Environmental changes in Sepetiba Bay, SE Brazil

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Abstract Sepetiba Bay is an example of an aquatic environment that has been severely impacted by human occupation and industrial activities in its basin. Some 400 industries including metallurgical, petrochemical and pyrometallurgical smelters, which emitted pollutants to air, soil and water, were established in Sepetiba Basin during the past 30 years. Apart from these point sources, changes in land use have also resulted in a large remobilization of pollutant deposition on Sepetiba Bay Basin. Studies have pointed out significant changes in sedimentation rates, concentrations of inorganic pollutants (Zn, Cd, Pb and Hg) and more recently, eutrophication, pointing to this area as an example of an impacted coastal zone. Notwithstanding local sources, Sepetiba Bay also suffers environmental impacts caused by diversion of river waters from adjacent basins, with some 30% of the total Hg flux to Sepetiba Bay and a 10-fold increase in water and sediment fluxes resulting from this. Decreasing environmental quality compromises both the large biodiversity and the potential economic uses of Sepetiba Bay, including fisheries and tourism. Monitoring of heavy metal levels in organisms (algae, mollusks, crustaceans and oysters) often shows concentrations well above the limits allowed following Brazilian legislation for food quality. Historical evolution of these concentrations suggests a worsening of the situation. Failure to monitor the effect of land-based activities, including those from other basins artificially associated with Sepetiba Bay has resulted in poor scenario construction and proper management planning.

Keywords Heavy metal contamination · Sedimentation · Water diversion · Land use change · Sepetiba Bay · Brazil

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Introduction

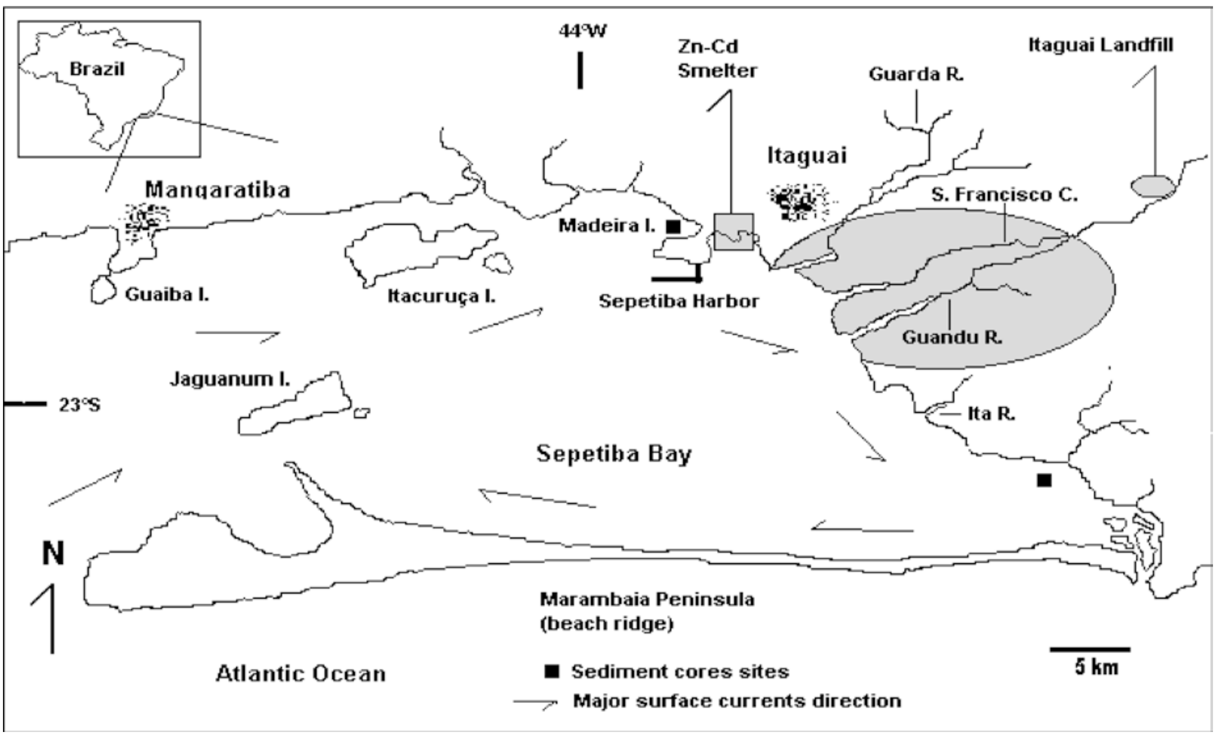
Coastal areas are particularly sensitive to regional environmental changes because they may be considered an ecotone where land and ocean interactions take place. Thus, they may suffer stresses originating from both, on site and land based activities that affect the flow of materials through the coastal zone. This is complicated because coastal areas are considered biologically the most productive regions of the sea and might compromise many activities, for example fish exploitation and consumption. A concentrated effort has been made by recent research to identify the main sources of environmental changes in coastal and marine areas around the world and has related possible effects on environments, such as bays, coral reefs, mangroves and seagrass beds, to on site anthropogenic activities. The observations, however, suggested that apart from direct impact from on site activities on coastal regions, other inland sources might also affect coastal seas through fluvial and atmospheric transport. Therefore, notwithstanding the different approaches used in such studies, the human dimension of fluxes, sediment fluxes and budgets, biogeochemical cycles, river drainage basin-coastal seas interactions, coastal ground water issues, among other factors, have to be taken into consideration in identifying and monitoring environmental changes (Buddemeier 1996; Smith et al. 1997; UNESCO 1998; Smith and Crossland 1999). Sepetiba Bay located in Southeastern Brazil, has been subject to various environmental modifications of physical, chemical and biological features due to on site and land-based human activities. Studies have identified significant changes in sedimentation rates (Forte 1996), concentrations of inorganic pollutants (Barcellos 1995) and more recently, eutrophication (Lacerda 1999), making this area a good example of an impacted coastal zone. Of great concern are impacts from increasingly large heavy metal loads being discharged from industrial emissions into the bay, to the local atmosphere and rivers (Barcellos et al. 1991; Pedlowisk et al. 1991a, 1991b; Mello 1993). Consequently high concentrations of heavy metals can be observed in most of the bay's compartments. However, since the bay suffers low influence of organic sewage and oil contamination (Fonseca et al. 1987), good environmental conditions still exist in most of the bay area, featuring various sensitive natural ecosystems that support the economically and socially important local fisheries. These sensitive systems are now increasingly exposed to high heavy metal concentrations (Lacerda et al. 1988b, 1988c). Notwithstanding the strengthening of environmental control policies on industrial point sources of heavy metals, the general contamination of the bay hasn't decreased as expected. Therefore, the knowledge of the land use changes occurring in the Sepetiba Bay watershed through time may help in explaining the continuous environmental changes presently occurring in the Bay.

