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Temporal and Spatial Variation on Heavy Metal Concentrations in the bivalve *Perna perna* (LINNAEUS, 1758) on the Northern Coast of Rio de Janeiro State, Brazil

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ABSTRACT

Heavy metal (Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn) concentrations were studied in Perna perna from three beaches (Barra do Furado, Buena and Ponta do Retiro) on the northern coast of Rio de Janeiro State by ICP-AES. The average concentration was 0.3 ± 0.04 (Cd); 1.9 ± 0.6 (Cr); 6.1 ± 0.7 (Cu); 1,130 ± 113 (Fe); 22 ± 3.2 (Mn); 9.3 ± 4.6 (Ni); 0.4 ± 0.2 (Pb); 44 ± 5.8 (Zn) $\mu g.g^{-1}$ dry weight. There were no significant difference for almost all the studied metals in relation to sex. Although, significant spatial variations ($p < 0.05$) occurred for Cr, Pb and Ni with higher values for Barra do Furado. Temporal variations were significant ($p < 0.05$) at the three beaches, however, no trends of accumulation were observed. The metal concentrations were similar to areas under low pollution impact, except for Fe, which was probably due to the local substrates enriched in iron oxides.

Key words: Heavy metals, mollusk, spatial variation and temporal variation

INTRODUCTION

Coastal waters are commonly situated as an endpoint of toxic and environmentally harmful chemicals by direct and indirect inputs (Moraes and Silva, 1995; Nybakken, 1997). Trace elements are natural components of the hydrosphere, although anthropogenic activities had already altered its geochemical cycles increasing people's attention. Since marine organisms accumulate and concentrate heavy metals in high levels, they are widely used as biomonitor organisms reflecting the extent of metal pollution in the coastal waters (Lacerda et al. 1985, Szefer et al. 1998). One of the most successful efforts is popularly known as the Mussel Watch Program (Cantillo, 1998).

Heavy metal toxicity to aquatic organisms in association with the capacity to entry and to keep on the trofic chain for long time justifies metal determination studies on aquatic organisms. However, it's hard to work with environmental contamination by heavy metals due the complex biogeochemistry of these elements on aquatic environments (Förstner and Witmann, 1983). Thus, it is necessary to monitor and to preserve the aquatic environments against the anthropogenic pollution by heavy metals through the knowledge of the base levels of these trace elements.

The aim of this work is to determine Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn concentrations in the bivalve *Perna perna* (LINNAEUS, 1758) collected on the northern coast of Rio de Janeiro

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State at three beaches: Barra do Furado (BF) (22° S; 41° W), Buena (B) (21° 29' S; 41° W) and Ponta do Retiro (PR) (21° 29' S; 40° 59' W), in order to evaluate the temporal and the spatial variation of these metals concentrations and to identify the critical elements in each sampled area.

The following hypotheses were tested: i) there is no difference in *P. perna* metal concentrations in relation to sex; ii) heavy metal concentrations are different among the sampled areas in accordance with local characteristics (different substrate, surface runoff, running coastal waters); iii) there is a seasonal variation on metal concentration along the year.

MATERIAL AND METHODS

Mussels with similar length (4.0 , 5.0 cm) were collected on the low-middle littoral at three beaches every 45 days approximately (from July/98 to May/99). The samples were washed with local seawater, and transported in plastic bags to the laboratory. In the lab, the soft tissues were removed from their shells and the sex was determinate.

The samples were grouped (n = 15) according to their sex, oven-dried at 60 °C, ground to a fine powder with a porcelain pestle and mortar. Aliquots (approximately 1.00 g of dried tissue) were digested (triplicate) in concentrate HNO_3, evaporated and redissolved in 0.5 N HNO_3 (Páez-Osuna et al., 1995; Riget et al., 1997). The extracts were analyzed by atomic emission spectrophotometry with induced coupled plasma (ICP-AES).

Standard reference material was analyzed in order to estimate the accuracy of the digestion method (Muscular Tissue of *Squalus acanthias* , DORM 1) supplied by the Marine Analytical Chemistry Standards Programs (Canada). The heavy metals recovery rates were 84 (Cr), 102 (Cu), 99 (Fe), 87 (Mn), 93 (Ni) e 88 (Zn) %.

