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Brown algae (*Phaeophyta*) for monitoring heavy metals at the Sudanese Red Sea coast

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Abstract This study aimed at monitoring some heavy metals at the Sudanese Red Sea coast using Brown algae (*Phaeophyta*) as biomonitor. The total contents of heavy metals in four species (*Turbinaria* sp., *Sargassum* sp., *Cystoseira* sp. and *Padina* sp.) as well as seawater were examined. Twenty-six algae samples were collected from seven locations. The ranges of concentrations (µg/g, dry wt.) of heavy metals in algae were 4.95–16.95 for Cr, 2.93–257.32 for Mn, 1.35–7.43 for Ni, 0.83–14.10 for Cu, 4.13–19.13 for Zn, 0.03–0.15 for Cd and 0.45–2.18 for Pb. The ranges of the pH and the salinity of seawater from the same locations were 8.11–8.82 and 38.00–41.00 PSU, respectively. The ranges of concentrations (µg/L) of heavy metals in seawater were 7.00–11.00 for Cr, 2.90–10.20 for Mn, 6.70–10.10 for Ni, 1.70–5.00 for Cu, 0.94–5.70 for Zn, 0.09–0.14 for Cd and 0.93–1.80 for Pb. No significant correlations between metal concentrations in algae and seawater were observed. Some locations in the study area recorded relatively high levels of heavy metals in algae indicating possible contribution from manmade activities. Cr recorded higher levels in the study area than those in other coastal areas in the word. *Padina* sp. and *Cystoseira* sp. were better bioindicator than *Turbinaria* sp., *Sargassum* sp. for their high metal uptake.

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Introduction

Monitoring of chemicals in the environment from both origins natural and anthropogenic requires efficient indicators. In marine environment, aquatic organisms are more efficient indicators than water and sediment because aquatic organisms demonstrate the bioavailable fraction of contaminants and hence reflect the direct risk (Phillips 1990; Chakraborty et al. 2014). Among other aquatic organisms, macroalgae have proven to be efficient in monitoring and sorption of metals because they act as sink for metals (Montazer-Rahmati et al. 2011; Lee and Park 2012). In addition, macroalgae can live in both systems clean and contaminated because they have the ability of adaptation to different environmental conditions (Rajfur et al. 2010). While green algae (*Chlorophyta*) are usually found in freshwater and terrestrial areas, red algae (Rhodophyta) and brown algae (*Phaeophyta*) are almost found in seawater (Hashim and Chu 2004).

Phaeophyta is a grand assemblage of macroscopic plants. More than 1500 *Phaeophyta* species have been identified (Davis et al. 2003). The brown color in *Phaeophyta* results from the large amounts of the carotenoid fucoxanthin, which masks other pigments (Hashim and Chu 2004). *Phaeophyta* has proven to be an effective bioindicator for metals due to the high density of carboxylic groups that present in alginate (the main component of their cell-wall). This feature results in high rate of metal accumulation (Phillips 2009; Hashim and Chu 2004; Försberg et al. 1988; Cazón et al. 2013). Electrostatic

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attraction and complexation take place in the cell-wall of *Phaeophyta* (Davis et al. 2003).

On the other hand, it is known that the coastal areas of the Red Sea are located in the arid zones. In addition, the Red Sea coastal areas suffer from the scarce of water sources that are fit for drinking and other purposes. Hence, the water of the Red Sea is the major source, after desalination treatment, for drinking, irrigation and industry in those areas. Accordingly, the monitoring of possible pollutants in the Red Sea water is of great of interest (Idris et al. 2007; Idris 2008).

The aims of this work were: (1) to determine the levels of Cr, Mn, Ni, Cu, Zn, Cd and Pb in *Phaeophyta* and seawater samples collected from the Sudanese Red Sea coast, (2) to study the *Phaeophyta* species variability in metal uptake and (3) to assess the levels of metals in different sites in the study area and to compare those levels against other coastal areas in the world.

Materials and methods

Study area

This study was conducted at the Sudanese Red Sea coast (Fig. 1). The Red Sea is land-locked; a feature that restricts water exchange and hence enhance the enrichment of contaminants including heavy metals. Port-Sudan and Sawakin, which lie along the Sudanese Red Sea coast, are the main harbors of Sudan and South Sudan countries. Port-Sudan and Sawakin harbors witness dense shipping and recreational boating activities. Port-Sudan is a large city, with the area of 218,887 km^2, and densely populated, with more than 520,000 residents. Port-Sudan also includes a wide industrial area with various processes. Swakin is a historical city in Sudan. It is more than 1000 years old.

