Mercury Distribution and Reactivity in Waters of a Subtropical Coastal Lagoon, Sepetiba Bay, SE Brazil

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As concentrações de Hg total e reativo, de carbono orgânico dissolvido (COD) e carbono orgânico particulado (COP) foram medidas em diferentes massas d’água na Baía de Sepetiba, SE do Brasil, com o objetivo de avaliar mudanças na reatividade do Hg ao longo do gradiente estuarino. As concentrações de Hg reativo foram mais baixas em águas da baía franca (0,09 ± 0,06 ng L\textsuperscript{-1}), seguidas pelas águas influenciadas por rios (0,19 ± 0,06 ng L\textsuperscript{-1}), e pelas de manguezais (0,29 ± 0,02 ng L\textsuperscript{-1}). As concentrações de Hg total foram menores em águas da baía franca (0,38 ± 0,20 ng L\textsuperscript{-1}), seguidas pelas águas de manguezais (0,56 ± 0,01 ng L\textsuperscript{-1}), e foram maiores nas águas influenciadas por rios (0,99 ± 0,29 ng L\textsuperscript{-1}). O percentual de Hg reativo em relação à concentração total foi menor nas águas influenciadas por rios (20,7%), seguidas pelas de águas da baía franca (31,2%), e foram maiores nas águas de manguezais (53,2%). O percentual de Hg reativo em relação à concentração total apresentou significativa correlação inversa com as concentrações de Hg total. Nenhuma correlação significativa foi encontrada entre as concentrações de Hg e a salinidade, o COP e o pH. Entretanto, as concentrações de Hg reativo apresentaram uma correlação positiva, significativa com o COD, mostrando que o controle da reatividade do Hg e, eventualmente, de sua biodisponibilidade na Baía de Sepetiba, deve-se principalmente a formação de complexos com a matéria orgânica dissolvida.

A survey of the reactive and total Hg concentrations in surface waters and of the quantity of particulate organic carbon (POC) and dissolved organic carbon (DOC), were performed along a transect from the major rivers mouths and mangrove dominated areas to the open bay waters of Sepetiba Bay, SE Brazil. The objective of the survey was to evaluate changes in Hg reactivity during the transport along the estuarine gradient. Reactive Hg concentrations were lowest in open bay waters, (0.09 ± 0.06 ng L\textsuperscript{-1}), followed by river influenced waters (0.19 ± 0.06 ng L\textsuperscript{-1}), and mangrove (0.29 ± 0.02 ng L\textsuperscript{-1}). Total Hg concentrations were lowest in open bay (0.38 ± 0.20 ng L\textsuperscript{-1}), followed by mangrove waters (0.56 ± 0.01 ng L\textsuperscript{-1}), and river influenced waters (0.99 ± 0.29 ng L\textsuperscript{-1}). The percentages of reactive Hg relative to the total Hg concentrations were lowest in river influenced waters, (20.7%), followed by open bay waters (31.2%), and mangrove waters (53.2%). The percent of reactive Hg to the total Hg concentrations showed a significant negative correlation with the total Hg concentrations following a potential equation. No relationship between reactive Hg concentrations and POC, salinity or pH were found, however, reactive Hg correlates significantly with DOC, suggesting that organo-complexes, rather than chloro-complexes, are the major controllers of Hg reactivity, and eventually bioavailability in Sepetiba Bay.

Keywords: mercury, Sepetiba bay, speciation, waters

Introduction

Mercury presents a non-conservative behavior during estuarine mixing, due to its particularly high reactivity. Along estuarine gradients, Hg may undergo strong speciation changes, resulting in different bioavailability\textsuperscript{1,2}. Thus, the study of Hg concentrations and of its variability in coastal waters has received large attention due to its direct relationship with fish Hg concentrations. Under estuarine conditions, Hg availability to biota may change due to changes in salinity regime\textsuperscript{3}, redox potential and sulfide concentration\textsuperscript{4}, properties of the dissolved organic matter\textsuperscript{5}, photochemical reactions\textsuperscript{6}, and sediment resuspension\textsuperscript{7}. Previous studies on the partitioning of Hg in surface estuarine waters have shown that from 50% to 80% of
the total Hg concentration at the upper portion of the estuarine gradient are strongly bound to organic matter and/or suspended particles. This characteristic may result in lower bioavailability and more efficient removal from the water column under estuarine conditions. However, during the transport through the estuarine gradient changes in the composition of suspended matter may occur, as well as degradation of particulate (POC) and dissolved (DOC) organic carbon, eventually releasing reactive Hg to the water column.

