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Baseline

Assessment of organotins and imposex in two estuaries of the northeastern Brazilian coast

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ABSTRACT

Butyltin compounds (BTs) were used worldwide, especially because of their properties as biocides. Due to its high toxicity, the use of tributyltin (TBT) in antifouling paints has been prohibited in most countries. The occurrence and impact of BTs were assessed in surface sediments and in *Stramonita rustica* populations of two tropical estuaries that host major ports in northeastern Brazil. ΣBT concentrations ranged from < LOQ to 542 ng Sn g^{-1} dry weight in sediments while imposex was not observed in *S. rustica*. This is in contrast to previous studies that reported high incidence of imposex at the same sites. Butyltin degradation index indicates recent input of TBT at levels that might trigger imposex in gastropod species more sensitive than *S. rustica*. These results emphasize the need of more rigorous controls by local authorities since Brazil has restricted the use of TBT-based antifouling paints.

In the 1960s, some organotin compounds (OTs) including tributyltin (TBT) began to be widely used as biocides in antifouling paints. Due to their high toxicity to organisms, several countries including the United Kingdom, France, Switzerland, the United States and Japan approved regulatory actions for the use of OT-based antifouling paints in the 1980s (Champ, 2000). In 2008, the convention on the control of harmful antifouling systems on ships (AFS Convention) banned the use of TBT-based antifouling paints all over the world (IMO, 2008). Currently, the AFS convention includes 73 member states representing 93% of the world's merchant fleet (IMO, 2017). Since the convention came into force, some studies have reported a decrease on OT levels in water, sediment and biological tissues at several coastal areas, including South Korea, the Netherlands, Germany, Belgium and the United Kingdom (Choi et al., 2009; Kim et al., 2011; Verhaegen et al., 2012; Law et al., 2012). Conversely, some studies have reported high OTs concentrations in countries such as Brazil, France, Venezuela and Chile, suggesting that TBT-based antifouling paints are still in use at some areas (Oliveira et al., 2010; Briant et al., 2013; Paz-Villarraga et al., 2015; Batista et al., 2016).

The most significant effect of these compounds is associated with endocrine disruption that affects reproduction of marine organisms, especially gastropod mollusks. In these animals, TBT may trigger imposex which is characterized by development of vas deferens and penis in female individuals. At advanced stages, imposex may lead to full obstruction of the pallial oviduct, resulting in sterilization (Bettin et al., 1996). Studies carried out worldwide have shown the effects of these pollutants on several marine organisms (Thain, 1986; Huang and Wang, 1995; McAllister and Kime, 2003; Abidli et al., 2012; Graceli et al., 2013; Batista et al., 2016).

In Brazil, the AFS convention came into force only in 2010 after a law was passed by the Federal Senate (Decree No. 797/2010). However, since 2007 the use of TBT-based antifouling paints is forbidden by the Brazilian Navy (DPC, 2007). Despite local authorities are struggling to control the use of TBT-based antifouling paints, recent assessments along the southern and southeastern coasts of Brazil have detected OTs in sites under influence of commercial ports, marinas and

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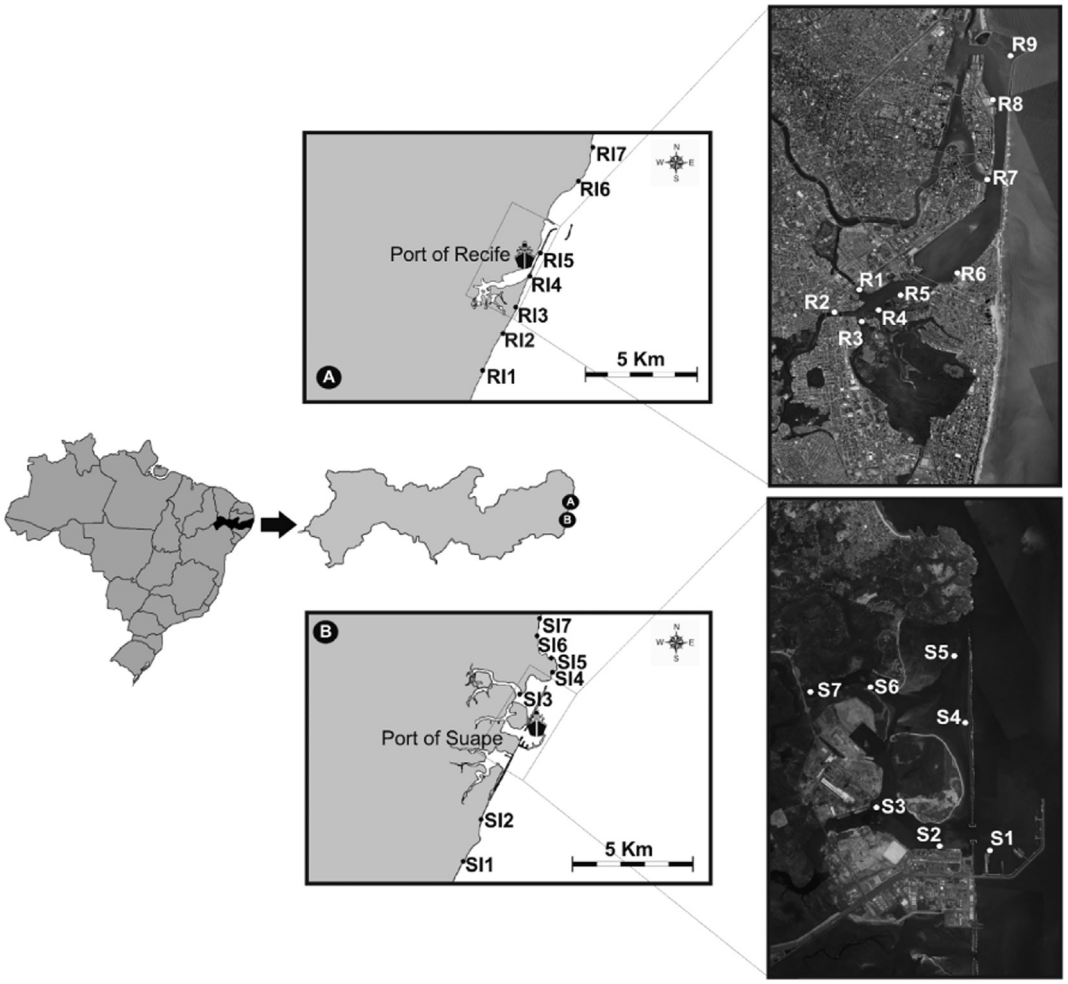

