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Sources and distribution of polycyclic aromatic hydrocarbons in a an urbanized tropical estuary and adjacent shelf, Northeast of Brazil



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ABSTRACT

Located in Northeastern Brazil, the Capibaribe Estuarine System is an important ecosystem that supplies food and protection for nursery of several species with ecological and economic importance. It is located inside an urbanized area, receives untreated domestic and industrial effluents, and houses some marinas and the Harbor of Recife, which are very important to national and international communities. The distribution and sources of PAHs were investigated in sediments from CES and adjacent shelf. Total PAH concentrations ranged from non-detectable to 497.6 μ g g⁻¹. Pyrolytic sources predominated in most of sites, but petrogenic PAHs were also recorded. The concentration decrease of PAHs toward adjacent shelf suggests that the main source of these compounds is in the upper portion of estuary, where there is an intense discharge of sewage, but atmospheric input also seems to be relevant to the area. The results reinforce the important role of an estuary in contaminant retention.

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Polycyclic aromatic hydrocarbons (PAHs) are among the priority organic contaminants studied, especially due to its wide distribution and high toxicity (Baumard et al., 1998; Magi et al., 2002; Boonyatumanond et al., 2006; Chen et al., 2013; Rahmanpoor et al., 2014). These compounds are lipophilic, easily absorbed by organism tissues (Tuvikene, 1995), and are associated to carcinogenic and teratogenic effects (ATSDR, 1995). PAHs may be originated from incomplete combustion of organic matter under high temperature (pyrolytic), or formed during diagenesis process and formation of fossil fuels under low temperature (petrogenic) (Boehm, 2005; Meire et al., 2007). In addition, some organisms such as plants, bacteria, and fungi can synthesize PAHs, even though in much lower concentrations compared to those processes mentioned above. These compounds may reach the marine environment through several ways, including atmospheric deposition, fluvial runoff, discharge of domestic, and industrial effluents and oil and its derivative spill (Zaghden et al., 2007; Lemos et al., 2014). Among marine ecosystems, estuaries are, generally, more susceptible to receive anthropogenic discharges and have been the focus of several studies to evaluate their contamination (Budzinski et al., 1997; Viguri et al., 2002; Bícego et al., 2006; Wagener et al., 2012; Sharif et al., 2014). The Capibaribe Estuarine System (CES) is located in the metropolitan area of Recife, Northeastern Brazil, and it is formed by the confluence of several tidal creeks, consisting of a natural drainage system. The CES receives continuously the discharge of domestic and industrial effluents, mostly untreated (Nascimento et al., 2003; Somerfield et al., 2003), in addition to several marinas that provide shelter and maintenance to fishing and recreation boats. The CES houses the Harbor of Recife that has a key role within the international community, including Mercosul (Southern Common Market formed by Argentina, Brazil, Paraguay, Uruguay, and Venezuela), North America and European countries (Porto do Recife, 2015). The general cargo handling in 2014 was 1.5 million tons (Porto do Recife, 2015). Furthermore, due to its location, the CES receives atmospheric input of chemicals from industrial activities, fossil fuel combustion, and agricultural burning (sugar cane fields surrounding Recife). There is an estimate of 620,000 cars circulating every day at the metropolitan area (CTTU Companhia de Trânsito e Transporte Urbano, 2015). The Pernambuco State has an important role in the Brazilian agricultural economy, and it is ranked as the fifth largest sugar cane producer. In the past two years, the state produced 13,352 tons of sugar cane (SINDAÇÚCAR, 2015).

Despite its economic, ecological and social role for the local and international communities, there is visible degradation of the estuary, and only a few studies of the extent of contamination have been

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performed (Sericano et al., 1995; Fernandes et al., 1999; Lima et al., 2002; Macedo et al., 2007; Oliveira et al., 2014). This study investigated PAHs in sediments from CES and the adjacent platform, to characterize the spatial distribution and the main sources of these compounds to the area. Due to their physical and hydrodynamic characteristics, estuaries generally act as sediment traps (Wolanski et al., 2006) and thus, this study evaluated the role of the CES in acting as a contaminant retainer.

Fourteen sediment samples were collected in September and December 2011, using a stainless steel "van Veen" grab sampler, distributed along the estuary (#1–10) and adjacent shelf (#11–14). The superficial sediment layer (2 cm) was collected and stored in aluminum containers and kept at -18 °C until the laboratory analysis (Fig. 1).

The extraction of PAHs was performed according to the method described in UNEP (1992). An aliquot of 20 g of sediment freeze-dried was Soxhlet extracted with a mixture of n-hexane/dichloromethane (1:1, v/v), for 8 h. The extracts were purified and fractionated by silica/alumina gel chromatography (5% deactivated) and eluted with a mixture of dichloromethane and n-hexane (3:7, v/v). The PAHs were analyzed in a gas chromatograph (Agilent Technologies 6890), equipped with an HP-5MS fused silica capillary column (30 m, 25 mm i.d. and 0.25 μ m film thickness) and coupled to a HP 5973 N Mass Spectrometer (MS), in the selected ion monitoring (SIM) mode. The carrier gas was H₂ ultra-pure (UP 5.0). The 16 priority PAHs listed by

the US Environmental Protection Agency (US-EPA) were identified (Table 1).

Replicate samples, certified reference materials (IAEA-417, from International Atomic Energy Agency) and procedural blanks were used as quality control procedures. The recovery of the surrogate standard varied between 45% and 104%. The detection limit was obtained from three times the standard deviation of seven replicates of sediment (Table 1). The results are reported on a dry weight basis and they were not corrected for recovery efficiencies.

The granulometric analysis (grain size and distribution) was performed according to Suguio (1973) and the results interpreted using Sysgran 3.0 software. Organic matter (OM) percentage was determined by gravimetry through the differences between dry weight before and after the oxidation of sediment with hydrogen peroxide 10% (Schumacher, 2002).

Data normality was verified using the Kolmogorov–Smirnov test. Pearson correlation was performed between the grain size, OM, and PAHs for each station studied and the Student's t test was used to assess the significance of the same ($\alpha = 0.05$). Student's t test was also used to evaluate differences between PAHs of different molecular weight. Analyses were performed by BioEstat 5.0 software.

The samples had a predominance of silt and clay, except at stations 3, 5, and 9, and samples located on the shelf (stations 11–14) (Table 1). The percentage of OM in sediments ranged from 3.0% to 14.1%, with

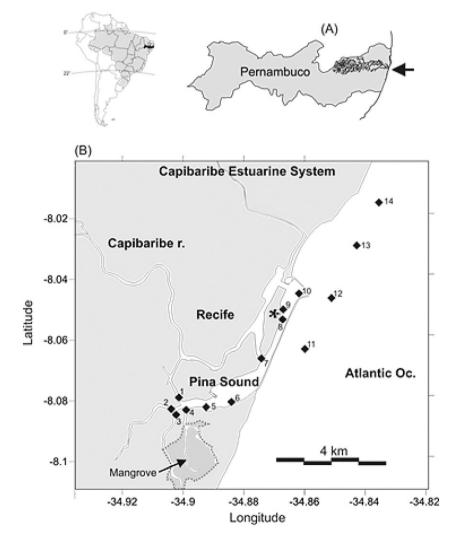


Fig. 1. (A) Capibaribe Estuarine System in South America and the state of Pernambuco. (B) Sampling stations within the Capibaribe Estuarine System (1 to 10) and adjacent shelf (11 to 14). (*) Harbor of Recife. Capibaribe r. = Capibaribe River.

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