Study area

Sepetiba Bay (Fig. 1) is a semi-enclosed water body connected to the sea in the east by a small, shallow inlet, with little water flow, which crosses extensive mangrove forests. In the west, a large natural channel, running between the large islands of Jaguanaum and Itacurussa, with a depth of 30 m, keeps a regular water exchange with the sea. The bay's area at high tide is about 447 km^2 while the minimum area at low tide is about 419 km^2 (Barcellos 1995). Mean water volume is 2.56×10^9 m^3, ranging from a maximum of 3.06×10^9 m^3 and a minimum of 2.38×10^9 m^3. Average depth is about 6 m. The tidal prism volume is 3.4×10^8 m^3, and the ratio between tidal prism and fluvial inputs is approximately 0.03, characterizing the bay as a well-mixed estuary. The turnover time of the water mass was estimated at around 6 days with maximum current velocity at peak tides ranging from 50 to 75 $cm.s^{-1}$. These results were based on a long time series of oceanographic data from two fixed stations at the bay entrance and a two dimensional hydrodynamic tidal model to simulate the circulation in the bay (Kjerfve 2001).

The bay climate is typically hot-humid tropical, with mean annual precipitation of 1,400 mm, and mean evaporation of 960 mm. Nine rivers drain the Quaternary plain at the north-eastern coast of the bay and are responsible for almost all the freshwater inputs to the bay, reaching an annual flow of 7.6×10^6 m^3. Among them, the São Francisco Canal, with an annual flow of 6.5×10^6 m^3, accounts for 86% of the total fluvial inputs. Circulation in the bay is driven by the winds and tides. Dominant winds are SW (250º), adding seawater from the Atlantic Ocean through the western channel. This water gets warmer in the inner portion of the bay close to river mouths, creating a clockwise current pattern, which drives fresh water and fluvial sediments southwards, and keeping water salinity around 30. However during strong NE (70°) winds this clockwise pattern is disrupted, and most of the fluvial inputs go directly through the main channel to the Atlantic

Fig. 1
Map showing the location and major features of Sepetiba Bay, SE Brazil



Ocean, lowering surface salinities to about 25 (Marins 1998). Apart from the marine ecosystem itself, the bay supports 40 km^2 of mangrove forests, which are best developed at the inner eastern end and play an important role in providing nursery and feeding areas for the bay's fisheries. The rocky northern shore borders the Serra do Mar Mountain chain, covered by tropical rain forests (Lacerda 1998).

The population inhabiting the Sepetiba Bay basin increased from 600,000 in 1978 to 1.2 million in the late 1980's, reaching about 1.7 million in 2000. Most of the population is living along the north side due to the industrial development of this region. Most industrial activities were recently introduced and centralized in the Queimados, Itaguaí, Campo Grande and Santa Cruz industrial districts. Beside industrialization and urbanization, the drainage basin also witnesses increasing agriculture (fruits and vegetables), which have transformed important fragments of the tropical rain forest on the Serra do Mar Mountain slopes; beach ridge and sand dune vegetation in the Marambaia peninsula, and mangroves along the east coast (Lacerda et al. 1988a).

Sediment flux and sedimentation rates

Studies of changes in sediment transport and sedimentation rates have proved valuable in understanding changes in aquatic environments because they may cause a large range of transformations in the physical structure of water bodies. For instance, a reduction of sediment transport and corresponding decrease in sedimentation rates may increase erosion in coastal zones. On the other hand, increasing sedimentation rates may silt up protected coastal sites such as estuaries, seagrass beds and mangroves.

One the most significant occurrences related to Sepetiba Bay coastal changes concerns the sediment transport and sedimentation rates in the bay resulting from anthropogenic activities within the drainage basin. Based on comparisons of nautical charts for the São Fransisco canal mouth, Leitão Filho and Patchineelam (1988) proposed that sediment accumulation rates near that region reached up to 0.5 cm/year; up to 2-m depth difference was measured between present day charts and charts made before 1950.

While these figures from analysis of charts are approximate, they are consistent with data based on ^{210}Pb dated sediment cores collected on the Northeastern shore of Sepetiba Bay. During the past 100 years, the sediment accumulation increased from about 32 to 321 mg cm^{-2} y^{-1} (Forte 1996) (Fig. 2). Landscape change analysis provides some reasons for this ten-fold increase in sedimentation rate.

At the beginning of the century, civil works were initiated in the bay's basin, mostly to control malaria, and included digging and straightening of river channels and the building of artificial canals. Although of relatively small scale, these engineering works more than doubled the sediment accumulation rates in the bay. In the 1950s, to supply water for the fast growing population and the development of industrial activities, the Paraíba do Sul River waters were diverged to Sepetiba Bay basin, through the Guandu River, and then distributed among the channels that discharge into Sepetiba Bay (Fig. 3).

