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Fucus spp. as a Mercury Contamination Bioindicator in Costal Areas (Northwestern Portugal)

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Abstract Mercury has been considered as one of the most important pollutants in coastal and estuarine areas. Efforts have been made to detect, as early as possible, the effects of this and other metals in several species. Macroalgae, particularly *Fucus* spp., have been widely used as biomonitors of metal pollution. In this study, three *Fucus* species (*F. spiralis*, *F. vesiculosus* and *F. ceranoides*) were collected from several sampling sites in Portugal. The concentrations of mercury were determined in three structural parts (holdfast, stipe and receptacles). Two different techniques were used to determine mercury concentrations. Almost all mercury concentrations (in sediments and in water) were below national and international standards. Mercury concentration in the specimens (0.012–0.061 μg g^{-1} for receptacles, 0.028–0.221 μg g^{-1} for stipe and 0.029–0.287 μg g^{-1} for holdfast) was always higher that those obtained for the sediment (0.001–0.112 μg g^{-1}). With few exceptions the contrary was found for receptacles. In general, a good agreement between concentrations of mercury in sediment and *Fucus* was found. The results indicate that *Fucus* accumulate mercury and may be a suitable species for use in risk assessment for coast and estuarine areas, by providing valuable information regarding the levels of mercury that will be available for the consumers of *Fucus* spp.

Keywords Northwestern coast of Portugal · Risk assessment · Mercury · *Fucus* · Bioindicator

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Anthropogenic discharges are currently one of the most important problems of pollution. Most of the substances present in the composition of these discharges are considerably toxic, persistent and have the potential of bioaccumulating in organisms (Abreu et al. 2000). Mercury is one of these substances, being considered one of the most important pollutants in coastal and estuarine areas, the principal sources of contamination being agriculture, industry and domestic wastes (Pereira et al. 1998a; Ferrat et al. 2003). In the aquatic environment, mercury is normally associated with suspended particulate matter (essentially organic in nature) even when released in its dissolved form. These particles, when present in the water, are later deposited on the surface of the sediment (Caçador et al. 1996). Therefore sediments are considered

as a natural reservoir of mercury and constitute a historical record of mercury released over a period of time (Ramalhosa et al. 2001). The surface layers of sediment are frequently re-suspended, making the mercury bioavailable. This re-suspension may be due to the activity of benthic organisms or dredging activities (Pereira et al. 1998b).

Chemical concentration data do not necessarily translate into impacts on aquatic biota. Each contaminant's potential for harm depends in great part on the environmental conditions, including presence of other contaminants, organic matter content, salinity, and pH (MacFarlane 2002).

Since the 1970s algae have been used as bioindicators of contamination by metals. However, the concentration in tissues show variations according to season, temperature, salinity, age of the stalk, place of fixation, affinity with other metals for the formation of complexes and growth rate (Barreiro et al. 2002).

Macroalgae, as primary producers of great ecological relevance, normally exhibit a high rate of accumulation of the metals present in the sediment and in the water. This suggests that macroalgae could play an important role in the metals transfer cycle throughout the food chain (Thompson-Roberts et al. 1999; Lee and Wang 2001).

Different species of *Fucus* are very abundant in the North Atlantic and in temperate coastal areas (Pearson and Davison 1994), showing consistent seasonality in reproduction, despite their variability (Berger et al. 2001). This genus is of enormous importance, from both ecologically and economical points of view (e.g. production of natural medicinal products), with a large distribution and existing in places exposed to discharges of urban and industrial effluents and to agricultural runoff (Macfarlane 2002). They also have other characteristics making them to be considered as good indicators of contamination by metals, such as wider distribution, ease of harvest, tolerance to wide variations of temperature and salinity and being present all year round (Martin et al. 1997; Coquery et al. 2000).

These algae are mainly found in rocky areas, where evident zonation occurs (Hurd and Dring 1990). According to Karez and Chapman (1998) competitive capacity is inversely related to the position the species occupy in the rocky substratum, and this can be seen by the fact that *Fucus vesiculosus* is more competitive than *Fucus spiralis*, given its inverse location in the rocky substratum. Studies carried out on this genus along the eulittoral showed that specimens present in the lower zone of the rocky substratum normally have a higher concentration of metals than those located in the middle and upper zones. However, this pattern of accumulation is dependent on the metal considered (Martin et al. 1997). When present in high concentrations, metals cause alterations (morphological, histological and biochemical) in *Fucus*. In extreme concentrations they may lead to mortality (Geisinger et al. 2001).