Changes in the availability of Hg may be more important to control Hg levels in fish than the total Hg content in the water column. In the Sepetiba Bay estuarine complex, an important fisheries area at the Rio de Janeiro coast, this may be the case. A recent survey on Hg content in the snapper *Micropogonias furnieri* showed highest Hg concentrations in fish collected far from major rivers, which are the principal source of Hg to the Bay. The explanation for this apparent paradox may be the increase in Hg reactivity along the estuarine gradient, where Hg faces different physico-chemical environments from different water masses, notwithstanding the decrease in total Hg dissolved concentrations.

To test this hypothesis, a survey of the reactive and total Hg concentrations in surface waters and of the quantity of POC and DOC were performed along a transect from the major river mouths to the open bay waters of Sepetiba Bay. Also, since about 35% of the Bay’s coastline is covered by mangrove ecosystems, an environment with distinct water chemistry, surface waters from a mangrove dominated inlet were sampled.

The objective of the survey was to evaluate changes in Hg reactivity and organic matter characteristics during the transport along the estuarine gradient, from river mouths and from mangrove, to open bay waters, and eventually explain the results found on fish Hg concentrations.

**Material and Methods**

*Environmental setting of Sepetiba Bay*

Sepetiba Bay is a semi-enclosed water body with 447 km² of area, average depth of 6.0 m and tidal range of less than 2.0 m. The Bay is approximately located at latitude 23°S and longitude 44°W, about 60 km south of Rio de Janeiro city (Figure 1). The region is under a hot-humid tropical climate, with mean annual precipitation ranging from 1,400 mm to 2,500 mm, depending on the location along the coast. Nine rivers draining the quaternary plain at the northeastern coast of the Bay are responsible for almost the totality of freshwater inputs, reaching an annual flow of 7.6 million m³. Annual atmospheric precipitation over Sepetiba Bay (0.5 to 10 x 10⁷ m³ yr⁻¹) equals and may even surpass the fluvial inputs, increasing the importance of atmospheric derived pollutants from local smelters.

The lowlands of the eastern coast of Sepetiba Bay, with good transport facilities, cheap and ample land, good fresh water supply and low population density, became interesting to industrial development, further facilitated by the building in the late 70’s of large harbor facilities. Population of the bay has raised from 600,000 people in 1978 to 1.2 million, mostly concentrated along the northeastern shore as a result of the industrial growth. This fast and unplanned development resulted in high contamination of the bay in direct conflict with the necessity of environmental conservation and the sustainable utilization of the bay’s natural resources.

Most trace metal loads, including Hg, to the bay are from fluvial inputs associated with suspended particles. Terrigenous matter are transported through surface currents and preferentially deposited along the NE shore of the bay, forming extensive mud flats covered by mangroves and salt marshes.

*Sampling and analysis*

All sampling and analytical ware were pre-cleaned according to accepted protocols. In two cruises (November, 1997 and March, 1998) surface water samples were collected by divers with 0.5 L pre-cleaned Teflon bottles and Teflon gloved hands avoiding the surface micro-layer, following careful procedures to avoid sampling contamination. Cruises lasted 2 to 4 hours during neap tides only. Care was taken to avoid alterations in water chemistry due to tidal change by not sampling during spring tide and sampling river and mangrove waters during the ebb period and open bay waters during flood periods. The sampling bottles were rinsed three times with the local water.
before being filled. The bottles were capped while immersed in the water to prevent contact with the atmosphere between sampling and handling. Bottles were double bagged in acid-washed plastic bags and transported in an ice box to the laboratory. Two samples from each of a total of seventeen (17) stations were collected for Hg analyses along the estuarine gradient of Sepetiba Bay (Figure 1). All samples were immediately analyzed after collection (within 4 to 6 hours maximum)\textsuperscript{2,16,17,18,19}. During the period between sampling and analysis samples were kept in an ice box.