Sampling

Twenty-six brown algae samples were collected during October and November, 2014. The samples were collected from the fringing reefs area with transect at c.50 m and depth in the range of 50–150 cm. Four species including *Turbinaria* sp., *Sargassum* sp., *Cystoseira* sp. and *Padina* sp. were identified. Samples were stored in polyethylene bottles until transferring to laboratory. Samples were then flushed by seawater that was collected from the relevant locations. The soft parts in algae samples were separated and oven-dried at 105 °C. Dried samples were crushed to fine powder for treatment.

Seawater samples were collected from the same locations of the algae sampled. Samples were collected from about 20 cm below the surface water to avoid floating matters. Samples were collected directly in polyethylene bottles. The bottles were previously cleaned with 10% (v/v) nitric acid, double distilled deionized water and seawater. Seawater samples were then preserved for analysis by adding 0.5 mL of high purity nitric acid to each liter of a

Fig. 1 Map showing sampling stations along the Sudanese Red Sea coast
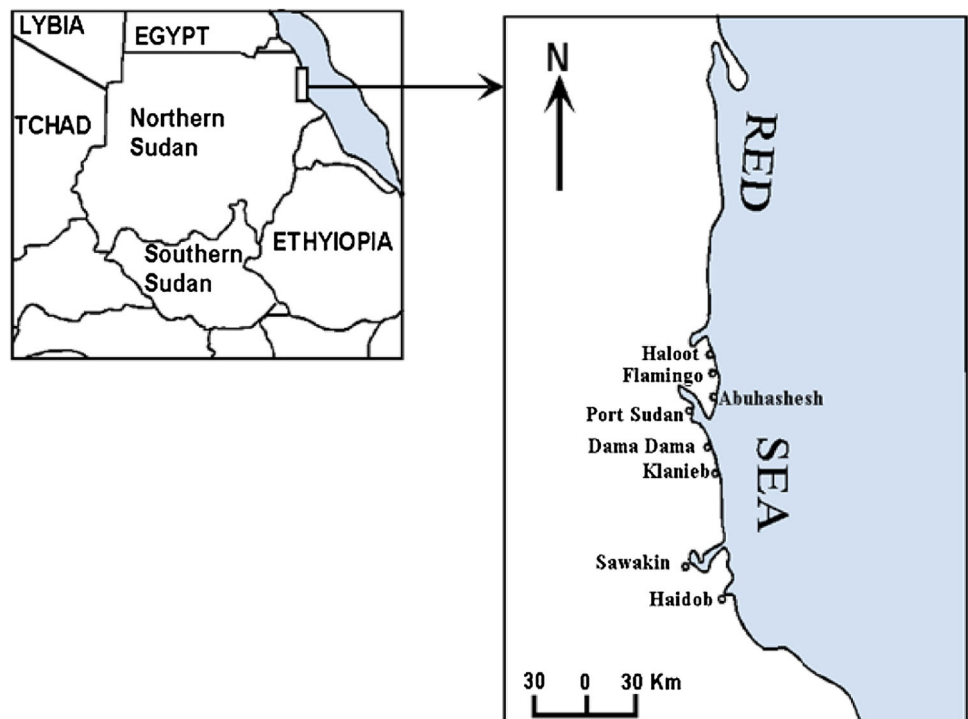
sample and stored at 25 °C. This acidification is believed to prevent the loss of metals by adsorption onto the walls of the polyethylene bottles.

Treatment of algae samples

Each alga sample was analyzed in triplicate. Wet digestion was applied for sample treatment. Seven mL of nitric acid were added to 1.0 g of dried homogenous powder sample in 100 mL beaker. The mixture was heated on a hotplate at 70 °C until dissolving and ceasing the evaporation of NO_2. The solution was allowed to be at room temperature. Thereafter, 2 mL of perchloric acid were added and the mixture was heated again at 70 °C on a hotplate until evaporating the excess acid and the mixture became as paste. The mixture was diluted by double distilled deionized water in 50 mL volumetric flask. Sample solutions were then filtered to be ready for AAS measurement. Blank samples were prepared in triplicate and treated in parallel with algae samples using the same treatment procedure. A certified reference material (CRM) was also analyzed in parallel with algae samples. The CRM was IAEA-V-10, which has been certified by the International Atomic Energy Agency, Vienna, Austria.

