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Partition of organochlorine concentrations among suspended solids, sediments and brown mussel *Perna perna*, in tropical bays

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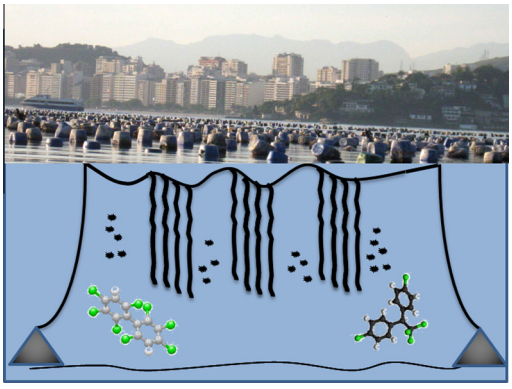
HIGHLIGHTS

- Pesticides and PCB concentrations were monitored in three environmental matrices.
- Suspended solids sampled by trapping method, sediments and mussels were analyzed.
- Sediment showed higher statistical relationships to the bioaccumulated by mussels.

GRAPHICAL ABSTRACT



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ABSTRACT

For evaluating the brown mussel *Perna perna* as a sentinel organism regarding environmental concentrations of organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs), the present study reports original data on the relationship between the concentrations of these chemicals in bottom surface sediments, suspended solids (SS) and concentrations bioaccumulated by this bivalve. Three *P. perna* cultivation areas, located at three bays in southeastern Brazil were used in this study. The three estuaries are under different degrees of environmental impact. Variations in the OCP and PCB concentrations bioaccumulated by the bivalves tended to be similar to those observed in the sediment, but differed from those found in SS. This latter difference might suggest that the SS trapping apparatuses should have been left in place for approximately 60 days (not only 15 days). This longer period would allow the integration of the environmental variability of the OCP and PCB burden adsorbed to this compartment. Authors encourage future studies to evaluate *P. perna* exposure to OCPs and PCBs through the evaluation of sediment concentrations.

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1. Introduction

When adopting a species as a biological model for studying contaminant dynamics in the environment, it is essential to establish the scope and limitations of the chosen tool. In a previous study (Galvao et al., 2012), we evaluated pesticide and Dioxin-like compound concentrations in the soft tissues of mussels (*Perna perna*; Linnaeus, 1758) from commercial cultivation areas in tropical bays. In that investigation, we observed distinct bioaccumulation profiles in mussels sampled in different periods and areas, suggesting their potential use as sentinel organisms for the monitoring of environmental contamination by organohalogen compounds (OHCs) (Galvao et al., 2012). However, the sentinel organisms that show efficiency for this type of approach present simple numerical ratios between concentrations in their tissues and in environmental compartments that act as exposure routes (Beeby, 2001). Organochlorine compounds (OCs), such as organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs), tend to form stable complexes with suspended solids (SS), due to their hydrophobicity (Dachs et al., 1996). This means that SS act as a transfer route of OCs to filter-feeding bivalves (Leadprathom et al., 2009). Sediment is identified as an abiotic compartment that integrates variations in OC levels in the environment (Föstner and Wittmann, 1979), mainly because solids are concentrated in this compartment before being taken into suspension.

To the best of our knowledge, no comparison of OC concentrations has been conducted among the three matrices, i.e., (1) suspended solids, (2) sediment and (3) marine bivalves, originated from the same system. The relationships between the variations in OC concentrations in a species and the same parameter in the corresponding abiotic compartments constitute valuable information. The comprehension of such interactions allows a better assessment of the suitability of the organism as a sentinel for environmental contamination by a given pollutant.

This study aimed to investigate the relationships between the OC concentrations (PCBs and OCPs) bioaccumulated by the brown mussel *P. perna* (Linnaeus, 1758) and those found in suspended solids and sediment from cultivation systems in tropical bays.

2. Materials and methods

Three commercially important bays were studied, comprising Guanabara Bay (GB); Sepetiba Bay (SB) and Ilha Grande Bay (IGB). All of them are located in the southeastern region of Brazil (Rio de Janeiro state). Detailed descriptions of the study area, as well as of the respective sampling sites, can be found in our previous publication (Galvao et al., 2012). Briefly, *P. perna* mussels, bottom surface sediment and suspended solids were sampled from each study site, in December 2008 and February 2009.

