



Decadal and spatial variation of Hg concentrations in sediments of a multi-stressor impacted estuary

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ARTICLE INFO

Keywords:

Mercury
Sepetiba Bay
Sediments
²¹⁰Pb dating

ABSTRACT

The present study has evaluated temporal and spatial mercury trends based on surficial sediment samples and ²¹⁰Pb-dated sediment profiles. The obtained results show that there are areas close to the main bay's tributary rivers where the Hg content has doubled during the last 15 years and regions where it has decreased by a factor of 2, mainly the area close to the navigation channel, which is submitted to periodic dredging. In the inner part of the bay, the most contaminated region, mercury shows a strong association with sulfide. In the same region, based on the ²¹⁰Pb results, it was possible to calculate the yearly increment on the Hg concentration in the surface sediment, 0.62 μg kg⁻¹ y⁻¹ to 0.29 μg kg⁻¹ y⁻¹, according to the distance to the bay's main tributary rivers.

1. Introduction

Sepetiba Bay, in the metropolitan area of Rio de Janeiro, exists in a conflicting situation. While an important recreational and touristic region, where fishery is still a significant economic activity, the bay's watershed harbors several large industries, including two steelwork plants, two commercial ship terminals for containers, and iron ore and coal transport, which require frequent large-scale dredging, and a population of approximately 2 million, served by poor or no sewage and solid waste treatment. These pollution sources contribute significant loads of sediment and toxic metals, in particular Zn, Pb, Cd and Hg. Sediment accumulation rates vary from 160 to 650 mg·m⁻²·yr⁻¹, from a total basin input to the bay of approximately 1 million tons of sediment annually (Molisani et al., 2006). Although no recent inventory is available, estimates of metals discharges in the past decade reach figures on the order of hundreds of tons annually (200, 7.7, 2 and 0.75 tons for Zn, Pb, Cd and Hg, respectively) (Molisani et al., 2004).

Between 2000 and 2005, the Rio de Janeiro state environmental authority (actual INEA former FEEMA) prepared a large environmental survey at Sepetiba Bay, including evaluation of heavy metals in surface sediments (FEEMA, 2006). Possibly due to the high sedimentation rate, as well as the decommissioning of the largest Cd-Zn smelter in the

region, the Inga Metallurgical, one of the major metal (Zn, Cd and Pb) contributors to the bay, a reduction of contamination levels of these metals relative to the late 1980s was observed. This trend was confirmed by biological monitoring of oysters (Lacerda and Molisani, 2006). Mercury was an exception to this trend, with an expansion of areas with sediments containing high Hg content. Veeck et al. (2007) observed that higher Hg concentrations were mostly localized close to river outfalls and related to fluvial sources and inputs, particularly in the northeast portion of the bay, in agreement with what was reported by Paraquetti et al. (2004) and Molisani et al (2007) who associated the Hg input to Sepetiba Bay with the transport of particulate material from rivers reaching the bay.

This paper reevaluates the present-day contamination by Hg of Sepetiba Bay sediments. Additionally, in order to test the Hg association with the organic matter content in sediments, as previously only reported based on statistical correlation, the stable isotope composition of carbon and nitrogen (δ(¹³C) and δ(¹⁵N)) and C/N ratio was determined, to detail this metal-organic association.

2. Material and methods

Bottom sediments were taken using a Petersen grab, stored in acid-

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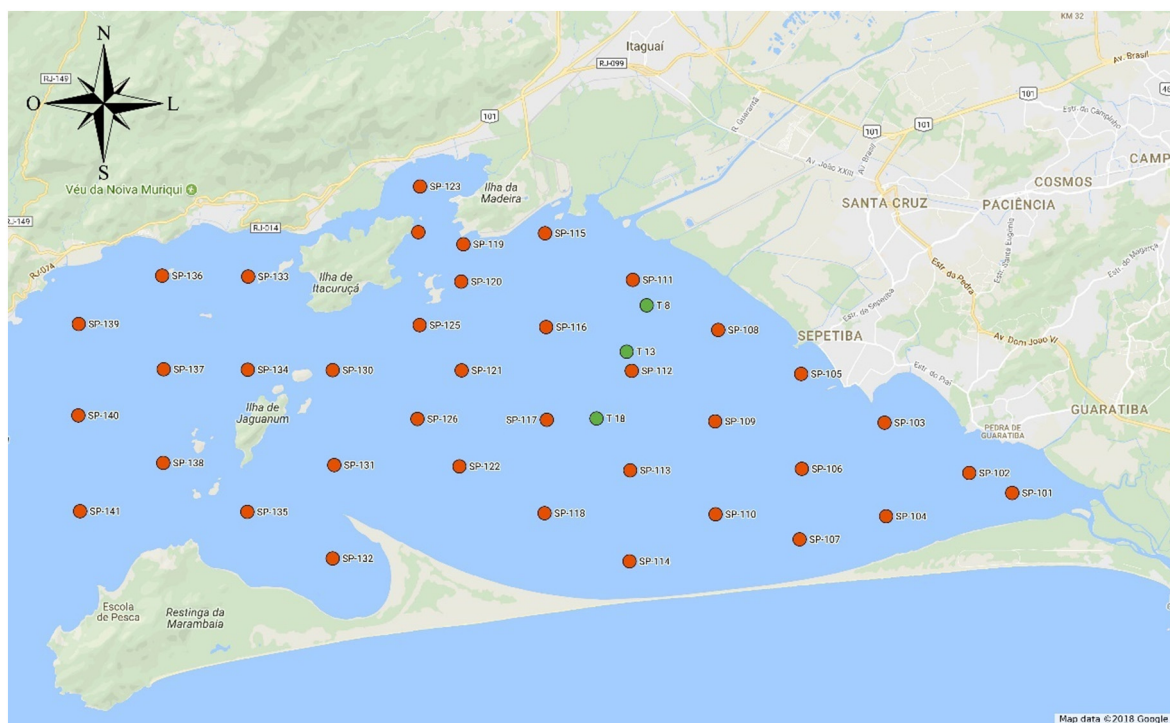


