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Mercury, zinc, and copper accumulation in mangrove sediments surrounding a large landfill in southeast Brazil

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"Capsule": *Mangrove sediments may prevent movement of metals to adjacent bodies of water.*

Abstract

The accumulation of Hg, Zn, and Cu was evaluated in mangrove sediments located between a large, 20-years-old landfill and waters of Guanabara Bay (southeast Brazil). The contamination history of the area provides substantial evidence that metal accumulation in the study site is influenced by past metal emissions from multiple sources (e.g. contaminated rivers and the landfill surrounding the site). At the southern part of the site, metal levels are up to 890 ng g^{-1} Hg, 850 µg g^{-1} Zn, and 58 µg g^{-1} Cu. Enrichment factors and excess (background-deducted) concentration inventories show a high spatial variability of metal contamination and storage in the site, with differences often by a factor higher than two within a sampling station and higher than five between sampling stations. These contrasts are attributable to a coupling between spatial variability of anthropogenic metal input and metal behavior and retention within the sediments. Results indicate that during the last few decades mangrove sediments retained a substantial part of metal emissions to the site, probably reducing the metal transport to Guanabara Bay waters, and suggest the suitability of mangrove ecosystems as physical and biogeochemical barriers to metal contaminant transport. © 2002 Elsevier Science Ltd. All rights reserved.

Keywords: Heavy metals; Metal remobilization; Mangrove rhizosphere; Contamination assessment; Mangrove management

1. Introduction

In many tropical and subtropical areas, e.g. Puerto Rico (Corredor and Morell, 1994), Australia (Mackey and Hodgkinson, 1995), Hong Kong (Tam and Wong, 1995), Saudi Arabia (Sadiq and Zaidi, 1994), Brazil (da Silva et al., 1996), and Taiwan (Chiu and Chou, 1991), mangrove ecosystems have been affected by solid wastes and wastewaters, which are important sources of heavy metals to the environment (Nriagu and Pacyna, 1988). Heavy metals are commonly retained within mangrove sediments (Harbison, 1986; Lacerda et al., 1993; Tam and Wong, 1996; Badarudeen et al., 1996), and several studies indicate that the anoxic, fine-grained, and organic-rich mangrove sediments can reduce the potentially deleterious effects of metal contamination because the low availability of metals accumulated within the sediments for remobilization and biotic uptake (Silva et

al., 1990; Lacerda et al., 1991; Chiu and Chou, 1991; Sadiq and Zaidi, 1994; Tam and Wong, 1995). However, the degradation of mangrove forests (e.g. physical habitat destruction due to dredging and harvesting of vegetation) tends to increase worldwide (Farnsworth and Ellison, 1997), which can promote the remobilization of sediment-trapped metals and facilitate its transport to adjacent areas (Peters et al., 1997; Lacerda, 1998).

Since mangrove sediments are potential sinks for anthropogenic metal contaminants (Harbison, 1986; Tam and Wong, 1996; Clark et al., 1997; Tam, 1998), we investigated the accumulation of Hg, Zn, and Cu in mangrove sediments between the landfill that receives most of the solid waste load of the Rio de Janeiro metropolitan area (the Gramacho Metropolitan Landfill) and the waters of Guanabara Bay, southeast Brazil. The possible role of mangrove sediments as a trap for heavy metals and the potential of mangrove ecosystems to be managed as barriers to heavy metal transfer to the adjacent waterbody are evaluated.

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2. Material and methods

2.1. Study area and sediment sampling

Metal accumulation was investigated in sediments from mangrove ecosystems of Jardim Gramacho (22°45′ S, 43°15′ W), located at the western margin of Guanabara Bay, municipality of Duque de Caxias, Rio de Janeiro State (Fig. 1). Guanabara Bay is a 384-km^2 eutrophic waterbody, which receives discharge from at least 7.8 million inhabitants and more than 6000 industries (Kjerfve et al., 1998). Guanabara Bay sediments show sufficient toxicity to affect benthic organisms, as experimentally observed for the shrimp *Panaeus schmitti* (Moraes et al., 2000).

