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Relative importance of nitrogen and phosphorus emissions from shrimp farming and other anthropogenic sources for six estuaries along the NE Brazilian coast

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Abstract

Shrimp aquaculture is a recent development of the Brazilian agribusiness but has increased by about 20% per year during the last decade along the semi-arid NE coast due to the optimal climate and environmental setting. The activity has been blamed to cause several environmental impacts mostly associated with the emission of large amounts of N and P to estuaries. Here we estimate, using an emission factor approach, the annual emissions of N and P from intensive shrimp farming and other anthropogenic sources and natural processes to six estuaries along the coast of Rio Grande do Norte State, NE Brazil, where about 40% of the shrimp farming area of the country is located. Emission factors for natural sources (atmospheric deposition and soil runoff) are 1 to 2 orders of magnitude lower than those from anthropogenic sources. Shrimp farming presents the largest average emission factors for N (1.9 t km⁻² yr⁻¹), followed by agriculture (1.3 t km⁻² yr⁻¹) and husbandry (0.7 t km⁻² yr⁻¹). For P, largest average emission factors are from husbandry (0.9 t km⁻² yr⁻¹) and agriculture (0.34 t km⁻² yr⁻¹), although shrimp farming also presents a significant emission factor per unit of area for P (0.23 t km⁻² yr⁻¹). Wastewaters and solid waste disposal and urban runoff present much lower emission factors per unit of area, due to the low level of urbanization and small population of the basins. Anthropogenic emissions of N and P are 20 to 50 times higher than natural emissions. Agriculture contributes with the larger fraction (40% to 63%) of the total annual N load to three of the rivers, whereas P emissions are dominated by husbandry (64% to 74%). Wastewaters contribute with significant loads of N and P to the more populated basin only (35% and 11%, for N and P, respectively). Urban runoff is practically negligible in all basins, (less than 5% of the total emission), with the exception of Guamaré basin where 22% of the N emission come from this source. Aquaculture is the most important source of N to the Açu basin (58%), where the largest pond surface is observed. In the other basins N contribution from aquaculture ranges from 2% to 22%. Aquaculture contribution to the total P emission is small in all basins, varying from 2% to 14%. Notwithstanding the small area covered by shrimp farms relative to agriculture or husbandry, the location of farms adjacent to estuarine areas makes possible

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direct inputs to estuarine waters, while most other emissions go firstly to soils before eventually being transported to surface waters. In general, hydrochemical proxies of nutrient loads were consistent with the estimated loads. © 2005 Elsevier B.V. All rights reserved.

1. Introduction

Most coastal ecosystems are submitted to environmental impacts resulting from the excess of nutrient loads from diverse natural processes and anthropogenic activities taking place in their watersheds. These include changes in community structure and food webs, harmful algal blooms, excessive seaweed and epiphyte growth, low oxygen level, and reduced biodiversity (Bricker et al., 1999, 2003). Most of these impacts result from a complex chain of events varying in space and time, but that can be attributable to an ultimate pressure: the accumulation of excess nitrogen and phosphorus in fluvial water in its way to the ocean (NRC, 2003; Tappin, 2002).

Several studies have correlated nutrient loads, in particular of nitrogen, with primary production and the eventual onset of the eutrophication process, in a manner similar to the correlation between actual nutrient concentrations in water and this ecological response (Rosenberg et al., 1990; Boynton et al., 1995; Brunner, 1998; EPA, 2002; NRC, 2003). Therefore, the use of nutrient loads instead of concentrations, particularly in areas were detailed studies on nutrient concentrations are unavailable, such as in the semi-arid coast of NE Brazil, may turn into a satisfactory approach to evaluate the sensibility of estuarine systems to further introduction of anthropogenic effluents.

Along the semi-arid coast of NE Brazil, major anthropogenic sources of nutrients are the untreated disposal of sewage; solid wastes and waste waters, urban runoff, use of fertilizers and other chemicals in agriculture and aquaculture. Industrial effluents are a minor source of nutrients along this part of the Brazilian coast. Apart from these, natural processes' contributions to the nutrient loads are atmospheric deposition and soil runoff.

Shrimp aquaculture is a recent development of the Brazilian agribusiness but has increased by about 20% per year during the last decade, particularly along the semi-arid NE coast of Brazil due to the optimal climate and environmental setting. Shrimp farms in Brazil occupied less than 100 ha by 1998. By 2003, however, farm area has increased to over 14,000 ha, 11,050 ha along the NE coast and with an average production of about 66,000 t yr^{-1} (ABCC, 2003). Worldwide the activity has been blamed to cause several environmental

impacts (Twilley et al., 1999), mostly associated with the emission of large amounts of N and P to estuarine waters (Burford et al., 2003). Experimental results form shrimp farms in Australia estimated emissions factors of N and P varying from 0.99 to 1.18 kg ha⁻¹ day⁻¹ and 0.06 kg ha⁻¹ day⁻¹, respectively resulting in annual emissions of about 290 and 16 kg ha⁻¹ yr⁻¹, for N and P, respectively (Jackson et al., 2003; Burford et al., 2003). At the Gulf of California, Mexico, Páez-Osuna et al. (1999, 2003) estimated an annual input of N and P from shrimp farms of 112 and 32 kg ha⁻¹, respectively. However, although these numbers are significant, no study to our knowledge has compared shrimp farming N and P emissions with other anthropogenic and natural sources in a given area.

In the present study we estimate, using an emission factor approach, the annual emissions of N and P from different anthropogenic sources and natural processes to 6 estuaries along the coast of Rio Grande do Norte State, NE Brazil, where shrimp farming is most developed, harboring about 50% of the shrimp farming area of the country.

2. Material and methods

2.1. Study areas

Rio Grande do Norte state is the largest producer of cultivated shrimp in Brazil, with about 5245 ha of shrimp ponds (47% of the entire NE shrimp farm area), an average productivity of about 6.2 t ha⁻¹ yr⁻¹ and a total annual production of about 35,690 tons (ABCC, 2003). The activity is developed in estuarine areas of low urban and none industrial development, seeking good environmental conditions, particularly of water quality for its proper development. However, most urban wastes are not treated, while agriculture and husbandry have recently developed fast along the coastal region. Therefore, some scattered data already suggest that some of these areas are showing signs of incipient eutrophication, potentially threatening the activity (Guedes, 2003).