The Mann-Whitney statistical analyze ($p < 0.05$) were applied in order to compare the metal concentrations between the sampled areas and sex. The Kruskal-Wallis ($p < 0.05$) variance test were applied to compare the temporal variation of the metal concentration in each sampled area.

RESULTS AND DISCUSSION

In general, Cd, Cu, Mn, Ni, Pb and Zn concentrations in *P. perna* at Ponta do Retiro, Buena and Barra do Furado beaches could be considered as natural background levels (Table 1). These results reflected the usual fluctuations of these metals and their availability for the biota incorporation on the northern coast of Rio de Janeiro State. Similar variability were observed at the same beaches by Rezende et al. (1997) and by Machado (1999) for Cr, Cu, Fe, Mn and Zn concentrations in two macroalgae species.

Comparing the present work with the data base of the International Mussel Watch Program (Table 1) (Cantillo, 1998), our results showed similar average concentrations for Cr (1.9 *vs* 1.6 $\mu g.g^{-1}$ d.w.) and Cu (6.1 *vs* 7.9 $\mu g.g^{-1}$ d.w.), higher for Ni (9.3 *vs* 2.2) and lower for Cd (0.3 *vs* 2.0 $\mu g.g^{-1}$ d.w.), Pb (0.3 *vs* 5.0 $\mu g.g^{-1}$ d.w.) and Zn (44 *vs* 130 $\mu g.g^{-1}$ d.w.). High concentrations for Cd and Pb at the Mussel Watch data came from programs specifically designed to sample "hot spots".

Specifically for Fe, the results showed concentration values close to the areas described under high impact pollution (Table 1), with average values equal or superior to 1000 µg.g-1 of dry weight (d.w.) at three beaches. These results could be related to the presence of this element on the local substrate which has been rich in iron oxides (Pedrini, 1984). This hypothesis was confirmed by Rezende and Lacerda (1986) who studied the capacity of P. perna to accumulate Fe in accordance with the colonized substrate type. The high capacity for Mytillidae to accumulate Fe were described by Regoli and Orlando (1994). They observed a rapid and an elevated Fe absorption as precipitated hydroxides within endocitycs vesicles and in the intertubular spaces.

The average of Cu (6.13 ± 0.5 $\mu g.g^{-1}$ d.w. or 0.9 ± 0.1 $\mu g.g^{-1}$ w.w.) and Zn (45 ± 6.8 $\mu g.g^{-1}$ d.w. or 6.3 ± 0.8 $\mu g.g^{-1}$ w.w.) concentrations were close to the values described for areas with low pollution impact (Carvalho et al., 1993; Szefer et al., 1998) and lower than the maximum concentration for human consumption established by Brazilian Federal Legislation (1978) (Table 1).

Table 1 - Average concentrations of Cd, Cr, Cu, Fe, Mn, Ni, Pb e Zn ($\mu g.g^{-1}$ dry weight) in mollusks of non-impacted (a) and impacted (b) areas for heavy metals MC = Maximum Concentration for human consumption in $\mu g.g^{-1}$ of wet weight (w.w.).

Sample area/Specie	N	Pollut.	Cd	Cr	Cu	Fe	Reference
B. Guanabara/*P. perna*	-	(b)	-	-	14.5	171.5	Rezende and Lacerda, 1986
Yemen/*P. perna*	-	(b)	0.6	-	27	900	Szefer et al., 1997
México/*M. strigata*	-	(a)	0.63	0.7	23	190	Szefer et al., 1998
Japão/*M. edulis*	-	(b)	3.2	5.1	-	314	Szefer et al., 1997
B. Sepetiba/*P. perna*	-	(b)	1.0	-	6.5	-	Carvalho et al., 1993
A. dos Reis/*P. perna*	-	(a)	1.0	-	8.9	-	Carvalho et al., 1993
Macaé/*P. perna*	-	(a)	0.3	1.3	5.1	675	Carvalho et al., 1998
El Salvador/*O. iridescens*	-	(b)	< 1.2	15.8	622	-	Michel and Zengel, 1998
B. Sepetiba/*C. brasiliana*	-	(b)	8.5	-	24.5	-	Gomes et al., 1991
A. dos Reis/*C. brasiliana*	-	(a)	1.7	-	227	-	Gomes et al., 1991
** World Mussel Watch	-	-	2.0	1.6	7.9	-	Cantillo, 1998
P. Retiro/*P. perna*	30	-	0.3 / 0.04	1.7 / 0.24	6.0 / 0.86	1190 / 170	* Present work, 1999
Buena/*P. perna*	30	-	0.3 / 0.04	1.4 / 0.20	6.7 / 0.96	1000 / 143	* Present work, 1999
B. Furado/*P. perna*	30	-	0.3 /0.04	2.5 / 0.36	5.7 / 0.81	1200 / 171	* Present work, 1999
MC (w.w.)	-	-	1	0.1	30	-	Brazilian Federal Legislation, 1978

