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# Levels and spatial distribution of trace elements in macroalgae species from the Todos os Santos Bay, Bahia, Brazil

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ABSTRACT

Concentrations of elements (As, Ba, Cd, Co, Cr, Cu, Li, Mn, Ni, Pb, V, and Zn) were determined in ten species of macroalgae collected from six sites in the Todos os Santos Bay, Brazil, between May and July of 2010. An optimized microwave-assisted digestion procedure was used to digest the samples. The elements were determined by inductively coupled plasma mass spectrometry (ICP-MS). A wide range of metal concentrations were observed between the species analysed. Somewhat higher concentrations of Cd (5.99 $\mu g\ g^{-1}$), Co (372 $\mu g\ g^{-1}$), Mn (640 $\mu g\ g^{-1}$), Ni (17.2 $\mu g\ g^{-1}$) and Zn (51.4 $\mu g\ g^{-1}$) were found in the brown macroalgae species *Padina* spp., whereas elements, such as As (19.7 $\mu g\ g^{-1}$) and Pb (8.27 $\mu g\ g^{-1}$), were mainly concentrated in the species *Sargassum* spp. and *Bostrychia montagnei*, respectively. Statistical analyses (ANOVA) of the *Padina* data showed significant inter-site differences for all metals examined except nickel.



The increasing problem of pollution in coastal metropolitan areas caused by the discharge of industrial wastes as well as domestic and atmospheric emissions has led to actions to estimate the effects of pollutants on marine communities (Amado Filho and Pfeiffer, 1998). Some trace elements are significantly accumulated by many marine and estuarine species. This accumulation gives rise to concerns regarding to the possible detrimental effects of toxic elements on coastal resources and their potential impacts on human health (Hatje et al., 2009).

Marine macroalgae are efficient biomonitors of trace metal pollution (Bryan, 1993). They are sedentary, widespread, with satisfactory dimensions, easy to sample and identify, have high affinities for several trace elements, and are relatively sensitive to ambient changes in metal concentrations in the water column (Breck, 1978; Lobban and Harrison, 1994). Moreover, they only respond to the soluble metal fraction and do not reflect metal levels associated with sedimentary or particulate material (Luoma, 1983; Luoma et al., 1982).

The Todos os Santos Bay (TSB) is located in the northeastern region of Brazil (13° S and 38° W). It is the largest tropical bay in the country, with an area of 1052 $km^2$, situated in the state of Bahia (Cirano and Lessa, 2007). The bay has 15 municipalities along its 184 km coastline perimeter and an urban population in excess of 3 million people (Wagener et al., 2010). The TSB shows remarkable reef formations and mangroves of great ecological and socio-environmental importance. These have suffered from the impact of the metropolitan area and industrial activity, including chemical and petrochemical plants as well as an oil refinery and harbor activities. The TSB also receives discharges from the Subae River, which drains an industrial area containing a lead smelter plant, a paper mill and alcohol distilleries (Amado Filho et al., 2008; Hatje and Andrade, 2009).

Although the TSB has a relatively rich flora of macroalgae (Alves, 2008; Alves and Moura, 2005; Alves et al., 2009, 2010, 2011, 2012, in press; Amorim et al.; 2008; Bandeira-Pedrosa et al., 2004; Barreto et al., 2004; Leite, 1982; Marins et al., 2008; Nunes and Paula, 2000; Nunes and Guimarães, 2008; Santos et al., 2008; Santos, 2010; Santos and Moura, 2011), trace metal levels in algae from TSB have not been well studied. Only one study was conducted (Amado Filho et al., 2008), employing *Padina gymnospora* (Kutzing), Sonder 1871 and *Sargassum* sp. collected in three areas (Tapera, Paranama and Botelho) of the TSB.

This preliminary study examined levels of 12 trace elements (As, Ba, Cd, Co, Cr, Cu, Li, Mn, Ni, Pb, V, and Zn) in ten species of macroalgae, from six coastal sites in the TSB, Bahia, Brazil. The primary objectives of the research were to obtain background data and evaluate inter-site and inter-samples differences in elemental

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concentrations. The algae considered are abundant and well representative throughout the study area and the metals examined are of importance from a biological and ecotoxicological view point.

Inductively coupled plasma mass spectrometry (ICP-MS) was selected for quantification because of its excellent detection power, wide dynamic range, relative freedom from matrix interferences and capability for rapid multielemental determination. The trace elements concentrations were compared with data reported in previous studies to provide information on the present status of heavy metal pollution in the coastal zone of the TSB.

Ten different macroalgae species (a total of thirty-five samples) of *Acanthophora spicifera* (Vahl) Boergesen, *Bostrychia montagnei* Harvey, *Dictyopteris jamaicensis* W.R. Taylor, *Padina* spp., *Sargassum* spp., *Ulva lactuca* Linnaeus, *Bryopsis plumosa* (Hudson) C. Agardh, *Caulerpa racemosa var. occidentalis* (J. Agardh) Boergesen, *Caulerpa scalpelliformis* (R. Brown ex Turner) C. Agardh and *Penicillus capitatus* Lamarck were collected in six sites from the TSB coasts between May and July of 2010 (Fig. 1). Specimens were identified according to Taylor (1960), Littler and Littler (2000) and Dawes and Mathieson (2008).

