



Evaluation of anthropogenic inputs of hydrocarbons in sediment cores from a tropical Brazilian estuarine system

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ABSTRACT

Sources and historical deposition of aliphatic and polycyclic aromatic hydrocarbons (PAHs) were investigated in sediment cores from the tropical Mundaú–Manguaba estuarine-lagoon system (MMELS), an important and representative ecosystem of Brazil. The MMELS has approximately 30% of its areas covered of sugar-cane monoculture. Anthropogenic inputs from sugar-cane processing and urban growth can pose environmental problems. The short cores from MMELS were collected in 2007 at four sites: Manguaba Lagoon (C03 and C09), Mundaú Lagoon (C07) and Mundaú River (C08). The analytical method was evaluated for *n*-alkanes and PAHs, the recoveries of the samples demonstrated good accuracy (mostly above 70% recovery) and precision (below 30%). The total aliphatic hydrocarbon (TAH) concentrations ranged from 27.8 to 139.5 $\mu\text{g g}^{-1}$. Only core C07 presented values above 100 $\mu\text{g g}^{-1}$, being considered contaminated by aliphatic hydrocarbon. The presence of the unresolved complex mixture (UCM), hopanes and steranes indicated petrogenic contribution. The characteristics of *n*-alkanes based on the terrigenous/aquatic ratio (TAR), carbon preference index (CPI) and average chain lengths (ACL) showed that the terrigenous input was predominant. The $\Sigma 16\text{PAH}$ concentrations ranged from 29.2 to 222.7 ng g^{-1} , and thus the sediments are considered slightly polluted. The abundance of perylene compared to other PAHs can be used as an indicator of natural origin of this compound. The MMELS cores showed concentrations of perylene greater than 10% of the total penta-aromatic isomers, indicating natural contribution of this compound. The lowest values of $\Sigma 16\text{PAH}$ and perylene were found for core C07, nevertheless the higher values of TAH, UCM, hopanes and steranes were observed. The PAH isomeric ratios reflected a pattern of mainly pyrogenic input (petroleum, coal and biomass combustion). The potential ecosystem risk assessment indicated for PAHs is considered relatively low in MMELS. Despite the industry and the sugar-cane monoculture, the system did not show elevated or alarming data.

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1. Introduction

The different sources of the individual hydrocarbons and their physico-chemical properties determine their fate in the marine environment [1]. Aliphatic and polycyclic aromatic hydrocarbons are associated with particulate and dissolved material and tend to be deposited in the sediments [2–5]. The composition of hydrocarbon compounds found in sediments reflects the relative contributions from different sources: biogenic, diagenetic, petrogenic and pyrogenic [6–9].

The study of hydrocarbons in estuarine systems is immensely important as these areas are highly productive and receive large amounts of pollutants from terrestrial drainage [10]. Estuaries are

partially enclosed water bodies with a restricted access to the sea [11]. Due to their hydrological-geomorphological configuration they retain, transform and accumulate a large fraction of terrestrial borne matter, with one of the material export terms being the ultimate burial in the sediments [12,13].

The hydrocarbons in sediment cores are used for source identification and the reconstruction of the historical records of these hydrocarbon inputs for environmental impact studies [7–9,14,15]. Oil pollution in sediments, and natural petrogenic inputs either from oil seeps or from the erosion of ancient rocks, are indicated by several features in hydrocarbon fractions. These features include the presence of hopanes and steranes, the unresolved complex mixture (UCM) and a lack of predominance of an even- or odd-carbon-number for *n*-alkanes [16]. In addition, the investigation of polycyclic aromatic hydrocarbon (PAH) concentrations in aquatic environments is needed to provide important information on anthropogenic impact on the environment and serve as an indicator of contaminant loading. Characterizing the sources and loadings of contaminants carried by

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rivers is fundamental to understanding the risks posed by human activities to freshwater and coastal marine environments [17].

The Mundaú–Manguaba estuarine-lagoon system (MMELS) is located in the State of Alagoas, northeastern Brazil (Lat. 9°35' S to 9°45' S and Long. 35°44' W to 35°58' W). It consists of two interconnected lagoons, the Manguaba (42 km²) and Mundaú (27 km²), and the mangrove lined channel system (12 km²) linked to the Southwest Atlantic Ocean. MMELS is a typical choked lagoon system characterized by a limited access to the sea, presents long residence times of water, a large potential for material recycling and retention [18]. The annual average fresh water discharge to Mundaú Lagoon by the Mundaú River is 33.5 m³ s⁻¹, to Manguaba Lagoon by the Paraíba do Meio and the Sumaúma Rivers is 17.6 and 5 m³ s⁻¹, respectively. Manguaba is generally oligohaline and Mundaú mesohaline, and both are characterized by eutrophic conditions and dominated by phytoplankton. The climate is tropical, semi-humid with a well-defined dry and rainy season. The average annual mean temperature is 25 °C and the wind regime at the coast is governed by trade winds in summer. In the river catchment area, sugarcane is the dominant land cover, with minor fragments of pristine vegetation, including Atlantic rainforest, mangrove and flood plains. MMELS has been subject to multiple anthropogenic activities, mainly from urbanization, sugarcane agriculture and chemical industries. The Mundaú Lagoon is further impacted by urban effluents from the city of Maceió (approximately 1 million inhabitants). Major sources of pollution for the Manguaba lagoon are cultivation of sugarcane and waste effluents from the sugar and alcohol industries [18–20]. Crop burning activities have been affecting the entire lower drainage basin and also Maceió city [21].

