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Assessing the Impacts of Organotin Compounds in Ilha Grande Bay, (Rio de Janeiro, Brazil): Imposex and a Multiple-Source Dispersion Model

F. M. Pinheiro†, M.A. Fernandez‡, M. R. Fragoso∞, J. P. Quadros¤, E.Camillo Jr.ζ and F. A. Dos Santos§

† Oceanography Dept., Rio de Janeiro State University, Rio de Janeiro, 20550-013, Brazil pinheiro_fernanda@hotmail.com

‡ Oceanography Dept., Rio de Janeiro State University, Brazil. hallfz@uerj.br

∞ Marine and Atmospheric Modeling Laboratory, Federal University of Rio de Janeiro, RJ, 21949-900, Brazil fragoso@acd.ufrj.br

¤ Oceanography Dept., Rio de Janeiro State University, Brazil. Jupq@hotmail.com

ζ Oceanography Dept., Rio de Janeiro State University, Brazil. Edcamillo@yahoo.com.br

§Marine and Atmospheric Modeling Laboratory, Federal University of Rio de Janeiro, Brazil falves@acd.ufrj.br

ABSTRACT

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Organotin compounds, especially tributyltin (TBT), have been widely used as antifouling paints on vessels' hulls since the early 1970s. Besides their efficiency, they are extremely toxic to non-target species. Some toxic effects include abnormal growth and thickening of oyster shells, high larval mortality, and an endocrine-disrupting syndrome in neogastropods called imposex, which consists of the development of male sexual characteristics in the females. As numerical models are useful tools to evaluate environmental impacts, this work reports a combined biomonitoring-numerical modeling approach to study organotin impacts at Ilha Grande Bay, a pristine region situated on the south coast of Rio de Janeiro, Brazil. It is the first attempt in Brazil to simulate the dispersion of organotin compounds from multiple sources. An inverse-in-time probabilistic trajectory model was also applied to verify possible sources of contamination in some particular polluted areas. Biological monitoring was realized using imposex development in *Stramonita haemastoma* in 44 sampling stations. The area proved to be an ecosystem heavily impacted by organotin compounds. These impacts are due to the presence of an oil terminal, a huge Brasfel's dockyard, and also to numerous small pleasure craft located in marinas in the region. A good agreement was found between the numerical dispersion model and the distribution of imposex intensities in the studied area. The results showed that biological data could be useful for calibration of this kind of model.

ADDITIONAL INDEX WORDS: *Tributyltin, biomonitoring, numerical modeling.*

INTRODUCTION

Tributyltin (TBT) has been the main organotin compound used as antifouling paints on vessels' hulls since the early 1970s. While global consensus on the efficiency of organotin compounds exists, they are also known to be highly persistent in some environments, bioaccumulate and be harmful to non-target species (HOCH, 2001). Some toxic effects include abnormal growth and thickening of oyster shells, high larval mortality, and an endocrine-disrupting syndrome in neogastropods called imposex. Imposex consists of the development of male sexual characteristics such as penis and a vas deferens on female gastropods (SMITH, 1971) and has been widely used as a biomarker of TBT contamination because of its dose-related response (EVANS *et al*., 1998; MATTHIESEN and GIBBS, 1998; STROBEN *et al*., 1995). In highly impacted ecosystems it can lead females to sterilization and consequently cause a decline on population levels.

The high toxicity of organotin compounds has led numerous countries to adopt national and regional measures to control its use (see review in CHAMP, 2000). Even though a decrease in impacts and concentrations of organotin has been widely observed (HARINO *et al*., 1999; MICHEL and AVERTY, 1999), contamination is still occurring at levels above the No Effect Level Concentration (NOEC; HOCH, 2001). A globally binding treaty has been adopted by the International Maritime Organization to regulate the use of anti-fouling systems and totally ban the presence of organotin compounds at ships' hulls by 1st January 2008 (www.imo.org).

Numerical models are useful tools to evaluate environmental impacts and have been applied to a variety of risk assessments from the most diverse contaminants (LIMA *et al.,* 2001; SELIGMAN *et al*., 1987).

