

ORGANOTIN POLLUTION AT ARRAIAL DO CABO, RIO DE JANEIRO STATE,  
BRAZIL: INCREASING LEVELS AFTER THE TBT BAN

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## A B S T R A C T

Imposex, an endocrine disruption phenomenon, is a biomarker of tributyltin (TBT) exposure in marine gastropod populations. The occurrence and intensity of the syndrome in populations of the neogastropod *Stramonita haemastoma* were ascertained at nine sampling stations in October 2008 at Arraial do Cabo, a very important tourist resort in Rio de Janeiro state (Brazil) and part of the Marine Extractive Reserve of Arraial do Cabo. The results obtained made it possible to evaluate the changes which had occurred since the last biomonitoring campaign, undertaken in 2001, at these same stations. Despite the ban on the use of TBT imposed by the Brazilian Navy in 2003, the controls on the use of this agent in Brazil resulting from the establishment of NORMAM 23 by the Navy in 2007 and the complete ban of organotin compounds in antifouling paint formulae by the IMO on a global scale in 2008, imposex was still detected. Instead of the expected reduction, an increase was observed in the areas affected by imposex. Furthermore, populations in the areas seriously affected by imposex in 2001 were absent in the 2008 sampling campaign. These results suggest an increase in TBT pollution in the study area, indicating the inefficacy of legislative measures.

## R E S U M O

O imposex, um fenômeno de desregulação endócrina, é um biomarcador da exposição ao tributilestanho (TBT) em populações de gastrópodes marinhos. A ocorrência e intensidade dessa síndrome nas populações do neogastrópode *Stramonita haemastoma* foram verificadas em nove estações amostrais em outubro de 2008 em Arraial do Cabo, uma área de grande importância turística no Rio de Janeiro (Brasil) e integrante da Reserva Extrativista Marinha de Arraial do Cabo. Com os resultados obtidos foi possível avaliar as alterações ocorridas desde o último biomonitoramento realizado em 2001, nessas mesmas estações. Mesmo após o banimento do TBT pela Marinha do Brasil em 2003, a entrada em vigor dos controles ao uso deste agente com o estabelecimento da NORMAM 23, no Brasil em 2007 pela Marinha, e o banimento completo do TBT nas fórmulas de tintas antiincrustantes pela IMO em escala global, em 2008, o imposex continua a ser detectado. Ao invés de um declínio esperado, observou-se um aumento da extensão das áreas afetadas pelo imposex. Além disso, observou-se a ausência de populações em áreas que em 2001 estavam gravemente afetadas pelo imposex. Esses resultados sugerem um aumento da poluição por TBT na área de estudo, indicando a ineficácia das medidas legislativas.

Descriptors: Imposex, *Stramonita haemastoma*, Forno Harbor, Organotins.

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## I N T R O D U C T I O N

Many chemical substances of different classes are currently being used to reduce the maintenance costs imposed by the biofouling of submerged structures. These substances include organometallic compounds such as Zyneb (Zn ethylene bisdithiocarbamate), Maneb (Mn ethylene bisdithiocarbamate) or Zn-Piritione, organochlorines like Sea Nine 211 and triazines like Irgarol 1501, among others (Fernandez and Pinheiro, 2007). Of the substances used as antifouling agents, TBT (tributyltin) was the most widely applied, as it was

used on some 90% of the world fleet in the 90's (Rivarolo et al., 1996). Due to the toxicity and persistence of organotin compounds, from the late 80's onwards, some countries, such as France, the United Kingdom and the USA, adopted local control measures, generally restricting its application to small boats (see the review by Champ, 2000). In 2001, the IMO (International Maritime Organization) established an international convention that came into force in September 2008, completely banning the use of these compounds in antifouling paints. In Brazil, the Brazilian Navy suspended the use of TBT-based antifouling paints on its vessels in 2003, and in 2007

enforced a regulation, the NORMAM 23, to restrict the use of organotin compounds in all the antifouling systems on all Brazilian registered vessels, from small leisure craft to ships. This ban produced positive initial effects in the Guanabara Bay region (RJ), where the most important Brazilian naval bases and shipyard are located (Fernandez et al., 2005). In this area, long-term biomonitoring studies have been conducted by our own research group and a reduction in the imposex intensity indexes at our fixed control station in Guanabara bay (on Vermelha beach) from 2001 on, and, more recently, a gradual recovery of some previously impacted sampling stations (Fernandez et al., 2006; our research group, unpublished data), have been observed.

