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Organotin contamination in South American coastal areas

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Abstract Organotin compounds (OTs) were used in antifouling paints for more than four decades. However, due to their widespread intensive use and high toxicity, undesirable effects in non-target marine organisms have been detected since the early 1980s. Consequently, the International Maritime Organization banned new maritime applications of these products on January 1, 2003 and their presence on ship hulls from January 1, 2008. Although extensively studied in Europe, North America, Oceania, and Asia, environmental levels and effects of organotin contamination are still poorly known for South America. Thus, the current review aimed to present the actual status of this problem in South America by summarizing and comparing the available data in the literature. An overview of the OTs concentrations in sediment and biota and their effects, mainly imposex in marine gastropods, are presented. This work showed that in Atlantic coastal areas of South America there are "hot spots" of OTs contamination, similar to that observed in industrialized countries of Northern Hemisphere. On the other hand, the number of accomplished studies in the Pacific coast is extremely low. Despite the limitation on studies about OTs environmental levels and their related effects, the available data pointed out for a widespread TBT contamination along the South American coastal areas. Therefore, the establishment of baselines of organotin contamination in the Pacific coast and the implementation of temporal trend studies in the South American coastal areas is crucial to verify the effectiveness of local regulations and OTs global ban, and to map the most sensitive areas related to present and future antifouling impacts.

Keywords TBT · South America · Contamination · Environment · Review

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Introduction

Antifouling paints have been used in structures exposed directly to seawater, including hulls of ships, aquaculture nets, offshore structures, and ducts (Champ 2000). According to Kotrikla (2009) the purpose of using antifouling paints are: (a) decreased frictional resistance between water and ship hulls, reducing fuel consumption; (b) decreased frequency of dry-docking, which results in lost of time and increase in maintenance costs; (c) reduction of ship hull corrosion and minimizing

the introduction of non-native species in the marine environments. Before the emergence of TBT-based antifouling paints, the world fleet had used paints containing copper or zinc oxides, which, however, had low durability (12 months; Almeida et al. 2007; Godoi et al. 2003a). Hence, more efficient antifouling paints became necessary.

Organotins compounds (OTs), particularly tributyltin (TBT) and triphenyltin (TPhT), have been used for several purposes for a long period of time. The industrial applications of OTs have been known since the 1920s, when they were used as fluid in transformers and capacitors. Their biocidal properties were discovered by the International Council of Researches on Paints during the 1950s, thereafter these compounds were commercially used in fungicides, acaricides, and other pesticides. In the elapsing 1960s, the OTs were widely used on many other products, such as stabilizers in the production of polyvinyl chloride and catalysts for several industrial processes. The use of organotin as active ingredient in antifouling paints have began in the 1960s (Clark et al. 1988; Godoi et al. 2003a), but became common in the 1970s (Almeida et al. 2007; Yebra et al. 2004).

The first antifouling paint based on organotin have simply mixed TBT to the paint (technology of soluble matrix), providing a very quick initial biocide release. However, these products were effective for up to 15 months (Almeida et al. 2007). Afterwards, TBT was used in self-polishing paints, compatible with steel and aluminum hulls. Such paints were based on acrylic copolymer with TBT groups bonded to the main polymer chain by ester bonds, in which the polymer is soluble in sea water. Since the dissolution can be controlled at a molecular level, it is possible to obtain a well-known self-polishing effect in that paint. As a result, these paints supplied an effectiveness of up to 7 years, releasing about 4 $\mu g/cm^2$/day of TBT in the seawater (Swennen et al. 1997). Due to its efficiency, in 1999 approximately 70% of all commercial shipping was protected by TBT-based paints, achieving direct savings of close to US$ 2,400 million a year in fuel and other costs (Almeida et al. 2007; Clark et al. 1988). Estimates showed that approximately 50,000 tons of organotin compounds were produced per year between 1990 and 2003 (Godoi et al. 2003a).

Due to its intensive use, several environmental problems involving organotin and undesirable effects in non-target marine organisms have appeared at the beginning of the 1980s. The first environmental damage related to antifouling TBT-based paints was reported in oyster cultures (*Crassostrea gigas*) from Arcachon Bay, France. This study observed that TBT caused a decrease in the number of juveniles recently fixed, and induced abnormalities in the larvae and shell malformations ("balling") in adults (Alzieu et al. 1986; Dyrynda 1992). Studies in cultivations of *Mytilus edulis* (Beaumont and Budd 1984), *Pecten maximus* (Davies et al. 1987, 1997; Paul and Davies 1986), *Perna viridis* (Kan-Atireklap et al. 1997), and the gastropod *Chorus giganteus* (Gooding et al. 1999) around the world have also reported pernicious effects related to TBT exposure. For the most, it has caused growth inhibition and increasing mortality rates during cultivations. Other environmental deleterious effects of organotin compounds have been reported worldwide, including imposex and population decline in gastropods, and immunosuppressant effects in marine mammals (Alzieu et al. 1986; Bryan et al. 1986; Beaumont and Budd 1984; Kannan et al. 1997). In fact, several other studies have reported harmful effects related to the environmental contamination by organotin in Europe (Barroso et al. 2000), North America (Evans et al. 2001), South America (Limaverde et al. 2007; Fernandez et al. 2005), Asia (Sudaryanto et al. 2000), and Oceania (Kannan et al. 1995; Wilson et al. 1993).

As a result of those environmental problems, since 1987 TBT-based antifouling paints have been restricted in many countries (Champ 2000), and in September of 2008, they were banned through the Convention on the Control of Harmful Antifouling Systems on Ships adopted by the International Maritime Organization (IMO 2008). However, it is well known that TBT-based antifouling paints are still widely used in several (mainly developing) countries (Shi et al. 2005). Specifically for South America, Bigatti et al. (2009) have mentioned the actual use of TBT-based antifouling paints in Argentina. The situation is not very different for the Brazilian coastal areas, where antifouling paints are still using TBT in formulations. Since pure TBT oxide

and chloride are freely produced and marketed in Brazil (CESBRA 2011), different formulations have been prepared mainly by amateurs such as fishermen and owners of recreational boats in small shipyards and private marinas.

Therefore, the current review aimed to assess the actual status of environmental organotin impact in South America based on the data available in the literature. An overview of the OTs concentrations in sediment and biota and their effects, mainly imposex in marine gastropods, throughout coastal areas of South America was presented and compared with worldwide results.