Sample area/Specie	N	Pollut.	Mn	Ni	Pb	Zn	Reference
B. Guanabara/*P. perna*	-	(b)	20	19	20.9	565.5	Rezende and Lacerda, 1986
Yemen/*P. perna*	-	(b)	60	-	5.2	40	Szefer et al., 1997
México/*M. strigata*	-	(a)	24	16	0.8	42	Szefer et al., 1998
Japão/*M. edulis*	-	(b)	-	-	-	1773	Szefer et al., 1997
B. Sepetiba/*P. perna*	-	(b)	18.4	7.4	< 1.3	205.3	Carvalho et al., 1993
A. dos Reis/*P. perna*	-	(a)	35.7	9.8	2	153	Carvalho et al., 1993
Macaé/*P. perna*	-	(a)	9.3	8.6	1.8	79	Carvalho et al., 1998
El Salvador/*O. iridescens*	-	(b)	-	10.6	< 1.2	2040	Michel and Zengel, 1998
B. Sepetiba/*C. brasiliana*	-	(b)	30.1	18.1	13.5	9500	Gomes et al., 1991
A. dos Reis/*C. brasiliana*	-	(a)	30.5	25	< 1.3	2349	Gomes et al., 1991
** World Mussel Watch	-	-	-	2.2	5.0	130	Cantillo, 1998
P. Retiro/*P. perna*	30	-	26 / 3.71	12 / 1.71	0.4 / 0.06	40 / 5.71	* Present work, 1999
Buena/*P. perna*	30	-	20 / 2.86	4 / 0.60	0.1 / 0.01	42 / 6.00	* Present work, 1999
B. Furado/*P. perna*	30	-	21 / 3.00	12 / 1.71	0.5 / 0.07	51 / 7.29	* Present work, 1999
MC (w.w.)	-	-	-	-	20	50	Brazilian Federal Legislation, 1978

* The present work concentrations are expressed in $\mu g.g^{-1}$ of dry weight and in $\mu g.g^{-1}$ of wet weight ($\mu g.g^{-1}$ d.w../ $\mu g.g^{-1}$ w.w.)
** World Mussel Watch data base , median concentration.

Moreover, comparing with oyster results described by Gomes et al. (1991) and Michel and Zengel (1998), the Cu and Zn concentrations in *P. perna* were especially lower. This trend was observed by Chapman et al. (1996) evidencing the high oyster capacity to accumulate this metals comparing with Mytilidae family members.

The Cr concentrations in *P. perna* (1.89 ± 0.6 $\mu g.g^{-1}$ d.w. or 0.3 ± 0.1 $\mu g.g^{-1}$ w.w.) were higher than the maximum consumption limit established by the Brazilian Legislation (1978) (Table 1). However, Lima (1997) studying the Cr distribution in different areas on the Rio de Janeiro coast observed that the maximum concentration

permissible for human consumption have been overpassed, even in natural waters. Similar results were observed by Carvalho et al. (1998) studying the metal concentrations in *P. perna* at Macaé coast, close to the present studied area. Our results, in accordance with these authors might be indicating the natural capacity of *P. perna* to concentrate elevated amounts of this metal in the environment.

