranged from 0.30 to 1.00 μg g^{-1} DW, was minimum in the *H. tuna* species and maximum in *E. compressa* species. The concentration of Cr ranged from 3.40 to 15.30 μg g^{-1} DW with a minimum concentration in *H. tuna* species and a maximum concentration in *E. compressa* species. For Cu, the concentration ranged from 8.31 to 20.50 μg g^{-1} DW and the minimum concentration was found in *H. tuna* species and the maximum concentrations were determined in the *C. racemosa* and *E. compressa* species. The concentration of Fe ranged from 15.70 to 90.10 μg g^{-1} DW with minimum and maximum concentrations in the *R. kochiamum* and *C. recemosa* species, respectively. The maximum concentrations of Mn, which ranged from 3.50 to 13.70 μg g^{-1} DW, were found in the *E. compressa* and *C. racemes* species, whereas the minimum concentration was observed in the *H. tuna* species. For Ni, the concentration ranged from 1.20 to 13.30 μg g^{-1} DW and the minimum concentration was detected in the *R. kochiamum* species and the maximum concentration was in the *E. compressa* species. The concentration of Pb ranged from 0.73 to 6.00 μg g^{-1} DW and the minimum concentrations were in the *R. Kochiamum* and *C. koiei* species whereas the maximum concentration was found in *E. compressa* species. The concentration V, which ranged from 0.30 to 1.20 μg g^{-1} DW, showed minimum concentrations in the *H. tuna* and *C. racemosa* species and maximum concentration in *C. koiei* species. The Zn concentrations ranged from 2.35 to 8.10 μg g^{-1} DW and the minimum concentrations were found in the *H. tuna*, *R. kochiamum* and *C. koiei* species and the maximum concentrations were observed in the *E. compressa* and *C. racemosa* species.

Phaeophyta

The concentrations of various heavy metals in the samples of different Phaophyta species are shown in Table 1 and Fig. 2b. The concentration of Cd was found to range from 0.35 to 0.76 μg g^{-1} DW and its minimum concentration was determined in the *T. elatensis* species and the maximum concentration in the *S. binderi* species. The concentration of Co ranged from 0.35 to 0.95 μg g^{-1} DW with a minimum concentration in the *T. elatensis* species and a maximum concentration in the *S. boveamum* species. The minimum concentration of Cr, which ranged from 2.20 to 11.00 μg g^{-1} DW was in the *P. boryana* species and the maximum concentrations were in the *S. boveamum* and *C. myrica* species. For Cu, the concentration ranged from 5.14 to 17.20 μg g^{-1} DW. The minimum concentration of Cu was observed in the *P. boryana* species and the maximum concentration was in the of *S. boveamum* species. The concentration of Fe ranged from 13.81 to 60.2 μg g^{-1} DW with a minimum concentration in the *T. elatensis* species and a maximum concentration in the *S. boveamum* species. The Mn concentration ranged from 2.20 to 7.90 μg g^{-1} DW and its minimum concentration was determined in the *P. boryana* species whereas the maximum concentrations were measured in the *C. myrica* and *T. elatensis* species.

