

# Anthropogenic sources and distribution of phosphorus in sediments from the Jaguaribe River estuary, NE, Brazil

Marins, RV., Paula Filho, F.J., Eschrique, SA. and Lacerda, LD.\*

Laboratório de Biogeoquímica Costeira, Instituto de Ciências do Mar, Universidade Federal do Ceará – UFC,  
Av. Abolição, 3207, CEP 60165-081, Fortaleza, CE, Brazil  
\*e-mail: ldrude@pq.cnpq.br

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## Abstract

This paper estimates annual P emission to the Jaguaribe River estuary, NE Brazil, responsible for approximately 30% of the country's farmed shrimp production. Emissions increased three times between 2001 and 2006 and reached 43.9 t.year<sup>-1</sup>. These emissions make aquaculture the third main source of P for this estuary and are much smaller than emissions from agricultural sources and slightly lower than emissions from wastewater. Their relative contribution also increased from 10.5% to 20.5%. Temporal variability of P concentrations in estuarine bottom sediments also suggest an increase, but not as evident as the emissions. The spatial distribution and speciation of P (total P and inorganic P) in sampling stations located upstream and downstream from the main aquaculture area confirmed the importance of this source. Inorganic P was the dominant form present in sediments downstream and showed an increase from 67% to 75% of the total P present in the sediment.

*Keywords:* nutrients, emission, estuary, shrimp culture, wastewaters.

## Emissão e distribuição de fósforo de fontes antrópicas no estuário do rio Jaguaribe, Nordeste do Brasil

### Resumo

É apresentada uma estimativa da emissão anual de P para o estuário do rio Jaguaribe, Ceará, Nordeste do Brasil, área responsável por cerca de 30% da produção total de camarão cultivado no país. As emissões aumentaram por um fator de três entre 2001 e 2006, atingindo 43,9 t.year<sup>-1</sup>. Essa emissão coloca a aquicultura como a terceira fonte principal de P para esse estuário, sendo bem menor que a emissão de fontes agrícolas e um pouco menor que as emissões por águas servidas. A contribuição relativa da aquicultura também aumentou de 10,5% a 20,5%, no mesmo período. A variação temporal das concentrações de P em sedimentos estuarinos sugere um aumento no período, porém não tão evidente quanto das emissões. A distribuição espacial e a especiação do P (P total e P inorgânico) em estações de coleta localizadas à montante e à jusante da principal área de aquicultura confirmaram a importância desta fonte. O P inorgânico foi a forma dominante presente em sedimentos à jusante e mostrou um aumento de 67% a 75% do total de P presente no sedimento.

*Palavras-chave:* nutrientes, emissão, estuário, carcinicultura, águas servidas.

### 1. Introduction

The accumulation of excess nutrients in waters in the continent-ocean continuum, particularly phosphorus, has been correlated with primary production and the eventual onset of eutrophication (Tappin, 2002). Among anthropogenic sources, intensive shrimp aquaculture accounted for environmental impacts associated with the emission of large amounts of P directly to estuarine waters (Burford et al., 2003; Jackson et al., 2003). For example, in northeast Brazil emission scenarios for 25 estuaries showed that shrimp farming P contribution varied from <1 to 13% of the total anthropogenic P emission and in

many areas compares with emissions from waste waters and agriculture (Figueiredo et al., 2005; Lacerda et al., 2008). In the Gulf of California, P derived from intensive shrimp farming reaches 1,600 t annually, representing 7% of the total P from the region's agriculture. Moreover, it is similar to P originating from wastewater of nearly 5 million inhabitants (Paéz-Ozuna et al., 2003).