Water was diverted from the Paraíba do Sul River into Guandu River, increasing the freshwater flow to Sepetiba Bay from <20 to 160 m^3/s (LIGHT, personal communication). As a consequence sediment fluxes to Sepetiba Bay also increased. Estimates of sediment fluxes from the Paraíba do Sul River in the wet season (January, February and March 2001) show that 718–801 t/day are discharged

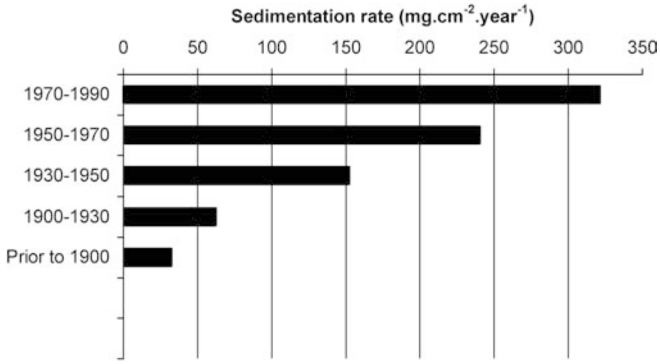


Fig. 2
Sedimentation rates estimated for the Eastern Sepetiba Bay coast, based on Forte (1996)

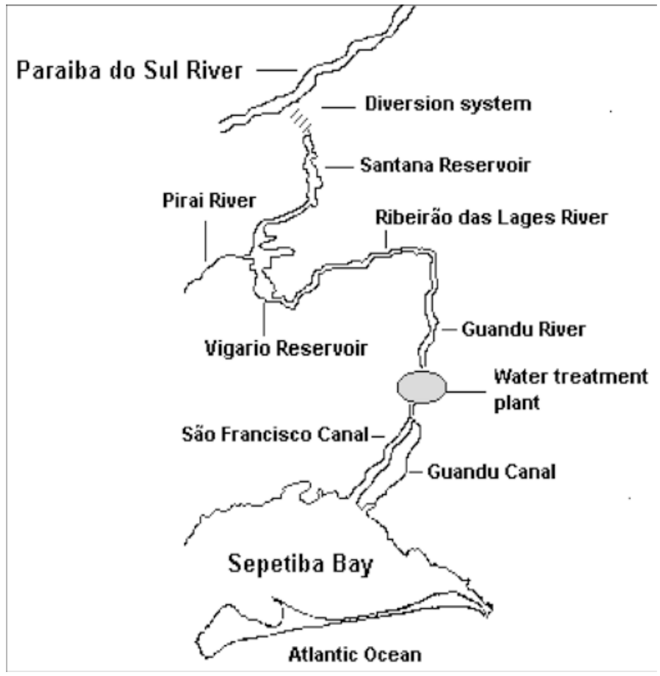


Fig. 3
Scheme of the water diversion system from the Paraiba do Sul River to Sepetiba Bay, SE Brazil

into Sepetiba Bay through the São Francisco Canal, comprising 23–31% of total suspended matter entering the bay (Molisani et al. 2002). This is probably an underestimate because part of the Paraíba do Sul River is diverged to the Guandu River water treatment plant to supply water for Rio de Janeiro City. The water cleaning process uses various techniques for settling suspended particles and organic colloids to the bottom of the reservoir, the sediments are discharged periodically into the Guandu Canal and then flow into Sepetiba Bay. This irregular load results in highly variable concentrations of suspended solids in the Guandu Canal waters that may vary from between 80 and 263 mg l^{-1} during summer months (Molisani et al. 2002). Sedimentation rates are greatest close to the mouth of the main tributary rivers. In other areas they are smaller and may reach zero at the main channel of the bay. Table 1 presents different sedimentation and sediment accumulation rates evaluated by several methods, for areas along Sepetiba Bay. Deposition patterns are spatially variable across the bay, being very high in sheltered places, such as the Saco do Engenho, where additional anthropogenic sediment loads from a Zn-Cd smelting plant contributes to the total fluvial load.

In the present analysis, landscape modifications not only increase by a factor of ten the amount of fresh waters reaching the bay, but also increase sediment accumulation rates to more than 250 mg cm^{-2} y^{-1}. After the 1970s, the creation of an industrial district and the population growth resulted in extensive deforestation of the basin increasing erosion and consequently leading to a further increase in sediment accumulation rates to the present level of 320 mg cm^{-2} y^{-1}. The planned dredging and expansion of Sepetiba Harbor will further augment the processes.

Pollutant distribution and sources

The development of industrial and urban activities in the Sepetiba Bay catchment has been the main cause of degradation of the bay's water quality. The lowlands of the eastern side of Sepetiba Bay, with good transport facilities, cheap and ample lands, good freshwater supply and low population density, have been favored sites for industrial development, further accelerated by the building of a large harbor in the late 1970s. In the last two decades, 400 industries, mostly metallurgical plants, have been built in the region. These include the second most important metallurgical plant in Rio de Janeiro State (COSIGUA – Companhia Siderúrgica Guanabara); a large petrochemical plant and two other large (>10,000 t/year production) pyrometallurgical smelters, plastic and rubber production and food and beverage plants (Lacerda et al. 1988b, 1988c). Industrial and domestic activities in the drainage basin discharge significant amounts of wastes to Sepetiba Bay. Of the many pollutants, heavy metals are the most important class of contaminants. Various studies showed strong contamination of Cd and Zn in areas influenced by freshwater input, with sediment concentrations ranging between 0.1–396 for Cd and 10 – 37,300 μg g^{-1} for Zn (Lacerda et al. 1988b, 1988c; Leitão Filho and Patchineelam 1988; Barcellos et al. 1991). At some sites concentrations reached about the same levels reported in irrigated soils by Jinzu River (Japan) where Cd contamination caused an Itai-itai disease outbreak (Kasuya et al. 1992). Patchineelam et al. (1989) determined the Pb distribution in bottom sediments of Sepetiba Bay and reported concentrations in most contaminated sites of up to 668 μg g^{-1}. The average values represented an enrichment factor around 2.8 relative to background levels. For Hg, bottom sediments concentrations ranged from 17–163 μg kg^{-1}, with higher values also observed near fluvial influence, reaching background levels (<20 μg kg^{-1}) seaward (Marins et al. 1998a).

