Environ Monit Assess (2008) 141:149–164
DOI 10.1007/s10661-007-9884-y

Estimating the importance of natural and anthropogenic sources on N and P emission to estuaries along the Ceará State Coast NE Brazil

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Received: 2 August 2006 / Accepted: 10 August 2007 /

Abstract The Northeastern semi-arid Brazilian region is experiencing rapid social and economic development based on improving water management and even in areas of low human occupation, anthropogenic emissions of N and P surpass natural emissions in at least one order of magnitude and these additional loads can alter the water quality of the receiving estuaries. This study estimates, using an emission factor approach, the annual emissions of N and P from natural processes and anthropogenic sources for estuaries along the Ceará State, NE Brazil. Emission factors from natural sources are one to two orders of magnitude lower than those for anthropogenic sources. Among the anthropogenic activities, the aquaculture is responsible for most N emission (0.52 t km^{-2} year^{-1}) followed by waste water and husbandry. For P, the largest average emission factors are from husbandry (0.30 t km^{-2} year^{-1}), waste water and agriculture.

Keywords Nitrogen · Phosphorous · Emission factor · Estuary · Northeastern semi-arid Brazil

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Introduction

The problems caused by nutrient over-enrichment in coastal areas are significant and likely to increase as human interference in the structure of the watershed/coastal zone continuum continues to intensify (NRC 1993, 2003). Presently, some coastal areas of NE Brazil are undergoing a rapid development based on river management which supports increasingly economical activities such as irrigation, agriculture, aquaculture, and urbanization. As human populates the watershed/coastal continuum, the deterioration of the such coastal environments has become a critical issue (Marins et al. 2002; Gaiser et al. 2003; Marques et al. 2004).

Preliminary studies carry out in some drainage basins of the region showed that even in areas of low human occupation such as the NE Brazilian coast, anthropogenic emissions of N and P surpass natural emissions in at least one order of magnitude and these additional loads can alter the water quality of the receiving estuaries (Lacerda et al. 2006). However, the lacks of detailed studies on nutrient concentration for most coastal areas from NE Brazil limit our understanding on nutrient over-enrichment in coastal waters. Thus, using nutrient loads instead of concentrations is a preliminary step to scale up and quantify the importance of natural and anthropogenic sources of nutrient emission to the coastal zone, and furthermore, to evaluate the sensibility of estuarine system to additional introduction of anthropogenic nutrient inputs.

In the present study we estimate, by using an emission factor approach, the annual emissions of N and P from different anthropogenic and natural sources to the estuaries along the coast of Ceará State, NE Brazil, which are presently witnessing most land use changes due to river management increasing sustainability of the economic development of these areas but on the other hand, potentially affecting water quality of the receiving coastal areas.

Material and methods

Study area

Figure 1 shows a map of Ceará State and the fifteen lower river basins and estuarine areas studied. The area is under an equatorial/semi-arid climate with annual rainfall at the coast varying from 828 to 1,428 mm (Table 1; FUNCEME 2005). The estuarine basins of the rivers are located within the “Tabuleiros Costeiros do Nordeste” formation characterized by Tertiary and Quaternary sediments forming coastal plains constituted 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; Lima et al. 2000). Most of rivers flowing into the studied estuaries are ephemeral or during dry season the freshwater inflow is much reduced. Generally, the freshwater discharge to most estuaries is also highly regulated by dam building across the drainage basins (Molisani et al. 2006).

Natural vegetation cover in most of the area has been converted to subsistence, non-mechanized agriculture and extensive husbandry. Major cultures in the sandy soils are cashew nut and coconut whereas the latosols are mainly used to bean and corn cultures and pasture. The human population is sparsely distributed along rural areas and small towns close to the sea though large density population centers are observed, for example, the Cocó river basin is located within a 2.5 million-inhabitant metropolitan area (Table 1). Total population along the coast increased from 6.8 millions in 1996 to about 8 millions in 2005 (IBGE 2006). The principal economic activities of estuarine areas in Ceará state become significant only during the past 10 years. First, shrimp aquaculture bloomed after 1998 to about 3,279 ha of shrimp ponds and total annual production of about 24,700 t (ABCC 2003). Second, tourism which also increased about 50% in the past 10 years reaching over 3 millions visitors in 2005. Agriculture and husbandry have recently grown fast along the coastal region based on increasing water availability due to river damming. Most urban and agribusiness wastes are not treated and therefore some scattered data already suggest