Measurements

The pH of seawater was measured on-site. The concentrations of heavy metals were measured in algae and seawater by atomic absorption spectrometer (AAS). The system was SpectrAA 220, which was supplied from Varian (Palo Alto, CA. USA). Air/acetylene flame and deuterium background corrector were used. The system was equipped with sample introduction device. Hollow cathode lamps were used for metal excitations, which were performed at the following wavelengths and currents: Cr (357.9 nm and 12 mA), Mn (279.5 nm and 15 mA), Ni (232.0 nm and 40 mA), Cu (324.7 nm and 25 mA), Zn (213.9 nm and 15 mA), Cd (228.8 nm and 20 mA) and Pb (217 nm and 12 mA). The instrument was calibrated using external mixed standard solutions, which were ready prepared by Sigma-Aldrich (Darmstadt, Germany). The AAS was calibrated for each metal using seven levels of concentrations.

Results and discussion

Table 1 shows the results of the quality control of the quantification of heavy metals. For CRM analysis, all measured values were within the confidence intervals. Acceptable recovery, with values in the range of 85–110%, were obtained for all metals. For algae and seawater analysis, the average values of the concentrations of heavy metals, which were obtained from triplicate measurements, were considered. The values of the relative standard deviation (RSD) of the triplicate measurements were less than 7% indicating acceptable repeatability.

The summary statistics of the pH and the concentrations of salinity, Cr, Mn, Ni, Cu, Zn, Cd and Pb in seawater are compiled in Table 2. The summary statistics of metal concentrations in the algae samples are compiled in Table 3. Metal uptake by algae is influenced by some conditions such as the pH and the salinity of seawater, in addition to metal concentration in seawater (Morrison et al. 2008). The values of pH and salinity of seawater showed narrow ranges and small coefficients of variation (<3%). The concentrations of heavy metals in seawater were in the following descending order: Cr > Ni > Mn > Cu > Zn > Pb > Cd. High coefficients of variation (>50%) were recorded in the concentrations of Mn and Zn in seawater. However, the concentration ranges of Mn (2.90–10.20 μg/L) and Zn (0.94–5.70 μg/L) were narrow. The levels of pH and salinity of seawater from the Sudanese Red Sea coast were comparable with those at two sites in the Irish coast (Morrison et al. 2008). Notably, the previous study considered one site was contaminated by heavy metals while the other site was considered free from contamination.

The average values of the concentrations of heavy metals in algae decreased in the following order: Mn > > Zn ≈ Cr > Cu ≈ Ni > Pb > Cd. This order has found to be similar with that recorded in brown algae collected from the Amursky Bay, Russia (Khristoforova and Kozhenkova 2002) and the Black Sea, Bulgaria (Jordanova et al. 1999; Strezov and Nonova 2003). Similar order of algae uptake reflects the correspondence affinity of all brown algae species toward the levels of metal accumulation. It has been reported that significant amounts of divalent metals were accumulated by brown algae, which was attributed to the high levels of binding polysaccharides and polyphenols (Chakraborty et al. 2014; Hashim and Chu 2004).

In the current study, the average concentration of Mn in brown algae is almost ten-fold higher than Zn and Cr concentrations and 20-fold higher than Cu and Ni concentrations. Because of their natural origin, Mn and Fe presented the highest amounts in sediment from the Sudanese Red Sea coat. Pb and Cd recorded the minimum concentrations among other metals (Idris et al. 2007; Idris 2008). The concentrations of Pb and Cd in marine macroalgae have a special concern because of their man-made origin, non-point source and high toxicity (Strezov and Nonova 2003; Duan et al. 2014).