Bottom sediments of Sepetiba Bay are important witnesses of metal contamination; hence some studies determined metal distribution in sediment cores to evaluate temporal evolution of the contamination in the bay. All studies reported increases in metal concentrations above background levels. Figure 4 shows Zn and Cd distribution in cores collected around Madeira Island (Fig. 1), an area showing very high Zn and Cd concentrations. Figure 5 compares the vertical distribution of these two elements with Hg, obtained from cores taken in a less contaminated site, Coroa Grande beach (Fig. 1). The accumulation profiles suggest that Hg contamination in the bay is more recent, coinciding with accelerated industrialization and urbanization processes in the 1970s. In comparison, the increase in Zn and Cd are thought to coincide with the beginning of the commissioning of the Ingá metallurgical plant in the late 1950s (Fig. 5).

To trace possible sources of heavy metals to Sepetiba Bay, emission factors have proved a useful tool to evaluate

Table 1
Sedimentation rates estimated for different areas in Sepetiba Bay, according to different methods

Locality	Method	Sedimentation rates (cm yr^{-1})	Accumulation rates (mg cm^{-2} yr^{-1})	Author
Mangroves, Enseada das Garças	^{210}Pb	0.12–0.18	240–360	Smoak and Patchineelam (1999)
East coast and the São Francisco Canal mouth	Bathymetric charts	0.5	400	Leitão Filho et al. (1995)
East coast, including São Francisco Canal mouth	^{210}Pb	0.2–0.8	160–640	Forte (1996)
Saco do Engenho	Sediment traps	1.3	650	Barcellos (1995)

Location of sites is detailed in Fig. 1

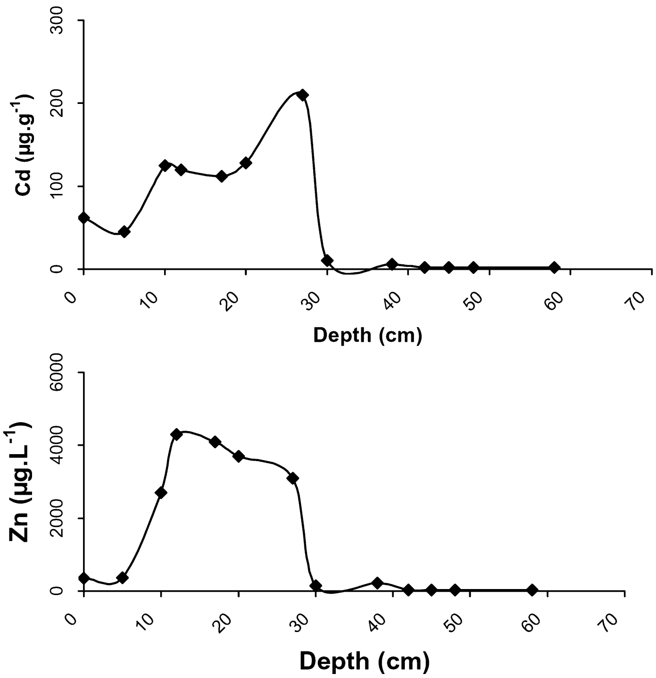


Fig. 4
Distribution of Zn and Cd in bottom sediment cores from a heavily contaminated site in Sepetiba Bay, Madeira Island coast, which receives effluent from a Zn-Cd smelting plant. See location of sampling site in Fig. 1

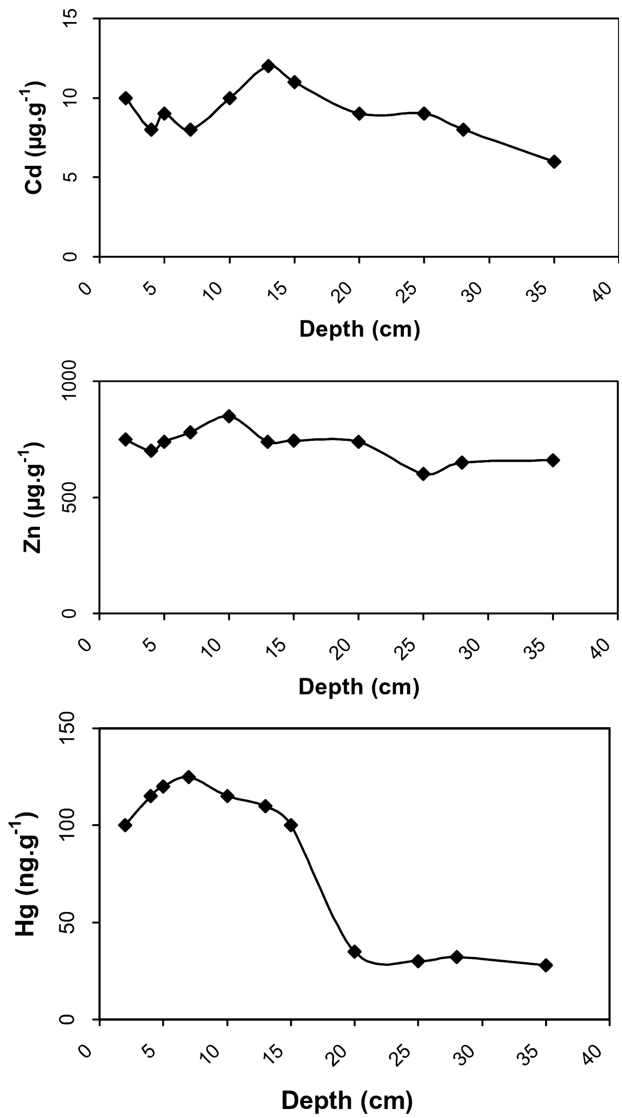


Fig. 5
Distribution of Cd, Zn and Hg in sediment cores form a moderately contaminated site in Sepetiba Bay, Enseada das Garças, which recieves river discharges transported by surface currents. See location of sampling site in Fig. 1

industrial and domestic inputs and pathways of heavy metals. They are estimated considering production technology use, waste treatment efficiency and the regional economy (Nriagu and Pacyna 1988). This approach was used to identify anthropogenic Zn, Cd, Hg and Pb emissions to Sepetiba Bay (Table 2) (Barcellos and Lacerda 1994; Lacerda et al. 1999; Marins et al. 1999).