Fig. 1 shows a map of Rio Grande do Norte State and the six estuarine areas studied. The lower basins of the six rivers are located within the "Tabuleiros Costeiros do Nordeste" formation characterized by Tertiary and Quaternary sediments forming coastal plains constituted



Fig. 1. Map showing the locations of the studied river basins in Rio Grande do Norte, NE Brazil.

by sandy soils closer to the coast and yellow-red latosols (mostly oxisols) inland. Small stretches of alluvial eutrophic soils occur along river valleys (Pedreira, 1971; RADAM-BRASIL, 1981; Silva and Mendonça, 1971; Silva, 1996; Hidroservice, 1998; IDEMA, 1999a,b; Lima et al., 2000). Natural vegetation cover in most of the area has been converted to subsistence, nonmechanized agriculture and extensive husbandry. Major cultures in the sandy soils are coconut, cashew nut and banana, whereas latosols are used mostly for sugar cane and pasture. Population density is generally low and concentrated in small towns close to the sea. Table 1 shows the major environmental characteristics of the six estuaries studied. The coastline studied can be roughly divided into a dry stretch along the west coast, including the Apodi, Açu, Guamaré and part of the Ceará Mirim basins, with annual rainfall varying from 400 to 800 mm, and a more humid stretch including part of the Ceará Mirin basin, the Guaraíras and Curimataú

basins, with annual rainfall varying from 1100 to 1500 mm (Table 1).

The six estuarine systems differ by a factor of 2 to 5 in basin area, from the larger Apodi (975 km²) and Açu (950 km²) basins to the smaller Ceará Mirim basin (200 km²), but differ over two orders of magnitude in volume of the estuary from 777×10^3 m³ in the Caerá Mirim basin to $27,120 \times 10^3$ m³ at the Açu basin. Area to volume ratios, which fairly reflect the estuary's dilution capacity, range from 19, at the high dilution capacity Guamarés estuary, to 257 at the low dilution capacity Ceará Mirim estuary (Table 1).

2.2. Emission factors approach

Nutrient loads to non-industrialized coastal watersheds are mostly from diffuse sources and therefore, are dominated by the particulate phase dependent on unpredictable events, such as storms, when most of

Table 1

major environmental enauteensites and simmip production statistics of the six estables statice along the coast of relo orange do rorte, rel blazi							
Estuary	on Sł nts) ^c ar	hrimp farm œa (ha) ^d					
Apodi	8	374					
Açu	16	579					
Guamaré	2	414					
Ceará Mirim		30					
Guaraíras	7	747					
Curimataú	10	070					
Curimataú		10					

Major environmental characteristics and shrimp production statistics of the six estuaries studied along the coast of Rio Grande do Norte. NE Brazil

^aHydroservice (1998), IDEMA (1999a,b), ^bMaia (2004), ^cIBGE (2000), ^dABCC (2003).

the load is transferred to waterways. Therefore, nutrient load is a difficult variable to be directly measured, so indirect approaches based on emission factors and inventories of natural processes and anthropogenic activities dimensions are the most applicable strategy for their estimation (Tappin, 2002). Estimates of nutrient loads to a given estuarine watershed are achieved by using emission factors based on production/consumption parameters of the different anthropogenic sources and the chemical balance of natural processes (Koudstaal, 1987; Nriagu and Pacyna, 1988; Howarth et al., 1996; Howarth, 1998). Most necessary variables can be estimated from surveys and inventories of these activities, such as population parameters, urban and rural areas per basin, agriculture production and fertilizer use, among others. Emission factors have been successfully used to estimate nutrient and pollutant load at the local (e.g., Barcellos and Lacerda, 1994; Binner et al., 1996; Marins et al., 1998, 1999); regional (Lacerda et al., 1995; Howarth et al., 1996; Lacerda and Marins, 1997; Howarth, 1998; NRC, 2003; Vaisman and Lacerda, 2003); and global (Nriagu and Pacyna, 1988; Nriagu, 1989; Pirrone et al., 1996, 1998) levels and have been adopted as standard methodology by various environmental agencies (EEA, 1999; EPA, 2002). In general, emission factors (EFs) used in this study were those available in the literature for each activity or process. However, all EFs were adapted to local conditions whenever necessary. For example, wastewater production per inhabitant is, in general, calculated based on water consumption parameters typical of urban areas. However, under the semi-arid condition of most of the studied basins, these factors were corrected by the actual water consumption rate of the population of each basin (Doll and Hauschild, 2002). Similarly, fertilizer utilization, shrimp farming technology and other agricultural parameters used in the present study were those of the actual sites. General global emission factors were used when data for the actual site were unavailable.

An expedite survey of key environmental indicators of water quality (total phosphorus, nitrate, ammonium and dissolved oxygen concentrations) were performed in water samples from each estuary. The results were used to rank the different estuaries according to these parameters and to compare with the ranking based on the estimated N and P loads obtained by the emission factors approach. Five sampling points were established along the saline gradient and samples were collected using Van Dorn bottles at about 1.0 m from the surface in all estuaries. Sampling took place during low tide periods when fluvial influence is maximum. Under such conditions influence of river catchments would be strongest by avoiding dilution with seawater. Samples were stored frozen and analyzed using established protocols (APHA/AWWA/WPCF, 1995; Esteves, 1998; Silva, 2004).

3. Results and discussion

3.1. Natural emissions: soil runoff

Two major natural processes contribute with N and P to the studied basins: soil loss and atmospheric deposition. Soil loss is a significant source of nutrients to rivers; it is highly increased by agriculture and depends on soil type and climate. Losses from agricultural land can range from 116 to 309 t km^{-2} yr⁻¹ of soil under temperate climates (Schlesinger, 1997; Gouldie, 1987) to 60 to 760 t km^{-2} yr^{-1} in tropical regions and averages about 130 t km⁻² vr⁻¹ for low declivity areas with lack of mechanized agriculture (Greenland and Lal, 1977) such as the coastal plains of northeastern. Brazil Based on a review on soil loss rates for soil types and environmental settings similar to those found in our study area, Gouldie (1987) also proposed a similar average soil loss rate of 128 t km^{-2} yr⁻¹, which will be then used for the calculations in the present study.