Fig. 1 Map of studied estuarine basins across the Ceará State

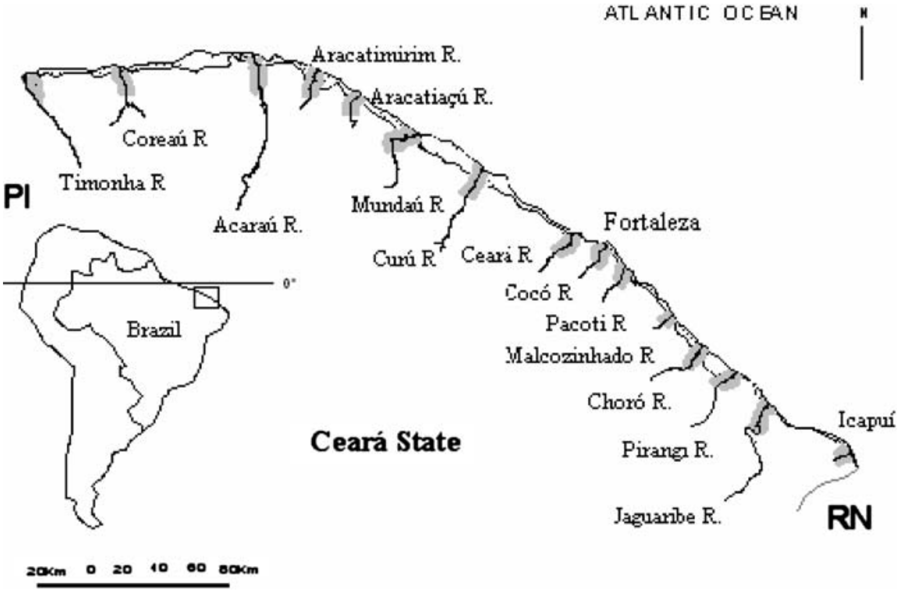

Table 1 Major environmental settings of the lower river basin and respectively estuaries studied along the coast of Ceará state, NE Brazil

	Basin area (km^2)[a]	Estuarine Volume (10^6 m^3)[a]	Area to volume ratio (m^2 m^{-3})	Annual rainfall (mm)[b]	Population (inhabitants)[c]
Timonha	616	39.7	15	1,122	26,084
Acaraú	3,002	1.36	2,213	1,083	132,082
Coreaú	4,678	7.66	610	1,092	123,913
Aracatiaçú	1,582	1.19	1,333	828	32,333
Aracatimirim	738	0.19	3,925	1,140	30,347
Curú	600	0.49	1,229	1,238	53,003
Mundaú	2,135	2.13	1,000	1,359	138,896
Ceará	1,640	0.41	4,000	1,311	405,344
Cocó	420	0.35	1,217	1,378	2,521,444
Pacoti	700	0.48	1,470	1,428	148,113
Malcozinhado	76	0.05	1,617	931	14,951
Choró	820	0.28	2,928	1,331	57,129
Pirangi	1,627	0.46	3,575	914	42,343
Jaguaribe	1,735	13.6	127	1,102	79,832
Icapuí	430	0.06	6,935	950	16,052

[a] ZEE (2005)

[b] FUNCEME (2005)

[c] IBGE (2006)

that some of these estuaries are showing signs of incipient eutrophication (Moreira 1994; Molisani et al. 2007).

The fifteen estuarine systems differ by a factor of about eight in basin area, from the larger Coreaú (4,678 km^2) basin to the smaller Malcozinhado basin (76 km^2). The magnitude of the estuarine volume ranges from 39×10^6 m^3 in the Timonha to 0.05×10^6 m^3 at the Malcozinhado. Basin area to volume ratios fairly reflects the estuary's dilution capacity (Bricker et al. 1999) and range from 6,935 at the low dilution capacity Icapuí estuary to 15 at the high dilution capacity Timonha estuary (Table 1).

Emission factors approach

Emission factors (EF's) rely on the correlation of emissions with one or more underlying performance parameters. This approach can be a useful tool for the nutrient emission estimates mainly for non-industrialized coastal watersheds where nutrient loads are mostly from diffuse sources and so difficult to be directly measured. Thus indirect approaches based on emission factors and inventories of natural processes and anthropogenic activities dimensions are the most applicable strategy for emissions estimation (Tappin 2002).