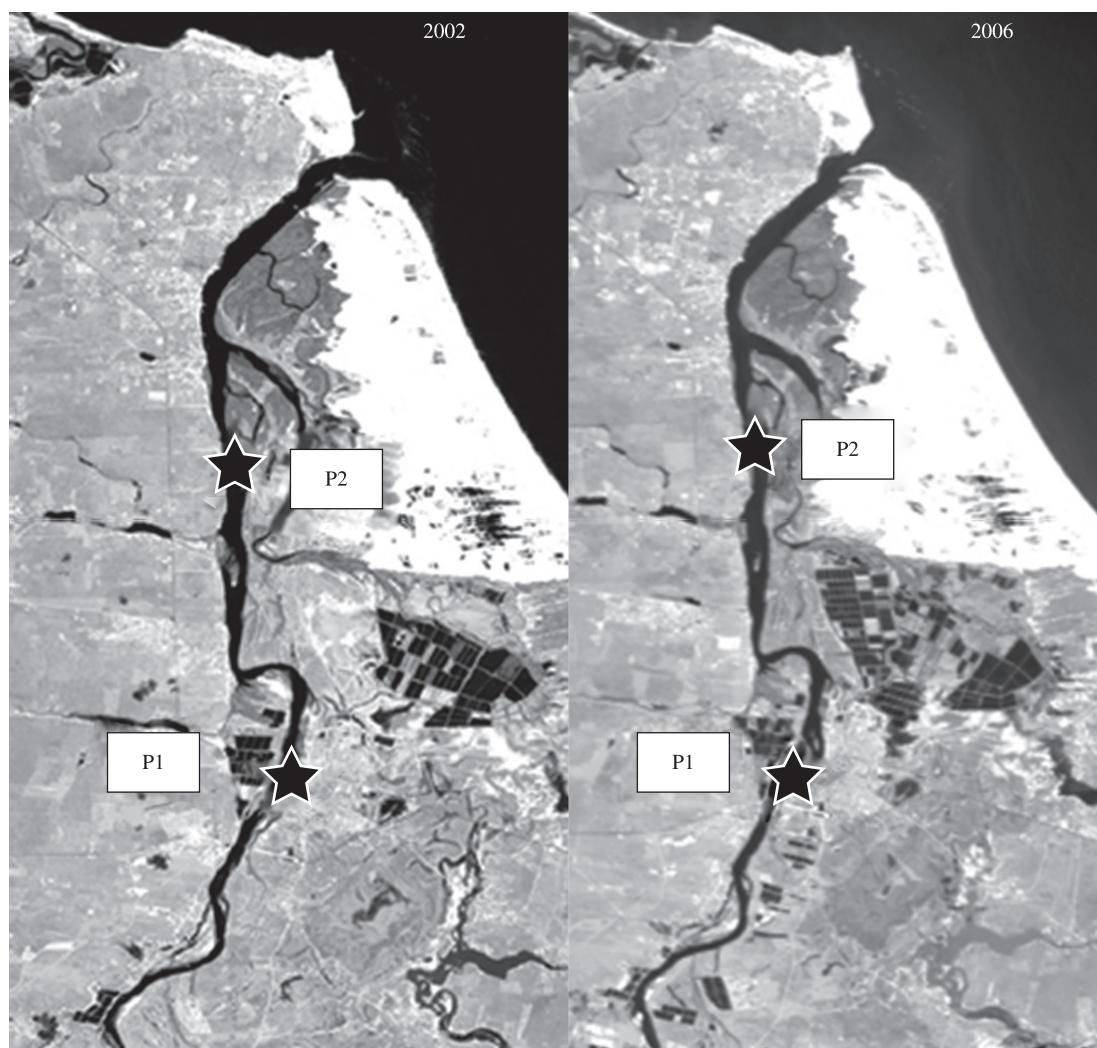
In the Jaguaribe estuarine basin, Ceará State on the Northeastern Brazilian semi-arid coast, freshwater discharge has been affected by the building of dams, water withdrawal for urbanization, agriculture and aquaculture decreasing

the dilution capacity of the estuary and maximizing the harmful effect of nutrient emissions from anthropogenic sources (Dias et al., 2009). In the estuary, the shrimp pond area increased from 820 ha in 2000 to 1,260 ha in 2003 and 1,640 ha in 2006, whereas the annual yield increased from 5,200 t in 2001 ( $6.3 \text{ t}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$ ) to 11,370 t in 2003 ( $8.2 \text{ t}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$ ). In 2004, there was an increase in the number of farms (98) and pond area (1,520 ha), but a decrease in the annual production (7,560 t) and productivity ( $5.2 \text{ t}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$ ) due to the spread of typical shrimp illness (Rocha and Rodrigues, 2004). In 2006, production remained around  $5.2 \text{ t}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$  and the total farm area increased to 1,640 ha (Figure 1). During this period, the population increased by 10% from 80,000 to 88,000 inhabitants. Although P emissions have also increased, the effects on environment P concentrations and distribution are still not reported. This study compares the annual P emissions from intensive aquaculture and other anthropogenic sources and natural processes to the Jaguaribe River estuary with

the temporal and spatial variation of P concentrations in bottom sediments of the estuary, covering the period between 2000 and 2006, when a sharp increase in shrimp farming area occurred in the region.