Emissions of N and P associated with soil loss also depend on their actual concentrations present in each soil type. Concentrations of these nutrients in soils of the region studied (see Table 2) are available for many NE Brazilian coastal basins and soil types (Silva, 1996; Ramalho and Sobrinho, 2001; Ramalho et al., 2001). The average concentrations of N and P in coastal plain soils of northeastern Brazil were used to estimate soil N and P loss to rivers. These concentrations range from 500 to 900 mg g^{-1} and from 100 to 500 mg g^{-1} for N and P, respectively, depending on soil type. Finally, the estimates of N and P losses were corrected taking into consideration the retention capacity for N and P of the soil, which also considered that part of the N emissions can be directly transferred to the atmosphere through denitrification and dissimilatory reduction of nitrates under sub-oxic conditions in waterlogged soils (Burford et al., 2003). For P, however, most of it can be transferred to rivers associated with the particulate load and eventually to coastal areas (NRC, 2003). Retention rates of N and P for agriculture soils have been reported as 70% and 65% for N and P, respectively (Malavolta and Dantas, 1980; Silva, 1996). Altogether, Table 2 presents the estimates of N and P emissions to surface waters of the six basins studied based on the

Table 2

Basin	Soil type ^b								
	Sands	Solonetz	Alluvial	Podsol/Latosol	Cambisols	Total			
Apodi									
Soil area (km ²)	50	664	170	91					
Nitrogen	3.3	43	19.8	9.9		76			
Phosphorus	0.7	43	11	0.9		55.6			
Açu									
Soil area (km ²)	100	210	140	260	250				
Nitrogen	5.8	13.6	16.4	30	28	93.8			
Phosphorus	1.2	13.6	9.1	3.4	3.1	29.4			
Guamaré									
Soil area (km ²)	350			100					
Nitrogen	22.8			6.5		29.3			
Phosphorus	4.6			1.3		5.9			
Ceará-Mirim									
Soil area (km ²)	160			40					
Nitrogen	10.4			2.6		13			
Phosphorus	2.1			0.5		2.6			
Guaraíras									
Soil area (km ²)	200			125					
Nitrogen	13.0			8.2		21.2			
Phosphorus	8.2			2.6		10.8			
Curimataú									
Soil area (km ²)	113	150	37	60					
Nitrogen	7.3	9.4	4.3	7.0		28			
Phosphorus	1.4	9.4	2.4	0.8		14			

Estimates of N and P emissions (t yr^{-1}) through different soil types, weathering and soil loss^a found in the studied basins along the Rio Grande do Norte coast, NE Brazil

^aSoil loss rate: $128-130 \text{ t km}^{-2} \text{ yr}^{-1}$ (Greenland and Lal, 1977; Gouldie, 1987). ^bSoil type area from RADAM-BRASIL (1981), Hidroservice (1998) and IDEMA (1999a,b). ^cAverage soil N and P concentrations (mg g⁻¹), respectively: Distrophic Quartz Sands: 500 and 100 (Silva, 1996); Solonetz: 500 and 500 (Silva, 1996); Cambisols: 900 and 100 (Ramalho and Sobrinho, 2001); Alluvials: 900 and 500 (Ramalho et al., 2001).

parameters described above. Soil type and basin area were considered the two major factors controlling nutrient emissions. Alluvial and Cambisol are the richer soil units regarding N and P concentrations, followed by latosols, solonetz and quartz sands.

Range of N emissions from soil loss under temperate climate and non-mechanized agriculture are from 75 to 230 kg km⁻² yr⁻¹, with an average of 133 kg km⁻² yr⁻¹ (Howarth et al., 1996; Howarth, 1998), whereas P emissions range from 5 to 50 kg km⁻² yr⁻¹ (Howarth et al., 1996; Valigura et al., 2000) and up to 230 kg km⁻² yr^{-1} in the Amazon Basin (Howarth et al., 1995). Based on the total emission presented in Table 2, rates of N and P losses per unit of area vary little among the studied basins and are in general within the lower range reported by the above-mentioned authors for mechanized agriculture. The estimated loss per unit of area depends on soil type distribution rather than basin area. Losses of N vary from 97.7 kg km⁻² yr⁻¹ in the Acu basin to 65 kg km⁻² yr⁻¹ in the Ceará Mirim, the Guamaré and the Guaraíras basins. Losses of P vary from 56.4 kg km^{-2} vr^{-1} in the Acu basin to 13 kg km⁻² vr^{-1} in the Ceará Mirim, the Guamaré and the Guaraíras basins. When the total basin area is considered, however, the larger basins, Açu and Apodi, present the largest inputs of N and P, irrespectively of soil type.

3.2. Natural emissions: atmospheric deposition

Atmospheric deposition is a function of the basin area, the annual rainfall and the concentration of N and P in the bulk deposition (dry and wet). The fraction of the deposition eventually reaching surface waters will also depend on the retention rate of the atmospheric-derived N and P in soils (Golley et al., 1978; Johnson and Lindberg, 1998; Silva Filho et al., 1998). Total N and P deposition has been monitored in coastal areas worldwide, showing a wide range of values depending on the anthropogenic sources present in a give region. Along the Brazilian coast deposition rates of these nutrients also vary depending on the degree of urbanization and industrialization of the specific sector of the littoral. Along the Brazilian coast, total N and P atmospheric deposition ranges from 80 to 300 mgN m⁻² yr⁻¹, and 4

to 10 mgP m^{-2} yr⁻¹, over pristine and heavy industrialized sites, respectively and with an annual rainfall of about 1000 mm (Silva Filho et al., 1998; Mello, 2001, 2003). These ranges of values are similar to those found for other coastal areas in the world under similar development conditions (Schlesinger et al., 1982; Johnson and Lindberg, 1998; Brunner, 1998; Tan and Wong, 2000). Taking into consideration the local low level of industrialization and urbanization we used as best an average estimate 100 and 8 mg m^{-2} vr^{-1} , for N and P, respectively, which are similar to the averages reported by other authors for natural or lowdeveloped areas (Golley et al., 1978; Burns, 2004). The estimated deposition rates, however, were corrected for the actual annual precipitation of each basin and the average soil retention rates observed for these two nutrients. For example, Burns (2004) estimated an average of 63% atmospheric N retention based on results from eight Mid West USA basins. Golley et al. (1978) reported 70% retention of atmospheric P in soils in the Central American coast. In coastal Brazilian soils retention rates of N and P are similar to those (about 65% and 70% for N and P, respectively) (Malavolta and Dantas, 1980; Silva et al., 2000) and these figures are used in the present study. The fraction retained in soils, however, will be included in the calculation of inputs from soil runoff, since it makes up part of the soil concentrations of N and P.