Unfiltered, non-acidified, sub-samples (250 mL) were analyzed for reactive Hg considered here as the fraction of the total Hg concentration that is reduced and therefore released from solution after the addition of 1.0 mL of acidic (2.6 mol L\(^{-1}\) HCl) 1.0\% SnCl\(_2\) solution\textsuperscript{16,17}. This fraction includes mostly ionic (Hg\(^{2+}\)) plus dissolved gaseous Hg (DGM), but also the Hg fraction weakly bound to inorganic and organic complexes\textsuperscript{2,16,17}. However, it does not include Hg leachable from particulate matter\textsuperscript{18}. Since we did not acidify the samples prior to analysis, our reactive Hg fraction does not compare with the reactive Hg fractions typically reported in literature which used acidified samples\textsuperscript{18,19}.

Total Hg was determined after oxidation with bromine monochloride (BrCl). After oxidation, 0.1 mL of 1\% ascorbic acid solution was used to reduce the excess BrCl; this was followed by a reduction with the same SnCl\(_2\) solution used for reducing the reactive Hg fraction\textsuperscript{20}. Total Hg includes all dissolved Hg species strongly bound to soluble inorganic and organic complexes, and Hg weakly bound to suspended particles.

The Hg\(^0\) resulting from the reduction reaction for water samples was purged from the sample with Hg-free Argon, at a flow rate of 40 mL min\(^{-1}\), for 3 min, and collected on a Au wool trap connected to the outlet of the reaction vessel. Two Au traps in the gas line stripped any Hg from the carrier gas. All connecting tubing was made of Teflon with glass-Teflon connections made of clean silicone tubing. Mercury determination was performed, in a Tekran Model 2500 Cold Vapor Atomic Fluorescence Spectrophotometer. Under the operational conditions described, the analytical detection limit of the method, based on three standard deviations of the average Hg concentrations of the reagents blank, was 0.02 ng L\(^{-1}\). All samples were analyzed in duplicate. Differences between duplicates remained below 15\% for all stations.

From each station during the second cruise (station 10 through 17), 250 mL sub-samples were collected for POC and DOC analysis. Particulate organic carbon (POC) were determined after filtration through a Millipore filter 0.45\(\mu\)m and POC concentrations determined in an Auto analyzer. Dissolved organic carbon (DOC) was analyzed in the filtered sample with a Dorhman DC 80, a wet oxidation analyzer. In this case an aliquot of 4.0 mL was sampled from the 250 mL bottle. Samples were acidified and purged immediately prior to the injection of CO\(_2\)-free nitrogen for inorganic carbon removal. Total oxidation was performed by simultaneous action of UV-irradiation and persulfate oxidation.

During the sampling, some major physical and chemical parameters were also measured \textit{in situ}. Salinity was determined using a SHIBUYA refractometer whereas pH and temperature, were measured with a DIGIMED pH-meter, with a combined glass electrode. Precision of such filed determinations was 0.1 units.