## 2. Material and Methods

Nutrient loads for coastal watersheds are difficult to measure properly; hence the use of indirect approaches based on emission factors which consider production/consumption parameters of the different anthropogenic sources and the chemical balance of natural processes associated with inventories of natural processes and anthropogenic activities (Tappin, 2002). The most necessary variables can be estimated from surveys and inventories of these activities, such as population parameters, urban and rural area extensions per basin, and agriculture production and fertilizer use. Emission factors have been successfully used to estimate nutrient and pollutant loads at local, regional



**Figure 1.** The Jaguaribe River estuary, NE Brazil in 2002 and 2006, showing the expansion of intensive shrimp farming and sampling points.

and global levels (Lacerda et al., 2006; Martinelli et al., 2002; NRC, 2003), and have been adopted as a standard methodology by various environmental agencies (EEA, 1999; EPA, 2002).

In the Jaguaribe river estuary, an extensive inventory of N and P emissions from natural and anthropogenic sources was carried out by Lacerda et al. (2006, 2008) based on statistics from 2001. In general, emission factors (EF) used in that inventory were those available in the literature for each activity or process, but adapted to local conditions whenever necessary. For example, wastewater production per inhabitant, generally calculated based on water consumption parameters of metropolitan areas, was corrected by the actual water consumption rate of the local population (Döll and Hauschild, 2002). Therefore, we use this summary of P emissions as a comparative basis for the present study. When necessary, as in the case of aquaculture, emissions from different sources were updated using the most recent statistics.

A survey of total and inorganic P concentrations in bottom sediments of the Jaguaribe river estuary were carried out in three campaigns in 2001, 2003 and 2006 to check for alteration in sedimentary P concentrations. Analyses were performed in sediment sampled by inserting two PVC tubes into the first 20 cm of the bottom sediments at depositional areas along the river banks located at two selected stations along the estuary. Station P1, at Aracati, is located downstream from the major urban area of the estuary but above the major shrimp farm areas. Station P2, at Raimundinho, is located downstream from the major shrimp farm areas (Figure 1). At station 2, two additional 80-cm-long cores were collected and divided into 10-cm layers to evaluate the temporal variability of P concentrations. All samples were analyzed in duplicate, averages presented are thus from  $n = 4$ . Samples were stored frozen and analyzed using established protocols (Marins et al., 2007). Total P was obtained after the sample ignition at 550 °C during 12 hours. Ashes were digested by HCl 1.0 M under stirring. Another sub-sample was directly treated with HCl 1.0 M at room temperature, and under constant stirring for inorganic P determinations. This method using individual subsamples reduces the material losses typical of sequential analysis of P fractions (Berner and Rao, 1994; Koch et al., 2001; Marins et al., 2007).

### 3. Results and Discussion

The results comparing emission inventories for 2001 and 2006 are summarized in Table 1 and show that today, emissions from natural sources ( $23.3 \text{ t}\cdot\text{year}^{-1}$ ) are 9 times lower than those from anthropogenic sources ( $191 \text{ t}\cdot\text{year}^{-1}$ ). Agriculture shows the largest load and emission yield ( $82.8 \text{ t}\cdot\text{year}^{-1}$ ;  $47.7 \text{ kg}\cdot\text{km}^{-2}\cdot\text{year}^{-1}$ ), whereas wastewaters ( $44.7 \text{ t}\cdot\text{year}^{-1}$ ;  $25.8 \text{ kg}\cdot\text{km}^{-2}\cdot\text{year}^{-1}$ ) and aquaculture ( $43.9 \text{ t}\cdot\text{year}^{-1}$ ,  $25.3 \text{ kg}\cdot\text{km}^{-2}\cdot\text{year}^{-1}$ ) rank second and third among P sources followed by husbandry ( $19.0 \text{ t}\cdot\text{year}^{-1}$ ;  $11.0 \text{ kg}\cdot\text{km}^{-2}\cdot\text{year}^{-1}$ ). Urban runoff plus solid waste disposal shows the smallest contribution ( $0.3 \text{ t}\cdot\text{year}^{-1}$ ;  $0.2 \text{ kg}\cdot\text{km}^{-2}\cdot\text{year}^{-1}$ ).