Among the numerous side effects of TBT on marine organisms (see review by Fent, 1996), the imposex response is the best-known. This sexual alteration is a characteristic response to TBT exposure and can be used as a specific biomarker (Matthiessen and Gibbs, 1998). Imposex has been described in more than 190 species on a worldwide basis (Shi et al., 2005). The neogastropod species *Stramonita haemastoma* (Kool, 1987) is found on all the coasts of Rio de Janeiro State, being the the region's most extensively studied species. The development of male sexual characters in this species is proportional to the intensity of navigation in the area concerned (Santos et al., 2009; Fernandez et al., 2005; Spence et al., 1990). Imposex induction by TBT and triphenyltin (TPT) has been demonstrated for this species in laboratory exposures (Limaverde et al, 2007) and imposex has also been shown to occur in snails transplanted to contaminated areas (Queiroz et al., 2007; Castro et al., 2005).

Arraial do Cabo District is a very important touristic area of the Rio de Janeiro State due to its transparent waters and great biological productivity due to a local upwelling system (Carvalho and Gonzalez-Rodriguez, 2004). Its coast is part of the Marine Extractive Reserve of Arraial do Cabo, the purpose of which is to preserve the natural and landscape resources while permitting sustainable exploitation of fishing by the local population. Various inlets are to be found in this area, among them being Anjos Inlet, where Forno harbor and a small marina are located. Forno harbor receives only about 0.02% of the total annual shipping tonnage of all Brazilian ports (ANTAQ – National Agency of Waterborne Transport). Due to the presence of these potential organotin sources, imposex has previously been measured in populations of *S. haemastoma* in the region to verify their influence on neighboring biota (Ribeiro, 2002). This present study aims to evaluate the changes which have occurred in the region since 2001, describing the present levels of imposex found in the area as a measure of the effectiveness of the legislation presently in force.

## MATERIAL AND METHODS

Specimens of *S. haemastoma* were collected at Arraial do Cabo (Rio de Janeiro, Brazil) in October 2008, at the same stations sampled in 2001 by Ribeiro (2002). The stations' spatial distribution is shown in Figure 1. Thirty adult animals were collected at each station by diving and were maintained in constantly aerated seawater until they were brought to the laboratory.

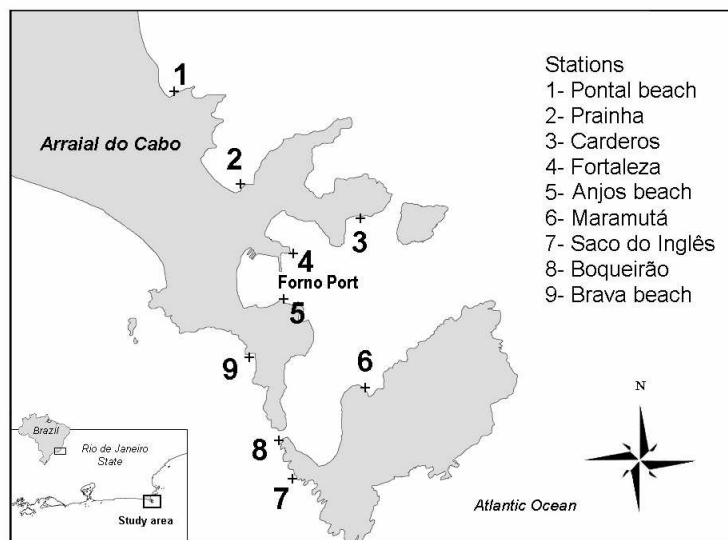


Fig. 1. Location of sampling stations in Arraial do Cabo, Rio de Janeiro, Brazil.

In the 2008 study, the specimens were narcotized in a 1:1 mixture of 7% MgCl<sub>2</sub> solution in distilled water and local seawater. After narcotization, the animals were observed by pulling the soft parts away from the shell opening and exposing part of the pallial cavity. The animals were identified as males or females, and those that showed vas deferens and/or penis, or, in other words, any sign of imposex, were considered as males (see explanation below). After analysis, the snails were transferred back to an aquarium with local seawater to revert the narcotization process and they were forthwith taken back to their original sampling areas.

According to the total numbers of females and males in each sampling station, the probability (P) of a sample with the observed "n" of healthy females being derived from a natural, 1:1 sex ratio, was calculated for a sample of "N" animals, through the algorithm developed by Fernandez et al. (2007).