This study aims to provide the temporal trends and sources of aliphatic and polycyclic aromatic hydrocarbons over the past few decades, as well as to evaluate anthropogenic inputs and the potential ecosystem risk assessment from this region.

2. Materials and methods

2.1. Sample collection

The short core samples were collected with a gravity corer at four sites: Manguaba Lagoon (C03, length = 41 cm; C09, length = 70 cm),

Mundaú Lagoon (C07, length = 44 cm; located close to the sewage outlet of Maceió city) and Mundaú River (C08, length = 30 cm) in 2007 (Fig. 1 and Table 1). In the field, the cores were sectioned at 2 cm and C08 at 4 cm intervals and stored at -20 °C. Prior to analyses of the organic extracts the sediment cores were oven dried at 60 °C until constant weight.

2.2. Chemical standards

Perdeuterated standards, pyrene-d₁₀ (98%) and *n*-tetracosane-d₅₀ (98%), were obtained from Cambridge Isotope Laboratories (Andover, USA). *n*-Tetracosane (99%), *n*-octacosane (97%) and perylene (Per) standards were from Sigma-Aldrich (USA), while *n*-eicosane (98%) was from Merck (Rio de Janeiro, Brazil). Stock standard solutions were prepared by weighing 10.0 mg of each standard and dissolving separately in 10 mL volumetric flask with dichloromethane and obtaining a final concentration of 1.0 mg mL⁻¹. These standard solutions were diluted in dichloromethane to give a series of working standard solutions with different concentrations, which were used in the analytical curve.

A reference mixture of 16 PAHs was from Supelco, Bellefonte, USA (catalog no. 48905-U) containing 2000 mg L⁻¹ of acenaphthene (Ace), acenaphthylene (Aceph), anthracene (Ant), benzo[a]anthracene (BaA), benzo[a]pyrene (BaP), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[g,h,i]perylene (BghiP), chrysene (Chry), dibenzo[a,h]anthracene (DBA), fluoranthene (Flt), fluorene (Fluo), indeno[1,2,3-cd]pyrene (InP), phenanthrene (Phen), naphthalene (Nap) and pyrene (Py). The PAH mixture was further diluted with dichloromethane to prepare calibration solutions for GC/MS analyses. Standard solutions were stored at -20 °C.

All solvents used for sample processing and analyses (dichloromethane, hexane and methanol) were chromatographic grade from Tedia Brazil (Rio de Janeiro, Brazil).

2.3. Analyses of grain-size and elemental composition

Sediment grain-size was determined with a particle laser CILAS 1004 analyzer and granulometric distribution curves established with the statistical program Gradstat 4.0. Prior to analyses the samples were treated sequentially for 2 weeks with 10% H₂O₂,

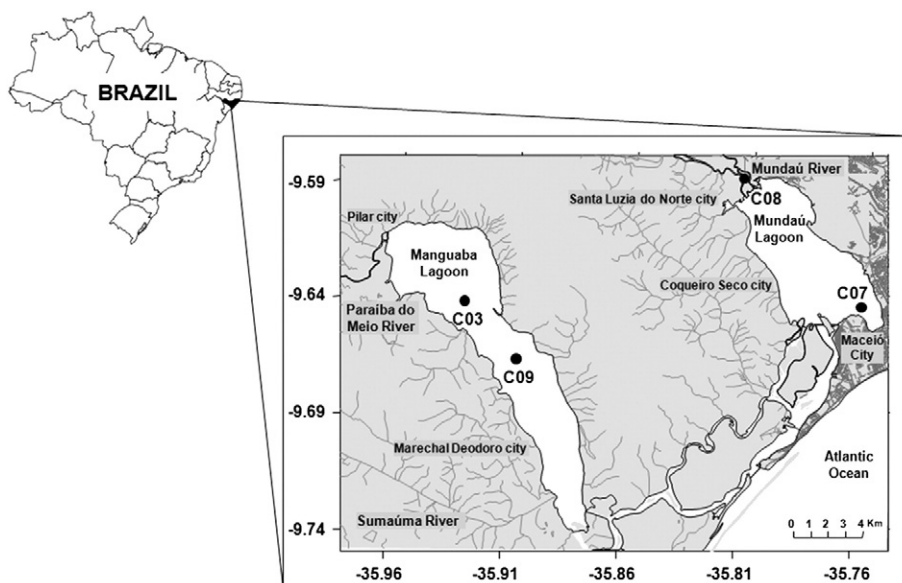


Fig. 1. The MMELS study site with location of core samples collected.

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