The main objective of this study was to determine the total concentration of mercury, both organic and inorganic, in different structures (receptacles, stipe and holdfast) of three species of *Fucus*, *F. ceranoides* L., *F. spiralis* (*F. spiralis* L. var. *spiralis*, *F. spiralis* L. var. *platycarpus* Batters) and *F. vesiculosus* L. var. *vesiculosus*, collected in different sites along the Portuguese Northwestern coast, from the mouth of the Minho river estuary to Aveiro Lagoon. The mercury concentration in water and sediments was also determined.

Materials and Methods

The three species of *Fucus* were collected from 11 sites along the Northwestern Portuguese coast, between the estuary of the Minho river and the Aveiro Lagoon (Fig. 1). A global position system (GPS, Magelan 2000XL) was used to determine sample site coordinates.

Information regarding sampling stations is provided in Table 1.

Fucus species were found fixed to rocky substratum, their distribution on the Portuguese coast being apparently first determined by the presence or absence of rocky substrata. Thus, their distribution was not homogeneous in the studied area. The species *F. ceranoides* appeared in four stations as the only species of this genus: S1, S2, S7 and S8. The species *F. spiralis* var. *platycarpus* was found in S3 and S6 as the only species of the genus, and in S5 (in Lima river estuary) with *F. vesiculosus*, this last being found in the lower coastal zone. In S9 (Cabo do Mundo) and S10 (Boa Nova), the two varieties of *F. spiralis* were present, *F. spiralis* var. *spiralis* being found on the upper coastal zone. *Fucus vesiculosus* var. *vesiculosus* was observed in S4 and S11, in association with *F. spiralis* var. *spiralis*, which was found as in the previous case, on the upper zone of the rock (Cairrão et al. 2004).

Samples of water, sediment and *Fucus* specimens were collected in September 2002, under diurnal ebb low tide conditions in the crescent moon phase. Sediment samples were collected from the surface (5 cm deep) with a grab and stored at –20°C until analysis. In the laboratory, the sediment was sieved through a 1 mm mesh, and left to dry at room temperature (at approximately 20°C) for five days (Pereira et al. 1998a). Water samples were collected by hand, using polyethylene gloves and glass bottles of 1 dm^3. After collection, water samples were immediately filtered in the laboratory using filters of 0.45 μm Millipore (HA). The dissolved fraction was acidified to pH < 2, with HNO_3 (concentrate, 'mercury free'), and maintained at 4°C until