Fig. 1. Study area and location of the sampling sites under the influence of Recife (A) and Suape ports (B), northeastern Brazil. Imposex was assessed at sites RI1 to RI7 (Recife) and SI1 to SI7 (Suape). Sediments were sampled at sites R1 to R9 (Recife) and S1 to S7 (Suape).

shipyards (Castro et al., 2012a; Buruaem et al., 2013; Sant'Anna et al., 2014; Artifon et al., 2016). In addition, studies along the Brazilian coast have reported the occurrence of imposex in gastropods, evidencing that the TBT issue is far from being solved (e.g. Oliveira et al., 2010; Castro et al., 2012a; Borges et al., 2013). In this study, OTs in surface sediments and imposex in *Stramonita rustica* at Capibaribe and Suape estuaries - including adjacent coasts (northeastern Brazil) - were appraised in the same area where imposex had been previously recorded 13 years ago (Castro et al., 2007).

Capibaribe and Suape estuaries are located in Pernambuco State, northeastern coast of Brazil, a tropical area under influence of the intertropical convergence zone (ITCZ). Typically, wet season is observed from March to August while dry season is observed from September to February (Araújo and Pires, 1998). There are two major commercial ports along the coast of Pernambuco: Suape (8° 23′ 39.42″ S, 34° 57′ 53.26″ W) and Recife (8° 3′ 20.03″ S, 34° 52′ 6.22″ W) which are located about 40 km apart (Fig. 1). Both ports receive national and international cargo.

Suape is the largest port along the northeastern coast of Brazil with over 19 million metric tons of cargo loaded/unloaded in 2015 (Porto de Suape, 2015). It is divided in two sections: outer port located offshore (see S1 in Fig. 1) and inner port located inshore (see S2–S3 in Fig. 1). One of the South America's largest shipyards is hosted in the inner port. Several marinas, fishing harbors and the Port of Recife, are located in the Capibaribe Estuarine System (CES), a complex mosaic of channels that drain one of the most populated urban areas along the eastern coast of South America. In 2015, 1.4 million metric tons of cargo was loaded/unloaded in the Port of Recife (Porto do Recife, 2015).

A total of 14 surface sediment samples were collected at Suape estuary, seven in July 2011 (wet season) and seven in February 2012 (dry season), while nine were sampled at CES in December 2011 (dry season) (Fig. 1). Samples were collected with a stainless steel sediment grabber, stored in previously combusted (450 °C for 4 h) aluminum containers, and frozen (− 18 °C) until analysis.

Up to 30 adult individuals of the gastropod *S. rustica* were manually collected in September 2012 at seven coastal sites near the Port of Suape and seven near the Port of Recife (Fig. 1). Samples were collected from the same sites where imposex had been previously reported (see Castro et al., 2007). After collection, animals were transported to the laboratory, narcotized with $MgCl_2$ solution (3.5% in a mixture of distilled water and seawater, 1:1, v/v), and shell lengths were individually measured with a calliper. Shells were then opened and soft parts were removed. Sex identification was confirmed by the presence of prostate gland in males, and albumen gland, sperm-ingesting and capsule glands in females. Penis length of males and imposex-affected females was also measured using a digital calliper. Imposex levels were assessed according to the protocol described by Gibbs and Bryan (1987) and modified by Fernandez et al. (2005) to be used in *Stramonita* spp. The following indices were calculated: percentage of imposex in females (I %), female penis length index (FPLI), relative penis length index (RPLI), and vas deferens sequence index (VDSI). Although VDSI scale was originally developed for assessing imposex in *Stramonita haemastoma*, it is also valid for *S. rustica* due to phylogenetic similarities between both species (Castro et al., 2012a).