Emission rates estimates suggest that the main Zn and Cd source to Sepetiba Bay are effluents from the Zn-Cd ore processing plant (Ingá Cia.), which contributed 24 t y^{-1} of Cd and 3,660 t y^{-1} for Zn into the bay's basin, until 1997 when the plant closed. How much of this contribution is still reaching the bay after the closure of the plant is unknown. Large amounts of tailings have been washed into the bay during heavy rains in 1999, and there are calls for urgent plans to manage these wastes. Other activities also dispose considerable amounts of metals, particularly iron and steel, plastic and rubber production (Table 3). Fluvial input is the main Cd and Zn pathway to Sepetiba Bay, followed by direct atmospheric deposition (Pedlowiski et al. 1991a, 1991b; Barcellos 1995).

It is estimated that 386 t y^{-1} of Pb reach Sepetiba Bay from anthropogenic sources through soils, atmosphere and water (304, 65 and 17 t y^{-1}, respectively). Iron and steel production are the main Pb sources to Sepetiba Basin (nearly 60% of total), represented by an important metallurgical plant, COSIGUA, where iron-work uses a lead galvanic bath for iron wires, which is one of their principal products. Zn ore processing contributes to about 17% of the total Pb entering Sepetiba Bay and basin.

Hg has been found in the bay sediments, although there is no notable source within the catchment. Marins (1998) and Marins et al. (1998b) showed Hg input into Sepetiba Bay mainly originated from fluvial discharge and atmospheric deposition, with sewage discharge, municipal solid waste, fertilizer use, plastic and rubber manufacturing, chemical production, metal manufacturing, iron and steel production and oil-fired plants being principal Hg sources (Table 4).

Total industrial and urban emissions of Hg range from 200–364 kg y^{-1}. Major pathways are atmospheric deposition (115–149 kg y^{-1}) and discharges to soils (84–215 kg y^{-1}). Direct inputs to waterways are very small (less than 1.0 kg y^{-1}), highlighting the importance of diffuse sources of Hg to Sepetiba Bay.

Watersheds account for nearly 95% of total Hg input to Sepetiba Bay, while direct atmospheric deposition

Table 2
Major activities that contribute to the total heavy metal loads to Sepetiba Bay

	Number of plants	Production (t year^{-1})	Number of employers
Metal smelting			
Fe	03	1.102.000	4.053
Al	02	98.500	925
*Zn	01	60.000	438 (until 1996)
Manufactures			
Paper	05	534.000	4.041
Chemical	16	176.900	3.887
Metallurgical	19	33.370	4.372
Plastic and rubber	03	30.900	1.722
Food	08	16.700	1.645
Others	13	7.200	1.996
Power station	01	160 MW	250
Fishing and agriculture		10.500	14.000
Harbor and navigation	01	19.000.000	300
Total	72		37.900

Only industries with more than 60 employers are included. (Source: Barcellos and Lacerda 1994).
*Companhia Industrial e Mercantil Ingá was the second most important Zn smelter in Brazil. It closed in 1997, however, around 2 million tons of metal enriched solid wastes are still deposited adjacent to the bay, being regularly leached during the rainy season

contributes with only 5%. Major rivers that flow into the bay were monitored to assess fluvial inputs. The data showed Hg fluxes reaching the bay ranged from 560–740 kg y^{-1}. This measurement was used to calibrate the emission rates and determine the Hg mass balance for the study area.
Significance of metal input through diffuses sources, such as soil leaching, highlights the importance of changes in land use across the basin as a major factor controlling heavy metal mobilization. The main processes are the remobilization of solid wastes by runoff or through percolation through the soil profile. Atmospheric input is an important pathway that has also been identified in monitoring programs because about 90% of the total emission is deposited within 10 km from the source (Marins et al. 1996). Table 5 compares estimates of Cd, Zn, Pb and Hg loads from fluvial and atmosphere inputs. It shows most inputs are of fluvial origin, although significant loads of Zn (29% of total) and Pb (39% of total) enter the bay through the atmosphere.
Comparison of estimated metals emissions with measured loads in Sepetiba Bay open many interesting questions of environmental significance. As expected, most metals emission estimates were higher than fluvial and atmospheric loads measured in situ. Fluvial and atmospheric inputs comprise only a minor fraction of the estimated emissions (10 for Cd, 3 for Zn and 2% for Pb). Only Hg did not follow this pattern and presented smaller emissions than the actual measured input to the bay, suggesting errors in emission estimates or sources not registered by the existing inventory (Marins et al. 1998b, 1999).
Mass balance estimates shows that two times more Hg enters into the bay than was emitted from main sources (Fig. 6). Hg flux that reach the bay's waters ranges from 560–740 kg.y^{-1} while Hg emission were estimated to vary from 200–364 kg y^{-1} (Marins et al. 1999). Additional sources located in Sepetiba Bay basin are unlikely, since survey to date have not identified any other source of Hg different from those listed in Table 2. However, the significant input of water and suspended load from the Paraiba do Sul River basin to Sepetiba Bay is likely to contribute with this additional Hg load.
Waters diverted from the Paraíba do Sul River to Sepetiba Bay may be considered a potential source for Hg and other heavy metals because it flows through one the most industrialized areas of Rio de Janeiro State, the middle Paraíba do Sul River (Fig. 3). Mello (1999) studied the Hg distribution in the Paraiba do Sul River prior to the diversion system and found it to be highly contaminated by Hg.
A mass balance, based on measurements of Hg concentrations in water and suspended particles from the diversion system intake to the mouth of the São Francisco Canal at Sepetiba Bay (see Fig. 3 for location of the river stretch sampled, Molisani et al. 2001), showed that the Paraíba do

Table 3
Emission loads of Cd and Zn from major sources to Sepetiba Bay and Basin in t year^{-1}