Cd (0.3 ± 0.03 $\mu g.g^{-1}$ d.w.), Mn (23 ± 3.1 $\mu g.g^{-1}$ d.w.), Ni (8 ± 4.8 $\mu g.g^{-1}$ d.w.) and Pb (0.4 ± 0.2 $\mu g.g^{-1}$ d.w.) presented concentrations near to low pollution impact areas for heavy metals as described by the literature (Table 1).

Nevertheless, comparing the Mn results with those of Carvalho et al. (1998) working near Barra do Furado beach (9.3 $\mu g.g^{-1}$ d.w.) high concentrations were observed in the present study. The same condition was observed for Fe. These findings showed different sources of this two metals in the northern coast of Rio de Janeiro State and it could be related to a singular terrestrial input in association with suspension particulate matter.

Metal concentrations and Sex

Many authors have highlighted the importance of sex concerning the variation on metal concentrations during the reproductive season. During this stage, proteins and carbohydrates contents, which have high affinity for heavy metals, are very recruited for gonad tissue production, energetic storage and consumption (Latouxe and Mix, 1982; Páez-Osuna et al. 1995; Lima, 1997).

The results showed no significant differences for Cr, Cu, Fe, Mn, Pb and Zn concentrations between *P. perna* male and female. This could be associated to restricted and short length of these organisms (4.0 , 5.0 cm), in accordance with Rezende and Lacerda (1986). The authors did not find differences on heavy metal concentration between males and females of Mytillidae within the same class length (5.5 , 6.0 cm). However, Lima (1997) showed significant differences ($p < 0.05$) on Cd, Cu, and Zn values among males and females of *P. perna* on Rio de Janeiro State coast with the highest one on males. Latouche and Mix (1982) reported that the gonadal stage could influence the heavy metal bioconcentration in bivalve mollusks. Coimbra and Carraça (1990) suggested the use of immature bivalves in environmental monitoring programs for heavy metals since the youngest mollusks showed reduced gonadal growth.

Significant differences ($p < 0.05$) for Cd and Ni among males and females of *P. perna* occurred in Buena beach with highest values always for females. These results suggested the need of additional studies in order to evaluate the natural fluctuations of this metals between both sexes at these site.

Spatial variation for heavy metal concentrations in *Perna perna*

No significant differences for Cd, Cu, Fe, Mn and Zn among the three studied areas were observed (Fig. 1). These results might be associated to a similar bioavailability of these metals to *P. perna*. Besides, all these elements (Cd in exception) are essential metals, which probably have intracellular regulatory mechanisms to keep their concentrations in equilibrium in the organism.

There were significant differences ($p < 0.05$) on Cr concentrations between Barra do Furado and the other beaches (Fig. 1). The highest values observed at the first could be related to a high availability of this metal in the area. Similar results at the same area were described by Machado (1999) for marine benthic algae. The author suggests that the dissolved fraction could be the major metal incorporation way for marine macroalgae and an additional source for Cr incorporation by mollusks.

Significant differences ($p < 0.05$) in Pb concentrations among Barra do Furado and the other beaches were observed (Fig. 1) with higher values at the former, although the concentrations were lower than the described for non-impacted areas (Phillips et al., 1982; Riget et al., 1997). The highest Pb concentrations observed at Barra do Furado could be associated with the presence of a shipyard neared the sampled area. Chiu et al. (2000) suggested that an increasing in shipping could lead to increase in Pb bioavailability for mariculture zones on coastal waters.