The Gramacho Metropolitan Landfill was constructed in 1978 in a mangrove area of Jardim Gramacho (Fig. 1). This 1.3-km^2 landfill receives a solid waste input of about 5000 ton day^{-1} (JICA, 1993). Leachate samples from four leachate ponds along the landfill area showed Hg, Zn, and Cu average ($\pm$SD, $n = 4$) concentrations of 8.8 ± 8.3 µg l^{-1} (ranging from
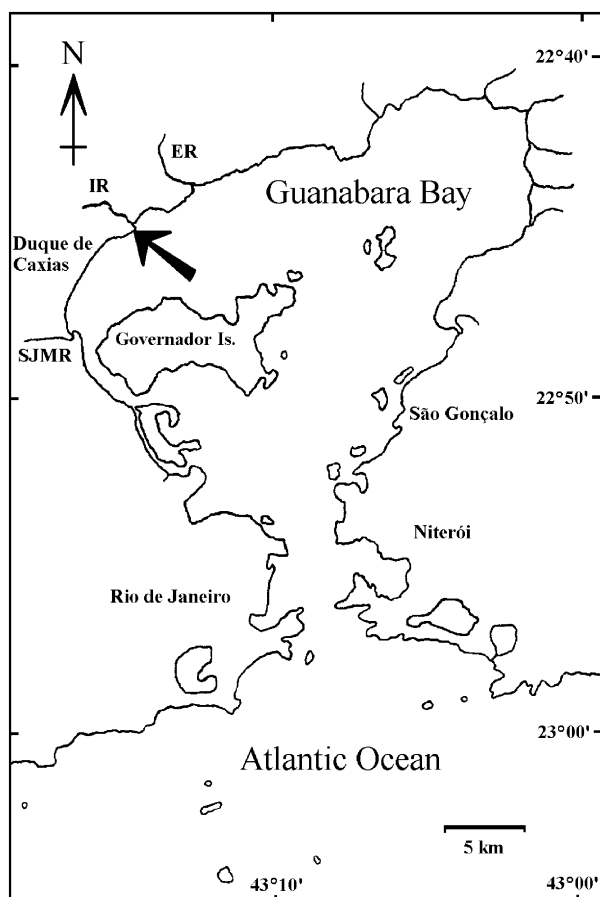

Fig. 1. Location of Guanabara Bay (Rio de Janeiro State, SE Brazil) and the study site at the western part of the bay (indicated by an arrow). IR, Iguaçú River; ER, Estrela River; SJMR, São João do Merití River.

2.5 to 20 µg l^{-1}), 323 ± 177 µg l^{-1} (ranging from 140 to 560 µg l^{-1}), and 118 ± 46 µg l^{-1} (ranging from 70 to 170 µg l^{-1}), respectively (Carvalho et al., 1999). The site possibly receives metals from bay waters, landfill surface runoff and groundwaters, and waters of the Iguaçú River (Fig. 1). This river is an eutrophic, frequently anoxic waterway impacted by industrial and urban effluents (de Souza et al., 1986). Rebello et al. (1986) identified the Estrela River (Fig. 1) as a Cu point source affecting an area within a radius of about 6–7 km from the river mouth, including Jardim Gramacho coast.

After a decline of mangrove ecosystems in the landfill surrounding area, mangrove stands now exhibit considerable regeneration and a patchy structure. Sampling stations included two mangrove stands dominated by *Laguncularia racemosa* (both nearly 100 m from the landfill edge), named stations FM1 and FM2. An additional station was located in the largest degraded area surrounding the landfill (nearly 120 m from the landfill edge), without plant cover, named station DGR. Sampling stations have muddy sediments with a high organic matter content, as indicated by a loss-on-ignition range from 16.8 to 43.0% (Machado, 2000). Between August 1998 and March 1999, two sediment cores were collected at each station using acid-cleaned PVC tubes (9 cm i.d.), 4 m from each other. In the laboratory, sediments were extruded, visually examined, and subsampled. Sediments next to tube walls (0.5 cm thick) were discarded to prevent contamination from the PVC.