Macroalgae samples were collected from rock and sand stones by hand in the sublittoral zone (0.5–3 m). At each station, different species of algae present during the season were sampled. The samples were washed at the sampling site in seawater and then transferred to the laboratory in polyethylene bags containing seawater. After delivery to the laboratory, all epiphytes and organic and mineral particles were manually removed, and each algae was washed with deionized water (Milli-Q grade). The sample was first washed with tap water and then rinsed in distilled water. Next, the samples were freeze-dried, homogenized and then reduced to a fine powder.

All closed vessels, polyethylene flasks and plastic containers were cleaned with $HNO_3$ (65% w/v, diluted 1/10 with high-purity water) for 24 h and rinsed with high purity water. Subsequently, all material was dried at 50 °C and stored under clean-air conditions. All plastic containers, polyethylene flasks, pipette tips, PFA Teflon digestion vessels (Milestone SRL, Sorisole, Italy) and reagents that came into contact with the samples or standards were checked for contamination. All solvents and reagents were of the highest commercially available purity grade.

De-ionized water with a resistivity of $\geqslant 18\ M\Omega\ cm^{-1}$ was obtained by means of a Milli-Q Plus pure water generating system from Millipore (Millipore, Molsheim, France) and was employed to prepare all standard and sample solutions. Analytical grade nitric acid (Merck, Darmstadt, Germany) was doubly distilled in a model duoPUR 2.01E sub-boiling system (Milestone, Bergamo, Italy). Monoelemental, high-purity grade stock solutions ($1\ g\ L^{-1}$) of As, Cd, Co, Cr, Cu, Mn, Ni, Pb, V, and Zn and a multielemental

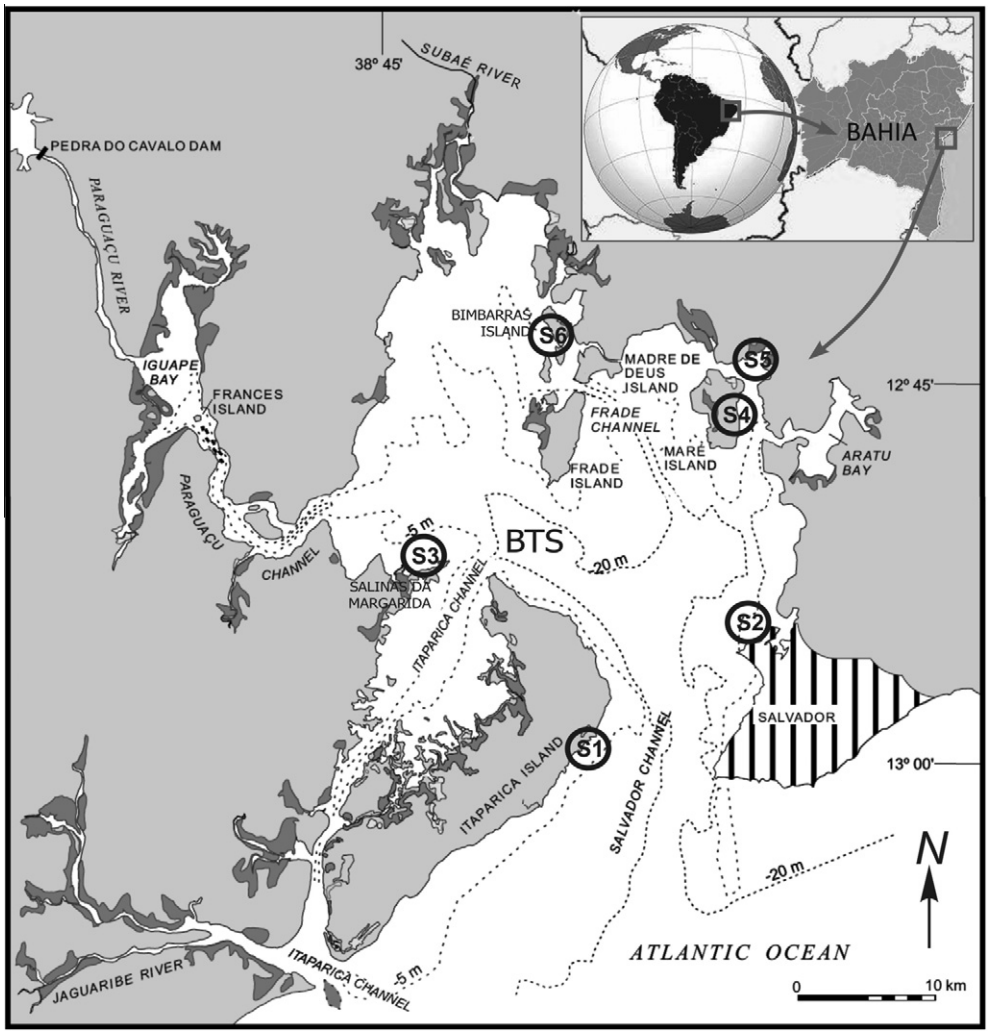


**Fig. 1.** Collection stations (S) in the Todos os Santos Bay, Bahia State, Brazil (S1 – Itaparica Island, Penha beach; S2 – Salvador, Ribeira beach; S3 – Salinas da Margarida; S4 – Maré Island, Botelho beach; S5 – Caboto and S6 – Bimbarras Island) (map based on Cirano and Lessa, 2007).