OTs contamination in abiotic matrices

Once released into the water from an antifouling coating, OTs are rapidly absorbed by organisms such as bacteria and algae, or adsorbed to suspended particles or dissolved organic matter (Gadd 2000; Konstantinou and Albanis 2004). Under favorable conditions, OTs may degrade through successive dealkylation producing dibutyltin or diphenyltin (DBT or DPhT), monobutyltin or monophenyltin (MBT or MPhT), and, ultimately, inorganic tin and becoming progressively less toxic in the process (Watanabe et al. 1992). The degradation of OTs through dealkylation to tin occurs via biotic and abiotic reactions (Maguire 1984). This mechanism of degradation is accelerated by UV radiation, increasing temperature, and biological activity, the latter being of greater importance (Clark et al. 1988). However, the biotic mechanism of OTs degradation is still not well understood (Gadd 2000).

In oxidized sediments, the degradation of OTs occur within a few weeks (Clark et al. 1988), whereas in cold and anoxic environments is significantly slower, ranging from 2 years to decades (Dowson et al. 1996; Clark et al. 1988; Mora and Phillips 1997). In those circumstances, the sediments (mainly the anoxic ones) can act as reservoirs of OTs and, due to their high affinity to particulate matter, act also as a source of these compounds back to the water column following processes (physical and/or biological) of remobilization (Axiak et al. 2000; Hallers-Tjabbes et al. 2003). Since anoxic sediments are predominant in areas with limited circulation (i.e., bays or estuaries; Michaud and Pelletier 2006), the very same area where ship activities are more intense, the processes involved in degradation and remobilization of OTs must be better understood to further assess the extent and intensity of contamination.

Organotin residues were recently detected in sediments from several areas associated with ship traffic in Europe (Chiavarini et al. 2003a; Nogueira et al. 2003; Díez et al. 2002, 2006), Asia (Harino et al. 2007; Nhan et al. 2005), Oceania (Haynes and Loong 2002), and North America (Landmeyer et al. 2004). In South America, ten studies determined organotins in sediments from 25 different sites associated to maritime activities (Fig. 1). The highest butyltin concentrations detected in surface sediments of South America ranged from a few (Todos os Santos Bay, Brazil) to 6,500 ng Sn g^{-1} (Mar del Plata, Argentina), which are comparable to other worldwide determinations (Fig. 2).

Regarding South America, only two recent studies have determined butyltins (BTs) in surface sediments from the Pacific coast. BTs were analyzed in six sites inside the Gulf of Guayaquil (Ecuador; Castro et al. 2011) and seven sites in San Vicente Bay (central Chile; Pinochet et al. 2009). In Ecuador, the values ranged from 12.7 to 99.5, 1.8 to 58.4, and 43.9 to 339.9 ng Sn g^{-1} dw, whereas in Chile, from 14 to 1,560, <20 to 1,170, and <27 to 470 ng Sn g^{-1} dw for TBT, DBT, and MBT, respectively. In both studies, the highest concentrations were found close to the harbor terminals.

In the Atlantic coast, Delucchi et al. (2006) analyzed six surface sediments (Villarino Viejo, Puerto Cuatrenos, Maldonado, Puerto Galvan, Puerto Ingeniero, and Puerto Belgrano) at Blanca Bay estuary (Argentina) and found a wide range of TBT (ranging from <1.1 to 3,227 ng Sn g^{-1} dw) and DBT (ranging from <0.2 to 1,645 ng Sn g^{-1} dw) levels. Puerto Galvan presented the highest values of TBT in the inner zone of the estuary, which is the most active harbor in the area. The results of this research suggested an intensive use of TBT-based antifouling paints on ships that navigated along the estuary. Additionally, the authors stated that ships careenage in dry docks inside the

Fig. 1 Areas assessed for imposex in gastropods (▽) and, organotin contamination in sediments (▬) and marine mammals (▲) of the South American coast (*asterisk* ongoing studies)

estuary may be one of the main sources of TBT in local sediments.

High TBT concentrations were detected in water and sediments of 12 sites from Mar del Plata and Mar Chiquita (Argentina; Goldberg et al. 2004). This study presented one of the highest TBT concentrations found so far for sediments (ranging from <80 to 6,500 ng Sn g^{-1} dw) and water (ranging from <80 to 8,000 ng Sn L^{-1}) of South America. These elevated concentrations in water and sediment indicated recent inputs of TBT in the area and suggest these areas as hotspots for OTs contamination in South America.

Another recent study conducted in Argentina analyzed TBT concentrations in sediment samples and correlated with imposex incidence in 12

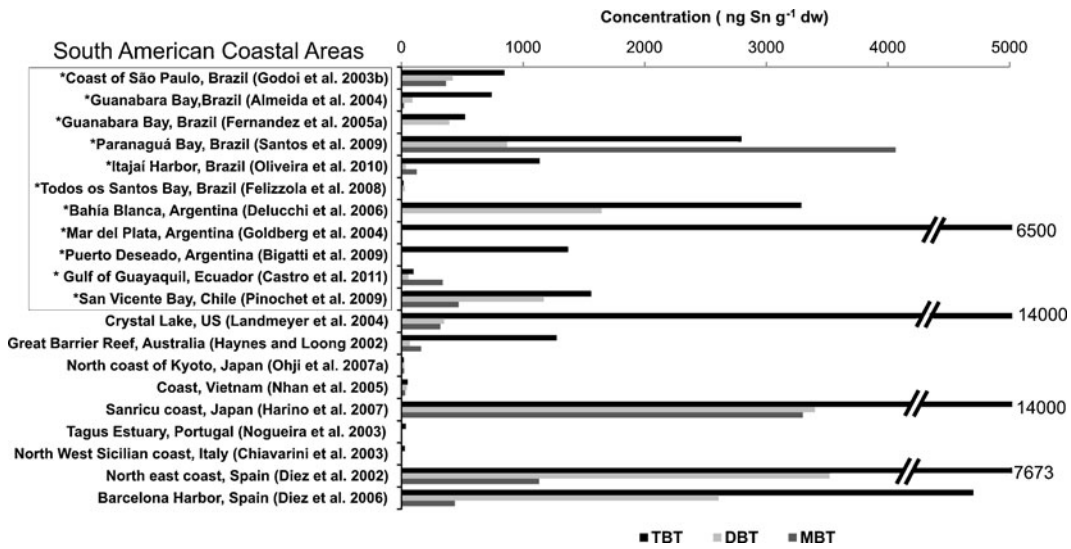

Fig. 2 Highest butyltin concentrations (ng Sn g^{-1} dw) detected in surface sediments of South America in comparison to other worldwide determinations

gastropod species from 40 localities along 4,700 km of Argentinean coastline (Bigatti et al. 2009). The localities with higher levels of TBT contamination were Mar del Plata harbor (728 ng Sn g^{-1}) and Puerto Deseado harbor (1,370 ng Sn g^{-1}), whereas low values were found in San Antonio Oeste (33.3 ng Sn g^{-1}), Punta Cuevas (1.9 ng Sn g^{-1}), Caleta Cordova (5.1 ng Sn g^{-1}), and Caleta Sara (5.6 ng Sn g^{-1}) located at the coast of Patagonia.