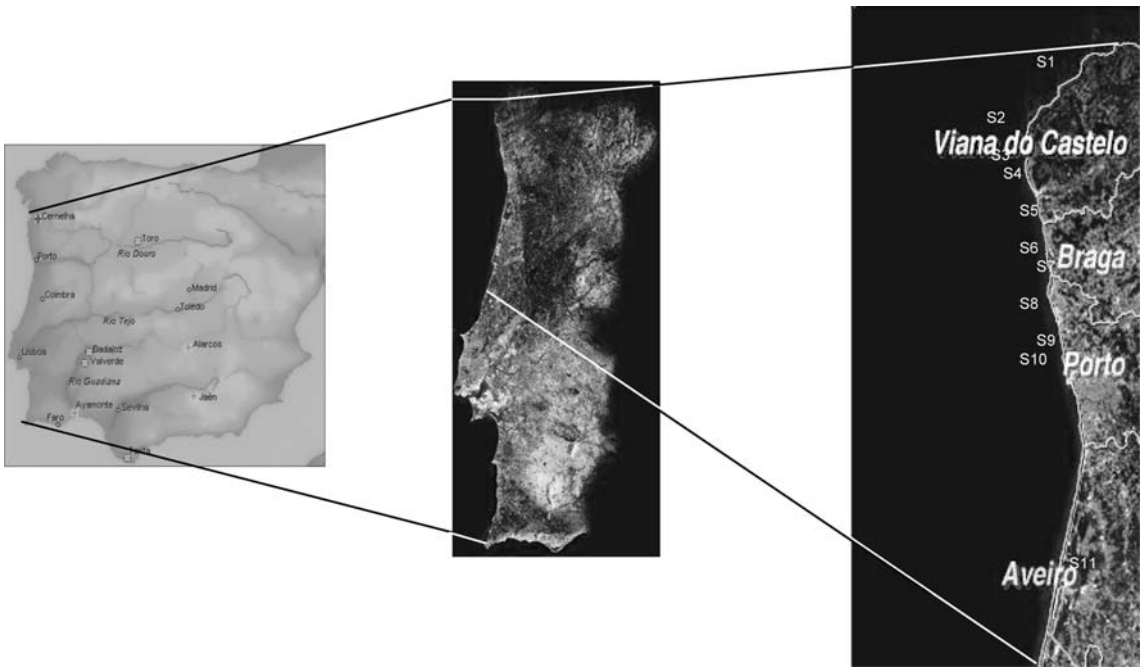

Fig. 1 Sampling sites in the Northwestern Portuguese coast. *S1* Minho river estuary near Seixas; *S2* mouth of Minho river estuary near Caminha; *S3* beach at Carreço; *S4* beach at Areosa; *S5* mouth of the estuary, river Lima near Cabelelo; *S6* beach at São Bartolomeu do Mar; *S7* Esposende, in the Cávado river estuary; *S8* Azurara, in the Ave river estuary; *S9* beach at Cabo do Mundo, Matosinhos; *S10* beach at Boa Nova, Matosinhos; *S11* Barra harbour in Aveiro Lagoon

Table 1 Localisation of the three species of *Fucus* and sources of pollution in the sampling stations

Sampling stations	*Fucus* species	Localisation	Main sources of pollution/pollutants
S1	*F. ceranoides*	Seixas – Minho river estuary	[a]
S2	*F. ceranoides*	Caminha – Mouth of Minho river estuary	Petroleum derived products (fluvial traphic) and urban effluents
S3	*F. spiralis* var. *platycarpus*	Carreço – coastal zone	[a]
S4	*F. spiralis* var. *spiralis* *F. vesiculosus* var. *vesiculosus*	Areosa – coastal zone	[a]
S5	*F. spiralis* var. *platycarpus* *F. vesiculosus* var. *vesiculosus*	Cabedelo – Mouth of Lima river estuary	Mainly petroleum derived products but also urban and industrial effluents
S6	*F. spiralis* var. *platycarpus*	São Bartolomeu do Mar – coastal zone	[a]
S7	*F. ceranoides*	Esposende – Cávado river estuary	Mainly industrial and urban effluents but also petroleum derived products
S8	*F. ceranoides*	Azurara – Ave river estuary	Mainly industrial and urban effluents but also petroleum derived products
S9	*F. spiralis* var. *spiralis* *F. spiralis* var. *platycarpus*	Cabo do Mundo – coastal zone	Urban, industrial and petroleum derived products (refinery)
S10	*F. spiralis* var. *spiralis* *F. spiralis* var. *platycarpus*	Boa Nova – coastal zone	Petroleum derived products (harbour, refinery and marine traphic)
S11	*F. spiralis* var. *spiralis* *F. vesiculosus* var. *vesiculosus*	Barra Harbour – Aveiro Lagoon	Petroleum derived products (harbour and traphic), industrial, agricultural runoff