**Table 1**
Instrumental parameters for elements determination using ICP-MS.

| Instrumental parameter | |
|---|---|
| RF incident power, kW | 1.3 |
| Plasma argon flow rate, L $min^{-1}$ | 13 |
| Auxiliary argon flow rate, L $min^{-1}$ | 0.7 |
| Nebulizer argon flow rate | 0.87 |
| Scanning mode | Peak jump |
| Resolution, ms | Standard |
| Dwell time | 10 |
| Sweeps | 100 |
| Number of readings per replicate | 3 |
| Conditions | $^{140}Ce^{16}O^{+}/^{140}Ce$ and $^{137}Ba^{++}/^{137}Ba^{+}$ < 2% |
| CCT mode gas flow | 6.5 mL $min^{-1}$ |

**Table 2**
Analysis of reference material BCR 279 (sea lettuce): certified values, measured values (mean ± S.D.) and recovery (%).

| | Certified (μg $g^{-1}$ dry wt.) | Measured (μg $g^{-1}$ dry wt.) | Recovery (%) |
|---|---|---|---|
| As | 3.09 ± 0.21 | 3.28 ± 0.15 | 106 |
| Cd | 0.274 ± 0.022 | 0.252 ± 0.019 | 92.0 |
| Cu | 13.1 ± 0.4 | 12.1 ± 0.4 | 92.1 |
| Pb | 13.5 ± 0.4 | 13.8 ± 0.5 | 102 |
| Se | 0.59 ± 0.04 | 0.649 ± 0.051 | 110 |
| Zn | 51.3 ± 1.2 | 44.2 ± 0.8 | 86.1 |

solution (100 mg $L^{-1}$) of Bi, Ge, In, Tl, Rh and Sc were purchased from Merck (Darmstadt, Germany). Plasma torch argon purity was higher than 99.99%.

Acid digestion of the macroalgae samples was performed using a commercial high-pressure laboratory microwave oven (Milestone Ethos 1600 Microwave Labstation, Sorisole, Italy), operating at a frequency of 2450 Hz, with an energy output of 900 W. This microwave digestion system was equipped with ten vessels made with perfluoroalcoxi polymer (PFA) with a volume of 100 mL. Maximum operating temperature and pressure were 300 °C and 100 bar, respectively. The following procedure was considered to be the most suitable for handling macroalgae samples to obtain extracts free from undissolved residues. Freeze-dried samples (200 mg) were digested with 7 mL of nitric acid doubly distilled and 1 mL of hydrogen peroxide. The heating programme was performed in four successive steps. In the first step the temperature was linearly increased up to 120 °C in 5 min. In the second step, the temperature was kept at 120 °C for 3 min. In the third step, the temperature was linearly increased up to 210 °C in 10 min and, in the fourth step the temperature was kept at 210 °C for 15 min. Three replicates of each sample were analyzed. Blank assays were carried out. The extracts were stored at −4 °C in PET-HD flasks previously cleaned with nitric acid.

A quadrupole ICP-MS XseriesII (Thermo, Germany) equipped with a hexapole collision cell (CC) was used for As, Ba, Cd, Co, Cr, Cu, Li, Mn, Ni, Pb, V and Zn determinations and the results were expressed in μg $g^{-1}$ (dry weight). Internal standards (Tl, In, Bi, Ge, Rh and Sc) were added to compensate for any acid effects and instrument drift. The instrument software allows the rapid switch between the standard mode (no gas, cell vented to mass analyzer chamber) and the CC mode, while continuously aspirating the sample. The pre-mixed gases $H_2$ (7%) in He ($H_2O$ and other impurities <5 ppm) were admitted into CC under flow control through stainless steel lines. The measurements were made with nickel sampler and skimmer cones (1.0 mm and 0.7 mm diameter orifices) and standard concentric nebulizer. A glass impact bead spray chamber cooled to 4 °C by a Peltier cooler and a shielded Fassel torch was used to minimize the plasma potential and thereby obtain a low and narrow initial ion energy distribution. Table 1 presents the ICP-MS operational conditions. The quality of the method was checked and confirmed by analysis of seaweed reference material (BCR 279-*U. lactuca*). Taking into account the agreement with the certified values, the following conditions were selected for analysis from the experimental uncertainties: CCT mode for the determination of $^{51}V$, $^{52}Cr$, $^{55}Mn$, $^{59}Co$, $^{63}Cu$, $^{64}Zn$, $^{75}As$ and STD mode for the determination of $^{7}Li$, $^{60}Ni$, $^{111}Cd$, $^{137}Ba$ and $^{208}Pb$.

All the analyses were performed using three replicates. The one-way analysis of variance (ANOVA) was used to compare average contents of the trace elements in algae between the sampling sites. For this statistical test, *Padina* spp. was chosen because it is the sample found in the largest number of collection points, from five the sampling sites (S1, S3, S4, S5 and S6). Principal component analysis (PCA), a multivariate technique, was used to create an ordination plot to demonstrate the variance between metal

**Table 3**
Concentrations of trace elements in the samples of macroalgae species collected from the sampling sites (mean ± SE).