Inputs to rivers from the atmosphere per unit of area estimated using the parameters above are 35 and 5.6 mg $m^{-2} yr^{-1}$ for N and P, respectively. As expected, total loads from atmospheric deposition resulted highly influenced by the basin area. The larger basins, Apodi and Açu receive about 24 and 1.6 t yr⁻¹ and 17 and 1.2 t

Table 3

Estimates of N and P inputs (t yr^{-1}) from natural sources to six estuaries along the coast of Rio Grande do Norte, NE Brazil

Estuarine basin	Soil runoff ^{a,d}		Atmospheric deposition ^{b,c}		Total natural input	
	N	Р	N	Р	N	Р
Apodi	76.0	55.6	24.0	1.6	100.0	57.2
Açu	93.8	29.4	17.0	1.2	111.0	30.6
Guamaré	29.3	5.9	12.0	0.7	30.0	6.6
Ceará Mirim	13.0	2.6	7.0	0.5	13.5	3.1
Guaraíras	21.2	10.8	14.0	1.2	32.2	12.0
Curimataú	28.0	14.0	12.0	1.1	40.0	13.1

^aFrom Table 2; ^bAverage N and P bulk atmospheric deposition (100 and 8.0 mg m⁻² yr⁻¹, respectively), based on concentrations in bulk atmospheric deposition from Mello (2001, 2003) and Silva Filho et al. (1998); ³Basin area and annual rainfall from Table 1; ⁴Soil retention rates of 65% and 70% for N and P, respectively, after Malavolta and Dantas (1980) and Silva (1996).

 yr^{-1} of N and P, respectively, whereas the smaller basin Ceará Mirim receives 7 to 14 and 0.5 to 1.2 t yr^{-1} of N and P, respectively (Table 3).

Natural loads of N and P from natural sources are summarized in Table 3. Notwithstanding the differences in N and P concentrations in soil types, basin area resulted the most important parameter controlling the natural loads of N and P to the basins studied. Maximum input was estimated for the larger Açu and Apodi basins (111 and 30.6 t yr⁻¹, and 110 and 57.2 t yr⁻¹ for N and P, respectively). In the smaller basins, Ceará Mirim and Guaraíras natural loads of N and P varied from 13.5 to 40 t yr⁻¹ and from 3.1 to 12 t yr⁻¹, for N and P, respectively. As expected, these loads are much smaller than those reported for more urbanized sites along the Brazilian coast (Mello, 2001, 2003), but are similar to those estimated for pristine environments (Golley et al., 1978; Burns, 2004).

4. Anthropogenic emissions of N and P to the lower basins of the studied rivers

The most important anthropogenic sources of N and P in the studied areas are agriculture, husbandry and aquaculture, urban wastewaters, disposal of solid wastes and urban runoff. Their relative contribution varies depending on the degree of urbanization, population and extension of agricultural lands. Invariably, none of these effluents receive any treatment before being released into the environment along this area of the Brazilian coast.

4.1. Agriculture

Leaching of agricultural soils has caused eutrophication in many water systems, from small catchments under subsistence agriculture to larger basins receiving effluents from mechanized agriculture, such as the Gulf of Mexico Basin receiving Mississippi River inputs (Rabalais, 2002). The fraction of N and P applied as fertilizers lost from agriculture soils range from 10% to 40% and 25% to 80% for clayey and sandy soils, respectively (Howarth et al., 1996). Emission factors for N and P loss from agriculture soils are available but mostly restricted to mechanized, large-scale agriculture and can reach 200 kg km⁻² yr⁻¹ (Sharpley and Syers, 1979; Sharpley and Rekolainen, 1997; Sharpley and Tunney, 2000). Unfortunately however, these emission factors cannot be applied to the agriculture practices taking place in our sites.

For non-mechanized agriculture average loss rates of P are in general much lower, N losses, however, are similar and are highly dependent on crop type (Silva et al., 2000). Results obtained from studies on this type of agriculture along the Brazilian coast provide loss rates relative to the amount of fertilizer applied varying from 6% to 20% for P and from 26% to 32% for N (Malavolta and Dantas, 1980; CFSEMG, 1989; Silva et al., 2000). The crop type also influences the loss rate of nutrients, since different cultures require different amount of fertilizer application. Table 4 summarizes the typical quantities of fertilizers used in agriculture in the northeastern Brazilian coastal region. Demands for N fertilization are particularly high for sugar cane (90 to 275 kg ha^{-1}), sweet potatoes (40–140 kg ha⁻¹), and citrics (120–180 kg ha^{-1}), for example. Whereas for P, particularly demanding cultures are beans and citrics (30-60 kg ha^{-1}), fruits (12-20 kg ha^{-1}) and corn (40-70 kg ha^{-1}) (Agrobyte, 2003; EMBRAPA, 2003; SEAGRI, 2003; CPAA, 2003). Major N and P losses from these cultures are from sugar cane and citrics (26-32% for N and 6-20% for P), followed by coconut, corn and beans (16-25% for N and 6-20% for P) (Malavolta and Dantas, 1980; CFSEMG, 1989). The estimated amount of nutrients loss in each basin is a function of these differences in culture type and their relative area of cultivation. As a result, the relatively small Curimataú basin (300 ha) receives more N from agriculture (749 t yr^{-1}), due to the dominance of sugar cane, than the larger Acu basin (950 ha), which receives only 45 t yr^{-1} of N, due to the relatively small cultured area and less-N demanding culture types typical of this basin (Table 5).