This work reports a combined biomonitoring-numerical modeling approach to study organotin impacts at Ilha Grande Bay, a pristine region situated in the south coast of Rio de Janeiro, Brazil. It is the first attempt in Brazil to simulate the dispersion of organotin compounds from multiple sources. The goals are: (1) to evaluate the possible environmental impacts from using these compounds in an area with great economic importance that supports activities such as tourism, traditional fishing and shipping; (2) verify the validity of the numerical modeling using the imposex response instead of water chemical analysis. The last approach is justified by the low costs of this kind of monitoring and the fact that this biological response is proportional to the mean water concentration of TBT (STROBEN *et al*., 1995). In this case, the animals act as integrators.

Study Area

Ilha Grande Bay is situated in the southeast coast of Brazil with its center at latitude 23°15'S and longitude 44°30'W (Figure 1). Its mountainous relief and the presence of numerous islands make it a very attractive place. An oil terminal and a nuclear power plant are located in the area, besides a great number of marinas and dockyards, as well as Angra dos Reis harbor.

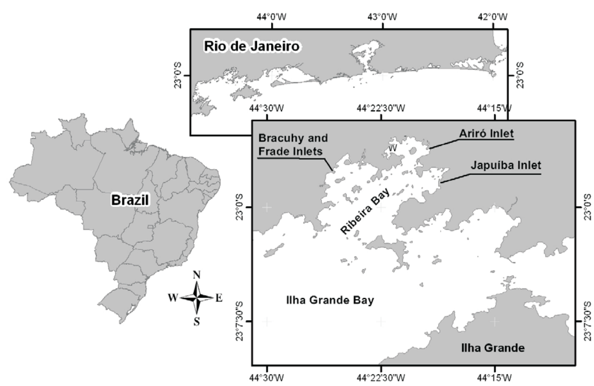

Figure 1. Ilha Grande Bay, Rio de Janeiro, Brazil.

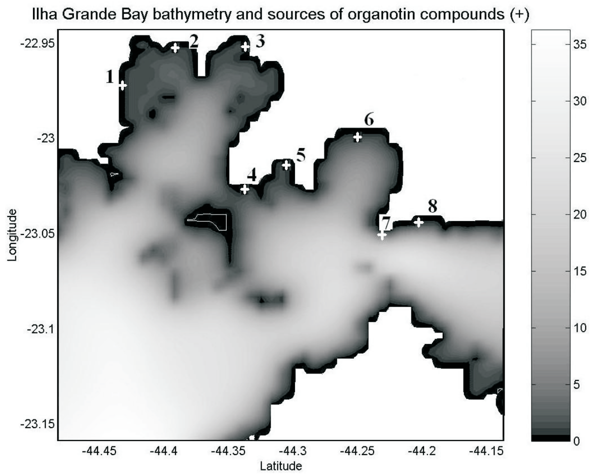

Figure 2. Ilha Grande bay bathymetry and main sources of TBT.

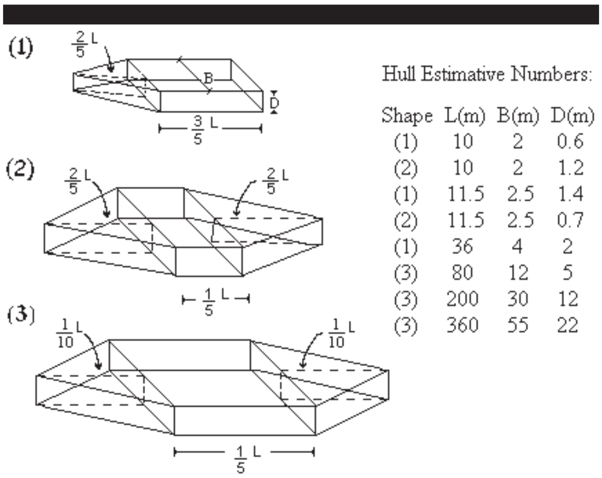

Figure 3. Ship Hull used for assessing crafts surface area in contact to the water (L = length, B = breadth and D = draft).

Tidal currents around 0.2 m/s during spring tides and 0.1 m/s in neap tides are observed (IKEDA and STEVENSON, 1980; SIGNORINI, 1980). According to these authors, there is also an eastward quasi-steady current in the channel between the island and the mainland. A natural oscillation period around 6 hours has been observed (SIGNORINI, 1980; FRAGOSO, 1999). The region is dominated by a wind-driven circulation, with the predominance of southerly winds (FRAGOSO, 1999).