| Sites | Samples | Li | V | Cr | Co | Cu | As | Cd | Zn | Ba | Pb | Mn | Ni |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| S1 | *Sargassum* spp. | 2.30 ± 0.05 | 14.1 ± 0.8 | 6.94 ± 0.10 | 189 ± 5 | 4.75 ± 0.21 | 19.7 ± 0.6 | 0.719 ± 0.028 | 26.6 ± 1.3 | 34.3 ± 1.1 | 3.02 ± 0.10 | 83.2 ± 1.3 | 9.93 ± 0.29 |
| | *C. scalpelliformis* | 0.688 ± 0.016 | 9.85 ± 0.24 | 5.29 ± 0.13 | 271 ± 9 | 4.82 ± 0.16 | 5.50 ± 0.16 | 0.262 ± 0.010 | 14.6 ± 0.8 | 3.97 ± 0.13 | 1.28 ± 0.05 | 16.7 ± 0.6 | 8.98 ± 0.27 |
| | *U. lactuca* | 0.321 ± 0.012 | 6.58 ± 0.32 | 3.35 ± 0.12 | 169 ± 8 | 3.13 ± 0.12 | 5.68 ± 0.16 | 0.265 ± 0.012 | 10.2 ± 0.5 | 25.2 ± 0.1 | 0.729 ± 0.012 | 10.6 ± 0.3 | 7.12 ± 0.24 |
| | *C. racemosa* | 1.57 ± 0.05 | 14.5 ± 0.6 | 6.16 ± 0.26 | 324 ± 20 | 4.05 ± 0.08 | 8.55 ± 0.23 | 0.675 ± 0.022 | 11.4 ± 0.3 | 9.50 ± 0.18 | 1.59 ± 0.14 | 33.1 ± 1.5 | 9.81 ± 0.73 |
| | *Padina* spp. | 1.37 ± 0.05 | 17.6 ± 0.3 | 7.67 ± 0.13 | 293 ± 9 | 4.65 ± 0.22 | 5.42 ± 0.38 | 0.736 ± 0.040 | 26.8 ± 0.7 | 26.0 ± 1.1 | 2.20 ± 0.19 | 88.1 ± 1.5 | 17.2 ± 0.4 |
| | *P. capitatus* | 2.06 ± 0.03 | 1.32 ± 0.04 | 3.20 ± 0.14 | 49.9 ± 0.7 | 2.01 ± 0.04 | 3.82 ± 0.16 | 0.541 ± 0.016 | 6.98 ± 0.11 | 9.02 ± 0.38 | 2.35 ± 0.14 | 37.1 ± 0.4 | 4.56 ± 0.08 |
| S2 | *B. plumosa* | 0.737 ± 0.048 | 8.66 ± 0.37 | 4.57 ± 0.20 | 189 ± 4 | 10.1 ± 0.2 | 21.3 ± 0.6 | 0.166 ± 0.009 | 20.3 ± 0.3 | 5.74 ± 0.11 | 2.63 ± 0.13 | 23.4 ± 0.5 | 8.01 ± 0.26 |
| S3 | *Padina* spp. | 2.81 ± 0.04 | 8.08 ± 0.23 | 5.79 ± 0.17 | 154 ± 7 | 5.21 ± 0.11 | 8.40 ± 0.13 | 1.12 ± 0.03 | 15.8 ± 0.6 | 31.2 ± 0.5 | 2.28 ± 0.11 | 159 ± 4 | 7.73 ± 0.18 |
| S4 | *Padina* spp. | 3.99 ± 0.22 | 9.78 ± 0.12 | 7.20 ± 0.14 | 88.4 ± 3.3 | 27.1 ± 1.2 | 8.98 ± 0.37 | 1.62 ± 0.06 | 51.4 ± 1.6 | 25.2 ± 1.0 | 3.83 ± 0.13 | 250 ± 6 | 8.07 ± 0.47 |
| | *Sargassum* spp. | 1.44 ± 0.03 | 25.1 ± 0.7 | 4.41 ± 0.08 | 178 ± 9 | 15.5 ± 0.4 | 16.1 ± 0.5 | 0.381 ± 0.014 | 25.1 ± 0.5 | 38.6 ± 1.0 | 3.12 ± 0.12 | 81.2 ± 1.6 | 7.47 ± 0.31 |
| | *D. jamaicensis* | 1.41 ± 0.05 | 9.99 ± 0.20 | 5.60 ± 0.28 | 216 ± 7 | 29.8 ± 0.5 | 9.85 ± 0.21 | 0.062 ± 0.05 | 25.9 ± 0.6 | 14.2 ± 0.6 | 3.34 ± 0.14 | 12.3 ± 0.4 | 7.94 ± 0.18 |
| S5 | *Padina* spp. | 1.92 ± 0.03 | 17.2 ± 0.6 | 6.42 ± 0.18 | 372 ± 17 | 103 ± 1 | 6.62 ± 0.10 | 1.08 ± 0.02 | 43.6 ± 2.5 | 42.5 ± 0.3 | 7.66 ± 0.09 | 640 ± 6 | 13.2 ± 0.3 |
| | *B. montagnei* | 3.65 ± 0.06 | 15.7 ± 0.5 | 7.42 ± 1.26 | 16.6 ± 0.5 | 126 ± 5 | 4.01 ± 0.24 | <0.019 | 33.0 ± 1.3 | 16.4 ± 0.4 | 8.27 ± 0.18 | 486 ± 16 | 7.88 ± 0.28 |
| S6 | *Padina* spp. | 2.90 ± 0.07 | 11.9 ± 0.4 | 7.38 ± 0.22 | 193 ± 13 | 13.2 ± 0.1 | 9.81 ± 0.25 | 5.99 ± 0.19 | 41.5 ± 1.0 | 41.2 ± 0.7 | 3.83 ± 0.15 | 163 ± 5 | 9.12 ± 0.54 |
| | *C. racemosa* | 3.38 ± 0.13 | 18.0 ± 0.6 | 8.871 ± 0.33 | 168 ± 9 | 10.3 ± 0.4 | 7.71 ± 0.21 | 1.60 ± 0.05 | 29.5 ± 1.2 | 43.9 ± 0.5 | 3.05 ± 0.08 | 126 ± 3 | 10.6 ± 0.4 |
| | *C. scalpelliformis* | 1.20 ± 0.02 | 4.17 ± 0.10 | 2.64 ± 0.12 | 30.1 ± 2.1 | 5.46 ± 0.15 | 6.22 ± 0.27 | 0.454 ± 0.016 | 12.4 ± 0.4 | 13.0 ± 0.3 | 2.86 ± 0.05 | 165 ± 3 | 3.28 ± 0.08 |
| | *A. spicifera* | 2.90 ± 0.07 | 17.5 ± 0.4 | 9.26 ± 0.29 | 195 ± 4 | 18.4 ± 0.3 | 8.69 ± 0.24 | 4.41 ± 0.21 | 25.7 ± 1.1 | 8.40 ± 0.49 | 2.25 ± 0.09 | 106 ± 2 | 15.9 ± 0.4 |
| | *P. capitatus* | 3.69 ± 0.09 | 6.80 ± 0.14 | 6.46 ± 0.27 | 45.7 ± 0.5 | 4.76 ± 0.12 | 3.59 ± 0.11 | 0.599 ± 0.017 | 12.5 ± 1.0 | 42.5 ± 1.4 | 3.89 ± 0.13 | 104 ± 1 | 5.43 ± 0.18 |