Table 4

Amount of N and P as fertilizer for the different cultures in the studied region and their respective loss rates

Crop	Fertilizer (kg	ha ⁻¹)	Loss (%)		
	N	Р	N	Р	
Sugar cane	90-275 ^{a,b}	20-40 ^{a,b}	$26 - 32^{h}$	6-20 ^h	
Beans	0-20 ^{e,f}	60 ^c	$16 - 25^{k}$	$0.7 - 1.4^{k}$	
Coconut	$40^{\rm e}$	$20^{\rm e}$	25 ¹	20^{1}	
Cassava	$0-40^{c,d}$	15-60 ^{c,d}	25 ¹	20 ¹	
Corn	20-60 ^{f,g,h}	$40 - 70^{g,h}$	26-32 ^h	$6 - 20^{h}$	
Banana	22 ^b	12 ^b	16 ^m	6 ^m	
Mango	40^{i}	20 ⁱ	16 ^m	6 ^m	
Sweet potatoes	$40 - 140^{j}$	20 ^j	$16 - 25^{k}$	$0.7 - 1.4^{k}$	
Citrics	$120 - 180^{i}$	$30 - 60^{i}$	$16 - 25^{k}$	$0.7 - 1.4^{k}$	
Pineapple	40^{i}	20^{i}	25 ¹	20^{1}	

^aAgrobyte (2003), ^bEMBRAPA (2003), ^cAbreu et al. (2003), ^dCPAA (2003), ^eFerreira (2003), ^fSilva et al. (2000), ^gCFSEMG (1989), ^hMalavolta and Dantas (1980), ⁱSEAGRI (2003), ^jDGPC (2003), ^kVollenweider (1968), ^lAverage limit for sandy soils after Howarth et al. (1996), ^mAverage limit for clay soils after Howarth et al. (1996).

Table 5

Estimated emissions	from ag	griculture	and	husbandry	for the	six	basins
studied (t yr^{-1})							

Basin	Agriculture ^a		Husba	Husbandry ^b		Total	
	Ν	Р	N	Р	N	Р	
Apodi	224	543	314	389	538	932	
Açu	45	111	88	90	133	156	
Guamaré	30	70	170	176	200	246	
Ceará Mirim	38	24	19	20	57	44	
Guaraíras	864	147	426	561	1290	708	
Curimataú	749	67	190	244	939	311	

^aAmount of N and P applied for each culture (see Table 4). Agriculture area and culture type (IBGE, 2000; IDEMA, 1999a,b). Average loss rates of applied N (30%) and P (15%) from Malavolta and Dantas (1980); CFSEMG (1989) and Silva et al. (2000). ^bAnimal stock from IBGE (1999). Volume of manure: kg animal⁻¹ day⁻¹, 10, 2.5, 1.0 and 0.18, for cows and horses, pigs, sheep and chicken, respectively (Boyd, 1971); Manure N and P concentrations in % of animal excrete: N=0.6, 0.5, 0.5 and 1.2; P=0.35, 0.3, 0.5 and 1.3 for cows and horses, pigs, sheep and chicken (Boyd, 1971; Esteves, 1998); Soil nutrient retention: N=65%; P=70% (NRC, 1993, 2003; Bouwman and Booij, 1998; Bouwman et al., 1997).

4.2. Husbandry

Husbandry is an important source of nutrients to soils and eventually to surface waters and can affect the hydrochemistry of estuarine areas. Nutrients are released to soils as animal excretes and their concentrations will depend on the concentrations found in animal feed and on the different animals grown in a given region. According to Boyd (1971) and Esteves (1998), average amounts of excrete ranges from 10 kg animal⁻¹ day^{-1} , for cows and horses, to 2.5 and 1.0 kg animal⁻¹ day^{-1} , for pigs and sheep, and chicken, respectively. Nitrogen and P concentrations in animal excrete are relatively constant varying from 0.6% for cows and horses and 0.5% for pigs and sheep for N; and 0.35%, 0.3% and 0.5% of P for cows and horses, pigs and sheep, respectively. For chickens, with excrete production of about 0.18 kg animal⁻¹ day⁻¹, N and P contents are about 1.2% and 1.3%. Once deposited in soils, rates of nutrient retention and uptake by plants will determine the fraction eventually released to waters, which are similar to those described for N and P retention of the natural inputs (see Table 2) (NRC, 1993; Bouwman et al., 1997; Bouwman and Booij, 1998; NRC, 2003). Although some livestock are reared in closed premises the large majority of the region's husbandry is extensive and dominated by bovine. Therefore, we considered that nutrients emitted from this practice will always pass through soils prior to reaching rivers.

Emissions of N and P to the studied estuaries from the region's husbandry are independent of basin area and ranged from 19 and 20 t yr^{-1} , for the Ceará Mirim basin, to 426 and 561 t yr^{-1} , for the Guaraíras basin, for N and P, respectively, being dominated by bovine cattle due to its larger number and amount of manure produced per capita. Nitrogen load is relatively smaller than the P load mostly due to ammonium loss to the atmosphere. It is important to note that manure is frequently used as fertilizers, which eventually may result in some export of nutrients in farm products out of our basins. This loss, however, is probably very small since this practice is typical of subsistence, familiar agriculture and is not estimated here.

4.3. Wastewaters

Wastewaters are one of the major sources of nutrients to coastal areas, particularly in urbanized estuaries. When no treatment plants exist, nutrient load from this source is directly proportional to population and the amount of water used per inhabitant (Smith et al., 1997), since N and P concentrations in wastewater vary within a narrow range (I.C. Consultants, 2001). Therefore emission factors can be easily derived from population parameters to estimate inputs from wastewater released to estuaries (Smith et al., 1997; Howarth, 1998; Bidone, 2000; Bidone and Lacerda, 2002; Martinelli et al., 2002). In our estimates we used the latest population census available for the year 2000 and a detailed inventory of water use by the local population published by Doll and Hauschild (2002). This survey showed water consumption varying from 82 to 125 (average of 85) L inhab⁻¹ day⁻¹, in rural areas along the coast, to 100 to 150 (average of 115) L $inhab^{-1} day^{-1}$ in urban areas, at the six studied basins. Nutrient concentrations in wastewater were those suggested by von Sperling (1996) based on Brazilian conditions. Finally, the assumption of no-treatment prior to release was used, since the availability of sewage treatment in the northeastern region of Brazil reaches only 10% of its population, basically restricted to metropolitan areas of state capitals, not included in this study.