[a] Apparently not subject to anthropogenic discharges

analysis. The pre-weighed filters used to retain the particulate matter were placed in an oven, at 60°C, for 12 h. To calculate the mass of the suspended particulate matter in the water an additional weighing procedure was performed. The filters were placed in a mixture of equal parts of 50 mL of HNO_3 4 M and HCl 0.7 M, for 2 h, in a sand bath at 60–70°C. These sub-samples were then placed in glass tubes and centrifuged at 6,000 rpm, for 20 min (B. Braun, model Sigma 4–10). The separated material was filtered, using 0.45 μm Millipore (HA) filters, placed in acid-cleaned

polyethylene bottles, preserved by acidification to pH < 2, with HNO_3 (concentrate, 'mercury free', Merch), and kept at 4°C until analysis (Pereira et al. 1998a). Blanks were prepared with ultrapure water using the same procedure, in order to examine for any possible contamination. Ultrapure water was obtained from a Milipore Milli-Q model 185 system. The variability of replicates for filtration was assessed through analysis of two replicates of each sample, analysed two to three times each; the coefficient of variation was in the range from 1.3 to 6.5. The method for mercury analysis in water has a mean analytical detection limit (defined as three times the standard deviation of the blank signal) of 0.79 ng dm^{-3}.

Fucus specimens were identified according to *Flora Phycologica Iberica* (Martí et al. 2001). Each specimen (always with receptacles) was randomly collected by hand with the maximum of care to prevent any loss of biomass, principally at the holdfast, which was fixed to the rocky substratum, to a total of about 0.5 kg of each of the existing varieties (to account for within site variability). The specimens were transported to the laboratory immediately, in a thermic container, where they were washed with distilled water and ultra-pure water to remove surface material (i.e., epiphytes, small crustaceans, and sediments debris) in order to evaluate only the mercury present in algae tissues (Coelho et al. 2005). After this procedure, the three structural parts to be studied (receptacles, stipe and holdfast) were separated, and placed in an oven for three days, at 30°C. After this period, they were homogenised and placed in sterilized glass containers. All material used in the procedures carried out for the determination of mercury was pre-washed in acid (12 h in HNO_3 at 25%), rinsed three times with distilled water and three times with ultrapure water (Pereira et al. 1998a).

Water samples and suspended particulate matter were analysed to obtain the concentration of reactive mercury, using Cold Vapour Atomic Fluorescence Spectrometry (CV-AFS) (cold vapour generator PSA, model 10.003, connected to a Merlin PSA model 10.023 detector). The determination of the concentration of mercury in sediments and in algae was carried out using a gold amalgamation pre-concentration procedure, thermal desorption and Cold Vapour Atomic Absorption Spectrophotometry (CV-AAS) (Perkin Elmer, model 3030 with a MHS-20 system). Accuracy was assessed by taking measures of certified reference materials (CRM). The CRMs used was MESS-2 (marine sediment) for sediments and Rye Grass reference material for *Fucus*. The results were corrected according to the daily recovery percentage of the CRM analyses. CRM analyses showed high efficiency (close to or above 80%) in mercury recovery for all the matrices, and excellent reproducibility. The total mercury concentration in standard marine sediment (MESS-2; 0.078 ± 0.001 μg g^{-1}, mean and standard deviation) does not differ significantly from the value in the reading, with confidence level of 84.7%, from the Rye Grass reference material (0.092 μg g^{-1}, n = 5). For the determination of total mercury in *Fucus*, the Rye Grass (Rye Grass – 0.0150 ± 0.0003 μg g^{-1}) reference material was used for comparison and was found to not significantly differ from the reference material (0.0205 μg g^{-1}, n = 15) (74.3% confidence level).

Data about mercury in the sediment and in the dissolved fraction in the water was analysed by one-way Analysis of Variance (ANOVA), using the SigmaStat Software package version 1.00. The Tukey multicomparison test was used to compare samples from different sites, for each parameter under analysis. For the analysis of dissolved fraction in water, Dunnets test was used to determine the significance between the samples and the blank. In suspended particulate matter data, the Kruskal–Wallis test was used. The Dunn method was later used to discriminate which sampling station was statistically different from each other (Zar 1996). In the analysis of the concentration of Hg in the different structural parts of the *Fucus* specimens, a two-way ANOVA was carried out to compare the three structural parts and sampling stations. A Tukey test was later performed. The significance level was 0.05.