Collection sites (S) in the Todos os Santos Bay, Bahia State, Brazil (S1 – Itaparica Island, Penha beach, S2 – Salvador, Ribeira beach, S3 – Salinas da Margarida, S4 – Maré Island, Botelho beach, S5 – Caboto and S6 – Bimbarras Island).

**Table 4**
Values of one-way analysis of variance in *Padina* spp. species for all sampling sites ($n = 6$) for metal concentrations with $p = 0.05$ (3.285).

| Analyte | *F* | Analyte | *F* |
|---|---|---|---|
| Li | 24.345 | Cd | 2247.8 |
| V | 5.4195 | Zn | 194.91 |
| Cr | 7.3040 | Ba | 50.821 |
| Co | 5.0408 | Pb | 44.792 |
| Cu | 400.76 | Mn | 32.349 |
| As | 5.3301 | Ni | 1.0592 |

concentrations in *Padina* spp. between each sampling station. This technique reduces the number of variables to a smaller set of orthogonal factors, which are easier to interpret when displayed with correlations existing between the original variables (DelValls et al., 1998).

Analytical results for the three subsamples used to calculate the mean elemental concentrations of macroalgae specimens showed coefficients of variation (CV) of 2–10%, demonstrating the homogeneity of the samples prepared of each macroalgae.

The method quantification limits (LOQs) obtained for As, Cd, Co, Cr, Cu, Mn, Ni, Pb, V and Zn were 0.026, 0.019, 0.018, 0.029, 0.034, 0.031, 0.046, 0.012, 0.049 and 0.034 μg $g^{-1}$ dry weight, respectively. The LOQs were determined as 10 SD of the 20 consecutive measurements of the reagent blanks multiplied by the dilution factor used for sample preparation.

The accuracy of measurements was tested using the certified reference material (CRM) BCR-279 Sea lettuce, *U. lactuca* (Institute