Although aquaculture accounts for only 20.5% of total P emissions, the location of farms adjacent to estuarine areas results in direct inputs to estuarine waters effectively increase environmental concentrations. Most other emissions go firstly to soils before eventually reaching the estuarine waters (David et al., 2009).

Natural processes contributing with P to the Jaguaribe River Estuary are soil loss and atmospheric deposition is dependent on the basin area, soil type and retention capacity and climate. Soil loss contributed to this estuary  $11.2 \text{ t}\cdot\text{year}^{-1}$  in the 2001 inventory whereas atmospheric deposition contributed with  $12.1 \text{ t}\cdot\text{year}^{-1}$ . Since no significant soil use or climate change occurred between 2000-2006, no significant changes in P emissions from these natural processes can be statistically predicted. The total natural load of P reaches  $23.9 \text{ t}\cdot\text{year}^{-1}$  ( $13.7 \text{ kg}\cdot\text{km}^{-2}\cdot\text{year}^{-1}$ ), with 55% from soil loss and 45% from atmospheric deposition. The relative contributions of natural sources to the total P load however, decreased from 13.1% in 2001 to 10.9% in 2006 (Table 1).

Their relative contribution of the anthropogenic P sources varies depending on the degree of urbanization, population size and extension of agricultural lands and shrimp pond area. Invariably, none of these activities' effluents receive any treatment before being released into the local environment. Non-mechanized agriculture present at the Jaguaribe River estuarine basin, is mostly dependent on crop type and size of cultivated area. Major P losses from the regional cultures are from cashew nuts, manioc and corn (Lacerda et al., 2008) and reach  $82.8 \text{ t}\cdot\text{year}^{-1}$  ( $47.7 \text{ kg}\cdot\text{km}^{-2}\cdot\text{year}^{-1}$ ). No significant changes in major crop types or cultivated areas occurred in the Jaguaribe river lower basin. Therefore, no difference in P emissions from agriculture occurred over the past 6-years period (Table 1).

Livestock in the Jaguaribe estuarine basin is reared extensively and is dominated by bovine. Therefore, the existing inventory considered that P emitted from this practice will always pass through soils prior to reaching rivers. Emission of P from the region's husbandry was  $10.7 \text{ t}\cdot\text{year}^{-1}$  in 2001 ( $6.2 \text{ kg}\cdot\text{km}^{-2}\cdot\text{year}^{-1}$ ). Unlike agriculture, increasing bovine and ovine cattle resulted in a 1.8 increase of P emission from this source to  $19.0 \text{ t}\cdot\text{year}^{-1}$  in 2006 (Table 1).

Urban areas are diffuse sources of P to surface waters. When no treatment plants exist, nutrient loads from this source are directly proportional to the population and the amount of water used per inhabitant as P concentrations in urban wastes, wastewater and runoff vary within a narrow range, resulting in emission factors (EFs) directly derived from population parameters (Smith et al., 1997). The population in the estuarine basin increased less than 10% from 2001 to 2006. Therefore, P emissions from wastewater varied little, increasing by about 5% only from  $42.5$  to  $44.7 \text{ t}\cdot\text{year}^{-1}$ . Urban runoff and solid waste disposal are relatively small sources of P compared to wastewaters, therefore any increase in P emission from these sources due to population increase is statistically undetectable, for

the short period considered and methods used, remaining at about 0.3 t.year<sup>-1</sup> only (Table 1).

Intensive shrimp aquaculture in the Jaguaribe River estuary uses large amounts of fertilizers and feed to maintain one of the highest productivity rates reported for the activity (about 6.2 t.ha.year<sup>-1</sup>). Super-phosphate is the major form of P fertilization with an application reaching 25 kg.ha<sup>-1</sup>.year<sup>-1</sup>, assuming the local typical food conversion rates of 1.5 to 1.8 (Lacerda et al., 2006). Despite the economic importance of the activity, there are few studies estimating P emission from these farms. However, since intensive shrimp farming procedures are similar worldwide, it is reasonable to use EFs estimated for other areas in the. Emission factors for P estimated that farms worldwide vary only 2.5 times reaching an average of about 20 kg.ha<sup>-1</sup>.year<sup>-1</sup>. This figure is very similar to the EFs derived from the Jaguaribe farms (Figueiredo et al.,