Six studies have analyzed organotins in sediments from areas under the influence of important harbors from the southern (São Francisco do Sul, Itajaí–Navegantes, and Imbituba (Oliveira et al. 2010); Paranaguá Bay (Santos et al. 2009); southeastern (Coast of São Paulo (Godoi et al. 2003b)); Guanabara Bay (Fernandez et al. 2005; Almeida et al. 2004); and northeastern (Todos os Santos Bay (Felizzola et al. 2008)) Brazil.

Butyltin compounds were determined in 17 samples of surface sediments collected from areas under the influence of three harbors (São Francisco do Sul, Itajaí–Navegantes, and Imbituba) in the coast of Santa Catarina state (southern Brazil). The BTs concentrations ranged from <130 to 1,137 ng Sn g^{-1} dw for TBT, <160 to 394 ng Sn g^{-1} dw for DBT and <25 to 312 ng Sn g^{-1} dw for MBT. The highest concentration of total BTs was found at Itajaí river dockyard, indicating intense inputs of organotin-based antifouling paints to the environment. However, the occurrence of heavy contaminated sites was observed to be relatively close to places with low to moderate BT levels in the three studied estuaries, which may be related to local sources such as the shipyards and the small boats that were still using TBT-based antifouling paints. Substantial amounts of the degradation products (DBT and MBT) were also determined in those studied areas, indicating that natural attenuation is contributing to the sediment remediation of these areas (Oliveira et al. 2010).

Additionally, TBT levels up to 2,800 ng Sn g^{-1} were found in sediments of Paranaguá Bay, whereas MBT levels were even higher (up to 4,000 ng Sn g^{-1}) in most of the 20 studied sites along the Bay (Santos et al. 2009).OTs levels up to 700 ng L^{-1} in suspended particulate matter (considering the water concentration of particulate matter) were also found in this study. These results indicated that Paranaguá Bay is the most contaminated area by organotin compounds among the studied areas throughout the Brazilian coastline. According to the authors, the high concentrations detected were related to the presence of two important harbors inside the bay

(Paranaguá and Antonina harbors) and also to the pattern of local circulation and tidal action. In addition, 17 out of 20 sediment samples were collected inside the navigation channel, and all samples were taken in July 2006 (before the TBT banishment). There were no regulations on TBT use and the control on antifouling paints use was virtually absent in Brazil at that time. The predominance of metabolites over the parent compound might be related to the favorable biotic and abiotic environmental conditions to degrade TBT in the sediments.

Organotin concentrations (TBT, DBT, and MBT) were analyzed in sediments of five different areas along the coast of São Paulo state: Ubatuba, São Sebastião, Guarujá, Santos, and Cananéia (Godoi et al. 2003b). Concentrations ranged from 17 to 847 ng Sn g^{-1}, for TBT, 14 to 421 ng Sn g^{-1} for DBT and 36 to 367 ng Sn g^{-1} for MBT, whereas the highest TBT levels were found in areas nearby marinas and harbors (i.e., Santos and Guarujá).

Guanabara Bay (Rio de Janeiro state) constitutes the most dramatic example of man-made degradation along the Brazilian coast (Almeida et al. 2004). The estuary houses the second most important commercial harbor in Brazil with 1,700 ships/year (CDRJ 2000). In addition to harbor activities, there are several shipyards, two naval bases and many marinas within the bay (Kjerfve et al. 1997). Two independent studies analyzed organotins in sediments of Guanabara Bay (Fernandez et al. 2005; Almeida et al. 2004). Almeida et al. (2004) studied butyltin speciation in five sediment cores sampled from different sites inside Guanabara Bay. Total tin concentrations were also analyzed to verify the final point of dealkylation. The TBT concentration in surface sediments ranged from 14 (environmental protection area) to 742 ng Sn g^{-1} (vicinity of a major shipyard). According to the authors, the space–time distribution of organotin species in the sediments of Guanabara Bay was very irregular and there is little evidence that extensive degradation has occurred after sedimentation. Fernandez et al. (2005) examined organotin concentrations in nine sites inside Guanabara Bay (Guapimirin, Paquetá island, D'agua island, Niterói mooring site, Ishikawajima shipyard, Niterói ferryboat, Rio de Janeiro harbor, Glória marina, and Botafogo inlet). The concentrations ranged from 10 to 520 ng Sn g^{-1} for TBT, <4.5 to 394 ng Sn g^{-1} for DBT, <3.9 to 39.4 ng Sn g^{-1} for TPhT, and <4.5 to 18.7 ng Sn g^{-1} for MPhT, while MBT and DPhT were not detected in this study. This was the first study to analyze phenyltins in the Brazilian aquatic environments. These results revealed that Guanabara Bay can also be considered as a "hot-spot" for TBT contamination in the South American coast.

With an area of the approximately 1,000 km^2, the Todos os Santos Bay is the largest bay in the Brazilian coast. Located in the northeastern region, Todos os Santos Bay has witnessed an increasing industrialization and a subsequent increase of shipping activities in the area since the 1950s. Felizzola et al. (2008) have analyzed OTs in 17 surface sediments of Todos os Santos Bay and have observed TBT contamination in all of the sites. The organotin concentrations observed in that study ranged from <5.4 to 15.9 ng Sn g^{-1} of TBT, while DBT and MBT were detected only in 1 out of 17 sampled sites (Bimbarras Sul, DBT = 28.8 and MBT = 4.0 ng Sn g^{-1}). These results were the lowest found so far for sediments from the Brazilian coast (Fig. 2), which could be related to the shallow (average of 6 m), highly transparent (relatively low concentration of suspended matter, 2.15–10.07 mg L^{-1}) and turbulent (caused by winds and tidal currents) water of Todos os Santos Bay. Thus, Todos os Santos Bay can be considered as a low to moderately impacted area for organotin contamination. According to the authors, the virtual absence of DBT and MBT in sediments of Todos os Santos Bay is probably due to the fact that the TBT found in paint particles is not easily degraded. However, considering the intense hydrodynamic conditions in Todos os Santos Bay, it is more likely that organotin compounds are rapidly degraded, which resulted in DBT and MBT concentrations below the detection limits of the analytical method used.