Typical performance factors include production/consumption parameters of the different anthropogenic sources and the chemical balance of natural processes (Koudstaal 1987; Nriagu and Pacyna 1988; Nriagu 1989; Howarth et al. 1996; Howarth 1998). Most necessary variables can be obtained 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, 2006; 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; US EPA 2002). In general, EF's used in this study were those available in the literature for each activity or process. However, all EF's were adapted to local conditions whenever necessary. Under the semi-arid conditions, typical consumption parameters for urban areas cannot be applied and thus, 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.

Results

Natural emissions: soil runoff

Soil losses are important natural process contributing with N and P transfers to coastal areas through the fluvial transport. This process is influenced by soil type and climatology. Soil loss is highly increased by urbanization and agriculture practices occurring through a drainage basin. Losses from urban and agricultural soils can reach rates of 116 to 309 t km^{-2} year^{-1} in areas under temperate climate (Schlesinger 1997) and 60 to 760 t km^{-2} year^{-1} in tropical regions (average of 128 t km^{-2} year^{-1}; Goudie 1987). An average of about 130 t km^{-2} year^{-1} was reported for low declivity areas with lack of mechanized agriculture in tropical Africa (Greenland and Lal 1977). This environmental setting is similar to the coastal plains of northeastern Brazil. Thus, average soil loss rate of 128 t km^{-2} year^{-1}, as proposed by Goudie (1987), will be used for the calculations of N and P emissions from soil loss in the present study.

The concentrations used to estimate soil N and P losses to rivers in the studied basins lie in the average concentration of these nutrients in coastal plain soils from northeastern Brazil (Silva 1996; Ramalho and Sobrinho 2001; Ramalho et al. 2001). 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 each soil type. In the studied area, N and P concentrations are higher in eutrophic alluvial and podsols soils followed by latosols, solonetz and quartz sands. Finally, the estimates of N and P losses were corrected according to 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 some waterlogged soils or for some site of alluvial soils along river valleys (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). Average 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).

Table 2 presents the N and P soil losses for each soil unit to the estuarine basins. The N and P loads are from 4.9 and 1.0 t year^{-1}, respectively, in the smallest Malcozinhado basin, to 223 and 201 t year^{-1}, respectively, in the largest Coreaú basin. N and P soil losses considered, larger basins present the largest emissions of N and P. Based on the total emissions presented in Table 2, rates of N and P losses per unit of area were, on the average, 66±2 and 44±20 kg km^{-2} year^{-1}, respectively. These ranges of emissions vary little among the studied basins and are in general within the range of soil loss reported for non-mechanized agriculture areas under temperate climate (75 a 230 kg km^{-2} year^{-1} for N, and 5 to 50 kg km^{-2} year^{-1} for P; Howarth et al. 1996; Howarth 1998; Valigura et al. 2000). Estimated loss per unit of area depends on soil type distribution and basin area. By comparing the nutrient losses and basin area for each estuarine basin, we observed lower mean P loss per unit of area in eastern estuarine basins which comprise Pacoti, Malcozinhado, Choró, Pirangi, Jaguaribe and Icapuí (0.022±0.009 t km^2 year^{-1}) than western estuaries including Timonha, Acaraú, Coreaú, Aracatiaçú, Aracatimirim, Curú and Mundaú (0.056±0.006 t km^2 year^{-1}). This pattern is attributed to increasing presence of quartz sand unit in eastern estuarine basins (total area of 4,120 km^2 against 1,511 km^2 for western basins) which has lower P concentration compared to other soil types (Table 2). On the contrary, N loss and basin area ratio between eastern and western estuaries is similar with values of about 0.66 t km^2 year^{-1}.

Natural emission: atmospheric deposition

Another important natural source of N and P is the atmospheric deposition which 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 the estuaries 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

Table 2 Estimates of N and P emissions (t year^{-1}) through different soil types, weathering ad soil loss found in the studied basins along the Ceará state, NE, Brazil