2.2. Sediment analysis

Total root content (live and dead roots) in sediments was estimated after wet-sieving via 1-mm mesh sieves and drying (50 °C for 48 h), which was composed by fine roots (<5 mm diameter). Roots were separated from the samples prior to chemical analysis using acid-cleaned plastic spatulas. Metal concentrations were determined on 1.0 g of dried root-removed sediments (50 °C for 24 h), after an acid digestion in 50% aqua regia at 70 °C for 1 h (Marins et al., 1998). This digestion extracts most metals, other than the refractory metals, from the sediment. Handling was performed in acid-cleaned vials and blank digestions were carried with all set of analysis. Mercury was analyzed by cold vapor atomic absorption spectrophotometry, following reduction with SnCl$_2$, whereas other elements were analyzed by flame atomic absorption spectrophotometry. Detection limits, as indicated by three-times the standard deviation of reagent blanks, were 4 ng g^{-1} Hg, 0.6 µg g^{-1} Zn, and 0.5 µg g^{-1} Cu. Triplicate metal analysis were conducted in 10–20% of samples from each core. Precision of this analysis, as indicated by relative standard deviations of replicates, was 13% Hg, 19% Zn, and 16% Cu. To determine the metal

recovery, standard reference sediment (NIST SRM 2704) was analyzed in parallel with the samples. Precision of this analysis, as indicated by relative standard deviations of six replicates, was within 8% for all elements. It is likely that our recovery data are biased low (averages of 88% Hg, 79% Zn, and 76% Cu), and results given below do not represent total metal concentrations.

Maximum enrichment factors (EFs) and excess concentration inventories were estimated to evaluate contamination and contaminant storage in sediments, respectively. Inventories and EFs were calculated for sediment layers above those used to calculate background levels. The average of low and relatively constant values in deep consecutive layers indicated the background value for a core. Because the studied sediments have a consistent fine texture, as previously demonstrated for the western part of Guanabara Bay (Kjerfve et al., 1998; Wasserman et al., 2000), textural corrections were not conducted to estimate background. Zinc and Cu distributions are consistent in all cores, while in most cores isolated Hg concentration peaks between background levels were excluded from the calculation. Average backgrounds ($\pm$SD) for the site, as indicated by average values of the six individual cores, measured 35 ± 5 ng g^{-1} Hg, 54 ± 8 μg g^{-1} Zn, and 5.6 ± 0.7 μg g^{-1} Cu. Maximum EFs were calculated by the ratio between maximum concentrations of each metal and the respective average background for each core. Metal excess inventory (I_{xs}) of a sediment profile was calculated according to the equation: $I_{xs} = \Sigma(M_{xs} \times \rho \times t)$, where M_{xs} is the metal excess (background-deducted) concentration in ng g^{-1} (Hg) and μg g^{-1} (Zn and Cu), ρ is the sediment density in g cm^{-3}, and t is the thickness (cm) of sediment-depth interval, for each depth layer. Density of studied sediments was low (0.24–0.52 g cm^{-3}; Machado, 2000), with the lower values in horizons richer in organic matter and roots.

3. Results and discussion

3.1. Metal distribution, contamination, and storage

Concentrations of all metals in sediments showed a pronounced subsurface increase followed by a concentration decrease to low-contaminated, or background levels in deeper layers (Fig. 2). There was a high variability in metal concentrations at different stations and between surface and subsurface sediments within a station. If the anthropogenic input dominates the metal distribution in sediments, the lower concentrations observed in surface layers of most cores in comparison to subsurface layers suggest a recent decrease of anthropogenic metal emissions for the study area (Godoy et al., 1998). Remarkably, the Hg and Zn concentration

decrease from subsurface to surface layers in station FM1 sediments is comparable to, or higher than maximum concentrations observed in other stations, whereas a similar strong contrast was not observed for Cu (Fig. 2).

