ARTICLE IN PRESS

Ocean and Coastal Management xxx (xxxx) xxx-xxx



Contents lists available at ScienceDirect

Ocean and Coastal Management



journal homepage: www.elsevier.com/locate/ocecoaman

Organic and inorganic contamination in sediments from Araçá Bay, São Sebastião, Brazil

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ARTICLE INFO

Keywords: Polycyclic aromatic hydrocarbons Trace metal Steroids Marine sediments Araçá Bay

ABSTRACT

The aim of this work is to verify the organic and inorganic contamination in Araçá Bay, which contains one of the last remnants of mangrove forest on the northern coast of São Paulo State, Brazil, to improve the environmental management of this area, which is part of the Marine Protected Area of the Northern Coast of São Paulo State (APAMLN) and the Environmental Protected Area of the Alcatrazes Municipality (APAMA). Surface sediments were analyzed to provide information on the levels, spatial trends and sources of trace elements, aliphatic hydrocarbons (AH), polycyclic aromatic hydrocarbons (PAH), and fecal steroids. Low levels of trace elements were observed, and the enrichment factors showed no evidence of contamination in the area. The highest concentrations of all organic contaminants were recorded in the samples collected in the external sublittoral stations, mainly in the São Sebastião Channel, which had the finest grain sizes and higher OC contents. With the exception of the samples collected in the inner part of the bay, most of the samples presented unresolved complex mixtures (UCM) of AH, which are indicators of petroleum and its derivative inputs. PAH concentrations were higher in the external stations, but the levels were below the limits of the sediment quality guidelines (SQG) recommended in Brazilian federal legislation on dredged sediments. The major sources of these compounds were from pyrolytic sources. Sewage contamination was detected mainly in the inner part of the bay, especially near the northern stream and the sewage outfall. The contaminant distributions were related to the hydrodynamics of the intertidal flat, with the most central dynamic areas presenting lower contaminant levels.

1. Introduction

An analysis of different contaminants in sediments is very useful for identifying the extent and main sources of anthropogenic inputs to a particular environment because sediments are a pool of organic and inorganic contaminants. Both hydrocarbons and trace elements are potential environmental contaminants, and some of them may have significant impacts on marine life (Mearns et al., 2016).

Hydrocarbons such as *n*-alkanes, isoprenoid hydrocarbons and PAHs (polycyclic aromatic hydrocarbons) are the main components of petroleum and its derivatives. PAHs also have other anthropogenic sources, including the incomplete combustion of fossil fuels (NRC, 1985). Some steroids can be indicators of domestic sewage. The fecal steroids coprostanol (5 β -cholestan-3 β -ol) and coprostanone (5 α -cholestan-3-one) are produced in the digestive tracts of mammals as a result of the microbial reduction of cholesterol (Murtaugh and Bunch, 1967; Hatcher and McGillivary, 1979).

Trace elements can originate from diffuse sources such as industrial

activities, dredging operations, domestic sewage and port activities (e.g., Al Rashdi et al., 2015; Ho et al., 2010; Wysocka and Vassileva, 2016; Vallejo-Toro et al., 2016). Some trace metals can affect marine organisms, the entire ecosystem and even human health through the processes of biomagnification (e.g., Bargagli et al., 1998) and bioaccumulation (Diop et al., 2015; Guan et al., 2016; Kim et al., 2016). In addition to anthropic activities, trace elements originate from natural weathering and sedimentation processes, which can make sources assessments in environmental studies difficult (Grygar et al., 2013; Wang et al., 2015; Xia et al., 2012).

Araçá Bay contains one of the last remnants of mangrove forest on the northern coast of São Paulo State, and it has high environmental complexity and biological diversity (Amaral et al., 2010; Mani-Peres et al., 2016). These forests are important ecosystems that have been impacted by different anthropogenic activities, including the introduction of hydrocarbons and trace metals (Lewis et al., 2011).

In the adjacent São Sebastião Channel (SSC), there are frequent oil spills and discharges resulting from the presence of the largest oil

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https://doi.org/10.1016/j.ocecoaman.2017.12.028

Received 31 July 2017; Received in revised form 6 December 2017; Accepted 25 December 2017 0964-5691/ © 2018 Elsevier Ltd. All rights reserved.

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terminal in Brazil (Silva and Bícego, 2010), and there is the potential for the continuous leaking of oil due to the operation of the terminal and São Sebastião harbor and of domestic sewage from the outfall of the Araçá, which has been in operation since 1990 and serves an average population of 21,200 inhabitants (Lamparelli and Ortiz, 2007).