For the sake of simplicity, it is easily inferred that the probability of obtaining a sample without a single healthy female from a binomially distributed,  $p = q = 0.5$  population, is less than one in a billion. This case, in organotin pollution monitoring, corresponds to a 100% imposex population, which is a very common feature in highly polluted areas. This was the case, for instance, at two stations in the previous study at Arraial do Cabo.

It should be noted that the variability in the observed sex ratios when compared to those expected in each sample could derive from either of two reasons: the natural effect of the small samples employed, or the artificial, highly biased populations observed in polluted areas. The criteria used to distinguish between these two conditions are indicated by the significance levels of the 'P' calculated. For  $P < 0.001$ , a station is considered impacted, for a deviation in sex ratio ranging from 0.001 to 0.05 ( $0.001 < P < 0.05$ ), the local environment is considered probably impacted, and for P values  $> 0.05$  significance, the station is considered not to be impacted.

The original data of Ribeiro (2002) on the number of healthy females, imposexed females and

males has been treated by the same statistical method to allow comparisons with the 2008 data obtained by the new, non-destructive method for imposex evaluation (Fernandez et al., 2007). Data on imposex incidence, calculated by the percentage of the imposexed females among the total number of females collected at each station, was also used for comparison.

## RESULTS

The occurrence of imposex in populations of *S. haemastoma* from Arraial do Cabo had spread in 2008. In 2001, all the sampling stations presented specimens of the species studied and imposex incidence was restricted basically to Anjos Inlet, where Forno harbor and the local marina are situated. In 2008, organisms were absent from three stations (Saco dos Carderos, Fortaleza and Anjos beach) despite the intense sampling effort (Table 1).

All the stations previously free of this syndrome now present imposex in their populations. The incidence of imposex at these stations now ranged from a minimum of 25% in the case of Saco do Inglês station to 47% at Prainha station, in 2008. The other stations presented even higher intensities. At Pontal Beach station, where 4% imposex was observed in 2001, a frequency of 83% was observed in 2008. The single exception to this tendency was Boqueirão station, where a small reduction of the incidence occurred (15% to 11%). A more radical change, however, was observed in the more seriously impacted stations, that presented up to 100% frequency of the syndrome as compared with 2001. Populations of this species were not to be observed at these stations in 2008. This absence of gastropod populations might be an indicator of serious TBT pollution, since this same pattern has been observed, for instance, in Guanabara Bay, highly affected by TBT. Population distribution about 1960 was reduced by two-thirds as compared with that of 2000-2001 (Fernandez et al., 2005). A slow later recovery is being observed, but the current distribution is still reduced (our research group, unpublished data).

Table 1. Sampling stations in Arraial do Cabo (%I= Percentage of affected females; N = sample size; ND = not determined).

Stations	% I 2001	% I 2008	N 2001	N 2008	Situation 2001	Situation 2008
Pontal beach	3.8	83.3	40	30	Not impacted	Impacted
Prainha	0.0	47.4	39	30	Not impacted	Probably impacted
Saco dos Carderos	86.4	ND	39	3	Impacted	-
Ponta da Fortaleza	100.0	ND	27	1	Impacted	-
Anjos beach	100.0	ND	30	0	Impacted	-
Maramutá	35.7	72.7	29	22	Probably impacted	Impacted
Saco do Inglês	0.0	25.0	22	31	Not impacted	Not impacted
Boqueirão	15.0	11.1	23	30	Not impacted	Not impacted
Brava beach	0.0	38.1	24	30	Not impacted	Not impacted

Through the probabilistic method, in which the ratio of males to females found in the sampled populations could indicate the imposition of male characteristics on the organisms, it was observed that in 2001, five stations were considered as not impacted, one as probably impacted and three as impacted. In 2008, these numbers had changed to three, one and five, respectively, considering as impacted the stations where populations of *S. haemastoma* were not found, but where the animals had been observed before (Fig. 2).

## DISCUSSION

The increasing severity of imposex and the total absence of *S. haemastoma* populations in some stations characterize a situation which is precisely the reverse of what would be expected in view of the restrictions on the use of TBT presently in force in Brazil (IMO, 2001; NORMAM 23, 2007). The enforcement of more restrictive controls on the application of organotins as biocides in antifouling paints has led to a reduction in imposex intensity in several areas around the world as, for instance, in Portugal (Sousa et al., 2009; Galante-Oliveira et al., 2009), England (Morton, 2009; Oliveira et al., 2009) or Alaska (Tallmon and Hoferkamp, 2009). In many

cases, a reduction in the severity of sediment contamination by organotins has also been observed (Choi et al., 2010). These findings clearly show that the regulations governing the use of organotins worldwide have been producing positive results. In Arraial do Cabo, a general reduction in imposex intensity was, therefore, to have been expected. Instead, there was an increase in imposex levels and possibly a local extinction of those populations that had already been severely affected by imposex in 2001. It is likely, then, that the organotin concentrations in the water are higher now than they were in 2001. This same pattern has been observed in other areas of Brazil, such as Angra dos Reis, where the absence of this bioindicator species has recently been reported in some places close to marinas and shipyards, where their existence had been recorded in 2002-2003 (Pessoa et al., 2009; Pinheiro et al., 2006).