Maximum concentrations, metal enrichment factors (EFs), and excess concentration inventories are shown in Table 1. Metal concentration, EF, and inventory analysis indicates a general trend of higher metal accumulation at station FM1. These results suggest a trend of higher Zn accumulation from the southern part (station FM1) to the northern part (station DGR) of the study site, while this trend was not evident for Hg and Cu, except for the higher metal accumulation observed in station FM1 (Fig. 2; Table 1). Maximum concentrations and EFs indicated comparable values for Hg and Cu accumulation in sediments from stations FM2 and DGR, and a higher Zn accumulation in sediments from station FM2 than from station DGR. There was some similar Hg$_{xs}$ and Cu$_{xs}$ inventories in cores from stations FM1 and DGR, while only Cu$_{xs}$ inventories showed a consistent difference between stations FM2 and DGR, with higher values in the latter. A high spatial variability in contamination and in contaminant storage was observed at the site, with differences often a factor higher than two within the same station and higher than five between sampling stations (Fig. 2; Table 1).

Although Perin et al. (1997) reported maximum HNO$_3$-extractable concentrations of 1660 μg g^{-1} Zn and 478 μg g^{-1} Cu in Guanabara Bay sediments, they showed median values of 242 μg g^{-1} Zn and 52 μg g^{-1} Cu for the whole bay. Maximum Zn concentrations in sediment cores (170–850 μg g^{-1}; Table 1) ranged from slightly below to about four times higher than median bay concentration, while maximum Cu concentrations in sediment cores (32–58 μg g^{-1}; Table 1) ranged from slightly below to close to the bay median concentration. Wasserman et al. (2000) reported that within an area affected by the past operation of a chlor-alkali plant at the São João do Merití River estuary (Fig. 1), Hg total (diluted aqua regia-extractable) concentrations in sediments are about 1000–3000 ng g^{-1}, reaching a maximum concentration of 37,200 ng