### Results and Discussion

Mercury concentrations of all samples and organic carbon concentrations of the second cruise samples are presented in Table 1. Reactive Hg concentrations were lowest in open bay waters, ranging from 0.04 to 0.22 ng L\(^{-1}\) (average 0.09 ± 0.06 ng L\(^{-1}\)), followed by river influenced waters, ranging from 0.13 to 0.26 ng L\(^{-1}\) (average 0.19 ± 0.06 ng L\(^{-1}\)). Mangrove waters presented the highest reactive Hg concentrations, ranging from 0.28 to 0.31 ng L\(^{-1}\) (average 0.29 ± 0.02 ng L\(^{-1}\)). Total Hg concentrations were also lowest in open bay waters, ranging from 0.08 to 0.66 ng L\(^{-1}\) (average 0.38 ± 0.20 ng L\(^{-1}\)), followed by mangrove waters, ranging from 0.55 to 0.56 ng L\(^{-1}\) (average 0.56 ± 0.01 ng L\(^{-1}\)). River influenced waters presented the highest total Hg concentrations, ranging from 0.44 to 1.30 ng L\(^{-1}\) (average 0.99 ± 0.29 ng L\(^{-1}\)). The percentage of reactive Hg relative to the total Hg concentrations were lowest in river influenced waters, with an average of 20.7\%, followed by open bay waters, with an average of 31.2\%. Mangrove waters presented the highest percentage of reactive Hg relative to the total Hg concentrations (average 53.2\%). Table 2 shows the mean and standard deviations of Hg concentrations in Sepetiba Bay waters.

Figure 2 summarizes graphically the results on the Hg distribution in the three different water masses. Reactive Hg concentration is the best parameter to differentiate water masses, separating clearly the three groups of samples at a P<0.05 level. Total Hg concentrations could only differentiate river influenced water masses (P<0.05), but not open bay waters from mangroves, due to the large variability of total Hg concentrations in open bay waters. On the other hand, the large variability found in the relative concentrations of reactive Hg in open bay waters could not clearly differentiate them from river influenced or mangrove waters.
Total and reactive Hg concentrations were highest close to rivers mouths, decreasing toward the open bay area. This is due to the high rivers water Hg content which has been shown to reach up to 10 times the concentrations measured in the bay 13. These highest Hg concentrations close to rivers mouths may also result from frequent dredging for harbor maintenance and the lower depths of these stations and may probably reflect ressuspension of bottom sediments, which typically present Hg contents ranging from 80 to 140 ng g⁻¹ in this sector of the Bay 13. Similarly, at Narraganset Bay 11, unfiltered water Hg concentrations decreased from fluvial influenced stations to open bay stations. Total Hg concentrations at Narraganset Bay decreased from 30.0 to 7.2 ng L⁻¹ along this gradient, whereas reactive Hg concentrations decreased from 3.7 to 1.1 ng L⁻¹. However, as in Sepetiba Bay, the percentage of reactive Hg relative to the total Hg concentrations also increased seaward 11.

The total Hg concentrations found in Sepetiba Bay are lower than in other coastal areas with significant rivers inputs and point sources of Hg. For example, total Hg concentrations ranged from 2.03 to 6.75 ng L⁻¹ in the Adriatic Sea and was higher close to rivers mouths 7, due to inputs from the mineralized region of Idria and the effluents of a chlor-alkali plant. In inshore marine waters of Australia, total Hg concentration average 15.0 ng L⁻¹, but may increase 2 to 4