Several studies have been conducted in the study area on the levels and effects of hazardous chemicals (e.g., Petracco et al., 2015; Pereira et al., 2007) and from those studies, it was possible to verify an indicator of the effects of pollution in the region. Some of the studies on organic and inorganic contaminants were focused along the SSC, the results of which indicated that the area near Araçá Bay is among the most contaminated area in terms of both petroleum hydrocarbons and sewage due to the influence of the PETROBRAS Maritime Terminal, the harbor of São Sebastião and the Araçá sewage outfall (Medeiros and Bícego, 2004; Silva and Bícego, 2010; Muniz et al., 2015). Thus, the aims of this study are to quantify and evaluate the spatial distributions of trace metals, hydrocarbons and fecal steroids in surface sediments from Araçá Bay to investigate the anthropic processes controlling the sedimentary contaminants and to contribute to the management of the area.

2. Methodology

2.1. Study area and sampling

Araçá Bay is located on the continental side of the SSC on the northern coast of São Paulo State (Brazil) (Fig. 1). It is part of the Marine Protected Area of the Northern Coast of São Paulo State (APAMLN) and the Environmental Protected Area of the Alcatrazes Municipality (APAMA). The area is mainly dominated by tidal currents that efficiently renew its waters with waters from the SSC (Amaral et al., 2010).

Thirty-seven sediment samples were collected from the intertidal (seventeen sampling sites) to sublittoral (twenty sampling sites) areas in Araçá Bay and the deeper adjacent areas, up to depths of approximately 25 m in the surrounding region of the SSC (Fig. 1a). Sampling was undertaken in October 2012 and was performed manually at the shallow sites and with multi-corer equipment at the deeper sites. The sediment samples (top 1–2 cm) were placed in glass containers to avoid external contamination and then stored at -15 °C to be freeze-dried and homogenized.

2.2. Organic contaminant analysis

The analytical procedure for the analysis was based on UNEP (1992) with minor modifications in the adsorption chromatography fractioning step for obtaining the fecal sterols. An amount of 20 g of dry sediment was Soxhlet-extracted with a 50% mixture of residue grade n-hexane and dichloromethane for 8 h. Before extraction, 1-hexadecene, 1-eicosene, naphthalene-d₈, acenaphthene-d₁₀, phenanthrene-d₁₀, chrysene d_{12} , perylene- d_{12} , and 5 α -androstanol were added to all the samples, blanks, and reference material from the National Institute of Standards and Technology (NIST- SRM 1944) serving as surrogates. The extracts were fractionated into F1 (aliphatics), F2 (polycyclic aromatic hydrocarbons), and F3 (steroids) by silica gel-alumina column chromatography. Fraction 3 was evaporated to dryness, and steroids were derivatized to form trimethylsilyl ethers using bis (trimethylsilyl) trifluoroacetamide (BSTFA) with 1% trimethylchlorosilane (TMCS) for 90 min at 65 °C. Aliphatic hydrocarbons and steroids were determined on an Agilent Technologies gas chromatograph 6890 with a flame ionization detector (GC-FID). PAHs were analyzed using an Agilent 6890 gas chromatograph coupled to a 5973N mass spectrometer (GC/MS) in a selected ion mode (SIM). Certified standards at five different concentrations of each group of compounds were used for calculations. PAH identification was based on the retention times of certified standards and their mass/charge ratios (m/z).

For quality assurance and quality control, a procedural blank was analyzed, and the interfering peaks did not coincide with the target compounds. The internal standard recoveries ranged from 64 - 117%. The sediment and blank samples (in duplicate samples) were spiked with a mixture of target compounds. The standard recoveries ranged from 66 - 115%, and the percentage relative deviation to evaluate precision was below 20%. Regular analyses of reference materials for organic compounds from the International Atomic Energy Agency (IAEA-417) produced satisfactory results.

The detection limits (DL), which were defined as three times the standard deviation of the signal with the same retention time as the target compounds in the blanks, ranged from 1.00 to 3.70 ng g⁻¹ for PAHs, 0.010 to 0.028 μ g g⁻¹ for steroids and 0.001 to 0.034 μ g g⁻¹ for aliphatic hydrocarbons.

Some ratios between individual PAHs have been calculated for the identification of PAH origins (Yunker et al., 2002) using the following

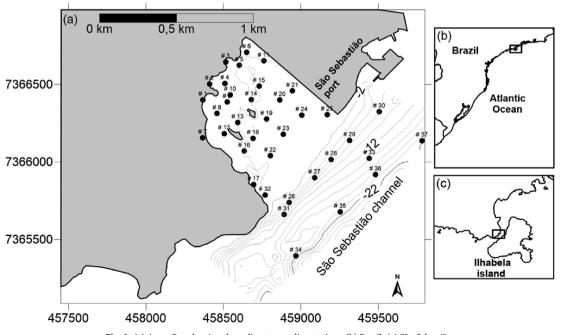


Fig. 1. (a) Araça Bay showing the sediment sampling stations; (b) Brazil, (c) São Sebastião.

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