Basin				Soil type[a]			
	Sand	Solonschak/solonetz	Aluvial/regosol	Podsol/latosol	Planossols	Total	Loss/area
Timonha							
Soil area (km^2)	61	25	–	405	101	592	
Nitrogen	1.90	1.63	–	26.33	6.57	36.42	0.061
Phosphorus	0.37	1.63	–	26.33	6.57	34.89	0.058
Acaraú							
Soil area (km^2)	135	71	150	2,082	357	2,795	
Nitrogen	8.74	4.60	17.57	135.4	23.26	189.6	0.067
Phosphorus	1.75	4.60	9.77	135.4	23.26	174.8	0.062
Coreaú							
Soil area (km^2)	419	165	–	2,132	692	3,408	
Nitrogen	27.2	12.7	–	138.6	45	223.4	0.065
Phosphorus	5.44	12.7	–	138.6	45	201.6	0.059
Aracatiaçú							
Soil area (km^2)	143	108	–	751	580	1582	
Nitrogen	9.29	6.99	–	48.8	37.7	103	0.065
Phosphorus	1.86	6.99	–	48.8	37.7	95.4	0.060
Aracatimirim							
Soil area (km^2)	171	103	–	450	14	738	
Nitrogen	11.13	6.64	–	29.24	0.96	48.0	0.065
Phosphorus	2.23	6.64	–	29.24	0.96	39.1	0.053
Curú							
Soil area (km^2)	239	51	55	255	–	600	
Nitrogen	15.6	3.3	6.4	16.6	–	41.9	0.070
Phosphorus	3.1	3.3	3.6	16.6	–	26.6	0.044
Mundaú							
Soil area (km^2)	343	27	187	950	613	2,120	
Nitrogen	22.2	1.7	21.8	61.7	39.9	147.3	0.069
Phosphorus	4.4	1.7	12.1	61.7	39.9	119.8	0.056
Ceará							
Soil area (km^2)	68	108	–	496	966	1640	
Nitrogen	4.4	7.0	–	32.3	62.8	107	0.065
Phosphorus	0.9	7.0	–	32.3	62.8	103	0.063
Cocó							
Soil area (km^2)	11	7	–	402	–	420	
Nitrogen	0.7	0.4	–	26.1	–	27.2	0.065
Phosphorus	0.1	0.4	–	26.1	–	26.6	0.063
Pacoti							
Soil area (km^2)	350	3	–	268	38	660	
Nitrogen	22.8	0.8	–	17.4	2.5	43.5	0.066
Phosphorus	4.67	0.8	–	17.4	2.5	25.3	0.038
Malcozinhado							
Soil area (km^2)	75.7	–	–	–	–	75.7	
Nitrogen	4.9	–	–	–	–	4.9	0.065
Phosphorus	1.0	–	–	–	–	1.0	0.013
Choró							
Soil area (km^2)	732	66	–	–	22	820	
Nitrogen	47.6	4.28	–	–	1.42	53.3	0.065
Phosphorus	9.51	4.28	–	–	1.42	15.2	0.018

Table 2 (continued)

Basin	Soil type[a] Sand	Solonschak/solonetz	Aluvial/regosol	Podsol/latosol	Planossols	Total	Loss/area
Pirangi							
Soil area (km^2)	1341	112	–	14	–	1,467	
Nitrogen	87.2	7.3	–	0.9	–	95.4	0.065
Phosphorus	17.4	7.3	–	0.9	–	25.6	0.017
Jaguaribe							
Soil area (km^2)	1259	168	26	132	66	1,735	
Nitrogen	81.9	10.2	8.8	4.3	4.3	123.6	0.071
Phosphorus	16.3	10.2	4.9	4.3	4.3	45.4	0.026
Icapuí							
Soil area (km^2)	363	–	–	48	–	411	
Nitrogen	23.6	–	–	3.2	–	26.8	0.065
Phosphorus	4.7	–	–	3.2	–	7.9	0.019

The nutrient loss and basin area ratio (loss/area) is expressed in t km^2 year^{-1}

Soil loss rate: 128–130 t km^{-2} year^{-1} (Greeland and Lal. 1977; Gouldie 1987)

Average 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); aluvials: 900 and 500 (Ramalho et al. 2001); planossol: 500 and 500 (Silva 1996)

[a] Soil type area from Estado do Ceará 2004

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. Total atmospheric deposition ranges from 80 to 300 mgN m^{-2} year^{-1}, and 4 to 10 mgP m^{-2} year^{-1}, over pristine and heavy industrialized coastal sites, respectively and with annual rainfall of about 1,000 mm (Silva Filho et al. 1998; Mello 2001, 2003). These 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 average 100 and 8.0 mg m^{-2} year^{-1}, for N and P respectively, which are similar to the averages reported by other authors for natural or low-industrialized 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. Burns (2004) estimated based on results from eight Mid West USA basins that an average of 63% of the total N deposition from the atmosphere is retained in soils. 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 estimates are used in the present study. The fraction retained in soils will be eventually included in the calculation of inputs from soil runoff, since it makes up part of the soil concentrations of N and P.