Two areas are relevant for this study, the inner inlets where the presence of numerous pleasure boats can be observed and the channel between Ilha Grande and the continent where is marked the route for the larger ships to the oil terminal.

METHODS

Biological Monitoring

Stramonita haemastoma was selected as bioindicator for this study because of its broad geographical distribution along the Brazilian coast, its high abundance in rocky shores, and because it appers develop imposex proportionally to the distance from sources (SPENCE *et al*., 1990; CASTRO *el al*., 2000; FERNANDEZ *et al.,* 2002). *S. haemastoma* has a planctonic larval stage, thus allowing areas heavily contaminated by organotin compounds, which lead females to sterilization, to be recolonized (SPENCE *et al*., 1990).

A total of 44 sites were analyzed for imposex during summer months, between October 2001 and January 2003. At each sampling station 20 organisms were collected and analyzed by conventional methods (GIBBS and BRYAN, 1987; FERNANDEZ 2001). The VDSI (Vas Deferens Sequence Index) applied is presented in table 1, and as this index has lower seasonal fluctuation (PINHEIRO *et al*., 1999) than the RPLI (Relative Penis Length Index) and RPSI (Relative Penis Size Index), it was compared to the dispersion model applied.

Numerical Modeling

Figure 2 shows a bathymetric map of the area used in the simulations and the major sources of TBT to the region analyzed in the dispersion model. The numbers are related to the major sources presented in table 1, where 1 to 5 and 8 are marinas, 6 is the major dockyard of the region and 7 is an oil terminal. The model spatial resolution is 300 meters in both horizontal directions.

Table 1. *Sequence of vas deferens development (VDSI).*

Stage	Description
0	Normal females
1	Presence of a vestigial penis
2	Presence of a developed penis (±1mm)
3	Presence of a completely developed penis which length can be measured and / or vas deferens incomplete
4	Vas deferens complete
5	Blockage of the oviduct
6	Presence of aborted capsules

Dispersion Model

The dispersion model is a numerical tool to study the behavior and approximate trajectory of passive constituents in water bodies. A two-dimensional numerical transport model developed by the Marine and Atmospheric Processes Modeling Laboratory (LAMMA) of the Federal University of Rio de Janeiro (UFRJ) (LIMA *et al.,* 2001) was applied to simulate the dispersion of organotin compounds from its sources, like marinas and dockyards. The governing equation of the model is the equation for advection and diffusion of passive constituents and is solved by finite difference technique.

The following steps were necessary in order to better represent the conditions simulated:

- Estimations of tributyltin inputs to the environment from leaching were obtained based the number of boats afloat in each source region (Table 2). In order to determine the surface area in contact to the adjacent water, a formulation based on estimations of mean length, breadth, and draft of the boats in each source was used. Figure 3 presents the scheme used, where (1) represents motor boats and (2) sail boats and (3) ships. It was also considered that a marina should have half of its boats as (1) and half as (2). Then, the leaching limit imposed by legislative measures in some countries of $4 \mu g.cm^{-2}.d^{-1}$ (CHAMP, 2000) was used to determine the theoretical mass of TBT available at each source. A fraction of boats coated with TBT was also considered according to CHAMP (2000), who suggests that 70% of the world fleet is still using these compounds.

- Marine current data for the region were acquired. These data were studied by FRAGOSO (1999), using POM (Princeton Ocean Model, BLUMBERG and MELLOR, 1987).

- Two 10-day periods were selected out of a 20-year wind time series of the region measured by Eletronuclear energy company (latitude of 23º00'19"S and longitude of 44º27'30"W). In case 1 we can observe the predominance of N winds, while in case 2 both N and S winds occur (Figure 4). The 10 day period was chosen based on the residence time of the TBT in the water (SELIGMAN *et al*., 1996) and the two cases were chosen based on typical (case 1) and critical (case 2) wind conditions of the region.

- Finally, a decay rate of 6% per day was applied following WALTON *et al.* (1986) that determined this rate based on biological experiments in San Diego bay. This decay is in agreement with more recent estimations (FENT, 1996).