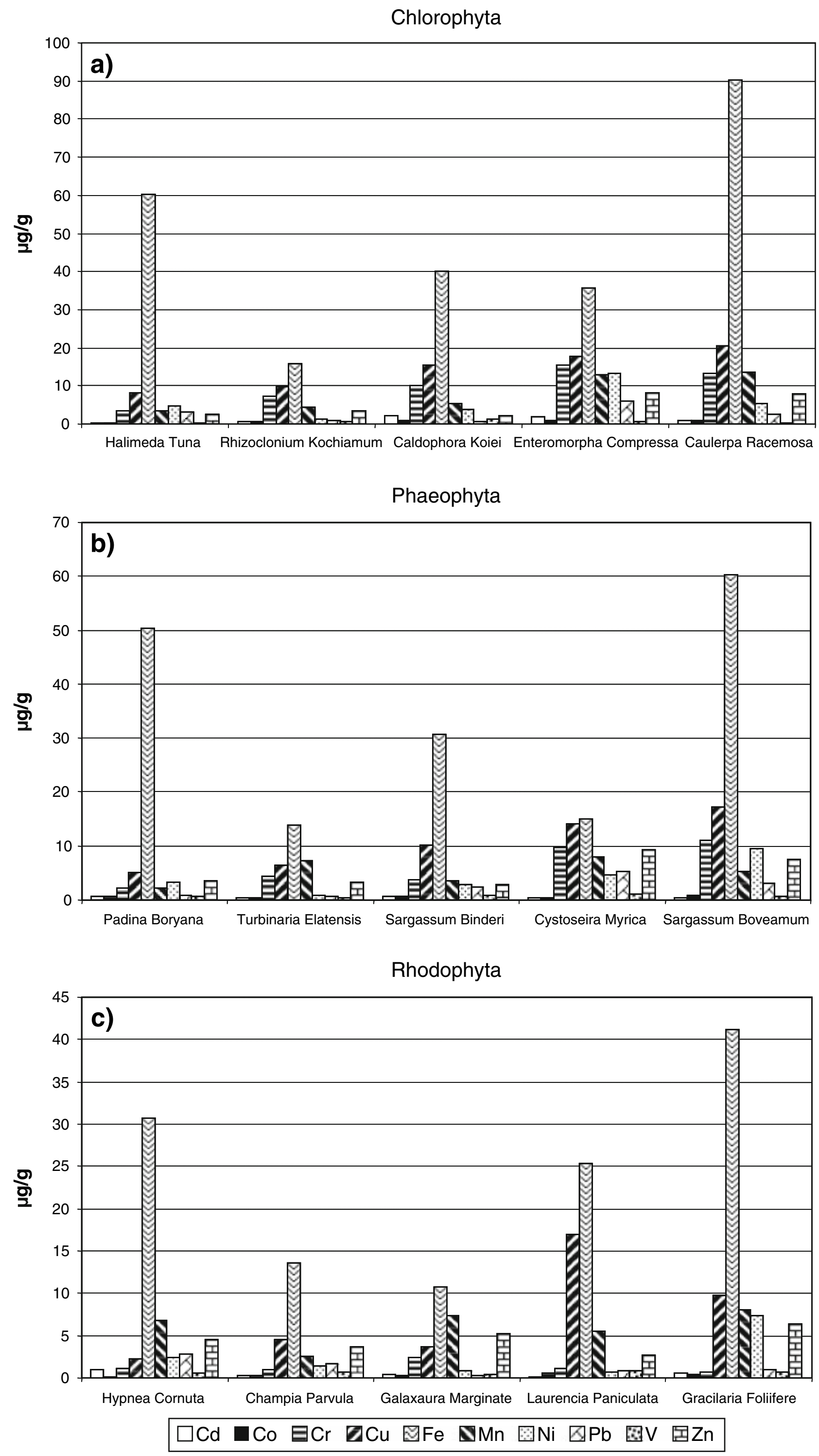

Fig. 2 The concentrations of different heavy metals measured in tissues of **a** Chlorophyta (green plants), **b** Phaeophyta (brown seaweeds) and **c** Rhodophyta (red seaweeds)

The concentration of Ni ranged from 0.90 to 9.54 µg g^{-1} DW with a minimum concentration in the *T. elatensis* species and a maximum concentration in the *S. boveamum* species. The Pb concentration ranged from 0.70 to 5.30 µg g^{-1} DW. The minimum concentrations of Pb were determined in *T. elatensis* and *P. boryana* species and the maximum concentration was found in the sample of the *C. myrica* species. The concentration of V ranged from 0.40 to 1.00 µg g^{-1} DW. The minimum concentration of V was found in the *T. elatensis* species, whereas the maximum concentrations were determined in the *C. myrica* and *Sargaussum binderi* species. The minimum concentration of Zn, which ranged from 2.80 to 9.20 µg g^{-1} DW, was found in the *S. Binderi* species and the maximum concentration was in the *Cystosoeira myrica* species.