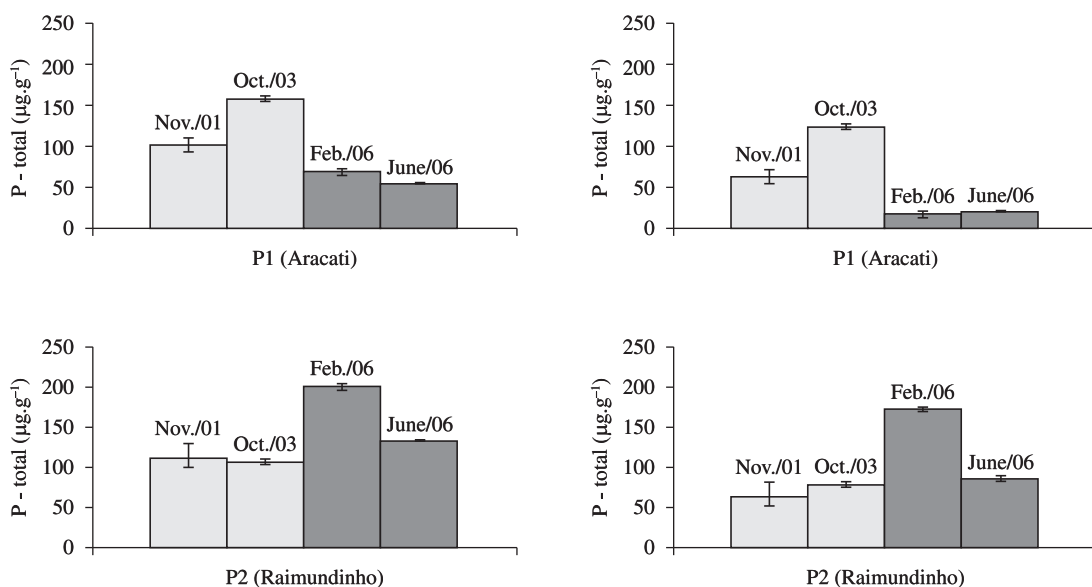
2005; Lacerda et al., 2006). From 2001 to 2006, P loads varied mostly following the increase in the total pond area. In 2004, however, increasing water renewal to prevent illnesses from 10 to 20% daily also resulted in larger EF. However, typical producing conditions were returned after 2004. The total P emission increased from 21.9 t.year<sup>-1</sup> in 2001 to 33.7 t.year<sup>-1</sup> in 2003 (54%) and 43.9 tP.year<sup>-1</sup> in 2006 (100%).

Marins et al. (2007) in a sedimentological survey of the Jaguaribe estuary characterized the sediments from the two sampling stations as siliciclastic (80-90% d.w.), suggesting a dominant contribution from terrestrial sources and particularly enriched in organic matter, especially at the station downstream from the creek receiving shrimp farm effluents (6.4 ± 0.2% d.w., n = 5) relative to the upstream station P1 (3.7 ± 0.4% d.w., n = 5). Figure 2 shows total and inorganic P in bottom sediments measured in 2001,

**Table 1.** Comparison of annual emission (t.yr<sup>-1</sup>), emission yields (kg.km<sup>2</sup>.yr<sup>-1</sup>) and relative contribution (RC %) of P for the Jaguaribe River Estuary, NE Brazil in 2001 and 2006.

Source category	2001			2006 <sup>a</sup>		
	Emission	Yield	RC	Emission	Yield	RC
Non-agriculture soils denudation	11.2	6.5	6.3	11.2	6.5	5.2
Atmospheric deposition	12.1	7.0	6.8	12.1	7.0	5.7
Agriculture	82.8	47.7	46.4	82.8	47.7	38.7
Husbandry	10.7	6.2	6.0	19.0	11.0	8.9
Wastewaters disposal	42.5	24.5	23.8	44.7	25.8	20.9
Urban runoff and waste disposal	0.3	0.2	0.2	0.3	0.2	0.1
Intensive shrimp farming	18.8	10.8	10.5	43.9	25.3	20.5
Total	178.4	102.9	100	214	123.5	100

<sup>a</sup> Updated using the newest available figure based on procedures in Lacerda et al. (2008).



**Figure 2.** Total and inorganic phosphorus distribution in sediments collected upstream and downstream the creek mouth receiving shrimp farming effluents at the Jaguaribe River estuary, NE Brazil, from 2001 to 2006. Error bars represents differences from duplicate samples (n = 4).