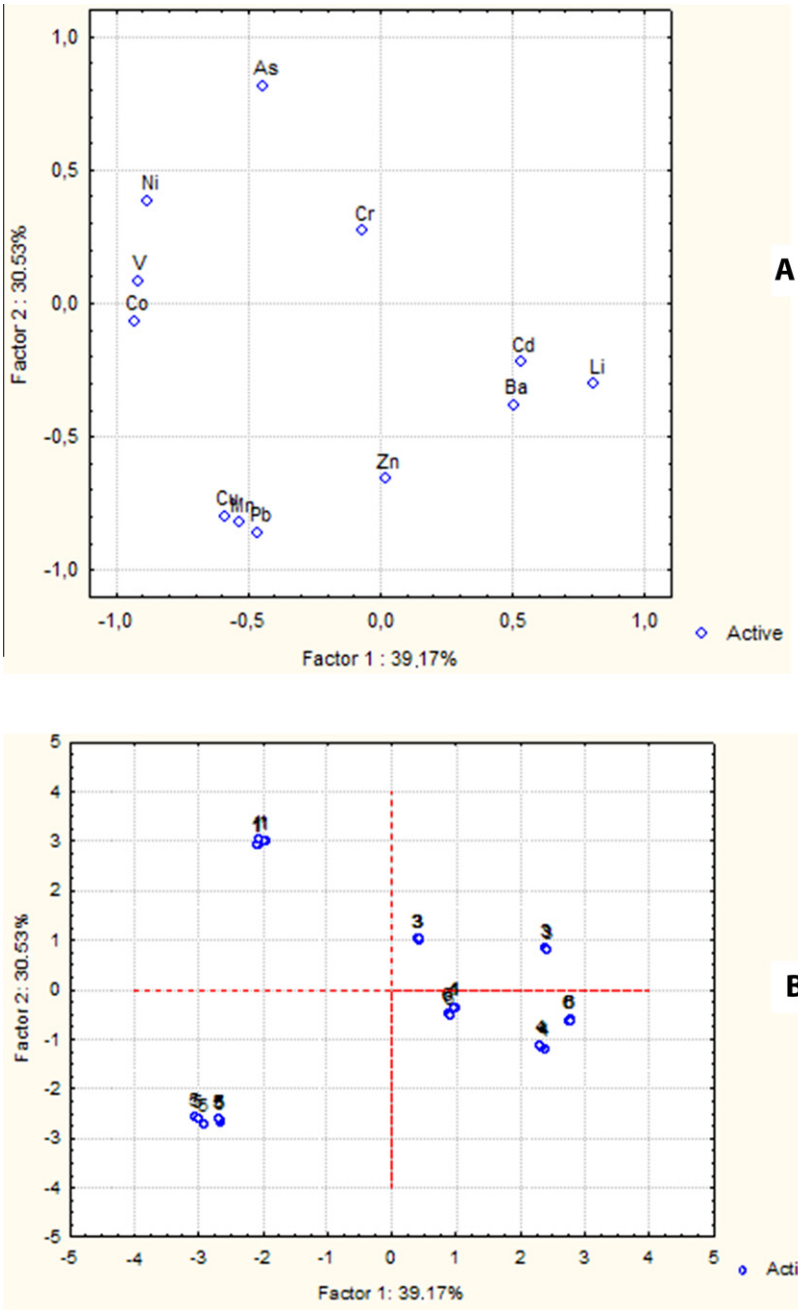


**Fig. 2.** Principal component analysis. (A) Plot of loadings – projection of the variables on the factor plane (1 × 2) and (B) plot of scores – projection of the cases on the factor-plane (1 × 2). PCA on to the first two principal components (PC1 = 39.17%, PC2 = 30.53% of the explained variance) for metal concentrations in algal genera *Padina* spp. analyzed from the collection stations (S) in the Todos os Santos Bay, Bahia State, Brazil (S1 – Itaparica Island, Penha beach; S3 – Salinas da Margarida; S4 – Maré Island, Botelho beach; S5 – Caboto and S6 – Bimbarras Island).

for Reference Materials and Measurements, IRMM, Brussels, Belgium). The results are shown in Table 2. The recoveries (%) by digestion procedures for As, Cd, Cu, Pb, Se and Zn ranged from 86% to 110%. The relative standard deviations were less than 10% for all investigated elements.

The trace element concentrations found in algae from the study area are listed in Table 3. The results revealed a high variability of metal concentrations among different species from the same sampling site. In fact, metal levels are dependent on both biotic parameters and structural differences among the algae species. Furthermore, different species of macroalgae have different affinities for trace elements, which may reflect competition between elements for binding or uptake sites in macroalga. The variable nature and sequestering capacities of metal binding ligands in