Rhodophyta

The concentrations of various heavy metals in different species of Rhodophyta vary as shown in Table 1 and Fig. 2c. The Cd concentration, which ranged from 0.17 to 93 µg g^{-1} DW, was minimum in the *L. paniculata* species and maximum in the *H. cornuta* species. The minimum concentration of Co, which ranged from 0.14 to 0.63 µg g^{-1} DW, was found in the *H. cornuta* species and the maximum concentration was in *L. paniculata* species. The concentration of Cr ranged from 0.70 to 2.35 µg g^{-1} DW with a minimum concentration in the *G. foliifera* species, and a maximum in the *G. marginate* species. For Cu, the concentration ranged from 2.31 to 17.01 µg g^{-1} DW and its minimum concentration was determined in the *H. cornuta* species and the maximum concentration was measured in the *L. paniculata* species. The concentration of Fe ranged from 10.75 to 41.2 µg g^{-1} DW. The minimum concentration of Fe was measured in the *G. marginate* species and the maximum concentration was in the *G. foliifera* species. The concentration of Mn, which ranged from 2.55 to 8.00 µg g^{-1} DW, showed a minimum concentration in the *C. parvula* species and a maximum concentration in the *G. foliifera* species. The Ni concentration ranged from 0.73 to 7.30 µg g^{-1} DW and its' minimum concentration was found in the *L. paniculata* species and its' maximum concentration was observed in the *G. foliifera* species. For Pb, the concentration ranged from 0.35 to 2.80 µg g^{-1} DW with a minimum concentration in the *G. marginate* species and a maximum in the *H. cornuta* species. The minimum concentration of V, which ranged from 0.44 to 0.87 µg g^{-1} DW, was determined in the *G. marginate* species and the maximum concentration was in the *L. paniculata* species. In the case of Zn, the concentration ranged from 2.71 to 6.35 µg g^{-1} DW. The maximum concentration of Zn was found in the *L. paniculata* species and the maximum concentration was in *G. foliifera* species.

Discussion

The concentrations of heavy metals in different algal plants vary between different species. Chlorophyta species were found to contain more heavy metals than phaeophyta and Rhodophyta species (Table 1, Figs. 2, 3). The uptake of heavy metals by different marine algal divisions is in the order of Chlorophyta > Phaeophyta > Rhodophyta (Fig. 3). Apparently, the uptake of Fe by these algal divisions is high compared to other heavy metals. The content of heavy metals in various algal species is in the order of Fe >> Cu > Mn > Cr > Zn > Ni > Pb > Cd > V > Co as shown in Fig. 3. This uptake order has been observed also in phytoplankton where the concentrations of heavy metals were found in the order of Fe > Zn > Mn > Cu > Cd > Ag > Pb > (Marin and Knuaer 1973).

It is obvious that the concentrations of heavy metals in marine algal species are several orders of magnitude higher than the concentrations of the same metals in seawater (Table 1; Donat and Dryden 2001; Gerlach 1976; Pytkowciz 1983; Turner et al. 1980; Whitfield 1979). They are lower in concentrations levels relative to their levels in sediments and similar in concentration levels determined in mollusks (Szefer et al. 1999), with the exception for Fe and Cu, which are higher in algal species. This indicates that these algal species are not contaminated and the sources of metals are natural. The presence of high concentrations of Fe and Cu in marine plants can be attributed to the fact that they are important micronutrients for various metabolic functions of the plants (Donat and Dryden 2001). The fact that metal concentration in these aquatic plants are typically several orders of magnitude higher than concentrations of the same metals in the water has led to support some speculation that metal may become progressively concentrated at higher trophic levels in aquatic food chains due to food chain magnification (Clark 1998; Laws 2000). The high level of Fe in all algal groups is consistent with the food chain magnification hypothesis (Laws 2000). The wide range of metal concentrations in different algal species reflects the importance of biochemical factors in affecting the relative tendency of different tissues to concentrate pollutants. Such biochemical or physiological differences may also play a major role in causing certain species to concentrate pollutants to a much higher levels than other organisms, regardless of the relative position of