Figures 2, 3, 4, 5, 6, 7 and 8 show the concentrations of metals in each brown alga species from each location along

Table 1 Results of quality control of heavy metals measurements by AAS

Metal	LOD[a]	LOQ[b]	Results of CRM[c]			
			MV[c]	RV[d]	CI[e]	Recovery (%)
Cr	0.0059	0.0190	7.01	6.5	5.6–7.1	108
Mn	0.0031	0.0103	48.50	47[f]	44–51[f]	103
Ni	0.0066	0.0220	4.50	4.2	3.8–4.9	107
Cu	0.0131	0.0438	7.98	9.4	8.8–9.7	85
Zn	0.0063	0.0211	22.80	24.0	23–25	95
Cd	0.0057	0.0191	0.032	0.03	0.02–0.05	107
Pb	0.0081	0.0269	1.76	1.6	0.8–1.9	110

[a] Limit of detection (µg/mL)

[b] Limit of quantification (µg/mL)

[c] Measured value (µg/g)

[d] Recommended value (µg/g)

[e] Confidence interval (µg/g) at 95%

[f] Information values and not recommended values

Table 2 Summary statistics of pH, salinity (PSU) and metal concentration (µg/L) in seawater from sampled locations at the Sudanese Red Sea coast

Parameter	Min	Max	Average	Median	SD[a]	CV%[b]
pH	8.11	8.82	8.64	8.72	0.23	2.61
Salinity	38.00	41.00	39.50	39.00	1.07	2.71
Cr	7.00	11.00	9.63	10.00	1.41	14.63
Mn	2.90	10.20	5.28	4.35	2.81	53.28
Ni	6.70	10.10	8.65	9.05	1.16	13.44
Cu	1.70	5.00	3.33	3.30	0.99	29.85
Zn	0.94	5.70	2.62	2.40	1.70	64.98
Cd	0.09	0.14	0.12	0.12	0.02	12.60
Pb	0.93	1.80	1.49	1.65	0.34	22.85

[a] Standard deviation

[b] Coefficient of variation

Table 3 Summary statistics ($n = 26$) of the concentrations (µg/g, dry wt.) of some heavy metals in brown algae along the Sudanese Red sea coast

Metal	Min	Max	Average	Median	SD[a]	CV%[b]
Cr	4.95	16.95	9.55	9.00	2.39	25.03
Mn	2.93	257.32	84.8	66.87	69.62	82.10
Ni	1.35	7.43	4.82	4.58	1.47	30.50
Cu	0.83	14.10	4.83	4.13	2.97	61.49
Zn	4.13	19.13	10.97	8.89	4.75	43.30
Cd	0.03	0.15	0.07	0.08	0.03	42.86
Pb	0.45	2.18	1.28	1.32	0.44	34.38

[a] Standard deviation
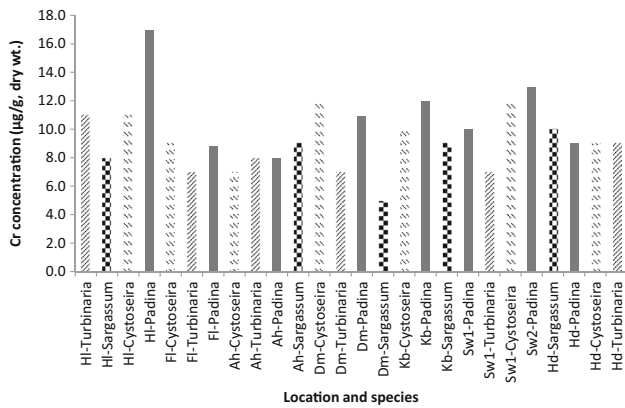
[b] Coefficient of variation