Although South America is constituted of developing countries and, therefore, is supposedly less industrialized, levels of organotins observed in most of the studied areas sediments were at a similar magnitude of those observed in the developed and highly industrialized countries, such

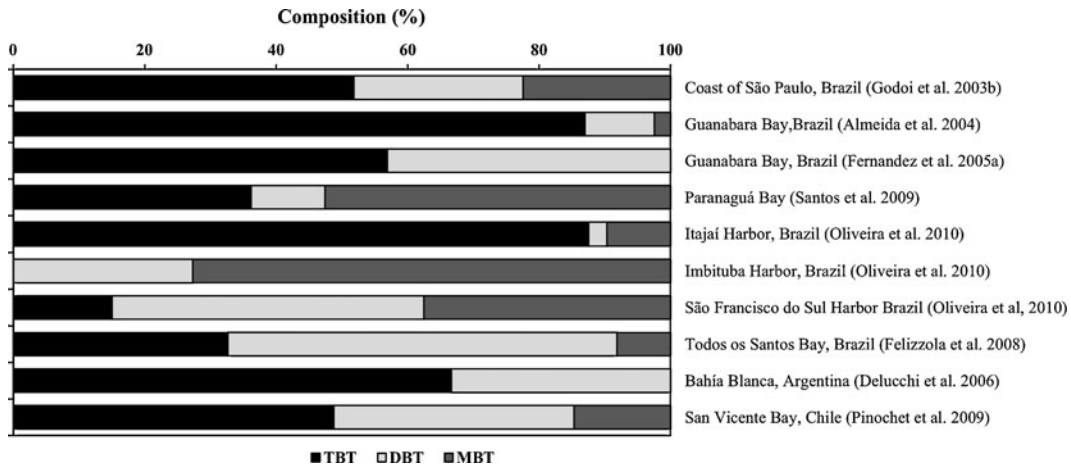

Fig. 3 Relative composition of butyltin compounds reported for sediments of South American coastal areas

as the USA (Landmeyer et al. 2004), Australia (Haynes and Loong 2002), Japan (Harino et al. 2007), Italy (Chiavarini et al. 2003b), and Spain (Díez and Bayona 2009) (Fig. 2). These findings are probably due to the absence of pre-banishment regulatory strategies (except Argentina) and a lack of inspections on the antifouling usage in South American countries. All studies emphasized that the highest OTs concentrations were found in nearby potential sources, such as shipyards, harbors and marinas, which was also demonstrated by other worldwide studies (Rato et al. 2008).

Based on the steps of BTs degradation (TBT→DBT→MBT and inorganic tin; Clark et al. 1988), the predominance or high levels of TBT indicated recent inputs to the environment. However, when DBT and/or MBT predominated in sediments, the contamination might be considered historic or, alternatively, a fast degradation process might have taken place (Oliveira et al. 2010). Figure 3 shows the organotin relative composition reported for sediments from several South American coastal areas. The degradation products (DBT and MBT) predominated over TBT concentrations in the sediments of Paranaguá Bay, São Francisco do Sul harbor, Imbituba harbor and San Vicente Bay, which might be already an indication a of a certain effectiveness of the TBT banning by IMO, since most of these data was collected after its implementation. However, these comparisons must be carefully analyzed since field data could be influenced by the local characteristics of several environmental factors. Additionally, in locations where high BTs concentrations were detected (i.e., Paranaguá Bay) the DBT and MBT levels could be related to both old and fresh inputs of TBT.

OTs in marine mammals

Marine mammals (mainly cetaceans) have been used worldwide as biomonitors of aquatic contamination (Tanabe and Subramanian 2006). Due to their apparently lower capacity to metabolize organotin compounds, when compared with other mammals (Tanabe 1999), cetaceans have shown significant accumulation and body distribution of OTs (Tanabe and Subramanian 2006). In South America, only one study has determined OTs in cetaceans (Castro, unpublished data). TBT, DBT, MBT, and TPhT were detected in all liver samples analyzed from five different dolphin species accidentally caught or found stranded along the coast of Paraná and south coast of São Paulo (southern Brazil) during 1997 and 1999. The OTs levels (ΣTBT, DBT, MBT, and TPhT) ranged from 180 to 7,300 (average of 2758) ng Sn g^{-1} (wet weight) for estuarine dolphin (*Sotalia guianensis*; $n = 15$), from 520 to 4,300 (average of 2,535) ng Sn g^{-1}

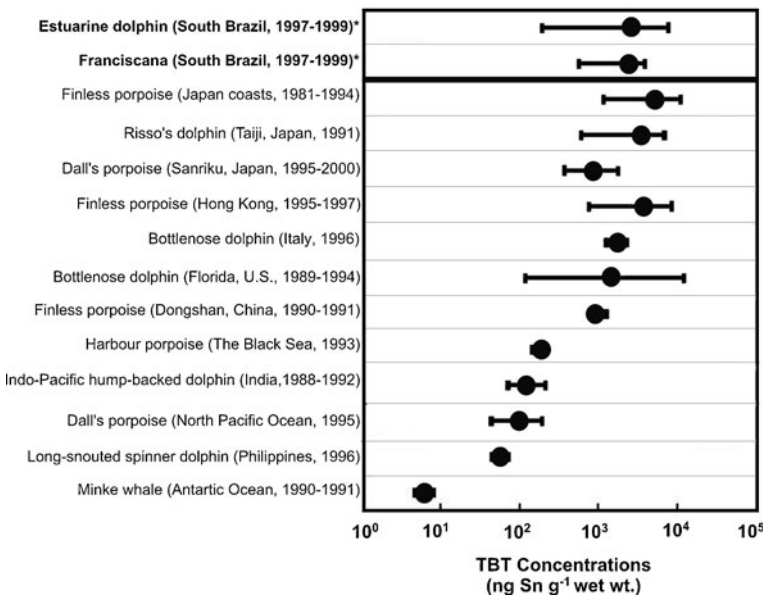

Fig. 4 Butyltin concentration (ng g^{-1} wet wt) in liver of different dolphins reported for South American coastal areas (*asterisk*) and several other regions of the world

(wet wt) for the Franciscana dolphin (*Pontoporia blainvillei*; $n = 21$), from 240 to 460 (average of 391) ng Sn g^{-1} (wet wt) for the Atlantic spotted dolphin (*Stenella frontalis*; $n = 2$), 440 ng Sn g^{-1} (wet wt) for the common dolphin (*Delphinus capensis*; $n = 1$) and 57 ng Sn g^{-1} (wet wt) for the striped dolphin (*Stenella coeruleoalba*; $n = 1$). The hepatic organotin concentrations (ΣOTs) found in estuarine and franciscana dolphins from southern Brazilian were in the same order of magnitude as levels found for cetaceans from very industrialized areas around the world (i.e., Japan, Hong Kong, Italy, and USA; Iwata et al. 1995; Kim et al. 1996; Tanabe et al. 1998; Takahashi et al. 2000; Kannan et al. 1996, 1997; Madhusree et al. 1997), and much higher than those observed for other developing countries such as Turkey, China, Philippines, and India (Madhusree et al. 1997; Tanabe et al. 1998; Fig. 4).