Activities	To soils		To atmosphere		To water		Total	
	Cd	Zn	Cd	Zn	Cd	Zn	Cd	Zn
Metal smelting								
Fe	1.0	168	0.33	30	0.1	17	1.43	215
Al	0.03	1.6	0.01	1.1	0.01	0.03	0.05	2.73
Zn	28	6.000	?	**1.200	2	120	30	7.320
Power station	0.05	3	0.02	0.5	0.01	0.6	0.08	4.10
Organic sewage					0.07	20	0.05	20
Solid refuse	0.4	42			0.04	4.2	0.42	46.2
Agriculture	0.01	0.15	0.01	0.1	0.01	0.01	0.03	0.26
Urban runoff	0.3	6			0.03	0.6	0.33	6.60
Harbor and navigation	0.5	95			0.05	10	0.55	105
Manufactures								
Paper	0.3	58	0.01	0.03	0.01	2.4	0.32	60.43
Chemical	0.4	35	0.01	0.2	0.04	0.02	0.42	35.22
Plastic and rubber	0.04	3.9	0.9	8.3	0.01	0.26	0.95	12.46
Metallurgical		10		1.0		1.1		12.1
*Total	31.1	6,432	1.3	241	2.4	176	34.8	6,750

* Values are rounded.
**Until 1997. Sources: Barcellos and Lacerda (1994), Lacerda et al. (1999)

Table 4
Emission loads of Hg and Pb from major sources to Sepetiba Bay and Basin in t year^{-1}

Activities	To soils		To atmosphere		To water		Total	
	Hg	Pb	Hg	Pb	Hg	Pb	Hg	Pb
Metal smelting								
Fe		165	0.1	55	?	8.3	0.1	228
Al	?	0.4	?	0.1	?	0.01	?	0.55
Zn	?	58	?	**7.2	?	1.2	?	66.40
Power station	?	4.4	0.05	1.1	–	0.88	0.05	6.40
Organic sewage					0.02	4.5	0.02	4.50
Solid refuse	0.3	18	?		?	0.5	0.3	18.50
Agriculture	0.01	–					0.01	
Urban runoff		2				0.1		2.10
Harbor and navigation		19				0.95		20
Manufactures								
Paper		29		0.03	?	0.25	?	29.30
Chemical		0.02		0.12	0.01	0.20	0.01	0.34
Plastic and rubber		7.7		1.24	0.01	0.01	0.01	9
Metallurgical	0.02	?			0.04		0.06	
*Total	0.33	304	0.15	65	0.08	17	0.56	386

Sources: Barcellos and Lacerda (1994); Marins et al. (1998b); Lacerda et al. (1999).
* Values are rounded.
** Until 1997

Table 5
Heavy metals inputs to Sepetiba Bay (t year^{-1})

Route/metal	Zn	Cd	Pb	Hg
Atmosphere	56	0.2	3	0.03
Fluvial	144	1.8	4.7	0.72
Total	200	2	7.7	0.75

Source: Lacerda (1983); Watts-Rodrigues (1990); Pedlowiski et al. (1991a, 1991b); Barcellos and Lacerda (1994); Barcellos (1995); Marins et al. (1996); Silva Filho et al. (1998); Marins et al. (1999)

Sul River waters contribute with 30% of the total fluvial Hg load (110 kg y^{-1}) to Sepetiba Bay (Fig. 6). The other 70% may be explained by sources along the stream course, like industrial plants in the Sepetiba Bay basin or domestic sewage input. The study also concluded that 90–99% of Hg is transported in particulate forms during periods with high suspended matter concentrations (>100 mg l^{-1}) during the rainy season period sampled. These results corroborate previous evaluations based on emission factors (Marins et al. 1999) that estimated Hg contribution from the Paraíba do Sul Basin represents about 30–45% of the total fluvial Hg input to Sepetiba Bay (this is roughly similar to the estimate based on the rainy season flux). Differences in metal distributions in bottom sediments suggest that the location of Zn and Cd hot spots is determined by proximity to sources. Zn and Cd hot spots in the northwestern coast (near to Saco do Engenho) lie close to the main source, a Zn-Cd ore processing plant; while Hg and Pb have high values close to rivers mouths. The dispersal and sedimentation patterns of fine sediments from fluvial sources are also an important factor controling metal distribution.

Although there is no study estimating metal export from Sepetiba Bay to the open sea, data on heavy metal concentrations in bottom sediments of the continental shelf in front of the bay's mouth and along its main

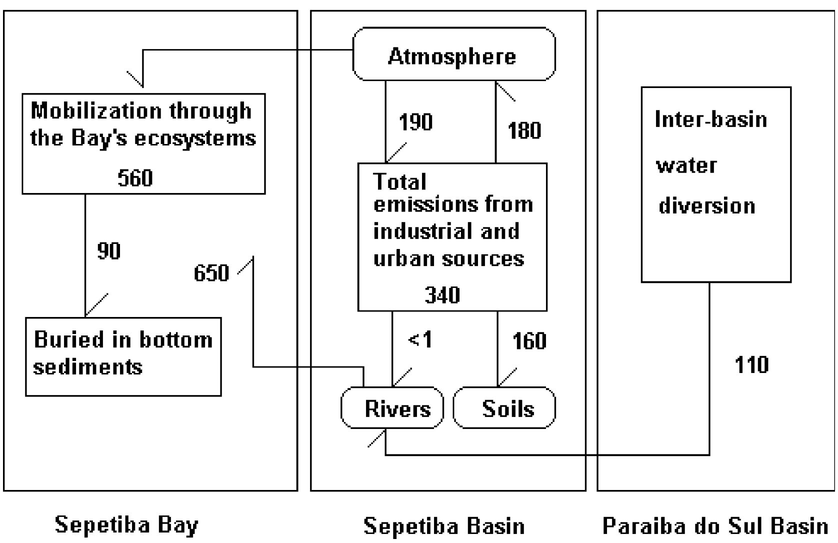


Fig. 6
Mass balance estimates showing the mercury dynamics in the Sepetiba Bay region