Probabilistic Model

An inverse-in-time probabilistic trajectory model was developed and applied to verify possible sources of

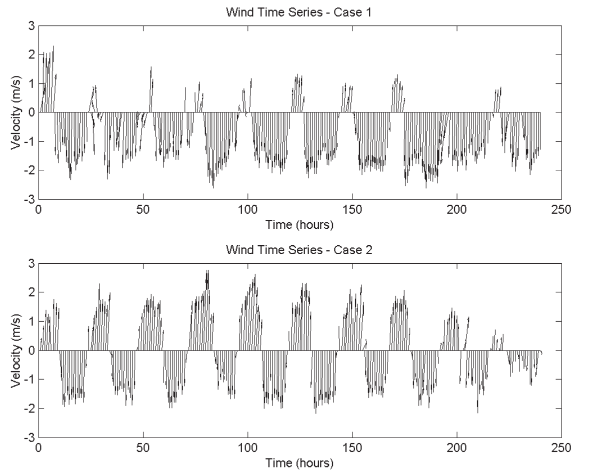

Figure 4. Wind time series used in the simulations.

contamination in some particular polluted areas. Its innovation refers to the possibility of combining a great number of wind and current sequences that can occur in the time series available, determining the possible trajectories in a 10-day period of a particle that reached a certain spot. This allows the observation of the areas most likely to influence that same spot, and therefore to evaluate possible contamination sources for that very place.

Current data were obtained from FRAGOSO (1999) and wind data from Eletronuclear. A sequence of 278 days of wind direction and velocity data was used in the simulation.

RESULTS AND DISCUSSION

Results of the imposex index, VDSI, are presented in figure 5. This index ranged from 0.1 in Mambucaba to 5.1 in Bracuhy, demonstrating that some areas of this ecosystem are heavily impacted by organotin compounds. A reference area, where no impact is observed, is not shown in the figure. It is located on the southern coast of Ilha Grande where UERJ's advanced research campus (CEADS) is located (44°11'W and 23°10'S).

The results show that besides the areas of Brasfels dockyard (4.9) and Angra dos Reis (3.8), the two largest marinas of the region (Bracuhy at 5.1 and Frade at 5) are the most impacted areas, as well as some stations of the inner inlets (Japuíba Inlet and Ariró Inlet) where samples of *S. haemastoma* were not found. This fact may be related to the high level of organotin compounds that might be adsorbed on local sediments, as these compounds have high partition coefficients and tend to associate to the particulate (LANGSTON and POPE, 1995), and these areas have very restricted water circulation. A similar observation was made in Botafogo Inlet, Guanabara Bay (FERNANDEZ, 2001; FERNANDEZ *et al*., 2002)

Minor impacts are observed outside Ribeira bay, going to the west, as station are located far from organotin sources, in an area of higher current velocities (MINCHIN and MINCHIN, 1997). The VDSI reaches 0.5 in Praia Brava and 0.1 in Mambucaba.

Westward the area of figure 2, another station was analyzed, and the levels of imposex found were critical and related to the presence of fishing boats in the area. However, this was considered another study case and not included here.

Sterile females were found in 11 stations, all of them in the vicinity of the major sources. The greatest number was found in the marinas of Bracuhy (8 sterile females) and Frade (6). Even though a considerable number of sterile females were found, it seems not to compromise most population levels.

The presence of females with vas deferens both incomplete and complete and the absence of a penis were frequently observed. This fact does not corroborate to the sequence of VDSI reported by SPENCE *et al.* (1990) who indicated that *S. haemastoma* followed the same scale proposed by GIBBS and BRYAN (1987) to *Nucella lapillus*, and is not reported even in the multi-route scale developed by STROBEN *et al*. (1995). However, another independent study has found similar females at Arraial do Cabo (RIBEIRO, 2002). These females were classified according to the vas deferens development and further studies are being conducted to clarify this observation.