Inputs from the atmosphere per unit of area estimated using the parameters above are 12 and 1.0 kg km^{-2} year^{-1} for N and P, respectively. As expected, total loads from atmospheric deposition resulted highly influenced by the basin area. The larger Coreaú basins receives about 51.1 and 4.09 t year^{-1} of N and P, respectively, whereas the smaller Malcozinhado basin receives 0.71 and 0.06 t year^{-1} of N and P, respectively (Table 3).

Estimates of N and P loads from natural sources are summarized in Table 3. Notwithstanding the differences in N and P concentrations in soil types, basin area results the most important parameter controlling the natural loads of N and P to the basins studied. Maximum input was estimated for the larger Coreaú basin (274 and 205 t year^{-1} for N and P, respectively) and minimum for the smaller Malcozinhado basin with natural loads of N and P varied from

Table 3 Estimates of total N and P inputs (t year^{-1}) from natural sources to the studied basins along the Ceará State

	Soil runoff[a, d]		Atmospheric deposition[b, c]		Total natural input	
	N	P	N	P	N	P
Timonha	36.4	34.9	6.91	0.55	43.3	35.5
Acaraú	189	174	27.5	2.20	217	177
Coreaú	223	202	51.1	4.09	274	206
Aracatiaçú	103	95.4	13.1	1.05	116	96.5
Aracatimirim	48.0	39.1	8.42	0.67	56.4	39.8
Curú	41.9	26.6	7.56	0.61	49.5	27.2
Mundaú	147	120	29.0	2.32	176	122
Ceará	107	113	24.2	1.94	131	115
Cocó	27.2	26.6	5.83	0.46	33	27.0
Pacoti	43.5	25.3	9.98	0.80	53.5	26.1
Malcozinhado	4.90	1.0	0.71	0.06	5.60	1.10
Choró	53.3	15.2	10.9	0.87	64.2	16.1
Pirangi	95.4	25.6	14.7	1.20	110	26.8
Jaguaribe	123	45.4	19.8	1.58	143	47.0
Icapuí	26.8	7.9	4.08	0.33	30.9	8.23

[a] From Table 2

[b] Average N and P bulk atmospheric deposition (100 and 8.0 mg m^{-2} year^{-1}, respectively) based on concentrations in bulk atmospheric deposition from Mello (2001, 2003) and Silva Filho et al. (1998)

[c] Basin area and annual rainfall from Table 1

[d] Soil retention rates of 65 and 70% for N and P respectively after Malavolta and Dantas (1980) and Silva (1996)

5.6 to 1.1 t year^{-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).

Anthropogenic emissions: agriculture

An important environmental concern related to agriculture is the nutrient losses to surface waters largely resulting from the application of inorganic and organic fertilizers. For non-urbanized areas this anthropogenic activity can be an important source of N and P to surface waters. Nutrient losses from agricultural soils vary according to soil type and agricultural practices. For some agricultural practices, fertilizer losses from soil reach about 10–80% (Howarth et al. 1996), which might represent losses of up to 200 kgP km^2 (Sharpley and Syers 1979; Sharpley and Rekolainen 1997; Sharpley and Tunney 2000).

Emission factors for N and P loss from soils due to agriculture are available but mostly restricted to mechanized and large-scale agriculture. These emission factors are inappropriate to be applied in the non-mechanized and small scale agriculture which is typically found in Northeastern Brazil. For these agricultural practices, average loss rates of P are in general much lower. N losses, however, are similar to those estimated for mechanized agriculture and highly dependent on crop type (Silva et al. 2000). Data from this type of agriculture along the Brazilian coast provide loss rates to the amount of fertilizer applied varying from 26 to 32% for N and from 6 to 20% for P (Malavolta and Dantas 1980; CFSEMG 1989; Silva et al. 2000). The crop type influences the loss rates of nutrients, since different cultures require different amounts of fertilizer application.