g^{-1} at the subsurface in the southern part of the estuary. These authors observed a Hg concentration range of 125–219 ng g^{-1} on the opposite side of the bay and indicated that Hg contamination from São João do Merití River is not dispersed throughout the bay. This agrees with previous results of Rebello et al. (1986) that indicated generally low Hg concentrations in Guanabara Bay sediments, except close to contamination point sources. Maximum Hg concentrations in sediment cores (260–890 ng g^{-1}; Table 1) were comparable to, or higher than those of sites far from the stronger contamination sources in the bay.
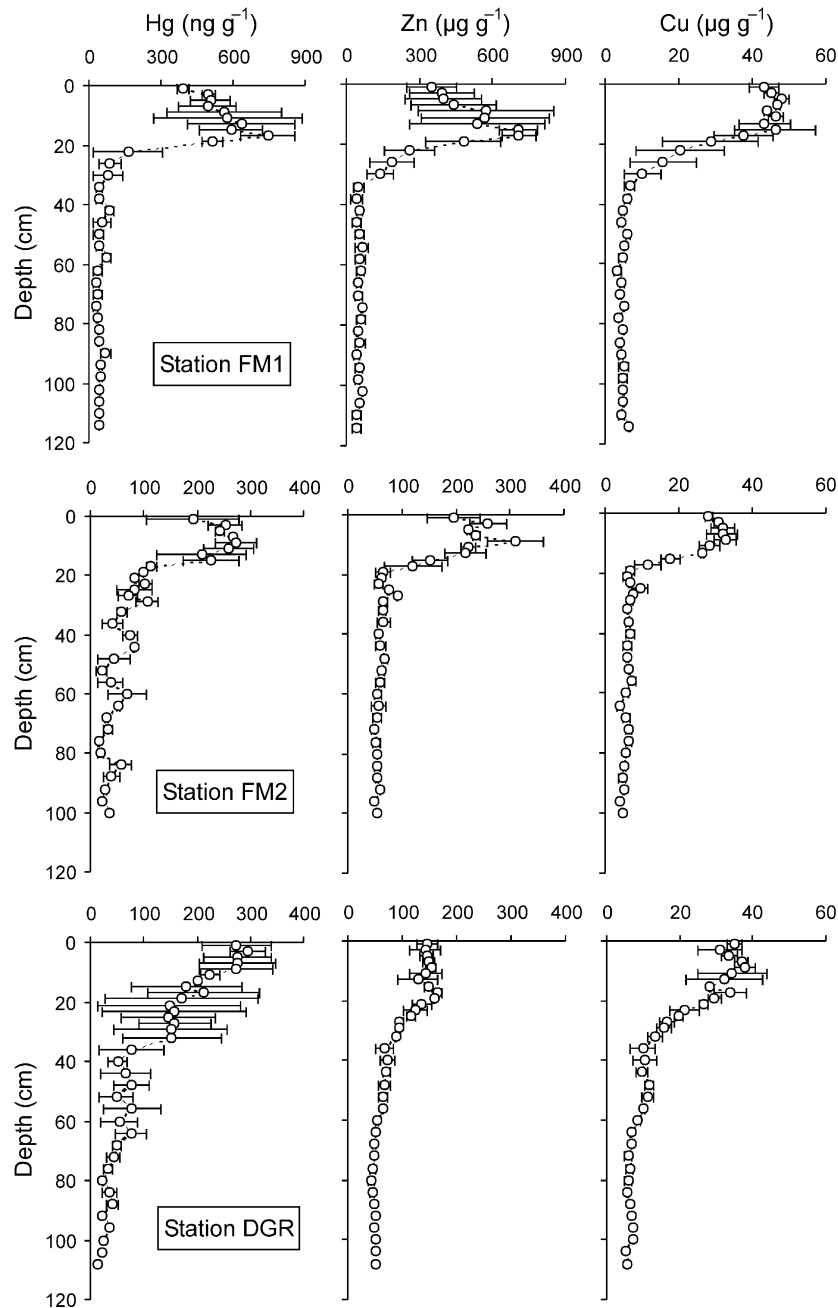
Fig. 2. Metal concentrations in sediments from mangrove forest stations (FM1 and FM2) and degraded area station (DGR). Note the scale differences of mercury and zinc data between station FM1 and other stations. Symbols indicate average values of duplicate core analysis and error bars indicate maximum and minimum values. Symbols of deeper sediment layers of station FM2 (below 94 cm) and station DGR (below 90 cm) indicate values of a single longer core.

3.2. Metal input for the study site

Godoy et al. (1998) investigated and reviewed studies on sedimentation rates in Guanabara Bay, showing that these are 1.6 ± 0.6 cm year^{-1} during the last 40–50 years, as indicated by Pb^{210} analysis. This variability suggest that depths of metal concentration increase in all sediment cores (about 18–34 cm; Fig. 2) correspond to periods less than 40 years before sampling. There was a contribution of metal emissions from industrial sources

for the sediment metal load in the study area since the late 1950s (Godoy et al., 1998), mainly through contaminated rivers (e.g. de Souza et al., 1986; Perin et al., 1997). In addition, the landfill runoff (diluting and transporting leachate) and the resulting wastewater overflow from the landfill to Guanabara Bay probably affected the metal input for the study site in the last two decades (Machado et al., 2000).