In respect to the numerical dispersion model applied there are minor differences between case 1 (Figure 6) and case 2 (Figure 7) simulations. In case 1, which has the predominance of N winds, the constituent seems to be more dispersed, while in case

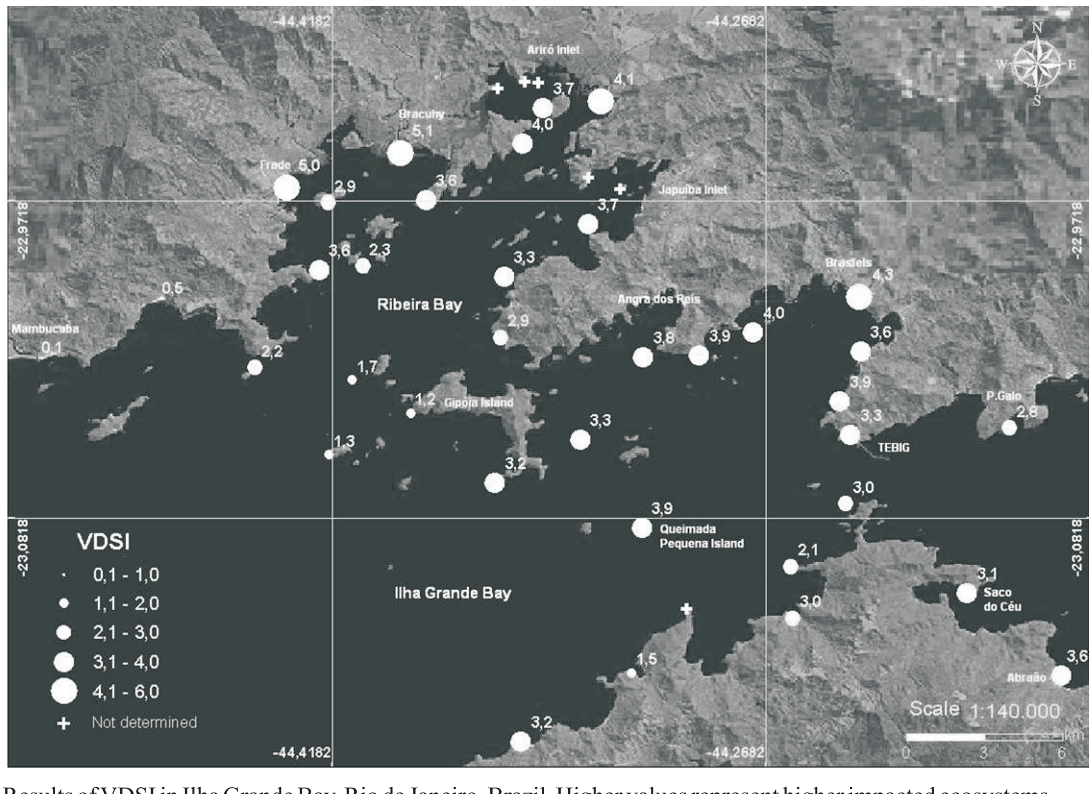

Figure 5. Results of VDSI in Ilha Grande Bay, Rio de Janeiro, Brazil. Higher values represent higher impacted ecosystems.

Table 2. *Major sources of organotin compounds in Ilha Grande Bay.*

Source Regions	Longitude / Latitude	N of crafts (mean size)	Estimations of TBT mass per day
(1) Frade, Santos Yacht Club	044.4334 W / 22.9722 S	120 (11.5m) + 1 (36m)	161.22 g / day
(2) Porto Marina Bracuhy	044.3929 W / 22.9519 S	650 (10m)	598.32 g / day
(3) Porto Marina 1	044.3367 W / 22.9519 S	18 (11.5m) + 2 (36m)	38.05 g / day
(4) Rio de Janeiro Yacht Club	044.3389 W / 23.0262 S	30 (10m)	27.61 g / day
(5) Pirata's Mall, Aquidabã, Marinas Club	044.3029 W / 23.0127 S	396 (11.5m) + 76 (10m)	399.14 g / day
(6) Brasfels Dockyard	044.2512 W / 22.9992 S	1 (360m) + 3 (80m)	1476.00 g / day
(7) TEBIG – Ilha Grande Oil Terminal	044.2318 W / 23.0509 S	1 (200m)	389.22 g / day
(8) Marina Porto Galo	044.2039 W / 23.0442 S	54 (38)	69.17 g / day

2, the major incidence of S winds seems to trap the constituent closer to the continent. However, ample a good dispersion can still be noticed. A limit of 1 µg of TBT was set up for the simulation. This value is a function of the kind of model employed, i.e. 2D dispersion of a non-conservative constituent, and it is not directly linked to real water concentrations. The dispersion pattern observed in the simulations is closely related to the lower imposex level boundary and theoretical masses were closer to these lower imposex values (Figure 5-7).