Results and Discussion

The total mercury concentration present at the surface (5 cm deep) of the sediments varied between 0.001 ± 0.000 and 0.112 ± 0.007 μg g^{-1} (mean ± standard error) (Table 2). The highest value was obtained in samples from the Barra harbour in the Aveiro Lagoon (S11). The existence of a chlor-alkaly industry discharging Hg rich effluent for two decades may have contributed to this situation. Despite new technology being adopted in 1992, which drastically decreased the amount of mercury in the effluent, despite that the majority of the metal is adsorbed in neighbouring sediments nearby the discharge location, specially in depth due to the temporal profile of the discharges, Ramalhosa et al. (2001) showed that the entire lagoon was affected and the resuspension of sediments (due to dredging or bottom erosion) originates episodes of renewed metal availability (Pereira et al. 1995; Costa and Jesus-Rydin 2001). Sediments from S7, from the Cávado river estuary, and S8, from the Ave river estuary, have high concentrations of mercury (0.088 and 0.079 μg g^{-1}, respectively) and the stations with lowest concentration of mercury are S10, S3 and S6. The sampling stations S2, S3, S5 and S6 have concentrations of mercury in the sediments that significantly differ ($F = 1825.1$; $df = 10, 32$; $p \leq 0.05$) from the other stations yet similar to each other. S4 and S9

Table 2 Concentration of mercury (μg g^{-1}) present in the superficial layer (5 cm deep) of sediment from 11 sample sites between the Minho river estuary and Aveiro Lagoon, along the Northwestern Portuguese coast

Sampling station	(Hg) (μg g^{-1})
S1	0.025 ± 0.001 d
S2	0.005 ± 0.000 b
S3	0.003 ± 0.000 b
S4	0.016 ± 0.000 c
S5	0.007 ± 0.000 b
S6	0.004 ± 0.000 b
S7	0.088 ± 0.004 f
S8	0.079 ± 0.002 e
S9	0.013 ± 0.001 c
S10	0.001 ± 0.000 a
S11	0.112 ± 0.007 g

Different letters indicate significant differences as calculated by the Tukey test at $p \leq 0.05$

Table 3 Concentration of reactive dissolved mercury (ng dm^{-3}) in the water column, from 11 sample sites between the Minho river estuary and the Aveiro Lagoon, along the Northwestern Portuguese coast

Sampling station	(Hg) (ng dm^{-3})
S1	6.17 ± 0.09 b
S2	25.88 ± 0.35 b
S3	4.68 ± 0.09 a
S4	4.12 ± 0.18 a
S5	9.30 ± 0.71 b
S6	5.18 ± 0.44 b
S7	4.68 ± 0.44 a
S8	4.80 ± 0.44 a
S9	5.18 ± 0.62 b
S10	3.31 ± 0.62 a
S11	11.45 ± 0.35 b
Blank	3.26 ± 0.26 a

Different letters indicates significant differences at $p \leq 0.05$

also show similar concentrations of Hg yet significantly differ from the remaining stations. The sampling sites which also contained considerable concentrations of mercury are located in S7, in the Cávado river estuary and S8, in the Ave river estuary (0.088 and 0.079 μg g^{-1}, respectively). Compared with USEPA standards for sludge (0.017 μg g^{-1}) these can be considered high concentration values (USEPA 2002). These estuaries are subject to continuous discharges of urban and industrial effluents which may carry mercury. These values are explainable by the lower dilution capability of estuaries when compared with coastal areas, as observed by Ramalhosa et al. (2001).

The concentration of reactive mercury present in the dissolved fraction in water varied between 3.31 and 25.88 ng dm^{-3} (Table 3). Sampling stations S1, S2, S5, S6, S9 and S11 differ significantly from the blank ($F = 383.8$; $df = 11, 24$, $p \leq 0.05$), containing reactive mercury, while the concentrations detected in the remaining stations are very close to the blank. The site with the highest value of reactive mercury in the dissolved fraction (25.88 ng dm^{-3}) was S2, well below the maximum permissible limit set by Portuguese law (50 μg dm^{-3}) and by the USEPA (2.1 μg L^{-1}). Stations S3, S4, S7, S8 and S10 (4.68; 4.12; 4.68; 4.8 and 3.31 ng dm^{-3}, respectively) showed values very close to the blank (3.26 ng dm^{-3}) indicating that no significant amounts of mercury are dissolved in the water. The low metal concentrations at stations S7 and S8 is probably due to the fact that mercury is transported in association with particulate organic matter (through organic ligands) or by the formation of complexes with various types of polysulphide and precipitated to the sediment (Pereira et al. 1998b). This seems to be supported by the relatively high concentrations found in the sediment (0.088 and 0.079 μg g^{-1}, respectively) and in the suspended particulate matter (0.94 and 1.16 μg g^{-1}, respectively) at these sites.