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<th>Salinity</th>
<th>Reactive Hg (ng L⁻¹)</th>
<th>Total Hg (ng L⁻¹)</th>
<th>Reactive Hg (%)</th>
<th>DOC (mg L⁻¹)</th>
<th>POC (mg L⁻¹)</th>
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Table 2. Mean and standard deviations and range of Hg concentrations in different water masses from Sepetiba Bay, SE Brazil. (n = number of samples collected and analysed in the respective water mass)
times at strongly contaminated sites\textsuperscript{21}. In 12 coastal lagoons\textsuperscript{17} of Rio de Janeiro, total Hg ranged from 7.3 to 57.0 ng L\textsuperscript{-1}. The lower values from Sepetiba Bay are due to the absence of Hg point sources in Sepetiba Bay basin, which receives this pollutant from atmospheric deposition and runoff from soils\textsuperscript{13,22}. On the other hand the values observed compare well with other coastal areas receiving Hg from diffuse sources only. In the Florida Everglades\textsuperscript{6}, for example, total Hg concentrations ranged from 1.8 to 2.55 ng L\textsuperscript{-1}. When compared to open ocean waters\textsuperscript{8,18,19,23}, where total Hg concentrations generally range from 0.1 to 0.4 ng L\textsuperscript{-1}, Sepetiba Bay waters showed total Hg concentrations 2 to 13 times higher, confirming the importance of anthropogenic sources to the region. On the other hand, unfiltered reactive Hg in the North Atlantic surface waters average about 0.19 ng L\textsuperscript{-1}, whereas total Hg averaged about 0.31 ng L\textsuperscript{-1}. The percentage of reactive Hg relative to total concentrations reached 61\% and was inversely correlated with total Hg concentrations\textsuperscript{23}. Reactive Hg concentrations in open bay waters of Sepetiba Bay are similar or slightly lower than in the open ocean. However, mangrove waters and, to a lesser extent river influenced waters are, in general, higher than those reported for open ocean waters. The greater importance of the reactive fraction in coastal waters relative to the open ocean is probably related to the higher DOC concentrations in coastal areas, particularly in estuaries. Fitzgerald & Lyons\textsuperscript{5} reported 50-60 \% of dissolved Hg in coastal waters as associated with organic matter. Andren & Harris\textsuperscript{3} reported that dissolved Hg at the Mississippi and the Everglades estuarine areas is associated with DOC. In the present study, no relationship between reactive Hg concentrations and DOC, salinity or pH were found. However, reactive Hg correlates significantly with DOC concentrations (Figure 3), confirming the importance of this parameter in increasing Hg reactivity. The highest reactive Hg concentrations obtained from the mangrove stations samples, where maximum DOC was also observed, support this hypothesis.

The percent of reactive Hg to the total Hg concentrations showed a significant negative correlation with the total Hg concentrations ($r = -0.956$). The relationship between these parameters followed a potential equation (Figure 4) with highest percentages, up to 75\% of the total concentration, occurring where lowest total Hg content was present (0.08 ng L\textsuperscript{-1}). Previous results from total digestion of rivers and estuarine waters from Sepetiba Bay showed that bromine chloride oxidation was unable to release all Hg present in water, suggesting that part of the Hg present was associated with clastic material, since aqua regia extraction resulted in higher Hg content\textsuperscript{13,24}. In open bay areas, deposition of suspended matter affect mostly the clastic material leaving mostly reactive Hg. Mercury is not readily exchangeable from POC in the transition between freshwater to sea water\textsuperscript{3}. Therefore, the decrease in total concentrations can be due to the scavenging of clastic particulate matter\textsuperscript{25}. Thus, the relationship found between the percentage of reactive Hg and the total dissolved Hg may be due to the deposition of suspended matter close to rivers mouths and the predominance of ionic and soluble organic-Hg in open bay waters relative to rivers and estuarine waters. This mechanism was suggested to explain the increasing reactive Hg concentrations seaward at Narrangasset Bay, by Vandal and Fitzgerald\textsuperscript{11}.\textsuperscript{11}
Conclusions

Mercury behavior in coastal areas is somewhat unusual, compared to most other heavy metals, due to the diversity of dissolved and particulate Hg species present in the water column. A preliminary budget for Hg in Sepetiba Bay concluded that about 15% of the total Hg entering the Bay is retained within the Bay sediments. At Haifa Bay, this figure reaches only 10%, whereas at Narrangasset Bay, sediment retention reached 50%. These mass balance studies highlight the importance of the Hg present in the water column in opposition to the Hg buried in sediments, as the major component of the Hg biogeochemical cycling in coastal areas.

The lack of correlation between Hg concentrations and salinity and the significant correlation between reactive Hg and DOC found in Sepetiba Bay waters suggest that organo-complexes, rather than chloro-complexes, are the major controllers of Hg reactivity, and eventually bioavailability in coastal waters. Thiol groups are most likely involved in the process, and the highest reactivity of Hg in mangrove waters, typically enriched in dissolved organo-sulfur compounds, confirms this hypothesis. Thus, the increasing reactivity of Hg seaward explains the relatively high Hg concentrations reported for fish caught in those areas.

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