Another study has determined total tin in liver samples of 13 cetacean species from Guanabara Bay and coast of Espírito Santo (southeastern Brazil; Dorneles et al. 2008). The authors assumed that hepatic tin in cetaceans is predominantly in the organic forms, thus levels of total tin in the livers reflected the anthropogenic inputs of organotin (Tanabe 1999). Based on that, the contamination levels of the studied species can be considered higher than or similar to those levels observed in cetaceans from developed countries. Concentrations of Sn as high as 4,803 ng g^{-1} were detected in specimens of *S. guianensis.*

The results for mammals together with those for sediments indicated that sources of OTs in the Southern Hemisphere (particularly in the southern Brazilian coast) can be comparable to those already experienced by developed countries from the Northern Hemisphere. That could be due to the quick industrial and harbor activities in specific areas of some South American countries in the last two to three decades.

Imposex in the South American coast

Imposex is defined as a superimposition of male sexual characters, such as penis and *vas deferens*, onto females of gastropods exposed to OTs (Gibbs and Bryan 1987). This phenomenon results from, a still unknown, metabolic perturbation in endocrinology regulation of prosobranch gastropods (Porte et al. 2006; Sternberg et al. 2010) that was firstly reported by Blaber (1970) in the caenogastropod *Nucella lapillus.* Nowadays, imposex is a widespread problem which was already been registered in approximately 170 prosobranch species (Shi et al. 2005). Imposex occurrence and intensity are proportional to the environmental

levels of OTs compounds, with a clear cause–effect relationship (Matthiessen and Gibbs 1998). Hence, this powerful biomarker has been used worldwide to map TBT pollution along marine environments (Sousa et al. 2009). Imposex levels are usually measured through biometric indexes such as: Relative Penis Size Index (RPSI), Relative Penis Length Index (RPLI), and Vas Deferens Sequence Index (VDSI), in addition to percentage of affected females (Gibbs et al. 1987).

The RPSI, which has been more used for animals with advanced imposex stages, is obtained by the equation RPSI = (mean penis length in females)^3/(mean penis length in males)^3 × 100, whereas RPLI = (mean penis length in females/mean penis length in males) × 100 has been used more during the initial imposex levels. These numerical indexes indicate the levels of imposex development in the studied sample (Bryan et al. 1987).

The VDSI was initially proposed by Gibbs et al. (1987) to be used in *N. lapillus* and later modified by several authors for the application with others prosobranch species (Fernandez et al. 2005; Horiguchi 1998; Queiroz et al. 2007; Stroben et al. 1995). In general, the VDSI is an individual verification of six different imposex stages considering the following parameters: the penis and vas deferens formation, the penis size, the penis and vas deferens ligation, the blocking of the vulva, and the presence of abortive capsules.

Due to its simplicity and low costs, imposex in gastropods has been used for assessing and monitoring the contamination by OTs in several areas along the South American coast. To our knowledge, 32 gastropod species were reported so far as imposex exhibitors in South America (Table 1).

Imposex in the Brazilian coast

The occurrence of imposex in *Stramonita haemastoma* and *Stramonita rustica* have been monitored to assess the impact of OTs along a large extension of the Brazilian coast (Castro et al. 2007a, b; Fernandez et al. 2005). Those studies included almost all of the largest harbors distributed along the 7,300 km of the Brazilian coast (Fig. 1).

In the northeastern Brazilian coast, the imposex levels were verified in 82 sites distributed over ten areas of intense shipping activities, such as: Pecém harbor (7 sites) and Mucuripe harbor (12 sites) in Ceará state; Natal harbor (9 sites) in Rio Grande do Norte state; Cabedelo harbor (9 sites) in Paraíba state; Recife harbor (7 sites), and Suápe harbor (7 sites) in Pernambuco state; Jaraguá harbor (10 sites) in Alagoas state; Sergipe river estuary (7 sites) in Sergipe state; and Todos os Santos Bay (Salvador and Aratu harbors; 14 sites) in Bahía state (Castro et al. 2007b). Although the study showed imposex occurrence in all monitored areas, its levels were related to the intensity of marine traffic in each area. For example, Suape harbor, which had recently started its operations at that time, showed imposex at just one of the seven monitored sites. On the other hand, busier harbors, such as a Salvador and Mucuripe, showed imposex in most of the sampled sites. The authors also emphasized that other factors may affect the imposex levels, such as local hydrodynamic and the harbor location (*in shore* or *off shore*).

In Guanabara Bay (Rio de Janeiro), imposex in *S. haemastoma* was strongly related to the potential organotin sources (harbors, marinas, and shipyards), where 10 out of 14 sites showed imposex (Fernandez et al. 2002). These results correlated well with organotin levels found in surface sediments of Guanabara Bay (see Item 2 above; Fernandez et al. 2005). Similarly, a good relationship was found between imposex indexes in the muricid *S. haemastoma* and the intensity of ship traffic in areas under the influence of four main harbors from southern Brazil (Paranaguá harbor in Paraná state, São Francisco do Sul and Itajaí harbors in Santa Catarina state, and Rio Grande harbor in Rio Grande do Sul state; Castro et al. 2007a). The highest imposex indexes were observed in Babitonga Bay (Santa Catarina state), which is under the influence of São Francisco do Sul harbor. Although this area did not hold the busiest among the studied harbors, it is a semi-closed bay that probably hinders the contaminant dispersion.