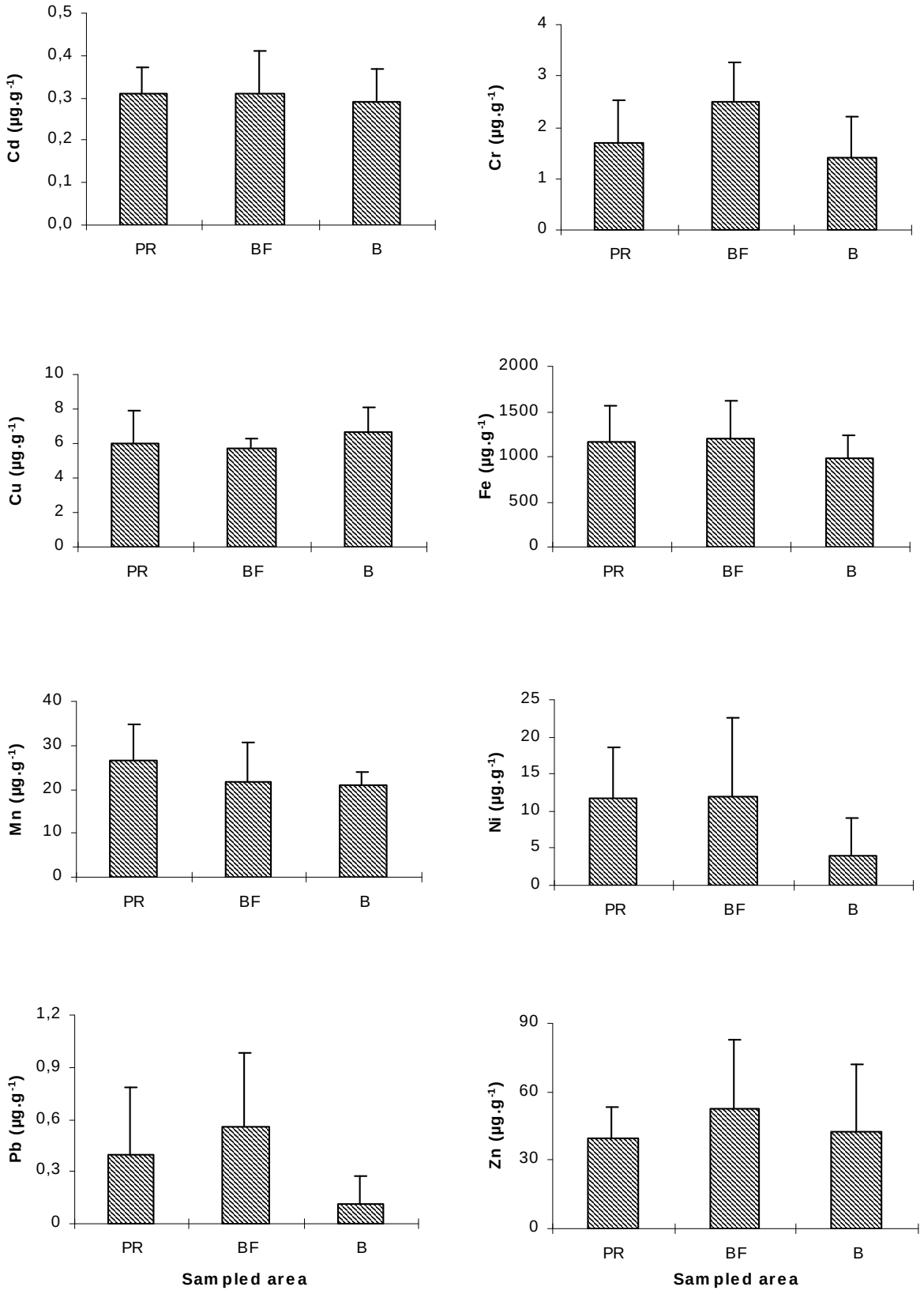

Figure 1 - Average and standard deviation of Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn concentrations µg.g^{-1} (dry weight) in each sampled area (PR = Ponta do Retiro; BR = Barra do Furado; B = Buena) (N = 30).