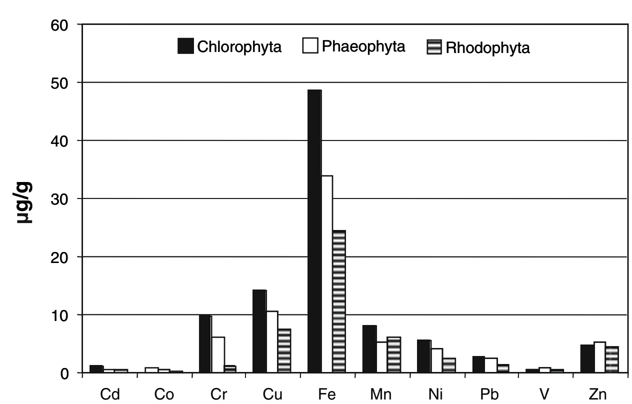

Fig. 3 The concentration of heavy metals in the three divisions of macroalgae

the species in the aquatic food chain (Millward and Turner 2001).

Additionally, the marine plants including algal plants are important in marine biogeochemical cycles not only because they are able to concentrate large quantities of elements (relative to seawater), but also because they can transport them in a variety of ways. These include: detritus sink and decomposition of organic matter in the sediments; direct release through plant tissue-water exchange, and part of aquatic food chain (Millward and Turner 2001).

The concentrations of heavy metals in various Chlorophyta species were found to be relatively high compared those in other two algal plants (i.e., Phaeophyta and Rhodophyta; Fig. 3). This indicates that Chlorophyta group has the tendency to uptake more metals than the other two groups. The uptake of heavy metals is high by *C. racemosa* followed by *H. tuna*, *C. koiei*, *E. compressa*, and *R. kochiamum*. However, they all show high levels of Fe and Cu, they are selective with respect to other heavy metals. For instance, for *H. tuna*, the concentrations are in the order of Fe >> Cu > Ni > Mn > Cr > Pb > Zn > Cd > Co > V and for *E. compress* they are in order of Fe >> Cu > Cr > Ni > Mn > Zn > Pb > Cd > Co > V.

In marine brown seaweeds (Phaeophyta), the concentration levels of heavy metals are less than in green plants (Chlorophyta). The concentrations are in the order of Fe >> Cu > Cr > Mn ≥ Zn > Ni > Pb > V ≥ Co > Cd. Except for Fe and Cu, different species of Phaeophyta division are selectively accumulate heavy metals (Table 1). The concentration levels of heavy metals in marine red seaweeds (Rhodophyta) are less than the concentrations in green plants (Chlorophyta), and brown seaweeds (Phaeophyta). The heavy metal concentrations are in the order of Fe >> Cu > Mn > Zn > Pb > Cr > V > V > Cd > Co. Also, different species of Rhodophyta division show selectivity toward the accumulation of heavy metals, except for Fe and Cu (Table 1).

Conclusion

It should be apparent that the concentration of metals associated with particulate materials including living species, is at least order of magnitude higher than the concentrations of metals dissolved in the water. It is also true in general that the concentrations of metals may differ greatly between one species to another and between different species in the same group.

In general, one ascribes these concentrations differences in the tendency of metals to bind to the various molecular groups found within the cells of each organism, as well as to the degree of the organisms exposure to the metal as influenced by its metabolic characteristics and its position in the food chain.

Apparently, marine algae may play a significant role in biogeochemical cycles of heavy metals in the coastal zones of Yemen.

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