axis, suggest that most of the metal loads are kept inside the bay. Mass balance calculations also suggest that after entering the bay metals are not immediately deposited in bottom sediments but suffer various cycles of deposition and resuspension due to relatively low depth, the strong tidal currents and strong wind mostly from the southwest. Zn, Cd, and Pb discharged into Sepetiba Bay, may undergo at least five to ten cycles of deposition and resuspension until being definitively buried in bottom sediments (Barcellos 1995). Marins et al. (1999) suggested that internal burial immobilizes around 90 kg of Hg annually in bottom sediments corresponding to only 15% of total Hg input. Therefore, about 85% (560 kg) of the total Hg input remains mobilized through the bay, being associated with soluble organic and inorganic complexes (Fig. 6) and is available for biological uptake (Marins et al. 2000a, 2000b; Lacerda et al. 2001a, 2001b). Larger Zn, Cd and Pb amounts (up to 60% of the annual input) are buried in bottom sediments (Pedlowisky et al. 1991a, 1991b; Barcellos 1995). Therefore, the "estuarine sink" of Sepetiba Bay is less capable of Hg retention than other metals of environmental importance for the region, enhancing availability for biota and dispersion through other coastal areas and the open ocean.

Pollutant transfer through biological systems

Mangroves are the dominant ecosystem along the Sepetiba Bay shoreline. Many studies (Lacerda 1998) have reported the contamination of Sepetiba Bay mangroves by heavy metals. Since most fisheries in the bay depend on mangroves as nursery grounds and as a food source, mangrove contamination by heavy metals is of high environmental concern.

Zn and Cd enrichment in sediments of Enseada das Garças and mainly in the Coroa Grande mangroves (Figs. 4 and 5) are related to the impact of industrial activities from the Zn-Cd ore processing plant (Lacerda and Abrão 1984; Aragon et al. 1986; Patchineelam and Bezera 1987). In some sites of the Itacuruça mangroves (Fig. 5), Hg shows enrichment of six times over background (Silva 2001).

These studies concluded that mangroves act mostly as sinks for heavy metals. However, changes in physical-chemical conditions associated with tidal water exchanges and the metabolism of mangrove plants may change metal speciation, enhancing their availability for biological uptake. Lacerda et al. (2001b) showed an increase in reactive Hg concentrations in tidal waters washed out from mangroves. Incoming tidal waters are enriched in organic-Hg when contacting with DOC enriched mud flat pore waters and this mechanism has been linked to relatively high Hg concentrations in some fish species dwelling in mangrove influenced areas (Marins et al. 2000b). The effect of root metabolism of *Spartina alterniflora* in mud flat sediments of Sepetiba Bay mangroves also mobilizes deposited Hg through exudation of oxygen that changes redox conditions of pore waters releasing Hg bounded to sulfides (Marins et al. 1997). Lima et al. (1989) also showed the ability of mangrove grass *Spartina alterniflora* to uptake Zn, Cd and Cr from contaminated sediments of the Coroa Grande mangroves increasing their subsequent potential availability to food chains through litter fall.

The potential availability of pollutants are critical mainly in a highly productive ecosystem such as mangroves (Twilley et al. 1986), which support great biodiversity and fisheries of economic importance. Mangroves of Sepetiba Bay are particularly rich in biodiversity, including important food-web zooplankton species, e.g., *Euterpina acutifrons*, *Acartia llijaborghi*, *Oithona ovalis*, and *Pseudodiatomus acutus,* around 20 species of crabs, e.g., *Cardisonma guaihumi, Ucides cordatus*, *Uca* spp., *Sesarma* spp., *Goniopsis cruentata,* and shellfish, e.g., *Mytela guyanensis*; edible bivalves, e.g., *Anomalocardia brasiliana*, *Tagelus plebeius, Macoma constricta* and *Iphigenia brasiliensis*, and gastropods, e.g., *Mellanpus cofeus*. Also many marine organisms spend at least part of their life cycles in mangroves and have expressive economical importance to Sepetiba Bay fisheries, such as shrimps, e.g., *Penaeus schimitii* and *P. brasiliensis*; crabs, e.g., *Callinectes danae* and *Callinects* sp., and a diversity of fishes, particularly mullet (*Mugil curema*), snapper (*Centropomus undecimallis*), sardines (*Sardinella aurita*), and anchovies (*Brevoortia tyrannus*), (Aveline 1980). These organisms use these areas as shelter to reproduce and feed. In doing so their exposure to heavy metals is enhanced.

Studies in Sepetiba Bay showed higher levels of Zn and Cd in benthic organism (algae, mollusks, crustaceans, oysters) than in other coastal areas of Rio de Janeiro State (Lima et al. 1986; Carvalho et al. 1991; Karez et al. 1994). Kurita and Pfeiffer (1991) and Pfeiffer et al. (1985) also showed Cr values in mollusks and fishes above the maximum permissible concentration (MPC) set by Brazilian legislation (Table 6).

Generally, higher concentrations for all metals are observed in mollusks, mainly filter feeders, that concentrate metals in dissolved and suspended forms. *Crassostraea brasiliana* was the organism that showed Zn values up to 80 times greater than the MPC for human consumption. This bivalve has been used as a biological monitor of metal concentration by many authors (Lima et al. 1986; FEEMA 1997), allowing the characterization of the variability of heavy metal contamination through time. Figure 7 illustrates, as an example, the temporal distribution of Zn in *Crassostrea brasiliana* from Sepetiba Bay.

Other mollusks show heavy metal concentrations above the maximum permissible by the Brazilian legislation, such as *Crassotarea brasiliana*, *Littorina angulifera* and *Thais haemastoma* which had enrichments of Cd and *Anomalocardia brasiliana* of Hg (Table 6). These results give importance to the monitoring of those species primarily consumed by local population, since contamination risk is real and immediate.