Environmental gradients of imposex indexes (decreasing levels with the increasing distance from possible sources) were found in most of studied areas along the Brazilian coast

Table 1 Prosobranch species from South American coastal areas which imposex was already reported

Species	Location	References
Acanthina monodon	San Antonio harbor, Chile	Huaquin et al. (2004)
Adelomelon ancilla	Peninsula Valdés, Argentina	Bigatti et al. (2009)
	Bahía San Gregorio, Argentina	
Adelomelon beckii	Mar del Plata harbor, Argentina	Arrighetti and Penchaszadeh (2010)
Adelomelon brasiliana	Mar del Plata harbor, Argentina	Penchaszadeh et al. (2001), Bigatti et al. (2009)
Adelomelon ferrusacci	Punta Quilla, Argentina	Bigatti et al. (2009)
Buccinanops cochlidium	Punta Quilla, Argentina	Bigatti et al. (2009)
Buccinanops globulosus	San Antonio Oeste, Argentina	Bigatti et al. (2009)
	Golfo Nuevo, Argentina	
Buccinanops monilifer	Mar del Plata harbor, Argentina	Penchaszadeh et al. (2001), Averbuj and Penchaszadeh (2010)
Chichoreus brevifrons	Margarita island, Venezuela	Miloslavich et al. (2007)
Chichoreus margaritensis	Margarita island, Venezuela	Miloslavich et al. (2007)
Chorus giganteus	Renocavi Bay, Chile	Gooding et al. (1999)
	San Vicente Bay, Chile	Bigatti et al. (2009)
Cymatium parthenopeum	Cabo Frio, Brazil	Castro et al. (2007c)
	Vitória harbor, Brazil	Costa et al. (2009)
Leucozonia nassa	Cabo Frio, Brazil	Castro et al. (2007c)
	Margarita island, Venezuela	Miloslavich et al. (2007)
Melongena melongena	Cispata Bay, Colombia	Hernandez and Stotz (2004)
Nassarius vibex	Fortaleza, Brazil	Lima-Verde et al. (2010)
	Rio de Janeiro, Brazil	Cardoso et al. (2009)
Nucella crassilabrum	Renocavi Bay, Chile	Gooding et al. (1999)
	San Vicente Bay, Chile	
Olivancillaria deshayesiana	Mar del Plata harbor, Argentina	Teso and Penchaszadeh (2009)
Odontocymbiola megallanica	Peninsula Valdés, Argentina	Bigatti and Penchaszadeh (2005)
Pareuthria plumbea	Caleta Sara, Argentina	Bigatti et al. (2009)
	Cro. Rivadavia, Argentina	
	Puerto Camarones, Argentina	
	Puerto Deseado, Argentina	
	Puerto San Julián, Argentina	
	Rada Tilly	
	Ushuaia, Argentina	
Prunum martini	San Antonio Este, Argentina	Bigatti et al. (2009)
Pugilina morio	Fortaleza, Brazil	(Lima-Verde, unpublished data)
Stramonita haemastoma	Northeastern Brazilian coast	Castro et al. (2007b)
	Guanabara Bay, Brazil	Fernandez et al. (2005)
	Southern Brazilian coast	Castro et al. (2007a)
Stramonita chocolata	Peruvian harbor areas, Peru	(Castro, unpublished data)
Stramonita rustica	Northeastern Brazilian coast	Castro et al. (2007b)
Thais deltoidea	Vitória harbor, Brazil	Costa et al. (2008)
Thais biserialis	Gulf of Gayaquil, Ecuador	Castro et al. (2011)
Thais brevidentata	Gulf of Gayaquil, Ecuador	
Thais kiosquiformis	Gulf of Gayaquil, Ecuador	
Trophon geversianus	Peninsula Valdés, Argentina	Bigatti et al. (2009)
	Puerto San Julián, Argentina	
	Punta Quilla, Argentina	
	Ushuaia, Argentina	
Voluta ebraea	Fortaleza, Brazil	Castro et al. (2008)
Xanthochorus cassidiformis	Renocavi Bay, Chile	Gooding et al. (1999)
	San Vicente Bay, Chile	
Ximenopsis muriciformis	Ushuaia, Argentina	Bigatti et al. (2009)

(Castro et al. 2007b). Similar results were observed in many other locations, such as the Mediterranean sea using *Hexaplex trunculus* (Axiak et al. 1995), Crok harbor, Ireland, using *N. lapillus* and *Littorina littorea* (Minchin et al. 1996), Phuket island, Thailand, using *Chicoreus capucinus* and *Thais distinguenda* (Bech et al. 2002) and Kyllibegs, Ireland, using *N. lapillus* and *L. littorea* (Minchin and Minchin 1997).

Imposex was also detected in punctual studies along the Brazilian coast. Gastropods populations of *S. haemastoma*, *Cymathium parthenopeum* and *Leucozonia nassa* from Cabo Frio (Rio de Janeiro state; Castro et al. 2007c); *Nassarius vibex* from Itacuruça island (Rio de Janeiro) (Cardoso et al. 2009), the endemic marine snail *Voluta ebraea* (Castro et al. 2008) and *N. vibex* (Lima-Verde et al. 2010) from Ceará state; and *C. partenopeum*, *T. deltoidea* and *L. nassa* from Espirito Santo state (Costa et al. 2009) were reported as impacted.

Imposex in the Argentinean coast

On the Argentinean coast, a large biomonitoring study was recently conducted (Bigatti et al. 2009). This was the first detailed monitoring study in South America which simultaneously analyzed imposex in snails and TBT levels in surface sediments. The results demonstrated that most affected areas were those with high marine traffic, such as commercial, fishery, and tourist ports. The localities with higher imposex incidence were Mar del Plata harbor, San Antonio Oeste, San Antonio Este harbor, Puerto Madryn harbor, Puerto Deseado harbor, Puerto Julián harbor, and Ushuaia harbor. Among the analyzed species, those inhabiting soft and mixed bottoms, such as *Buccinanops globulosus*, *Buccinanops cochlidium*, *Pareuthria plumbea*, *Ximenopsis muriciformis*, *Prunum martini*, *Adelomelon brasiliana*, *Adelomelon ferrusacci*, *Adelomelon ancilla*, and *Odontocymbiola magellanica*, showed better imposex response for areas with low or medium TBT contamination. On the contrary, no imposex was detected in the species inhabiting intertidal hard bottoms (*Trophon geversianus* and *Crepidula aculeata*) with low to medium contamination levels. In conclusion, the results of imposex in gastropods and the sediment analyses demonstrated that all the studied areas of the Argentinean coast were contaminated by TBT.