The concentration of reactive Hg present in the suspended particulate matter in the water column ranged from 0.38 μg g^{-1} at S9 to 1.38 μg g^{-1} at S6 (Table 4). Significant differences ($p \leq 0.05$) were found between S6 and S2 and the two sample sites situated in Matosinhos (S9 and S10). S2 is significantly different ($p \leq 0.05$) from all other sampling sites. The station showing the highest concentration of Hg in suspended particulate matter in water was S6 (1.38 μg g^{-1}). This station was located in a predominantly agricultural area. One source of mercury pointed out in the USEPA's report to the Congress (USEPA 1997) is

Table 4 Concentration of suspended particulate matter (μg g^{-1}) in the water column, from 11 sample sites along the Northwestern Portuguese coast, between the Minho river estuary and Aveiro Lagoon

Sampling station	(Hg) (μg g^{-1})
S1	0.69 c
S2	0.25 a
S3	0.62 c
S4	0.74 c
S5	0.62 c
S6	1.38 c
S7	0.94 c
S8	1.16 c
S9	0.38 b
S10	0.38 b
S11	0.69 c

Different letters indicate significant differences at $p \leq 0.001$

agricultural burning. At this time of the year farmers start burning weeds that invade the fields during spring and summer, to clean and reuse the nutrients presents. This could explain such high and unexpected levels of mercury in particulate suspended matter. Another possible factor is the lack of a municipal sewage system in this area. This leads to direct dumping of a large number of residential septic tank holding directly into the sea (INAG 2000).

In this study different concentrations of mercury were found in distinct parts of *Fucus* specimens, which is in good agreement with previous studies performed by Martin et al. (1997) in the Severn estuary. The values ranged from 0.012 to 0.061 μg g^{-1} for receptacles, 0.028 to 0.221 μg g^{-1} for stipe and 0.029 to 0.287 μg g^{-1} for holdfast. In all species and sites analysed, receptacles had the lowest concentration of Hg. No significant statistical differences (F = 15.5; df = 2, 151, $p \leq 0.05$) were found between the holdfast and stipe when all the values corresponding to the various sample stations are analysed.

Fucus ceranoides L. had the highest concentration of Hg in sampling station S7 (0.190 μg g^{-1}) and the lowest in S1 (0.046 μg g^{-1}), the differences between all sample sites being significant (F = 521.0, 497.3, 81.4; df = 3, 2, 6, 35; $p \leq 0.05$). The concentration of Hg, at each sampling site in receptacles differs significantly from other structural parts. The stipe and holdfast also show these significant differences, with the exception of S8, where the concentrations in the holdfast and the stipe were similar. The concentration of Hg between all sampling stations of this species, in the three structural parts of the *Fucus* was always statistically different, except in receptacles where the value of the concentration of Hg was similar in S1 and S2 (0.025 and 0.032 μg g^{-1}, respectively), S7 and S8 (0.061 and 0.059 μg g^{-1}, respectively) (Fig. 2). *Fucus spiralis* L. var. *platycarpus* Batters, showed the highest concentration of Hg in S5 (0.060 μg g^{-1}) and the lowest in S10 (0.027 μg g^{-1}), with differences occurring among all stations, with the exceptions of S6 and S10, where the mean values for the three structures were similar (0.031 μg g^{-1}). Under statistical comparison the concentration of Hg in the three structural parts of *Fucus* is different (F = 806.7, 1954.8, 376.9, df = 4, 2, 8, 49; $p \leq 0.05$) in all of the stations analysed (Fig. 2). For *F. spiralis* L. var. *spiralis*, the highest concentration of mercury was registered in S11 (0.069 μg g^{-1}) and the lowest in S10 (0.040 μg g^{-1}), this difference being statistically significant (F = 235.5, 522.3, 94.6; df = 3, 2, 6, 35; $p \leq 0.05$). Through analysis of the values of the concentration of Hg in the three structural parts of *Fucus*, it can be verified that there is a lower concentration of Hg in the receptacles in all sampling sites (Fig. 2). *Fucus vesiculosus* L. var. *vesiculosus*, has a higher concentration of mercury in S11 (0.113 μg g^{-1}), and lower in S4 (0.060 μg g^{-1}), the differences being significant (F = 521.8, 785.9, 41.7; df = 2, 2, 4, 29; $p \leq 0.05$). The value of the concentration of Hg in S4 and S5 between the stipe and the holdfast are not significantly different (Fig. 2). Species which co-exist in the same location were compared and it was verified that the mean mercury concentration differed between all of the species. All three structures showed higher mercury concentration for *F. vesiculosus* than *F. spiralis*. When comparing the two varieties of *F. spiralis*, present in the same location, a higher concentration was observed in *F. spiralis* var. *spiralis*. This is an unexpected outcome since *F. spiralis* var. *platycarpus* spends more time submerged with higher periods of contact with waterborne Hg (dissolved and particulate fractions).