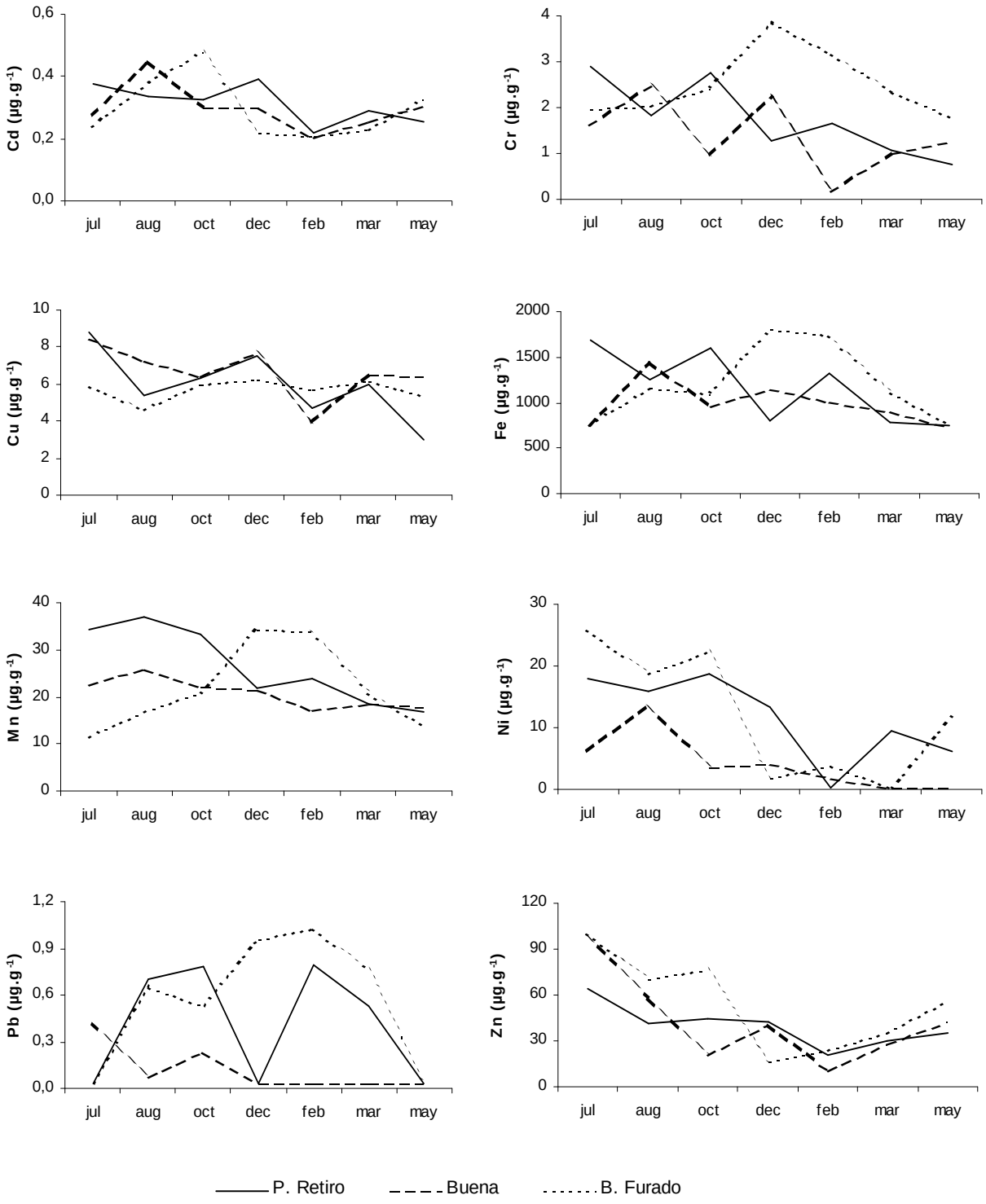

Figure 2 - Temporal variation of Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn concentrations µg.g^{-1} (average in dry weight) in each sampled area (Ponta do Retiro; Barra do Furado; Buena).

Temporal variation for heavy metal concentration in *Perna perna*

Barra do Furado

Significative temporal variations ($p < 0.05$) were registered in Barra do Furado (Fig. 2) for all the metallic elements, except for Cu, an important and essential microelement acting as a respiratory pigment of marine invertebrates and which could be metabolically regulated by mollusks (Förstner and Wittmann, 1983). Similar findings were observed by Wong et al. (2000) studying the heavy metal concentrations in *Perna viridis* from Hong Kong.

Cr, Fe, Mn, Pb and Zn concentrations presented higher concentrations on December/98 and February/99. This behavior is probably indicating that these elements, that are reaching the coastal environment, are from the same source, and follow a similar distribution pattern. C.I.D.E. (1997) explained that the north of Rio de Janeiro State have humid floodplain areas enriched with organic and vegetable material in different degradation stages with concomitant heavy metal liberation, mainly Fe and Mn (Salomão, 1997). During the rainy season (December , March), this inundated areas might be releasing these metallic elements linked to the suspended particulate matter (Kremiling, 1988) by superficial runoff until the local rivers and lakes, as the Flechas creek, an important communication way between the continental inundate areas and the coastal environment of Barra do Furado. Consequently these compounds would be ready for the marine organisms uptake.