In another study, imposex was detected in *O. magellanica* collected from 12 sites inside the Golfo Nuevo (Patagonia, Argentina). This investigation also observed that the more intense the marine traffic zones the higher the imposex levels (Bigatti and Penchaszadeh 2005). Other punctual studies in Mar del Plata pointed out imposex in other gastropod species, such as *A. brasiliana* (Penchaszadeh et al. 2001), *Buccinanops monilifer* (Averbuj and Penchaszadeh 2010), *Adelomelon beckii* (Arrighetti and Penchaszadeh 2010), and *Odontocymbiola deshayesiana* (Teso and Penchaszadeh 2009).

Imposex in other South American coastal areas

Others gastropod species reported as imposex exhibitors in South America coastal areas were: *C. giganteus*, *Xantochorus cassidiformis*, and *Nucella crassilabrum* collected from San Vicente Bay (central Chile; Gooding et al. 1999), *Acanthina monodon* from San Antonio harbor and Renocavi Bay (central Chile; Huaquin et al. 2004); *Chicoreus brevifrons*, *C. margaritensis* and *L. nassa* from Margarita island and continental coast of Venezuela (Miloslavich et al. 2007); *Melongela melongela* from Cispata Bay (Colombia; Hernandez and Stotz 2004); and *Thais biserialis*, *Thais brevidentata*, *Thais kiosquiformis* from coastal areas of Ecuador (Castro et al. 2011). In most of these, the imposex was accidentally found during studies about the reproductive system of mollusk species. It is worth mentioning ongoing studies on the Peruvian coastal areas that also found imposex in *Stramonita chocolata* (Castro, unpublished data).

Legislation

The French Government was the first to prohibit the use of organotin-based antifouling paints after the oyster industry collapse in Arcachon Bay in the early 1980s (Alzieu et al. 1986; Alzieu 2000). Since small boats were blamed to cause the problem in coastal waters, the use of these paints on vessels (<25 m in length) was banned in January 1982

Table 2 Overview of restraints concerning the use of TBT (modified from IMO 2008)

Country	Year	Regulations
Argentina	1998	Prohibited the use of TBT-based paints on vessels
Australia	1989	Prohibited the use of TBT-based paints on vessels smaller than 25 meters (m) in length. Maximum leaching rate of 5 μg/cm^2/day for vessels greater than 25 m in length. All dry-docks must be registered in the Environmental Protection Agency due to discharges. All antifoulants must be registered
Austria	1989	Banned the use of TBT antifouling paints in fresh-water lakes
Brazil	2007	Prohibited the use of TBT-based paints on large vessels
Canada	1989	Prohibited the use of TBT-based paints on vessels less than 25 m in length, except for aluminum-hulled vessels. Maximum leaching rate of 4 μg/cm^2/day for vessels greater than 25 m in length. All antifoulants must be registered
Chile	2001	Only regulates values regarding metallic tin (Sn^0), where the limits allowed in areas of coast protection is 0.5 mg L^{-1}. Legislation by National Institute of Normalization
Commission of the European Community (EC)*	1991	Prohibited the use of TBT-based paints on vessels smaller than 25 m in length TBT antifoulants available only in 20 L containers
Non-EC members	Varies	Prohibited the use of TBT-based paints on vessels smaller than 25 m in length (most countries)
Finland	1991	Prohibited the use of TBT-based paints on boats smaller than 25 m in length
France	1982	Prohibited the use of TBT-based paints on vessels smaller than 25 m in length, except for aluminum-hulled vessels
Germany	1990	Prohibited the use of TBT-based paints on vessels smaller than 25 m. Ban on retail sale. Ban on their use on structures for mariculture. Regulation for the safe disposal of antifouling paints after removal
Hong Kong	1990	All TBT antifoulants must have a valid permit for import/supply. All antifoulants must be registered
Japan	1990	TBT banned for all new vessels
	1992	TBT banned for all vessels
New Zealand	1989	The application of TBT copolymer antifouling paint was banned with three exceptions: aluminum-hulled vessels, the aluminum outdrive, or any vessel greater than 25 m in length. The application of TBTO free-association paints is banned. Maximum leaching rate of 5 μg/cm^2/day for vessels greater than 25 m in length. All antifoulants must be registered
Norway	1989	Prohibited the use of TBT-based paints on vessels smaller than 25 m in length
South Africa	1991	Prohibited the use of TBT-based paints on vessels smaller than 25 m in length. TBT antifoulants available only in 20-L containers. All antifoulants must be registered
Sweden	1989	Prohibited the use of TBT-based paints on vessels smaller than 25 m in length
	1992	Maximum leaching rate of 4 μg/cm^2/day for vessels greater than 25 m in length. All antifoulants must be registered
Switzerland	1987	The use of TBT-based antifouling paints is banned in fresh water lakes. All antifoulants must be registered
The Netherlands	1990	Prohibited the use of TBT-based paints on vessels less than 25 m in length. Washing/blasting slurry used to prepare TBT antifoulants may be treated as hazardous waste. TBT antifoulants available only in 20 L containers. All antifoulants must be registered
United Kingdom	1985	Sale of TBT-based paints restricted, effective bar on TBTO free-association paints
	1987	Prohibited the use of TBT-based paints on vessels less than 25 m in length and on fish-farming equipment. TBT antifoulants available only in 20 L containers. All antifoulants to be registered as pesticides; Advisory Pesticides Committee must approve sale and use. Washing/blasting slurry treated as hazardous

Table 2 (continued)

Country	Year	Regulations
United States	1988	Prohibited the use of TBT-based paints on vessels less than 25 m in length, except for aluminum-hulled vessels
	1990	Maximum leaching rate of 4 μg/cm^2/day for vessels greater than 25 m in length. All antifoulants must be registered. TBT-based antifouling paints can only be applied by certified applicators
Worldwide ban (IMO)	2003–2008	Proposed ban for January 1, 2003—no reapplication of TBT
		January 1, 2008. No ships or structures shall bear TBT. To be ratified if 25% of shipping tonnage or 25 of the worlds shipping nations sign

by the Ministry of Environment of France (Evans et al. 2000). Additionally, in October 1992, antifouling paints containing TBT were forbidden for the general public and were only available in special packaging for shipyards (Alzieu 1998). Similar legislation were introduced between the late 1980s and early 1990s in, the United Kingdom, United States of America, Australia, Canada, Netherlands, Japan, Denmark, and Hong Kong (Champ 2000). Austria and Switzerland, which have no direct access to the ocean, banned TBT in their lakes and rivers to minimize the direct effects on freshwater communities and indirect impacts on marine habitats downstream (Becker Van-Slooten and Tarradellas 1994; Fent 1996). Table 2 summarizes the nations (including South American countries) that have prohibited the use of TBT over the last few years.