Fig. 2 Cr concentration in brown algae species collected from locations at the Sudanese Red Sea coast; the full names of locations are *Hl* Haloot, *Fl* Flamingo, *Ah* Abuhashish, *Dm* Dama–Dama, *Kb* Klaineb, *Sw* Sawakin and *Hd* Haidoob
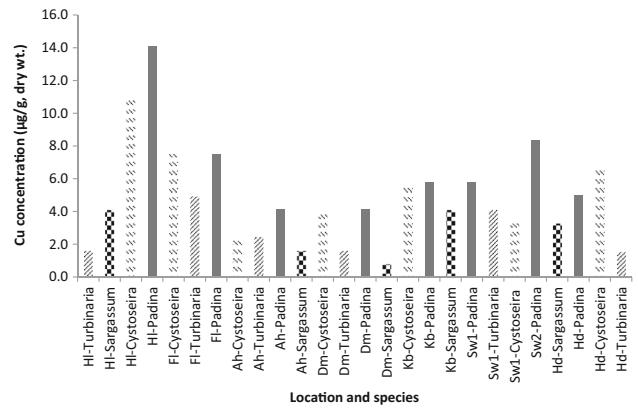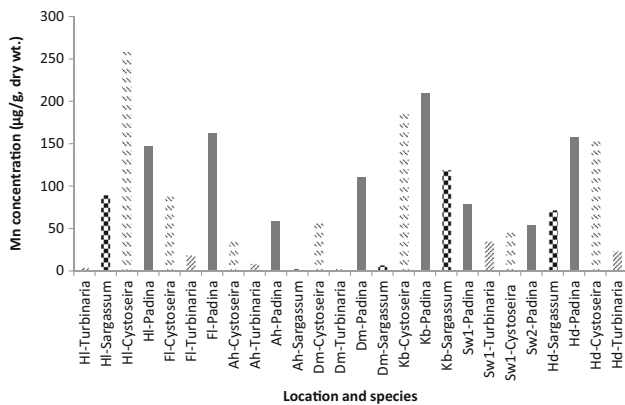

Fig. 5 Cu concentration in brown algae species collected from locations at the Sudanese Red Sea coast; the full names of locations are *Hl* Haloot, *Fl* Flamingo, *Ah* Abuhashish, *Dm* Dama–Dama, *Kb* Klaineb, *Sw* Sawakin and *Hd* Haidoob

Fig. 3 Mn concentration in brown algae species collected from locations at the Sudanese Red Sea coast; the full names of locations are *Hl* Haloot, *Fl* Flamingo, *Ah* Abuhashish, *Dm* Dama–Dama, *Kb* Klaineb, *Sw* Sawakin and *Hd* Haidoob
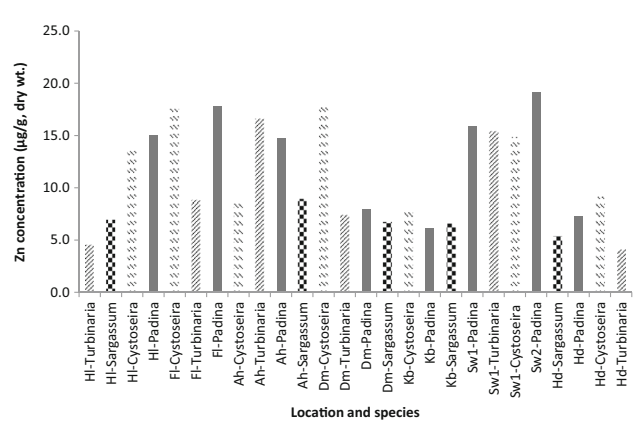

Fig. 6 Zn concentration in brown algae species collected from locations at the Sudanese Red Sea coast; the full names of locations are *Hl* Haloot, *Fl* Flamingo, *Ah* Abuhashish, *Dm* Dama–Dama, *Kb* Klaineb, *Sw* Sawakin and *Hd* Haidoob
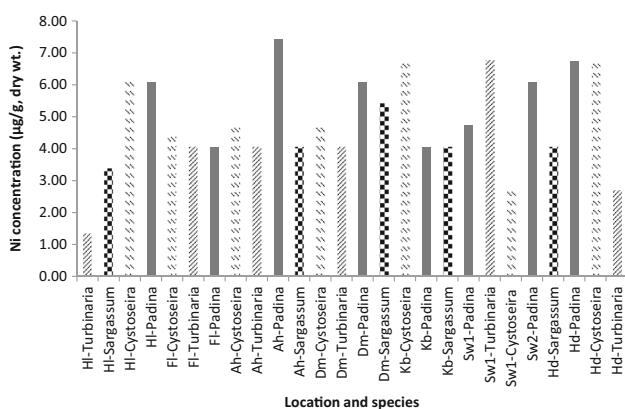

Fig. 4 Ni concentration in brown algae species collected from locations at the Sudanese Red Sea coast; the full names of locations are *Hl* Haloot, *Fl* Flamingo, *Ah* Abuhashish, *Dm* Dama–Dama, *Kb* Klaineb, *Sw* Sawakin and *Hd* Haidoob