Fig. 1. Sepetiba Bay and the surface sediment sampling points (red dots) and sediment cores (green dots). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

cleaned glass jars, kept cool using a cooler and ice for transport to laboratory, where samples were preserved in a refrigerator (3 °C). Sampling stations are shown in Fig. 1 and follow the same grid established by the Rio de Janeiro State Environmental Agency (FEEMA, 2006) involving 38 stations. Details on Sepetiba Bay hydrodynamics and geographical settings can be found in Cunha et al. (2006).

Half gram wet sediment samples were analyzed for total Hg concentrations by cold vapor atomic absorption, following the procedure described by Limaverde Filho and Campos (1999), in a RA-915 M Lumex Instruments Hg analyzer. The achieved detection limit was $0.002 \mu\text{g g}^{-1}$. Acid volatile sulfide (AVS) was determined following EPA-Method-376.2 and the APHA 4500-S²⁻ colorimetric determination method. Particle size determination was performed using a Gilas 1190 Particle Size Analyzer. All results were corrected for the wet/dry weight ratio. Distribution plots were drawn using Ocean Data View version 4 and the GEBCO1 Southern Ocean-Atl map.

All sediments were initially dried by lyophilization and homogenized, and approximately 8 mg was placed in tin vials to determine total carbon and nitrogen (TC and TN, respectively) and nitrogen isotopic composition. Then, approximately 8 mg was placed in silver vials, and 25 μL of HCl 2.0 mol L^{-1} was added slowly, 3 times, until the reaction finished, to determine organic carbon (OC) content and organic carbon isotopic composition. After that, the vials were dried in an oven at 110 °C (boiling temperature) for 24 h. Samples were then combusted on a continuous flow elemental analyzer (Flash 2000 Organic Elemental Analyzer), which measured elemental concentrations of C and N, coupled to a stable isotope ratio mass spectrometer (IRMS Delta V Advantage, Thermo Scientific, Germany), which measured isotopic composition of C and N. Pee Dee Belemnite (PDB) and atmospheric N were used as standard values for C and N analyses, respectively. The analytical precision was $\pm 0.1\%$ for $\delta^{13}\text{C}$ and $\pm 0.2\%$ for $\delta^{15}\text{N}$, and the accuracy for elemental and isotopic compositions were determined by certified standard (Protein OAS/Isotope Cert 114,859; Elemental Microanalysis).

Sediment dating was carried out on three sediment profiles (T-08, T-13 and T-17; Fig. 1). The sediment cores were sampled by applying a

6.0 cm diameter UWITEC gravity sediment corer, and two-centimeter-high slicing was applied. Lead-210 was determined as described by Godoy et al. (1998), and the sedimentation rate was calculated applying the Constant Rate-Constant Flux (CR-CF) and the Constant Rate of Supply models.

3. Results and discussion

Concentrations of Hg found in surficial sediments from all sampled stations in Sepetiba Bay are listed in Table S1 in the Supplementary Material. There is no Brazilian guideline for sediment quality relative to Hg, and as such, it is typical to apply the limits established by the National Environmental Council (CONAMA) resolution 454/2012, which are based on the “Canadian Sediment Quality Guidelines for the Protection of Aquatic Life”. In this interim sediment quality guideline (ISQG), the Hg concentration limit is $130 \mu\text{g kg}^{-1}$. Considering this limit, only one station, SP-111, had a Hg concentration higher than the ISQG (Table S1).

Descriptive statistics on sediment Hg concentrations are shown in Table 1, with a comparison to previous values reported in the literature. Concentrations are quite variable, since all these previous studies, as well as this one, covered the entire area of the bay. There is no evidence of any temporal trend from the 1990s to the present, at least when considering the entire area of the bay. However, for sediment samples collected from stations close to river mouths, present-day maximum concentrations are nearly double those reported in 1998.

Table 1
Mercury concentrations in surficial sediments from Sepetiba Bay – Descriptive statistics and comparison to the literature (values in $\mu\text{g kg}^{-1}$).

Reference	Mean value	SD	Range	Median
Present work	57	54	0.04–290	62
Veeck et al. (2007)	57	26	18–108	58
FEEMA (2006)	91	36	40–200	100
Marins et al. (1998)	–	–	17–163	–

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