High levels of the constituent are observed in the channel between Ilha Grande and the continent and the prevailing eastward currents can be noticed in both cases. Consequently higher levels of contamination can be found at stations located to the east of the sources. These results corroborate the imposex levels (2.1 and 3) found in some areas of Ilha Grande. Lower levels of TBT are found in the western portion of the island where the VDSI decreases from 3 to 1.5. However, an increase in VDSI is observed in the westernmost station of Ilha Grande, but it is known that an anchorage zone for tanker waiting for berth is located at 23°09'S and 44º24.5'W (TRANSPETRO, 1996). Those vessels can release huge quantities of TBT to the adjacent waters and this possible source was not included in the simulations.

In Ribeira Bay, a decrease of imposex levels from approximately 5 to 2.9 and 2.3 occurred from Bracuhy and Frade source areas to some close islands that are not even more than 5km away, and this decrease are not represented in the dispersion simulations. This can be a result of the model resolution, which may not represent very well some particularities of the area, specially the inner inlets.

MINCHIN and MINCHIN (1997) observed a decrease of imposex levels in *Nucella lapillus* with increasing distance from sources, and also related it to the circulation patterns of the area, where in areas of higher currents low levels of imposex levels in *Nucella lapillus* with increasing distance from sources, and also related it to the circulation patterns of the area, where in areas of higher currents low levels of imposex were observed within 2 km from sources while in more restricted areas this decrease was observed 6 km away from the sources.

RIBEIRO (2002) performed a 24 hours simulation of the dispersion of organotin compounds from a port in Arraial do Cabo, Brazil, finding a good correlation with imposex levels in *S. haemastoma*.

The probabilistic model played a complementary role to the dispersion models applied. An example of this is presented in Figure 8, which shows the area that can influence Queimada Pequena Island in a period of 10 days considering a variety of wind and current combinations. It is noticed that Brasfels dockyard may cause more influence at the island than can be seen in the dispersion model and that the zone located southwest from the island may also influence it, being where the anchorage zone for ships is located. The same way, Figure 9 shows the area that can influence Caieira Island. Not only waters from Japuíba and Ariró inlets can influence the island, the former having the most influence, but also that waters from Angra dos Reis can do so. This observation is important because some areas along the coast from Japuíba inlet in the direction of Angra dos Reis have a high level of imposex, which does not correspond exactly to the dispersion simulations. Even though the simulation shows the zone that influences the area analyzed, it must be kept in mind that the particle analyzed can be contaminated or not.

Good agreement was generally found between the numerical dispersion model and the distribution of imposex intensities in the studied area. These results show that biological data could be useful for calibration of this kind of model, instead of chemical data. In the case of TBT some problems of chemical data, other than the costs itself, are that TBT can cause biological responses at concentrations which are close to the detection limit of the methods used (1-2 ng/l). Furthermore, it may be released into the environment in pulses (e.g. from dry docks) in biologically harmful amounts which could be missed by periodic sampling (FOALE, 1993).

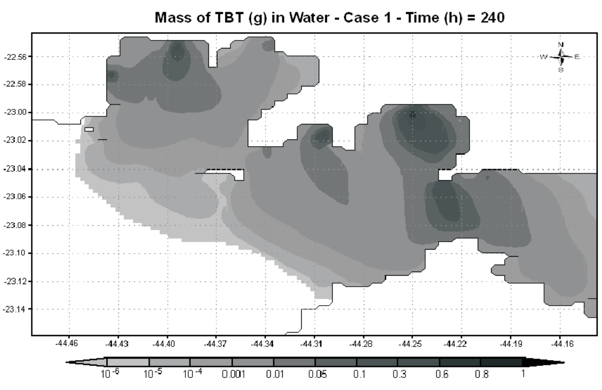

Figure 6. Dispersion model results for N winds in 10 days.