Table 2 shows a comparison between estimated metal input for the Gramacho Metropolitan Landfill and the

Table 1
Metal maximum concentrations, maximum enrichment factors (EF), and excess concentration inventories in sediment cores

	Station FM1	Station FM2	Station DGR
Maximum concentration			
Hg (ng g^{-1})	760 (630/890)	290 (270/310)	305 (260/350)
Zn (μg g^{-1})	820 (780/850)	310 (260/360)	170 (170/171)
Cu (μg g^{-1})	53 (48/58)	34 (32/36)	40 (35/44)
Maximum EF			
Hg	20.1 (14.1/26.1)	9.4 (7.9/10.8)	9.2 (7.7/10.6)
Zn	16.5 (13.5/19.5)	5.6 (4.5/6.7)	3.2 (3.2/3.3)
Cu	11.0 (9.7/12.3)	6.1 (5.7/6.5)	6.2 (5.4/7.0)
Excess concentration inventory			
Hg$_{xs}$ (mg m^{-2})	43.1 (30.5/55.7)	13.8 (9.7/17.9)	20.0 (10.2/29.8)
Zn$_{xs}$ (g m^{-2})	40.6 (34.7/46.4)	11.2 (8.8/14.3)	8.6 (8.2/9.1)
Cu$_{xs}$ (g m^{-2})	3.3 (2.6/4.0)	1.5 (1.4/1.6)	2.4 (2.2/2.6)

Values are averages of two cores analysis for each station; results of individual cores are showed in parentheses.

studied mangrove site. Although we can not exclude the possibility of the estimated metal inputs for the study site underestimate past emissions (e.g. because the possible no retention of the total metal load emitted for the site or a post-depositional remobilization of stored metals), this approach indicates metal inputs for the landfill area 2–3 orders of magnitude higher than those for the mangrove area (Table 2). If a substantial wastewater flux (by runoff, or groundwater transport) from the landfill occurred, the approach suggests that past metal inputs for the landfill are sufficient to explain the metal accumulation in the study site. Based on these assumptions we speculate that a coupling between past metal emissions from industrial and urban sources for bay waters and past wastewater transport from the landfill accounted for most of the metal load and distribution in the site. It might be expected that metal inputs to the landfill are mostly maintained within the landfill area since these are related to solid wastes, presumably non-susceptible to substantial transport. This may explain why the estimated metal inputs to the landfill area are so greater than those estimated for the mangrove area.

3.3. Metal behavior in mangrove sediments

Besides the input from natural and anthropogenic sources, heavy metal distribution in sediments can be affected by factors, e.g. as sediment chemical composition and diagenesis (Salomons and Förstner, 1984). Clark et al. (1998) presented a model that recognized the development of oxidation and reduction horizons, water table fluctuations, and bioturbation as major factors controlling the metal behavior within mangrove sediments. Burrowing fauna (polychaeta species, e.g. *Heteromastus filiformis* and *Naeonereis culveri*) were

Table 2
Metal input (g m^{-2}) to Gramacho Metropolitan Landfill area and studied mangrove area[a]

	Hg	Zn	Cu
Landfill area	17–50	2300–6200	850–2500
Mangrove area	0.01–0.06	8.6–46	1.4–4.0

[a] Metal inputs for the landfill are based on a landfill area of 1.3 km^2, 20 years of age, 365 days a year^{-1} operation, 5000 ton day^{-1} urban refuse input, and urban refuse Hg, Zn, and Cu concentrations of 0.6–1.8 g ton^{-1} (EPA, 1993; SWA, 1993), 80–220 g ton^{-1} (Nriagu and Pacyna, 1988), and 30–90 g ton^{-1} (Nriagu and Pacyna, 1988), respectively; excess concentration inventories are assumed as the anthropogenic metal input to the site.