In the present study, an aggravation of the imposex situation was clearly to be observed at some of the stations and this is a cause for concern. Several hypotheses may be raised to explain the increased levels of TBT between 2001 and 2008: an increase in shipping activity in this period; a release of organotins from the bottom sediments; or even an increase in activities such as naval repairs in shipyards.

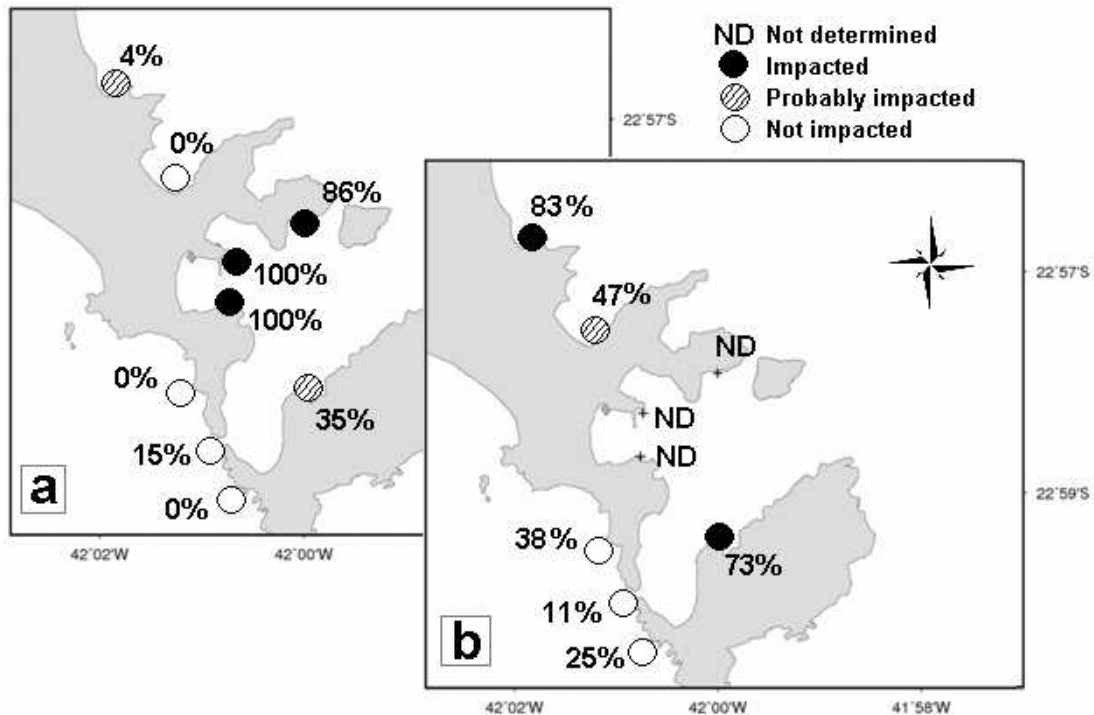


Fig. 2. Distribution of percent of imposex affected females (%I) and respective statistical evaluation in 2001 (a) and 2008 (b).

As regards the first of these hypotheses, in the last few years, Arraial do Cabo has undergone a substantial population growth, but with a marked reduction in fisheries in this period. This has been a reflection of a general reduction in these activities in Rio de Janeiro State, there having been a reduction of from 2,258 to 1,086 in the number of people directly employed in fisheries between 1996 and 2006. Similarly, the number of people involved indirectly in fishing, in the preparation of the catch for retail sale, transportation, frozen storage, and so on, shrank from 4,097 to 716 over the same period. At Arraial do Cabo, this kind of activity was almost completely discontinued, with the exception of retail sales (Soares, 2009). Local people informed the authors that most local youngsters now work in the offshore petroleum complex at Macaé, further up the coast, or just use their small boats for tourist activities. Thus, the number of small boats in the local marina has probably not changed much, with touristic activities today accounting for nearly all the local boating. This number of small boats is about two hundred. Anyway, these boats are not allowed to use organotins as antifouling by the current Brazilian regulations (NORMAM 23, 2007). So, at least theoretically, they cannot have contributed to the present situation.