The lowest concentrations of mercury were found in receptacles, when absolute values for all stations and each specimen were analysed. This structure is the one that is present on a periodic basis (reproductive periods) in the context of the species, given that the other two studied structures are considerably more resilient. This phenomena is regulated by diverse factors (ecological, geographical and physicochemical). The exception was observed in station S4 in *F. spiralis* var. *spiralis*, where the values for the concentration of Hg between the receptacle and the holdfast were similar (0.034 and 0.037 μg g^{-1}) and the organisms were smaller in size. This sampling station has a very characteristic typology, with a long strip of rocks defending the beach (were the *Fucus* are attached), which leads to a reduced hydrodynamic force. Hurd (2000) noted that specimens more exposed to wave action are of larger dimensions (within certain limits) in order to more effectively resist the drag effect. On the other hand, the hydrodynamic force is able to tear parts of the algae, specially receptacles, an effect that leads to a lower life expectancy for this structural part. This species is located in a zone closer to the beach, where wave action is less intense, possibly resulting in specimens of a smaller size. Furthermore, under these conditions receptacles are not easily destroyed or removed and would have the same opportunity as the holdfast to bioaccumulate mercury. This is supported by the results obtained for *F. spiralis* var. *platycarpus* which is present in the same location but near to the wave breaking action. In this variety a significant difference between the concentration in receptacles and holdfast was found, probably due to a more frequent removal of the receptacles. Values between the holdfast and stipe showed significant variations, however no consistent pattern was observed.

Comparisons to reference material IAEA–140 (*Fucus* sp.) (0.038 ± 0.006 μg mg^{-1}) (Coquery et al. 2000), showed that values obtained for the holdfast and stipe were always slightly higher in our study, with the exceptions of S2, S5, S7, S8 e S11 where values were significantly higher