**Table 5**
Metal concentration ($\mu g\ g^{-1}$, dry weight) in differents macroalgae from various coastal ecosystems.

| Area | Macroalgae | As | Cd | Co | Cr | Cu | Mn | Ni | Pb | V | Zn | References |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Antartic | 8 species | 6–91 | 1.4–11.7 | – | 0.6–13.1 | 1.3–49 | 2.8–154 | 1–15.4 | 0.5–21.5 | – | 26–131 | Runcie and Riddle (2004) |
| Ghana | 6 species | <LOD-15.73 | <LOD-6.52 | – | – | – | <LOD-126.96 | – | – | <LOD-39.60 | 11.83–32.00 | Serfor-Armah et al. (2001) |
| Antartic | 9 species | 6–152 | – | – | – | – | – | – | – | – | – | Farías et al. (2007) |
| San Jorge Gulf | *Ulva* sp. | 2.98–5.61 | 0.17–1.03 | 0.27–0.73 | 0.84–1.14 | 1.74–3.81 | 8.10–51.4 | 0.99–4.11 | 0.82–1.72 | 2.24–5.57 | 17.4–31.3 | Pérez et al. (2007) |
| Delmarva Peninsula, USA | *Ulva lactuca* | 3.70–6.00 | – | 2.30–7.54 | 6.43–9.35 | 0.65–0.69 | 22.40–50.45 | 0.68–1.60 | 1.63–5.05 | – | 6.86–12.95 | Chaudhuri et al. (2007) |
| São Miguel Island | 6 species | – | 0.03–2.34 | 0.05–2.19 | – | 0.24–7.49 | 3.54–98.05 | 0.3–19.85 | 0.09–0.83 | – | 7.50–740.0 | Wallenstein et al. (2009) |
| Coast of China | *Sargassum* spp. | 65.3–182.8 | – | 0.238–2.07 | 0.699–6.05 | – | 15.4–124 | – | – | – | 11.8–38.7 | Hou and Yan (1998) |
| Coast of Spain | *Ulva* spp. | – | – | – | – | 5.85–14.6 | – | 1.55–6.24 | – | – | 17.03–40.1 | Villares et al. (2002) |
| Arabian Gulf | *P. gymnospora* | – | 1.51–1.95 | – | – | 7.05–11.23 | – | – | 16.00–18.60 | – | 38.76–47.30 | Al-Homaidan (2006) |
| South East India | *Ulva lactuca* | – | 21.7–62.0 | – | 10.5–45.7 | – | – | – | 4.6–20.3 | – | – | Kamala-Kannan et al. (2008) |
| Canary Islands | *Sargassum vulgare* | – | 1.08–1.47 | – | – | – | – | – | 5.03–18.6 | – | – | Lozano et al. (2003) |
| Canary Islands | *Padina pavonica* | – | 0.57–1.66 | – | – | – | – | – | 3.99–22.1 | – | – | Lozano et al. (2003) |
| Egyptian coast | *Sargassum dentifolium* | – | 0.98–2.1 | 1.8–3.5 | 0.6–1.2 | 1.5–5.2 | 6.7–27.1 | 2.1–8.7 | 4.5–25.8 | – | 4.5–55.3 | Abdallah et al. (2005) |
| Egyptian coast | *Padina pavonia* | – | 1.1–6.6 | 4.6–7.4 | 1.0–5.9 | 2.9–10.7 | 5.5–40.9 | 4.9–13 | 7.4–38.9 | – | 5.7–78.4 | Abdallah et al. (2005) |
| Egyptian coast | *Ulva lactuca* | – | 0.8–5.3 | 4.5–6.8 | 6.8–9.2 | 3.5–8.3 | 12.5–16.9 | 2.9–11.5 | 11.3–37.2 | – | 12.4–40.7 | Abdallah et al. (2005) |
| Black Sea | *Ulva lactuca* | – | 4.04–21.8 | 2.91–32.2 | 0.5–1.04 | 4.95–9.52 | 9.98–17.2 | 2.06–2.72 | 1.54–1354 | – | 6.50–19.1 | Tuzen et al. (2009) |
| Southern Baltic | *Enteromorpha* sp. | – | <LOD-1.08 | – | – | 1.82–11.60 | 30.7–499.9 | 0.13–10.20 | 0.90–10.39 | – | 13.6–175.9 | Zbikowski et al. (2006) |
| Turkish coast | *Ulva lactuca* | – | <0.02 | <0.05 | <0.06 | 3.87–13.8 | 12.5–45.1 | <0.1–9.7 | <0.1 | – | 9.6–394.4 | Topcuoğlu et al. (2003) |
| Aegean Sea | *Ulva lactuca* | – | 0.24–1.1 | – | – | 7.0–14.5 | 25.3–182 | <LOD-52.6 | 0.02–2.8 | – | 16.4–88.0 | Sawidis et al. (2001) |
| Aegean Sea | *Padina pavonica* | – | 1.2–1.6 | – | – | 3.0–3.7 | 180.5–202.5 | 18.3–32.3 | 0.02–2.1 | – | 19.3–26.3 | Sawidis et al. (2001) |
| Aegean coast | *Ulva* sp. | – | 14.9–56.9 | – | 0.78–4.78 | 6.49–13.9 | – | – | 0.94–5.65 | – | 40.4–81.0 | Akcali and Kucuksezgin (2011) |
| Aegean coast | *Padina pavonica* | – | 17.9–147.6 | – | 0.73–3.19 | 3.73–8.22 | – | – | 1.22–4.73 | – | 26.3–75.1 | Akcali and Kucuksezgin (2011) |
| Antartic | 11 species | 5.0–112 | <0.1–10.4 | 2.4–357 | 1.6–12.1 | <0.2–15.2 | 0.3–6.5 | 1.8–8.8 | <0.6–7.6 | 0.1–38.4 | <0.2–15.0 | Farías et al. (2002) |
| Loreto Bay, Mexico | 7 species | <LOD-41.4 | – | 0.6–7.19 | 0.99–36.2 | – | – | <LOD-200 | – | – | 8–81 | Sánchez-Rodriguéz et al. (2001) |
| Rio de Janeiro | *Padina gymnospora* | – | 0.61–2.70 | – | – | – | – | – | – | – | 101–899 | Amado Filho et al. (1999) |
| Rio de Janeiro | *Sargassum stenophyllum* | – | 0.30–2.01 | – | – | – | – | – | – | – | 76–580 | Amado Filho et al. (1999) |
| BTS | *Padina gymnospora* | – | 1.01–1.64 | – | 5.5–7.2 | 6.6–32.4 | 350.1–709.1 | 7.8–11.7 | 6.1–11.4 | – | 18.4–54.3 | Amado Filho et al. (2008) |
| BTS | *Sargassum* sp. | – | 0.40–1.45 | – | 1.5–9.0 | 6.0–16.8 | 93.7–334.9 | 8.5–9.7 | 6.2–11.1 | – | 13.5–27.1 | Amado Filho et al. (2008) |
| BTS | *Ulva lactuca* | 3.80–7.56 | 0.232–0.298 | 20.9–316 | 1.01–5.70 | 2.59–3.67 | 9.48–11.7 | 2.59–11.7 | 0.450–1.01 | 1.21–11.9 | 9.86–10.5 | Present study |
| BTS | *Padina* spp. | 5.42–11.3 | 0.736–6.13 | 17.0–372 | 5.73–8.63 | 3.41–103 | 88.1–640 | 5.20–17.2 | 1.83–7.66 | 6.20–17.6 | 11.2–56.5 | Present study |
| BTS | *Sargassum* spp. | 9.95–29.5 | 0.264–0.907 | 168–191 | 2.77–7.77 | 4.33–16.1 | 42.9–123 | 6.11–10.1 | 1.70–4.33 | 11.1–30.2 | 25.0–26.9 | Present study |
| BTS | 7 species | 3.59–29.5 | <LOD-6.13 | 16.6–412 | 1.01–9.33 | 2.01–126 | 9.48–640 | 2.59–17.2 | 0.450–8.27 | 1.21–30.2 | 6.98–56.5 | Present study |

the algal cell wall are also important, at least for some trace elements (Lobban and Harrison, 1994; Phillips, 1977; Schintu et al., 2010).