On the other hand, we should take into account the particular case of Anjos beach and of Ponta da Fortaleza, where the harbor and the local marina are located side by side in the inner part of Anjos inlet, and thus act as a single source of antifouling to the whole area. So, the first hypothesis and the present results may indicate a greater impact of the Forno harbor than before. According to ANTAQ, in 2001, for instance, the harbor received 45 ships, equivalent to the transport of 529,249 tons of cargo, while in 2008 these figures fell to 123,532 tons transported in 8 ships, representing a great reduction in the volume of shipping in the period. Besides this reduction, there has been another important change. The available capacity of Forno harbor has been used in recent years to receive support vessels and undertake the maintenance of the floating platforms of the off-shore oil industry of the Campos Basin. This might be a problem, as not all countries are enforcing the global ban on organotins (only 45 countries with about 74% of the total world tonnage were officially doing so in September 2008 – IMO, 2010). But, as determined by NORMAM 23, foreign vessels registered to operate in Brazilian waters should also have organotin-free coatings. Again, theoretically, the harbor is thus not expected to be a source of the organotins in the water column.

As for the second hypothesis, another possible source of the organotins in the local water column was the desorption process of contaminated sediments. The long half-life of TBT in sediments

provides a persistent source for the marine environment and could lead to a continued effect on biota. In some countries, a reduction in imposex severity was ascertained after the implementation of controls on TBT use, indicating a lower concentration of organotin compounds available in the water column, despite the presence of high concentrations in sediments and mussels. This process was seen, for example, in Portugal after the 2003 EU ban (Sousa et al., 2007) and in Korea (Choi et al., 2010).

Although TBT has great affinity with sediments, with the decline in input, losses can occur by desorption and aerobic degradation. However, it persists for years (Choi et al., 2010; Langston and Pope, 1995). Under these conditions, without any new input, a progressive decrease occurs in the desorption rate, as a consequence of the 'aging effect' in the slow adsorption phase due the activation energy of bonds and diffusive transfer limitations (Ruiz et al., 2008; Burton et al., 2006; Pignatello and Xing, 1996), and it results in a progressively higher distribution coefficient ( $K_d$ ), that means a still higher concentration in the sediments. So water concentrations would become progressively lower and the imposex response would also be reduced. So, within this perspective, a further reduction of imposex intensities would be expected if the local sediments were the only source of TBT in the area.

Regarding the third hypothesis, the discussion is simpler. Shipyards are known to be sources of organotin pollution in these times of global TBT ban, due the replacement of the antifouling systems that, in many instances, have their resulting contaminated wastes transported without proper handling (Kotrikla, 2009). As there are still no shipyards in the Arraial do Cabo region, however, no impact from this kind of activity is to be expected.

From the above discussion, it is clear that the animals should have presented a much better condition, as they have done elsewhere. However, their present condition implies a recent input of organotins in the area. Recent inputs of organotins into coastal environments have still been observed elsewhere, as demonstrated by the ratio between TBT and its degradation products in sediments, water and tissues, in post-legislative scenarios (Oliveira et al., 2009; Tallmon and Hoferkamp, 2009; Sousa et al., 2009a; 2009b; 2007). Such inputs are also likely to be occurring in the Arraial do Cabo region, but as we do not have any chemical data from the area, this hypothesis may be suggested, but not proved.

Recent, localized inputs of TBT might be derived from three main sources: foreign vessels from countries that do not enforce the Antifouling Systems Convention, illegal use of organotins on Brazilian registered vessels or on small local craft. Presently, the real contribution of each one of these possible sources

cannot be determined with certainty. Another possible source is the use of organotins in platforms, as small floating platforms have been observed in the inlet after the first sampling performed in 2001.

In any case, our results pinpoint the need to undertake further detailed studies in order to elucidate the situation observed and to evaluate the possible impact of the enlargement of Forno harbor, located on Anjos Inlet and close to areas where gastropod specimens are now becoming rare.

It should also be remembered that this is a region possessing many natural resources. The maintenance of the health of the marine environment is, therefore, very important both from the preservation and the economic points of view. The impacts which are now appearing will exact their toll in the future. A well-managed tourist industry would generate resources and employment for an indefinite period of time. This question certainly deserves further study.

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