Ponta do Retiro

Significant temporal variations ($p < 0.05$) occurred for all metals in Ponta do Retiro (Fig. 2). Cr, Cu, Fe, Mn and Zn concentrations were higher during July/98 and October/98, possibly due to a common origin for these metals. At the same time lower rain levels were registered (average of 34.2 mm) comparing with December/98 to May/99 (average of 77.1mm). These results indicated that the rainy season in Ponta do Retiro beach, in contrast with Barra do Furado, was not related to an elevation of heavy metal bioavailability to *P. perna*. However, in accordance to Kavun et al. (2002), an increasing filtration rate could be produced in response to regionally low levels of chlorophyll-a, seston and suspended matter which probably influence the metal accumulation in *P. perna*.

The Cd, Ni and Pb elements didn't show high concentration peaks during the same months in Ponta do Retiro. This high variability could be indicating that the bioavailability and the dynamic of these elements have a complex regulation level that makes difficult an identification of the origin of these elements, mainly if we considered the reduced natural abundance of these metals.

Buena

Overall, there weren't significative temporal variation in Mn and Cu concentrations in Buena beach (Fig. 2), suggesting a good metabolic regulation of these metallic elements for *P. perna*.

Cd, Cr, Fe, Ni, Pb and Zn showed a tendency to accumulate during July and August/98. These results, as in Ponta do Retiro, were supposed to be related to an elevated filtration rate, which was generally accompanied by a rise in the assimilation of heavy metals from food sources. Fe showed significant temporal variation, with different incorporation periods. These results might reflect the singular capacity of *P. perna* to concentrate and to regulate this metallic element. It did not show a similar behavior at the three beaches, although the close proximity of Buena and Ponta do Retiro beaches (12 Km approximately).

Variations on metal concentrations in mussels during the year ware possibly influenced by several factors (i.e. metal bioavailability, season and the physiology of the mussels (Lacerda et al. 1983; Wong et al. 2000). Suspended particulate matter, a heavy metal source for marine environment (Pfeiffer, 1980; Páez-Osuna et al. 1995; De Gregori et al. 1996) could also provide valuable information to understanding the metal dynamic on the northern coast of Rio de Janeiro State, especially if we considered the local influence of fluvial inputs and the distinct pluviometric fate between the sampled areas.

Finally, we recommend the use of younger specimens of *Perna perna* to monitor the heavy metal concentrations, particularly Cr and Fe, in Ponta do Retiro, Buena and Barra do Furado beaches since this organism showed high capacity to accumulate these metals: satisfactory length, wide distribution and great abundance.

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RESUMO

Este trabalho descreve as concentrações de Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn em *Perna perna* das praias de Barra do Furado (BF), Buena (B) and Ponta do Retiro (PR) na costa Norte do Estado do Rio de Janeiro. As concentrações dos metais foram determinadas em ICP-AES. As concentrações médias foram 0.3 ± 0.04 (Cd); 1,9 ± 0.6 (Cr); 6,13 ± 0.7 (Cu); 1.130 ± 113 (Fe); 22 ± 3,2 (Mn); 9.3 ± 4,6 (Ni); 0.4 ± 0.2 (Pb); 44 ± 5,8 (Zn) $\mu g.g^{-1}$ de peso seco. Não ocorreram diferenças significativas para a maioria dos metais, em relação ao sexo. Variações espaciais significativas ($p < 0.05$) ocorreram para Cr, Pb e Ni com os maiores valores em BF. Variações temporais significativas ($p < 0.05$) ocorreram nas três praias, contudo, nenhuma tendência sazonal de acumulação foi observada. As concentrações dos metais foram similares aquelas áreas descritas sob baixo impacto de poluição por metais pesados, exceto para o Fe, cujos altos valores provavelmente estão associados a presença de substratos locais ricos em óxido de Fe.

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