Fig. 7 Cd concentration in brown algae species collected from locations at the Sudanese Red Sea coast; the full names of locations are *Hl* Haloot, *Fl* Flamingo, *Ah* Abuhashish, *Dm* Dama–Dama, *Kb* Klaineb, *Sw* Sawakin and *Hd* Haidoob
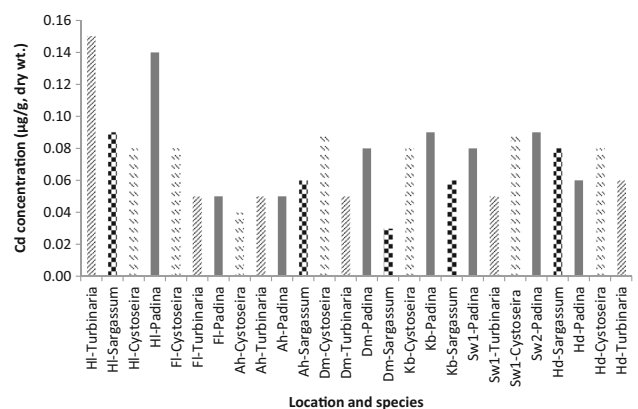
Fig. 8 Pb concentration in brown algae species collected from locations at the Sudanese Red Sea coast; the full names of locations are *Hl* Haloot, *Fl* Flamingo, *Ah* Abuhashish, *Dm* Dama–Dama, *Kb* Klaineb, *Sw* Sawakin and *Hd* Haidoob
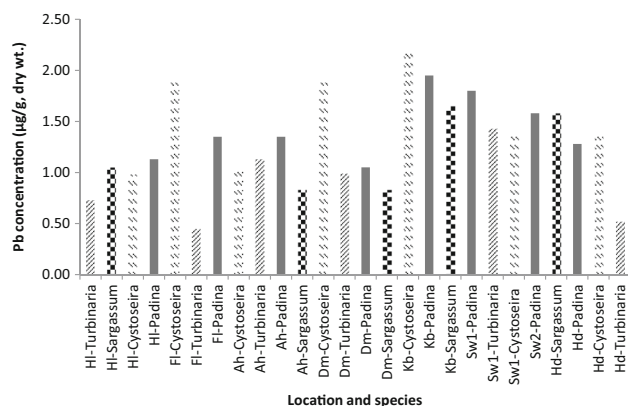
the Sudanese Red Sea coast. In general, different metal concentrations were observed in different species from one location. This issue confirms the variation in species affinity toward metal uptake. As shown in Fig. 2, *Padina* sp. and *Cystoseira* sp. recorded higher Cr uptake than *Turbinaria* sp. and *Sargassum* sp. Irrespective of algae species, the highest Cr uptake was observed at Haloot location, which may indicate Cr contribution from anthropogenic sources. *Padina* sp. and *Cystoseira* sp. also recorded higher Mn uptake than other species (Fig. 3). High uptake of Mn by *Cystoseira* sp. was observed at Haloot, Klaineb and Haidob locations. This issue could also indicate possible Mn contribution from anthropogenic sources. For Ni (Fig. 4), the highest uptake was also observed by *Padina* sp. and *Cystoseira* sp. However, no

location recorded significant elevated levels of Ni. In Fig. 5, *Padina* sp. and *Cystoseira* sp. from Haloot locations recorded the highest Cu uptake. On the other hand, Haloot, Flamingo, Abuhashish and Sawakin locations may be enriched by Zn from man-made activities, a finding that could be observed by high uptake by *Padina* sp. and *Cystoseira* sp. (Fig. 6). Cd (Fig. 7) significantly accumulated in *Turbinaria* sp., *Sargasum* sp. and *Padina* sp. from Haloot location. For Pb (Fig. 8), *Cystoseira* sp. recorded the highest uptake at all locations. On the other hand, no significant elevated levels of Pb were observed at all locations.

The correlation coefficients between the concentrations of the same metal in seawater and brown algae are listed in Table 4. No significant correlation was recorded. Metal concentrations in seawater showed wide fluctuations. Also, brown algae examined in the current study recorded different affinities toward metal uptake. The uptake of metals by algae from seawater behaves bimodal process, i.e., rapid adsorption and slow absorption (Benkdad et al. 2011).

On the other hand, the correlation coefficients between different metal concentrations in brown algae are presented in Table 5. Positive significant correlations were recorded between Cr-Cu, Cr-Cd and Mn-Cu. The coefficients of these correlations were above 0.5 at confidence levels of 0.05, which reflects significant variations in metal uptake by brown algae species.