The estimates presented in Table 6 shows N inputs varying from 11 to 534 t yr⁻¹ and P inputs varying from 1 to 188 t yr⁻¹, with maximum values at the Apodi basin and minimum in the low populated Ceará Mirim basin. Comparisons of the emission factors calculated for this study (4–8 g inhab⁻¹ day⁻¹ and 0.6–2.9 g inhab⁻¹ day⁻¹, for N and P, respectively) with others from the literature show our values in the lower range of reported factors, since emissions are much lower than those reported for urbanized sites, due to the higher water

Table 6

Estimated emissions from wastewaters and urban runoff, including leaching of solid wastes disposal sites for the six basins studied (t yr^{-1})

Basin	Waste waters ^a		Urban runoff ^b		Total	
	Ν	Р	N	Р	N	Р
Apodi	267-534	37-188	48	7	449	120
Açu	64-128	9-45	7	1	103	27
Guamaré	11-23	1 - 8	76	11	93	15
Ceará Mirim	21-43	3-15	<1	<1	33	10
Guaraíras	64-129	9-45	10	2	99	29
Curimataú	38-76	5-27	7	1	64	17

^aNutrient concentrations in waste waters: $N=35-70 \text{ mg L}^{-1}$; $P=5-25 \text{ mg L}^{-1}$ (von Sperling, 1996). Population data from IBGE (2000). Water consumption: 85 L inhab⁻¹ day⁻¹, in rural areas and 115 L inhab⁻¹ day⁻¹ in urban areas of NE Brazilian coastal area (Doll and Hauschild, 2002).

^bPopulation parameters from IBGE (2000). Annual rainfall from Hydroservice (1998). Includes leaching of solid wastes disposal sites, solid waste production per inhabitant from ABES (1983). Average constructed housing unit of 50 m². Average N and P concentrations in runoff from NRC (2003) and Binner et al. (1996).

consumption rates verified in metropolitan regions compared with the predominantly rural areas studied here. (Howarth, 1998; Bidone, 2000; Bidone and Lacerda, 2002; Martinelli et al., 2002).

4.4. Urban runoff and solid waste disposal

Large metropolitan areas can be significant as diffuse sources of contaminants to the environment, providing large quantities of contaminants and nutrients to surface waters through leaching of urban surfaces and solid wastes disposal. Major parameters controlling nutrient loss by runoff from urban areas are the area of impermeable surfaces, the number and dimensions of habitations and annual rainfall. For solid wastes disposal, the main parameters controlling contaminant emissions are population size and per capita production of solid wastes and their disposal method (Davis et al., 2001), as well as the average concentrations of N and P in runoff waters from landfill sites and the local rainfall levels (NRC, 2003). In the studied basins, large urbanized areas are absent. Also, most villages and cities present low levels of soil impermeability. Solid waste disposal is mostly restricted to solids present in wastewaters, already mentioned. Therefore, N and P emissions from this source are relatively small, ranging from <1 kg yr⁻¹ (for both nutrients) in the Ceará Mirim basin to 76 and 11 kg yr^{-1} , in the Guamaré basin, for N and P, respectively (Table 6), representing less than 10% of the emissions from wastewaters.

4.5. Aquaculture

Intensive shrimp farming is a major activity along the studied coast, harboring about 5000 ha of operating ponds in 2003, although up to 10,000 ha of ponds exist in the area under different states of construction or waiting authorization from environmental authorities to start operations (ABCC, 2003). The area covered by the present study includes about 4814 ha of ponds, which accounts for nearly 92% of the total operating shrimp farming area of Rio Grande do Norte State. This activity, when developed under non-sustainable conditions, has been related to negative environmental impacts, particularly when displacing mangrove areas (Twilley et al., 1999; Lacerda, 2002). Shrimp farms are large exporters of nutrients to the coastal environments (Burford et al., 2003). Intensive shrimp culture in Rio Grande do Norte uses large amounts of fertilizers and feed to maintain one of the highest productivity rates reported for the activity (about 6.2 t ha yr^{-1}) (ABCC, 2003). Typical fertilizers application reaches 40 and 10 kg ha^{-1} production cycle⁻¹ (100 and 25 kg ha⁻¹ yr⁻¹) of urea and superphosphate, respectively, assuming food conversion rates of 1.5 to 1.8 (Nunes, 2001).

Notwithstanding the economic importance of the activity, there is no study estimating the emission factors of nutrients from these farms. However, since there is a tendency to uniform shrimp farming processes worldwide, it is reasonable to use emission factors estimated for other areas in the world. Comparatively, we calculated emission factors for the local farms by analyzing monitoring data from a one-year period in a typical producing farm in Ceará State, NE Brazil (Abreu et al., 2003). Unfortunately monitoring data included total P, but only the dissolved inorganic species of N $(NO_3^-, NO_2^- \text{ and } NH_3 + NH_4^+)$ hampering the direct calculation of N emission factors, since up to 70% of the N present in the effluent could be in the form of organic particulate N (Burford et al., 2003). Therefore, we corrected the values of the monitored N by a factor of 3.3 to estimate the emission factor from shrimp farms.

Total excess P concentrations in outgoing waters during the monitoring year varied from 0.06 to 0.18 mg L^{-1} . Considering the water renewal time of the farm, which generally varies from 5% to 10% per day, the estimated average emission factor for P was about 0.05 kg ha⁻¹ day⁻¹ (14 kg ha⁻¹ yr⁻¹, considering 2.5 production cycles per year, the typical number of production cycles of shrimp farms of the NE Brazil). Measured excess N concentrations in effluent waters (including NO₃⁻, NO₂⁻ and NH₃+NH₄⁺ only) varied from 0.16 to 0.39 mg L⁻¹ (Abreu et al., 2003), resulting in total N concentrations, when corrected to include particulate organic N, of 0.52 to 1.29 mg L^{-1} . Under these conditions, the studied farm showed an average emission factor for N of about 0.47 kg ha⁻¹ day⁻¹ resulting in an annual emission of about 110 kg ha⁻¹.

Experimental data from Australian farms reported total N and P concentrations in effluent waters of 0.98 and 0.13 mg L⁻¹, respectively (Burford et al., 2003), in farms with the same technology used in our study area. These concentrations resulted in emission factors of 0.99–18 and 0.06 kg ha⁻¹ day⁻¹, and annual emissions of 290 and 16 kg ha⁻¹, for N and P, respectively (Jackson et al., 2003; Burford et al., 2003). In the Gulf of California, Mexico, Páez-Osuna et al. (1999, 2003) estimated N and P export from the local shrimp farms of about 112 and 32 kg ha⁻¹ yr⁻¹, respectively. A comparison of existing N and P emission factors for intensive shrimp farming is presented in Table 7.