Since shipping is a worldwide activity, controls on antifouling systems by individual nations are not effective enough to prevent pollution, even in their own territorial waters. Therefore, the problem was brought to the IMO. The IMO is a specialized agency of the United Nations with 167 member states and three associate members, which aim to develop and maintain a comprehensive regulatory framework for shipping, including safety and environmental concerns. Environmental issues are normally deliberated by the Marine Environmental Protection Committee (MEPC) that, for several years, collected information on the effects of organotin compounds on the marine environment and human health. Therefore, the MEPC approved a draft resolution of the Assembly banning new applications of organotin-based antifouling paints from January 1, 2003 and their presence on ship hulls from January 1, 2008 (IMO 2008).

In Brazil, resolution 357 of the National Environment Council (CONAMA 2005) established the maximum concentrations of TBT in water. Allowed levels vary according to the water category (fresh, brackish, or sea water) and determined use. Levels up to 10 and 370 ng L^{-1} are tolerated for classes 1 and 2, respectively, of brackish and sea waters. There are no regulations on TBT sedimentary levels though. The use of organotin-based antifouling paints was definitively banned in November 2007 by means of NORMAM–23/DPC. This regulation also determined inspection and registration standard procedures for all antifouling systems, as well as for management of residues of these compounds (Marinha do Brasil 2007). In Argentina, the institution that regulates maritime transportation (Prefectura Naval Argentina) prohibited the use of TBT-based antifouling paints in all types of vessels back in 1998 (Resolution 04/98). Although this resolution also regulated procedures for coatings replacement (DPMA 1998), organotin-based paints are still commercialized in the country (Bigatti et al. 2009). In Chile, the National Institute of Normalization regulates only values for metallic tin (Sn^0) in water (INN 2001); the limit allowed in areas of coastal protection is 0.5 mg L^{-1}. To our knowledge, there is no legislation on the use of antifouling paints in the other South American countries.

Future trends

Due to the environmentally harmful effects of TBT-based self-polishing paints and their consequent worldwide banning (IMO 2008), manufacturers had been forced to urgently study and develop new antifouling paints that were more environmentally friendly (Almeida et al. 2007). As a result, 16 products were homologated by the IMO International Convention on the Control of Harmful Anti-fouling Systems on Ships to be used as antifouling biocides (Thomas et al. 2001). These booster biocides are: Diuron, Irgarol 1051, Sea-Nine, Chlorothalonil, Dichlofluanid, Thiram, Busan (TCMTB), TCMS Pyridine, Pyridine-triphenylboron, Zinc pyrithione, Cuprous pyrithione, Ziram, Maneb, Cuprous oxide, Cuprous thiocyanate, and Copper naphthenate. Currently, the utilization of coatings containing copper combined with one or more of these booster biocides have been a usual alternative to organotin (Konstantinou and Albanis 2004). However, toxicity and deleterious environmental effects have already been detected for some of these biocides (Zhang et al. 2008; Manzo et al. 2008).

In order to develop antifouling systems of high performance, the naval industry have tried to develop more efficient, durable and less toxic products. According to IMO, the ideal antifouling biocide should have: broad spectrum activity, low mammalian toxicity, low seawater solubility, low bioaccumulation in the food chain, very low persistency (ideally not persistent) in the environment, compatibility with paint raw materials and good price (Takahashi 2009). Therefore, new and more environmentally friendly approaches have been investigated. For example, natural products produced by sessile fouling-free marine organisms (i.e., terpenes, phenols, steroids, and others) have showed similar antifouling properties to TBT, generating expectations that these compounds could be used as antifouling agents in near future (Omae 2006). Additionally, biocide-free antifouling systems are entering the market. These antifouling systems are based on silicones, fiber coats, epoxides, polyurethane and polyvinyl that reducing the adhesion capacity of aquatic organisms onto ship hulls (Watermann et al. 2005). However, ecotoxicological studies showed that biocide-free antifouling systems can also be toxic (Karlsson and Eklund 2004).

Another important issue is the replacement of TBT-based antifouling systems for new paints after the implementation of IMO resolution (IMO 2008). The paint particles removed during the replacement process could have possible caused a large recent worldwide input of organotin compounds into coastal environments (Kotrikla 2009). This could be confirmed by new studies in areas under the influence of shipyards. In addition, it is reasonable to assume that previously organotin contaminated sites will be the same ones thereafter affected by the new antifouling biocides. Furthermore, monitoring of the OTs contamination (using imposex in gastropods and/or chemical determinations) has shown to be an extremely efficient tool for mapping affected areas by past, present, or future use of antifouling biocides (Konstantinou and Albanis 2004).

The restrictions imposed by developed countries and IMO on TBT usage have had some positive effects. Recovery has been reported in marine gastropods populations severely affected by TBT (Lahbib et al. 2009) and reductions in environmental concentrations of TBT have been reported in Europe and Asia (Choi et al. 2010; Galante-Oliveira et al. 2009). Although studies evaluating the effectiveness of TBT global banning are still not available for South American countries, the existing data and the ongoing surveys in the Atlantic coast are targeting the understanding of temporal trends on TBT contamination levels and imposex.

Conclusions

Imposex occurrence was observed in gastropod populations of several sites from South American coastal areas and high concentrations of OTs in sediments from areas under the influence of ship activities from the Atlantic coastal areas, particularly in Paranaguá Bay (Brazil), Guanabara Bay (Brazil), Mar del Plata harbor (Argentina), Bahia Blanca (Argentina), and Puerto Deseado (Argentina). High OTs levels in liver of dolphins from the southern Brazilian coast were also

observed. Despite no available data for coastal areas of Suriname, Guyana, French Guyana, and Uruguay (Atlantic coast), and very limited data for the Pacific coast, even considering ongoing studies on the Peruvian coastal areas, the widespread TBT contamination along South American coastal areas is evident. Although there is a clear relationship between organotin contamination and imposex in gastropods, it is difficult to compare on a global scale results obtained with different South American species, since each one presents biological particularities which affect the intensity of imposex (Bech 1999). The lack of a unique species to cover larger areas is a limitation; even though, the available data pointed out some "hot spots" in coastal areas under the influence of South American main harbors, and that OTs contamination showed a similar distribution pattern seen for countries of Northern Hemisphere. Therefore, the establishment of baselines of organotin contamination in the Pacific coast and the implementation of temporal trend studies in the South American coastal areas is crucial to verify the effectiveness of local regulations and OTs global ban, and to map the most sensitive areas related to present and future antifouling impacts.

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