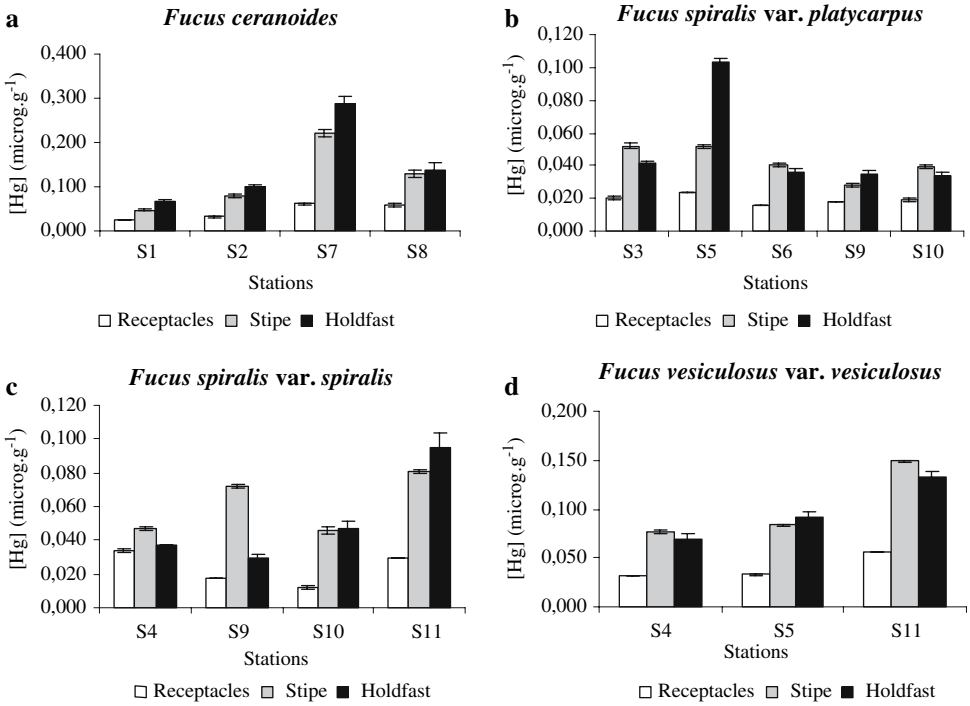

Fig. 2 Mercury concentration (μg g^{-1}) in three different structural parts (receptacles, stipe and holdfast) of **a** *F. ceranoides*, **b** *F. spiralis* var. *platycarpus*, **c** *F. spiralis* var. *spiralis* and **d** *F. vesiculosus* var. *vesiculosus* from 11 sampling stations along the Northwestern Portuguese coast, between the Minho river estuary and the Aveiro Lagoon

than those of the reference material. Mercury concentration in water and sediment at these stations showed the highest values. Naturally, these would be the stations were mercury availability was highest. The receptacle mainly registered lower levels than the reference with the exception of S7, S8 and S11. The reference value for mercury (Coquery et al. 2000) is considered low and this would suggest from our results that the Northwestern coast of Portugal is suffering generalised contamination. However if we consider the admissible maximum value for fish by USEPA (1 μg mg^{-1}) (USEPA 1997) is easy to conclude that the observed specimens (despite being much lower in the trophic chain) are still a great deal distant from concerning levels of contamination. For all 11-sample stations and all species analysed, the concentrations in sediment were lower than those found in holdfast and stipe. The same is not true for the receptacles in sites S1, S7, S8 and S11.

Fucus species are distributed according to a eulittoral gradient, with a well-known characteristic position for each species (Karez and Chapman 1998). *F. cerenoides* always occurs as the representative for the genus. The remaining species appear in unevenly distributed throughout the sampling sites. Martin et al. (1997) quantified the concentration of cadmium, copper, iron, nickel and zinc in *Fucus serratus* located in a characteristic gradient of rocky substratum and observed that specimens nearer the water had higher concentration levels of metals. In both stations S9 and S10, *F. spiralis* var. *spiralis* had higher values of mercury than *F. spiralis* var. *platycarpus*. In S4, S5 and S11, where *F. spiralis* and *F. vesiculosus* co-exist, the latter showed higher values than the former. These results clearly show that the accumulation of metals depends not only on the location of the species in the rocky substratum, but also on different species-specific rates of accumulation of metals by the algae which, in turn, are dependent, through zonation, on environmental factors, including the concentration of nutrients, salinity and temperature (Hurd and Dring 1990). Genetic characteristics of the different species in study could also account for these results (Scott et al. 2001).

In conclusion, the mercury concentrations for the three structural parts of *Fucus* spp. demonstrated that analyses cannot be restricted to a single part of the algae. On one hand receptacles provide information of recent metal pollution, since this structure is only present on a periodic basis. On the other hand, the other two structures provide information on the bioavailability of mercury released over a longer period of time. These results showed consistency with studies by Myklestad et al. (1978) and Lobban and Harrison (1997). At all studied sites, the concentration of mercury was always higher in *Fucus* specimens than in sediments or water. This indicates that *Fucus* species accumulate this metal in their tissues. For this reason, *Fucus* can be considered as a relevant bioindicator species, providing information regarding the concentration of mercury at which *Fucus* predators will be exposed through ingestion. This information may be of a great value for studies of transferance and/or biomagnification of mercury in a coastal trophic web.

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