2.1. Sampling

Fifteen *P. perna* individuals were sampled from each location. The soft tissue was removed to form composite samples from five individuals, comprising three pools by location and sampling month (18 composite samples in total).

Sediment samples were collected with an Eckman dredge. Only the top sediment layer was removed (<10 cm). Three dredge samplings were conducted in each study area, along mussel longline cultivations. One replica was obtained for each sampling, resulting in three replicates per location. The suspended solid (SS) collectors used in this study were of the sediment trap type, made from Polyethylene Terephthalate (PET) bottles. The bottoms of the PET bottles were removed and a glass tube was attached to each bottleneck, so that the SS collector worked as a funnel. In addition, a weight was attached to the bottles in order to maintain them in an upright position. Fifteen SS collectors were installed at each study site, at a water depth similar to that of the mussel cultivations (1.5–2.0 m), where they remained for 15 days. At the end of this period, suspended solids, sediments and mussels were collected. Composite samples from three to five SS collectors were generated.

Mussel, sediment and SS samples were frozen at −80 °C prior to freeze-drying. Mussels were crushed and homogenized in a mill homogenizer and aluminum cup (MA345 – Marconi®). Only the <74 μm fraction (stainless steel sieve) of the sediment and suspended solids was separated for analysis. Sample flasks were kept under refrigeration (5 °C).

2.2. OCP and PCB quantifications and data processing

The determination of PCB and OCP concentrations followed the procedure described in previous publications (Wang et al., 2009; Galvao et al., 2012). Briefly, the contaminants were extracted from 1 g of the samples in an Accelerated Solvent Extractor System (ASE 200, DionexGmbH), purified on a silica and alumina column, followed by purification on a modified silica gel C18 column. The extracts were injected in a high-resolution gas chromatograph coupled to a high-resolution mass spectrometer (HRGC–HRMS) system (Thermo Finnigan MAT95 or MAT95S, Bremen, Germany). Samples were spiked with internal ^{13}C-labeled OCP and PCB standards before extraction, and recovery standards were added to the extracts before injection into the HRGC–HRMS. The environmental sample results were corrected for blank values and the Method Detection Limits (MDL) were determined as three times the standard deviation of the blank, and, in the cases where blank sample signals were not detected, the signal to noise ratio of 3:1 was used. All data are reported in dry weight (d.w.) basis and they were rounded to three significant digits.

The multiple regression test was applied with the intent of exploring the data and not to establish a direct relationship of cause and consequence concerning the concentrations observed in mussels and the abiotic environmental compartments (sediment and suspended solids). For this, we assume that OCP and PCB concentrations observed in sediment and SS are representative of what is bioaccumulated by the mussels. The multiple regression assays was performed for every target compound, considering mussel concentration as the independent variable and sediment and SS as dependent variable. The adopted significance level was $p < 0.05$ and the statistic package used was the software Statistica 7.0 (StatSoft®). The box plots graphics were achieved using the software GraphPad Prism 5.0 (GraphPad Software Inc.®).

3. Results and discussion

Data on the presence and distribution of PCB and OCP concentrations in these study areas are scarce. Regarding the environmental matrices analyzed in this study, only data on OCPs in mussels (Xavier de Brito et al., 2002) and sediment (Japenga et al., 1988; Souza et al., 2008) were found for GB. Concerning the SB area, only one study on sediment was found (Japenga et al., 1988). To the best of our knowledge, no datum on OCPs and PCBs in sediment, suspended solids or bivalves is available for the IGB area. This limits the possibility of discussion, but highlights the relevance of this study. The OCP and PCB data about the three environmental matrices investigated (mussels, suspended solids and sediments) are presented on tables in supplementary material.

3.1. OCP and PCB concentrations in P. perna mussels

The pesticides hexachlorobenzene, octachlorostyrene, trans-heptachlorepoxide and methoxychlor were below MDL (median