The results presented in Table 7 suggest a good consistency of the estimated emission factors of N and, in particular for P for shrimp farms in different parts of the world, an expected result due to the similarity of the technological processes used. In fact, we consider the emission factors for this source are the most reliable among all other sources evaluated in this study. Whereas for most other sources typical emission factors range by a factor of 10 to 100 (Nriagu and Pacyna, 1988; Nriagu, 1989), emission factors for shrimp aquaculture vary only 2 to 3 times. Based on this range of emission factors, it was possible to estimate the total N and P contribution from shrimp farming to the different basins studied in Rio Grande do Norte State (Table 8). Emissions of N and P varied directly according to the total pond area in each basin. Nitrogen and P emissions

Table 7

Comparison of emission factors (kg $ha^{-1} day^{-1}$) of N and P from shrimp farms based on instantaneous data on nutrient concentrations in effluent waters

Location	Ν	Р
Ceará State, Brazil ^{a,b}	0.47 ^c	0.05 ^d
Australia ^e	$1.08^{\rm f}$	0.06
Gulf of California, Mexico ^g	0.58	0.12
Range of emission factors	0.33-1.08	0.05-0.12

^aThis study and Abreu et al. (2003). ^bWater renewal rate 5% to 10% dia⁻¹, pond depth 1.0 m, volume exchanged: 5×10^5 1 ha⁻¹ day⁻¹ (Nunes, 2001; Abreu et al., 2003). ^cN estimates based on approximate speciation given in Burford et al. (2003), i.e., 70% of the total N being particulate organic N and actual concentrations of NO₃, NO₂ and NH₄ (Abreu et al., 2003). ^dBased on actual concentrations in effluent waters monitored during one year (Abreu et al., 2003). ^gPaez-Osuna et al. (1999, 2003).

Table 8 Estimates for N and P emissions from shrimp farms^a to the studied estuaries in the State of Rio Grande do Norte, NE Brazil ($t yr^{-1}$)

Basin	Pond area ^b (ha)	N emission ^{c,d}	P emission ^d
Apodi	874	109-357	12-28
Açu	1.679	208-686	22-54
Guamaré	414	52-169	6-13
Ceará Mirim	30	3.7-12.3	0.4 - 1.0
Guaraíras	747	92-305	10-24
Curimataú	1.070	133-405	14-35

^aFrom emission factors presented in Table 5, and average production cycle per year of 2.5 and 30 initial days without water exchange and further exchange volume of 5% per day, from ABCC (2003) and Nunes (2001). ^bFrom ABCC (2003). ^cAssuming 70% of the total N in the effluent discharge being particulate organic N. ^dFrom emission factors in Table 7.

vary from 3.7 to 12.3 tN yr^{-1} and 0.4 to 1.0 tP yr^{-1} at the Ceará Mirim basin, with only 30 ha of ponds, to 208 to 686 tN yr^{-1} and 22 to 54 tP yr^{-1} at the larger Açu basin, which harbors about 1679 ha of ponds. Considerable emissions from shrimp farms also occur in the Curimataú and Apodi basins.

5. Discussion

Table 9 compares the emission factors, standardized to the same units per area, of the different natural and anthropogenic sources of N and P obtained under the specific conditions of the six basins studied. Emission factors for natural sources (atmospheric deposition plus soil runoff) are 1 to 2 orders of magnitude lower than those from anthropogenic sources. Aquaculture presents the largest average emission factors for N (2.67 t km^{-2} yr^{-1}), followed by agriculture (1.3 and t km⁻² yr⁻¹) and husbandry (0.7 t km⁻² yr⁻¹). For P, largest average emission factors are from husbandry (0.9 t $\text{km}^{-2} \text{ yr}^{-1}$) and agriculture $(0.34 \text{ t km}^{-2} \text{ yr}^{-1})$, although aquaculture also presents a relatively high emission factor (0.23 t $km^{-2} yr^{-1}$). Wastewaters and urban runoff, including solid waste disposal present much lower emission factors per unit of area, as a result of the low level of urbanization and low population of the basins studied.

The total loads of N and P for each studied basin are presented in Table 10. Anthropogenic emissions of N and P surpass natural emissions (Table 3) in all basins, being up to 50 times higher for N in the Guaraíras basin, for example. Similarly, anthropogenic P emissions are 10 to 20 times higher than natural emissions in the same Guaraíras basin. Among the anthropogenic sources, notwithstanding the high emission factors observed for aquaculture, the larger loads of nutrients are due to agriculture and husbandry, which occupy much larger areas. Agriculture contributes with the larger fraction of the N load to three of the rivers: Curimataú with 59%, Guaraíras (54%) and Ceará Mirim (39%), whereas P emissions from agriculture dominate the total P load in the Apodi (51%), Açu (42%) and Ceará Mirim (44%). Husbandry dominates the N emission in the Guamaré basin only (43%), but is the major source of P to the Guaraíras (74%), Curimataú (69%) and Guamaré (64%) basins. Wastewaters contribute with significant loads of N and P to the more populated basin (Apodi) which harbors over 240,000 inhabitants (34% and 11%, for N and P, respectively) and the Ceará Mirim basin, 33% and 18% for N and P, respectively. Urban runoff is practically negligible in all basins, representing less than 5% of the total emission, with the exception of the Guamaré basin where 28% of the N emission come from this source. Aquaculture is the most important source of N to the Acu basin (65%), where the largest pond surface is observed. In all other basins N contribution from aquaculture range from 28% at the Guamaré basin, to only 8% at the Ceará Mirim basin. Aquaculture contribution to the total P emission is small in all basins, varying from 2% at the Apodi and Ceará Mirim basins to 14% at the Acu basin. However, although the area covered by shrimp farms is small relative to agriculture or husbandry, the location of farms adjacent to estuarine areas makes possible direct inputs to estuarine waters, while most other emissions go firstly to soils before eventually being transported to surface waters.