detected above root-dense subsurface horizons in all cores, suggesting that bioturbation possibly affected the metal distribution in upper sediment layers. Mangrove sediments showed mottled subsurface layers with brown, orange, and gray hues, abundant in live and dead mangrove roots. Surface and deeper sediments were gray, or black, with scarcity, or absence of roots. Subsurface brown, or orange hues indicate partially oxidized conditions due to release of O_2 by live roots, while black, or gray hues indicate reduced conditions due to microbial sulfate-reduction (Harbison, 1984; Clark et al., 1998). Sediments at station DGR did not show evidence of oxidized horizons, presenting only gray hues with depth, and dead roots in subsurface layers. The development of redox and acidity gradients in rhizospheres may affect the trace metal distribution and geochemical partitioning in sediments (Lacerda et al., 1993; Clark et al., 1998), that can promote a higher metal accumulation than in surrounding sediments (Caçador et al., 1996; Doyle and Otte, 1997). Furthermore, the presence of mangrove plant cover may reduce the physical disturbance of sediments and preserve the metal sedimentary record better than adjacent unvegetated environments (Wasserman et al., 2000). Since there are frequent storms in the Guanabara Bay area (Kjerfve et al., 1998), it might be expected that plant cover reduces sediment disturbance, resuspension, and consequent oxidation associated to these events, that can increase the remobilization of metal contaminants (Salomons and Förstner, 1984).

The disagreement between EF and inventory trends indicate that caution is necessary to interpret the spatial variability of metal accumulation in diagenetic environments. For example, sediment bioturbation, or post-depositional redistribution of metals can mask original concentration peaks, affecting the assessment of degree of contamination by EFs without affecting the assessment of contaminant storage, except when metal losses from the sediment occur. Moreover, because metal diffusion from the sediments occurs in uppermost layers, this may decrease the metal inventory within the sediment column without affect concentration peaks and

respective EFs at subsurface and deeper layers. These processes may account for the contrast of metal accumulation observed within and between sampling stations. Since elements may show different behaviors during diagenesis (Salomons and Förstner, 1984), vertical distribution and accumulation of different metals within and between sediment profiles may be partially explained by different responses to authigenic processes.

Sediment cores from different stations have a variable root content, reaching total root mass of 0.81 and 1.16 kg m^{-2} in station FM1, 0.16 and 0.26 kg m^{-2} in station FM2, and 0.12 and 0.18 kg m^{-2} in station DGR. Oliveira et al. (1998) showed that roots of *L. racemosa* trees, growing in sediments with a variable degree of contamination by Zn (70–482 µg g^{-1}) and Cu (14–61 µg g^{-1}) along the Guanabara Bay coast, have 24 to 62 µg g^{-1} Zn and 7.8–20 µg g^{-1} Cu, suggesting a relatively low uptake of Zn and Cu by *L. racemosa* even at high sediment contamination levels. The relatively low metal uptake by plants seems to be a general pattern for most mangrove ecosystems (Peters et al., 1997; Lacerda, 1998). If this trend is valid for Jardim Gramacho mangrove stands, there is probably a low contribution of the metal uptake and decay of root materials to the observed metal accumulation. Thus, we hypothesized that the main mechanisms whereby mangrove plants may affect metal behavior in sediments are the physical trapping and stabilization of sediment particles and the development of redox gradients in rhizosphere environments.

4. Conclusions

Jardim Gramacho mangrove sediments appear to retain a substantial part of past metal contaminant emissions to the study site. Metals are probably derived from point sources surrounding the site (contaminated rivers and Gramacho Metropolitan Landfill) and diffuse sources in Guanabara Bay area. This suggests an apparent high metal retention by mangrove sediments within at least the past few decades. Different metal accumulation rates between and within the sampling stations are attributable to a spatial variability of anthropogenic metal input in the study site and diagenetic behavior within the sediments. Mangrove ecosystems surrounding the Gramacho Metropolitan Landfill appear to be a metal sink in the study area, retaining and possibly reducing the remobilization of metals in sediments, that can consequently reduce the metal transport to Guanabara Bay waters. Because sediment-bound metals are immobilized by physical and biogeochemical processes, these environments seem to constitute and may potentially be utilized as a barrier to metal transport in the study site. Further investigations should be conducted to evaluate the management of mangrove ecosystems as barriers to metal transport, including the evaluation of the conservation of remaining mangrove stands and mangrove restoration practices.

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