Trace elements concentrated in *Padina* spp. (Table 3) decreased in the order Co > Mn > V > Ba > Cr > Zn > As > Cu > Ni > Pb > Li > Cd in S1, Mn > Co > Ba > Zn > As > V > Ni > Cr > Cu > Li > Pb > Cd in S3, Mn > Co > Zn > Cu > Ba > V > As > Ni > Cr > Li > Pb > Cd in S4, Mn > Co > Cu > Zn > Ba > V > Ni > Pb > As > Cr > Li > Cd in S5 and Mn > Co > Zn > Ba > Cu > V > As > Ni > Cr > Cd > Pb > Li in S6. The levels of Mn and Co concentrations quantified are high for all sampling areas, suggesting natural accumulation in these species and therefore that these species are good bioindicators. It has long been well established that *Padina* spp. are useful bioindicators for trace metals and numerous references exist in the literature to that effect dating as far back as the mid 1960s (Stevenson and Ufret, 1966). That said, Mn is an element that is thought to be regulated to some extent in brown algae (Morris and Bale, 1975).

ANOVA (log-transformed data) was applied to test the differences between *Padina* spp. taxa concentrations in different sites. Significant differences among stations were found for all metals, except for Ni, in *Padina* spp. (Table 4). These results suggest that the contamination for each element determined in the TSB differs in distinct samples at sampling sites, likely due to anthropogenic features existing next to each collection point. The absence of any significant inter-site differences in Ni levels in *Padina* suggest that this element is homogeneously distributed throughout the study area.

Principal component analysis (PCA) was used to compare concentrations of the elements and the collection points for the algae *Padina* spp. Furthermore, PCA facilitated the examination of variations in the composition of metal in the algal genera studied around the bay (Fig. 2). Principal component 1 (PC 1) explained the majority of variance (39.17%), and principal component 2 (PC2) explained 30.53% of the variance. The combined explained variance of these two components is nearly 70%.

The levels of Cu, Pb and Mn in the algae *Padina* spp. exerted the main influence over the samples of Caboto. For the samples from Penha beach, As exerted the main influence. The samples collected in Salinas da Margarida, Maré Island and Bimbarras Island suffered major influences of Li, and the elements Cd and Ba also mainly influenced Bimbarras' samples.

Mean trace elements concentrations determined among the sampling stations indicated that minimum of V, Cu, As and Zn were obtained in *P. capitatus*; Cr, Ba and Ni in *C. scalpelliformis*; Li, Pb and Mn in *U. lactuca*; and Co and Cd in *B. montagnei*. Most of these values were found in samples collected in Penha beach. Maximum levels of Li, Co, Cd, Zn, Mn and Ni were found in *Padina* spp.; Cu and Pb in *B. montagnei*; V in *Sargassum* spp.; Ba in *C. racemosa*; Cr in *A. spicifera*; and As in *B. plumosa.* Highest concentrations were determined in samples from Caboto.

In order to verify the species of algae with the highest accumulation capacity of the elements, was studied the location Penha beach (S1) due to the greater number of algae sampled. For this region, green algae showed the lowest levels, mainly in *P. capitatus* for most of the elements studied. In contrast, brown algae *(Padina* spp. and *Sargassum* spp.) stood out for the highest concentrations in this locality, for most analytes, except for Cu. Similarly, the location of Bimbarras Island (S6), with 5 different algae collected, green species had the lowest values of concentration for most elements studied, with emphasis on *C. scalpelliformis* followed by *P. capitatus*. Algae of the genus *Padina* spp. showed the highest values for most analytes. Red alga *A. spicifera* showed characteristics intermediate between green and brown algae in the accumulation capacity of the analytes.

Concentration ranges in different algae collected in various geographic areas are shown in Table 5. Comparing different localities of the same gender with algae, among the algae *Padina*, this study also showed that the elements Cr, Cu, Mn and especially Co, showed higher values than the other areas. However, ranges of Cr and Mn were similar to those found in the same area in the work of Amado FIlho et al. (2008). For *Sargassum*, in general, the elements found presented within the range of other locations. For Mn and mainly Pb were obtained lower concentrations than those found for samples in the same locality (Amado Filho et al., 2008). Comparing the locations with algae of the genus *Ulva*, Cd presented concentrations lower than most regions presented. Although they are different species of *Ulva* being compared, it can be considered that they follow a pattern of affinity for elements because they have similarities in the ecology and physiology (Littler and Littler, 1980). The results of this study also demonstrate that the mean concentrations Co and Cu were higher than in the previous study, although for Co similar results were found in Antarctica. Cu contamination found in the S5 is mostly due to the influence of the industrial complex in the estuary that release high concentrations of this element in the environment, not affecting significantly the other BTS locations.

The information obtained in this preliminary investigation supports the assumption on the selective ability of macroalgae to accumulate trace elements from TSB seawater. The results show a high variability in and between species and sampling sites. The concentration of trace elements in macro algae samples encompassed a wide range from <0.019 $\mu g\ g^{-1}$ (Cd) to 640 $\mu g\ g^{-1}$ (Mn). Based on the results obtained in this study, new monitoring programs will be planned.

The use of bioindicators was very valuable for the study of a coastal area with fairly significant basal contamination levels. Despite the great dynamic that occurs in the BTS, the locality of Caboto was revealed as the area most affected by most of the elements studied, mainly for Cu, Pb and Zn. In contrast, Penha beach and Salinas da Margarida were the least impacted areas. The brown algae was clearly the most accumulated elements, followed by red and green, with emphasis on *Padina* and *P. capitatus*, with higher and lower concentration levels found, respectively, for the most studied elements. In general, Cr, Cu and Co were the elements that stood out in the BTS when compared with other geographical areas.

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