In an attempt to compare the estimates obtained in this study we ranked the six estuaries studied according to the concentrations of selected environmental indicators (total N and P concentrations in waters) and estuary volume with the P and N yield (the load per km²) for each basin (Table 11). This evaluation is not expected to result in linear relationships between nutrient yields and their concentrations in the water column, due to the many variables affecting the transport of nutrients from sources to estuarine waters. However, if the estimates presented

Table 9

Range and average (in parenthesis) of emission factors from the different anthropogenic sources of Nitrogen and Phosphorus standardized to t km⁻² yr⁻¹, for the specific conditions existing in the six estuarine basins studied along the coast of Rio Grande do Norte State, NE Brazil

Source	Ν	Р
Natural sources	0.05-0.09 (0.07)	0.01-0.06 (0.04)
Waste water	0.03-0.41 (0.22)	0.01-0.12 (0.06)
Husbandry	0.09-1.31 (0.70)	0.09-1.73 (0.91)
Agriculture	0.05-2.65 (1.35)	0.12-0.56 (0.34)
Urban runoff	< 0.01-0.14 (0.07)	< 0.01-0.02 (0.01)
Aquaculture	1.25-4.09 (2.67)	0.13-0.32 (0.23)

Table 10		
Average estimates of N and P emissions (t yr^{-1})	from anthropogenic sources in the studied basin along the Rio Grande do Norte coast, NE	Brazil

Basin	Source								
	Waste waters	Husbandry	Agriculture	Urban runoff	Aquaculture	Total			
Apodi									
Nitrogen	400 (34)	314 (26)	224 (18)	48 (4)	233 (18)	1219			
Phosphorus	112 (11)	389 (36)	543 (51)	7 (<1)	20 (2)	1061			
Açu									
Nitrogen	97 (14)	88 (13)	45 (7)	7 (1)	447 (65)	684			
Phosphorus	27 (10)	90 (34)	111 (42)	1 (<1)	38 (14)	267			
Guamaré									
Nitrogen	17 (4)	170 (43)	30 (8)	76 (19)	111 (28)	394			
Phosphorus	5 (2)	176 (64)	70 (25)	11 (5)	9 (4)	271			
Ceará-Mirim									
Nitrogen	32 (33)	19 (19)	38 (39)	<1 (<1)	8 (8)	98			
Phosphorus	9 (18)	20 (36)	24 (44)	<1 (<1)	1 (2)	55			
Guaraíras									
Nitrogen	97 (6)	426 (27)	864 (54)	10(1)	199 (12)	1596			
Phosphorus	27 (4)	561 (74)	147 (19)	2 (<1)	17 (3)	754			
Curimataú									
Nitrogen	57 (4)	190 (15)	749 (59)	7 (1)	269 (21)	1272			
Phosphorus	17 (5)	244 (69)	67 (19)	1 (<1)	25 (7)	354			

The relative contribution (%) of each individual source appears in parenthesis.

here are correct, at least the basins receiving the largest emissions per area and lower dilution capacity should present higher nutrient concentrations in their waters.

Nutrient concentrations in surface waters of these estuaries were quite variable. The highest average concentrations of total dissolved N and P were measured in the Guaraíras estuary (7.86 μ molP L⁻¹ and 6.3 μ molN L⁻¹), followed by the Ceará Mirim (7.32 μ molP L⁻¹ and 1.2 μ molN L⁻¹). The Guaraíras basin showed the largest nutrient yield, whereas the Ceará Mirim, although presenting the smallest nutrient yield, shows a very small dilution capacity due to small a volume and very high area to volume ratio (Maia, 2004). Apart from

the Ceará Mirim estuary, which nutrient concentrations seems dependent on hydrological parameters rather than the nutrient emissions, the estimated emissions are in agreement with the nutrient concentrations reported for these estuaries. The lack of a direct relationship of each individual nutrient source, including aquaculture with direct discharges at the estuary, suggests that nutrient levels are a function of the integration of all sources in a given basin and their hydrological characteristics. Therefore, the approach used in the present study seems to be useful for application in areas, such as northeastern Brazil, where actual concentration data are scarce or non-existent.

Table 11

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Basin	Estuarine volume $(\times 10^6 \text{ m}^3)$	Area/volume ratio (m ² /m ³)	Total P input rate (kg ha ⁻¹)	Total N input rate (kg ha^{-1})	P input from aquaculture (t yr ⁻¹)	N input from aquaculture $(t yr^{-1})$	Total P $(\mu mol L^{-1})$	Total P (µmol L ⁻¹)
Apodi	12.9	10.9	10.9	15.1	20	233	0.78 ± 0.24	$0.28 {\pm} 0.25$
							0.4 - 1.0	0.1 - 0.7
Açu	27.1	2.8	2.8	7.8	38	447	1.58 ± 0.81	0.18 ± 0.04
							0.5 - 2.7	0.1 - 0.2
Guamaré	23.5	4.9	4.9	8.6	9	111	0.7 ± 0.1	0.22 ± 0.18
							0.6 - 0.8	0.1 - 0.5
Ceará Mirim	0.8	2.8	2.8	6.2	1.0	8	7.32 ± 0.27	$0.56 {\pm} 0.5$
							6.9-7.6	0.1 - 1.4
Guaraíras	2.4	23.2	23.2	61.6	17	199	$7.86 {\pm} 2.87$	$2.58 {\pm} 0.5$
							5.4-12.6	1.9-3.1
Curimataú	17.5	11.8	11.8	51.7	25	269	1.03 ± 0.7	_
							0.9 - 1.7	

6. Conclusions

The results presented here show that even in areas of low human occupation such as the NE semi-arid coast of Brazil, anthropogenic emissions of N and P surpass natural emissions in at least one order of magnitude. Although all studied basins present low population densities, inputs from agriculture, husbandry and shrimp farming are considerable and can alter the water quality of the receiving estuaries. Intensive shrimp farming presents the largest emission factors for N and P per unit of area in the region studied. However, only in one estuary (Acu), where pond area reaches 1680 ha, it is the most important source of N. Shrimp farm contribution to the total P emission is relatively small in all estuaries. Since emissions from this source are directly disposed to estuarine waters the response of coastal ecosystem metabolism to shrimp farm effluents, however, may be more rapid than from other sources.

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