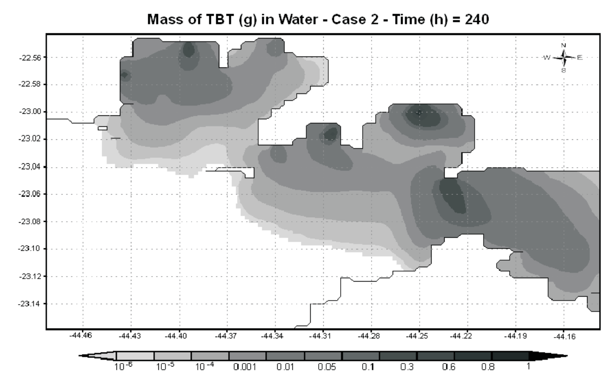

Figure 7. Dispersion model results for N and S winds in 10 days.

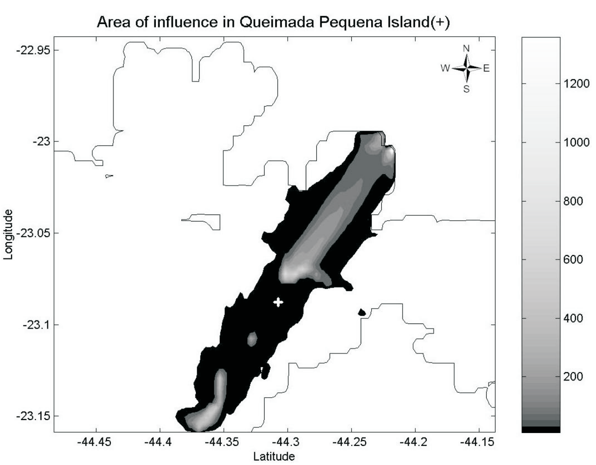

Figure 8. Probabilistic model applied to Queimada Pequena Island (VDSI = 3.9, Figure 5).

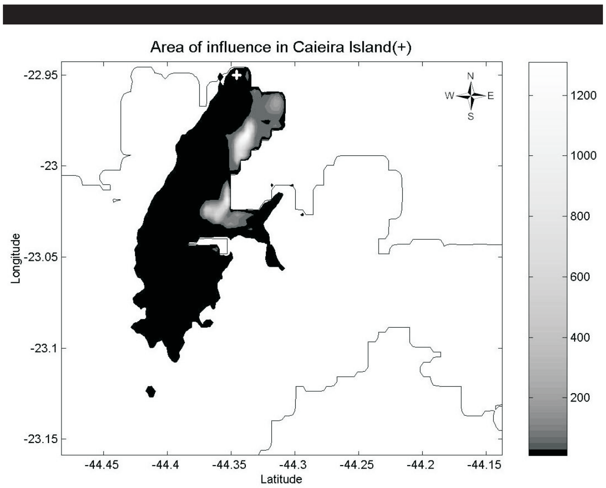

Figure 9. Probabilistic model applied to Caieira Island (VDSI = 3.7, Figure 5).

CONCLUSIONS

Several factors contribute to the impact of antifouling paints on the environment. In assessing this hazard, one must consider chemical properties of the paint in question, concentration of the active ingredients at the source region, populations of aquatic organisms exposed to the biocide, exposure regimes and effects of exposure on populations and ecosystems (DIRKX *et al.*, 1993). This is an attempt to simulate some of these conditions. A good correspondence was found in the biomonitoring-numerical modeling results, even though some approximations were made.

The studied area is an ecosystem heavily impacted by organotin compounds. Biomonitoring of imposex and the numerical models applied confirm this statement. However, the lack of basic ecotoxicological data on native species made impossible the evaluation of the exact magnitude of this impact. An example is the fact that organotin compounds can also cause high larval mortality (HORIGUCHI *et al.,* 1998). As the magnitude of these impacts can not be assessed, a precautionary approach should be kept in mind.

The dispersion model could successfully be validated by the biomonitoring of imposex and a more sophisticated 3D model for concentration could also, in the future, be calibrated in this way.

Attention is now turned into the possible impacts of the enter into force of the new convention. The need for safe removal, treatment and disposal of anti-foulants deemed harmful by the treaty and the disposal of TBT-contaminated port and harbor sediment raised serious concern (CHAMP, 2003). Therefore it is foreseen that worldwide impacts of TBT will last for many years.

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