For a compartive study, Table 6 shows the ranges of metal concentrations in brown algae from the Sudanese Red Sea coast and other areas in the world. Cr concentrations in the Sudanese Red Sea coast were found higher than those in other areas in the world. The levels of Zn, Cd

Table 4 Correlation coefficients between the same metal concentrations in seawater and brown algae

Metal	Cr	Mn	Ni	Cu	Zn	Cd	Pb
Correlation coefficient	0.256	0.140	0.155	−0.055	0.084	0.264	0.407
Significant (2-tailed)	0.299	0.814	0.195	0.819	0.799	0.345	0.077

Table 5 Correlation coefficients between different metal concentrations in brown algae

	Cr	Mn	Ni	Cu	Zn	Cd	Pb
Cr	1.000						
Mn	0.409	1.000					
Ni	0.023	0.413	1.000				
Cu	0.650[a]	0.690	0.433	1.000			
Zn	0.282	0.053	0.292	0.497	1.000		
Cd	0.810[b]	0.237	−0.226	0.435[a]	0.027	1.000	
Pb	0.299	0.453	0.313	0.287	0.361	0.146	1.000

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

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

Table 6 Metal concentrations (μg/g, dry wt.) in brown algae from the Sudanese Red Sea coast and other areas in the world

Area	Species	Cr	Mn	Ni	Cu	Zn	Cd	Pb	References
Sudanese Red Sea coast	*Turbinaria, Sargassum, Cystoseira, Padina and Cystoseira*	4.95–16.95	2.93–257.32	1.35–7.43	0.83–14.10	4.13–19.13	0.03–0.15	0.45–2.18	Current study
Saudi Arabian Gulf coast	*P. gymnospora, C. myrica, H. triquetra and S. angustifolium*	NR[a]	NR[a]	NR[a]	3.95–11.23	15.70-75.25	0.64–1.95	8.84–18.84	Al-Homaidan (2006)
Kutch Gulf, India	*Padina*	2.0–2.3	20.30–26.70	0.60–3.8	3.3–3.7	29.0–282.0	6.4–8.2	0.28–1.80	Chakraborty et al. (2014)
Rabta Bay, Algeria	*Dictyota*	0.89–0.91	NR[a]	NR[a]	2.84–3.14	4.93–5.23	0.098–0.133	0.94–1.48	Laib and Leghouchi (2012)
Black Sea, Bulgaria	*Cystoseira*	NR[a]	NR[a]	NR[a]	NR[a]	NR[a]	0.13–0.20	0.56–1.47	Manev et al. (2013)

[a] Not reported

and Pb in brown algae from the Arabian Gulf (Al-Homaidan 2006) were higher than those from the Sudanese Red Sea coast. High levels in the Arabian Gulf could be attributed to more manmade activities, in particular oil industry, than in the Red Sea. Subsequent studies in the Arabian Gulf showed a trend of increasing of metal concentrations (Naser 2013). Mn and Cu concentrations, which were recorded in some previous studies (Chakraborty et al. 2014; Laib and Leghouchi 2012), were within the ranges those found in the current study. The Kutch Gulf, India (Chakraborty et al. 2014) recorded higher levels of Zn, Cd and Pb than those reported in the current study as well as the Rabta Bay (Laib and Leghouchi 2012) and the Black Sea (Manev et al. 2013).

Conclusions

The uptake of heavy metals by brown algae at the Sudanese Red Sea coast was examined. Metal uptake was in the following descending order: Mn >> Zn ≈ Cr > Cu ≈ -Ni > Pb > Cd. Different affinities of brown algae species toward metal uptake were recorded. *Padina* sp. and *Cystoseira* sp. recorded higher metal uptake than *Turbinaria* sp. and *Sargassum* sp. Accordingly, *Padina* sp. and *Cystoseira* sp. are recommended to be used for metal uptake examination. On the other hand, brown algae from Haloot location recorded the highest uptake of Cr, Mn, Cu, Zn and Cd, which may indicate possible contamination. High Mn uptake was also recorded in algae from Klaineb and Haidob locations. Additionally, elevated Zn contents in algae from Flamingo, Abuhashish and Sawakin locations were observed.

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