2003 and 2006 at the two sampling stations (see Figure 1 for location). Inorganic P is believed to better represent anthropogenic sources. Station P1, is located downstream from the major town of the estuary, Aracati, with nearly 70,000 inhabitants and showed total P ( $101 - 157 \text{ mg.g}^{-1}$ , in 2001-2003) and inorganic P ( $62 - 125 \text{ mg.g}^{-1}$ , in 2001-2003) concentrations in sediments actually lower in 2006 ( $18 - 55 \text{ mg.g}^{-1}$  and  $20 - 70 \text{ mg.g}^{-1}$ , for total P and inorganic P respectively), notwithstanding the inputs from wastewaters, agriculture and husbandry located upstream of this station. Downstream from the canal which drains the shrimp farms, P concentrations were higher from 2001 to 2003 ( $106 - 110 \text{ mg.g}^{-1}$  and  $65 - 78 \text{ mg.g}^{-1}$ , for total P and inorganic P respectively) than in 2006 ( $134 - 201 \text{ mg.g}^{-1}$  and  $86 - 174 \text{ mg.g}^{-1}$ , for total and inorganic P respectively). The observed difference between February (rainy season) and June (dry season) in 2006 may be explained by different sedimentary and hydrological dynamics between the two seasons and be responsible for the differences observed in P concentrations. However, more detailed studies are needed to clarify these differences. Partitioning between P species was also different at the two sites. Inorganic P was the dominant form in sediments downstream from the shrimp farm and increased from 67% to 75% between 2001-2003 and 2006. At station P1, inorganic P accounted for 70% of the total P in 2001-2003, but was lower (~30%) in 2006.

Phosphorous distribution in sediment cores collected downstream from the creek mouth receiving shrimp farm effluents close to station P2 confirms the results by confirming superficial sediments. The site presents an average sedimentation rate (excess  $^{210}\text{Pb}$  method) of  $0.31 \text{ cm.year}^{-1}$  (Marins et al., 2007). The vertical distribution of P concentrations is in agreement with the results found in the surface sediments. Concentrations of total P averaged  $170 \text{ mg.g}^{-1}$  in the core's bottom layer at 60-70 cm, around 170-200 years ago, roughly corresponding with the establishment of the first urban nucleus in the region. 10 cm deep from the surface, concentrations increased and remained relatively constant around  $185 \text{ mg.g}^{-1}$ . From 10 cm to the surface, roughly corresponding to the past two decades, total P concentrations increased further and reached an average of  $205 \text{ mg.g}^{-1}$ . Similarly, concentrations of inorganic P were relatively constant (average  $100 \text{ mg.g}^{-1}$ ) throughout the core from the bottom to about 10 cm of depth. In the superficial 5 cm layer (corresponding to the past 15 years), P concentrations increased by about 15% to  $115 \text{ mg.g}^{-1}$ .

Differences of concentration between upstream and downstream stations, as well as between the bottom and surface sediments of the core collected in station 2, reinforces the importance of aquaculture as the dominant P source for the estuary, even considering that the activity contributes with 20.5% of the total P emissions when all the other anthropogenic sources are taken into account. This suggests that releasing effluents directly into the estuary may be a determining factor for the importance of shrimp farming.

Biao et al. (2004) showed that inorganic P exports from shrimp farms in China is not diminished by sedimentation ponds resulting in a direct relationship between P concentrations in receiving creek waters and the pond management strategy, mostly the water renewable time. They also reported continuous increases in the P concentration after installing it in shrimp farms. Increasing P concentrations have also been recorded from five major aquaculture sites in the Philippines (David et al., 2009). Distribution along dated sediment cores provided evidence similar to our study showing aquaculture as recent, but already responsible for a significant part of the P contribution, relative to other anthropogenic sources and probably responsible for the increase in P concentrations observed in the sediments in the estuary.

Shrimp farm P emission increased from 10% to 20% of the total in the Jaguaribe estuary between 2001 and 2006, relative to the other anthropogenic P sources. The release of effluents directly to mangrove creek waters draining into the Jaguaribe River estuary makes shrimp farm effluents more important in terms of the total load of P to the estuary and is probably the main cause of the direct response to the increase in sediment P concentrations to shrimp farm areas. On the other hand, the release of P from wastewater in open air and/or underground aseptic sinks, and from agriculture and even husbandry whose emissions nearly doubled in the period, released in soils prior to reaching river waters, cannot be directly correlated to changes in the sediment P content.

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