Among the organisms of economical importance, the shrimp, *Penaeus schimitii,* showed values above of the

Table 6
Cd, Zn and Pb concentrations (μg g^{-1}, wet weight) in organisms from Sepetiba Bay

Organism	Cd	Zn	Hg	Pb	Author
Crassostrea brasiliana (oyster)	1.2–3.3	72–1.258		1.0–1.3	Pfeiffer et al. (1985)
Crassostrea brasiliana (oyster)	0.4–5.1	870–4.030			Lima et al. (1986)
Crassostrea brasiliana (oyster)	<0.1–1.5	1.100–4.460			FEEMA (1997)
Littorina angulifera (leaf-snail)	2.8	1.100		0.8	Carvalho et al. (1994)
Thais haemastoma (rock-snail)	2.8	625		1.7	Carvalho et al. (1994)
Anomalocardia brasiliana (cockle)	0.3–0.6	15–28		0.5–1.4	Pfeiffer et al. (1985)
Anomalocardia brasiliana (cockle)	0.2–0.7	12–20		0.1–0.6	Kurita & Pfeiffer (1991)
Penaeus smithii (shrimp)	0.01–0.6	6.5–288		0.2 –18	Pfeiffer et al. (1985)
Penaeus smithii (shrimp)	0.1	20		1.8	Carvalho et al. (1994)
Penaeus smithii (shrimp)	<0.1–2.2	10–32			FEEMA (1997)
Cardisoma guanhumi (crab)	<0.1–1.4	14–106			FEEMA (1997)
Callinectes danae (crab)	0.2–0.5	24–96		0.8–3.9	Pfeiffer et al. (1985)
Callinectes danae (crab)	<0.1	27.6		<0.1	Carvalho et al. (1994)
Callinectes danae (crab)	<0.1–0.2	26–64			FEEMA (1997)
Anomalocardia brasiliana (cockle)			0.06–0.74		Coimbra (pers.comm)
Micropogonias furnieri (snapper)			0.02–0.25		Kehrig (1995)
Micropogonias furnieri (snapper)			0.01–0.03		Marins (1998a)
Centropomus undecimales (robalo)			0.01–0.02		Marins (1998a)
Paralichthys brasiliensis (flounder)			0.02–0.11		Marins (1998a)

Maximum permissible concentration (MPC) for human consumption by the Brazilian legislation MPC: Cd=1.0; Zn=50; Pb=8.0; Hg=0.5

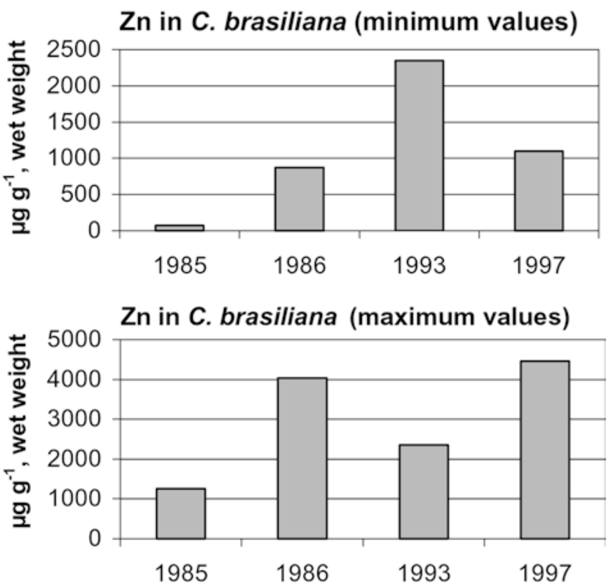


Fig. 7
Temporal variability of Zn concentrations in the oyster *Crassotraea braziliana* from Sepetiba Bay, SE Brazil (μg g^{-1}, wet weight)

maximum permissible for Zn and Pb, at least in one study (Pfeiffer et al. 1985), and for Cd in all studies. The crab, *Cardisoma guanhumi*, also presented concentrations higher than the maximum permissible for Cd and Zn, while *Callinectes danae*, at least, for Zn. However, Hg levels in fishes, such as *Micropogonias furnieri*, *Centropomus undecimalis* and *Paralichthys brasiliensis* are in permissible concentrations. Marins et al. (1998a) found significant correlations between Hg concentrations and fish size in Sepetiba Bay, suggesting methyl-mercury availability and accumulation through food webs.

This scenario of fisheries contamination is alarming because Sepetiba Bay is of immense economical and social importance for fisheries and relying communities, producing around 1,000 to 2,000 t/year (Lacerda et al. 1988b, 1988c). Although the production is distributed among some cities outside of the bay area, most fish are consumed locally increasing human exposure to heavy metals, a situation similar to the scenario observed in some areas, e.g., Hg, in Minamata Bay (Minamata Disease Municipal Museum 1997). In Sepetiba Bay many studies had already stressed the possibility of human contamination by heavy metals, both from occupational and environmental routes (Pfeiffer et al. 1985; Rio de Janeiro/SES 1988; Barcellos et al. 1992).

Conclusion

The environmental change scenario described here is likely to get worse, even considering the strengthening of pollution control policies applied to heavy metal point sources. The constantly high and even increasing concentrations verified in many biological monitors suggest that controlling point sources does not significantly affect heavy metal content in the Sepetiba Bay biota. The large amount of heavy metals already deposited in Sepetiba Bay Basin, the atmospheric transport and deposition of heavy metals from adjacent areas and the diversion of the Paraíba do Sul River are, probably, more significant sources of heavy metals than previously thought. The remobilization of this large reservoir will increase as land use changes accelerate with further urban and industrial development of the region. Augmenting the frequency of dredging due to expansion of harbor activities, deforestation and increasing soil erosion due to land reclamation for urbanization, increasing water demand from adjacent, contaminated watersheds such as the Paraíba do Sul River, among other probable scenarios, will likely become the major factors controlling the input and availability of heavy metals to the local biota. As a result, fisheries and other activities depending on good water quality will probably decrease and the relocation of people involved with these activities will further cause more changes in the watershed.

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