In the studied basins, some reports have summarized the usual quantities of fertilizer used in most present crops (Table 4). Demands for N fertilization are particularly high for sugar cane (90 to 275 kg ha^{-1}) and citric fruits (120–180 kg ha^{-1}) whereas for P, particularly demanding culture is corn (40–70 kg ha^{-1}; Agrobyte 2003; EMBRAPA 2003; SEAGRI 2003; CPAA 2003). Major N and P losses are from sugar cane (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;

Table 4 N and P losses by fertilizer application to different crop type

Crop	Fertilizer (kg ha^{-1})		Losses (%)	
	N	P	N	P
Caju	20[a]	30[a]	20[b]	25[b]
Cotton	22[a, c]	12[a, c]	16[b]	6[b]
Sugar cane	90–275[a, c]	20–40[a, c]	26–32[b]	6–20[b]
Bean	0–20[d, e]	60[d]	16–25[f]	0.7–1.4[f]
Coconut	40[g]	20[g]	25[h]	20[h]
Cassava	0–40[d, e]	15–60[d, e]	25[h]	20[h]
Corn	20–60[b, i, j]	40–70[i, j]	26–32[b]	6–20[b]
Banana	22[c]	12[c]	16[k]	6[k]
Mango	40[l]	20[l]	16[k]	6[k]
Sweet potato	100[l]	30[l]	20[k]	10[k]
Citric	120–180[l]	30–60[l]	16–25[k]	0.7–1.4[f]
Pineapple	100[l]	30[l]	20[m]	10[h]

[a] Agrobyte (2003)
[b] Malavolta and Dantas (1980)
[c] EMBRAPA (2003)
[d] Abreu et al. (2003)
[e] CPAA (2003)
[f] Vollenweider (1968)
[g] Ferreira (2003)
[h] Average limit for sandy soils after Howarth et al. (1996)
[i] Silva et al. (2000)
[j] CFSEMG (1989)
[k] Average limit for clay soils after Howarth et al. (1996)
[l] SEAGRI (2003)
[m] DGPC (2003)

CFSEMG 1989). The estimated amount of nutrients lost to estuaries in each basin is a function of these differences in culture type and their relative area of cultivation. Taking these considerations, the N and P emission estimated from agriculture practices in the studied basins range from 4.1 and 1.9 t year^{-1}, respectively, in the urbanized Cocó basin to 471 and 611 t year^{-1}, respectively, in the intensively cultivated Acaraú basin (Table 5).

Anthropogenic emission: husbandry

Animal husbandry is responsible for excessive emission of nutrients that accumulate in soils and eventually loss into groundwater and surface waters. Accumulation of nutrients in soils is related to animal excretes and their concentrations 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. N 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, incorporation by plants and losses to atmosphere (for N) will determine the fraction of nutrients from animal excretes transferred to surface and coastal waters, which are similar to those described for N and P retention of the natural inputs (see Table 2). Published estimates suggest that about 20–35% for

Table 5 Estimated emissions from agriculture and husbandry for the studied basins along the Ceará State (t year^{-1})

Basin	Agriculture[a]		Husbandry[b]		Total	
	N	P	N	P	N	P
Timonha	191	39	81	49	272	88
Acaraú	471	611	595	368	1066	979
Coreaú	133	101	530	324	663	425
Aracatiaçu	71	72	146	92	217	164
Aracatimirim	159	161	97	60	256	221
Curu	520	115	215	159	735	274
Mundaú	411	373	701	526	1112	899
Ceará	71	25	368	274	439	299
Cocó	4.1	1.9	157	132	161.1	134
Pacoti	84	27	690	584	774	611
Malcozinhado	62	11	136	113	198	124
Choró	81	52	318	262	399	314
Pirangi	159	251	320	257	479	508
Baixo Jaguaribe	146	168	145	90	291	258
Icapuí	88	131	55	32	143	163

[a] Amount of N and P applied for each culture (see Table 4). Agriculture area and culture type (IBGE 2006; 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)

[b] Animal stock from IBGE (2006), 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 percentage 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)

N and 35–60% for P present in animal excretes reaches the surface waters (NRC 1993; Bouwman et al. 1997; Bouwman and Booij 1998; NRC 2003).

A large proportion of cattle in the studied basins consist of bovine and therefore large number of individuals and amount of manure production by bovine will be the major source of N and P. Emission of N and P to the studied basins from region's husbandry is independent of basin area and range, respectively from 55 and 32 t year^{-1}, in the Icapuí basin, to 690 and 584 t year^{-1}, in the Pacoti basin, respectively (Table 5). It is important to note that manure is frequently used as fertilizer, 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 considered here.

Anthropogenic emissions: waste waters

Many coastal areas suffer from considerable pollution due to untreated waste water inputs. Waste waters are an important anthropogenic source of N and P mainly in urbanized areas where insufficient wastewaters treatment is observed. In these areas, nutrient loads from this source are 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). The waste water emission is the primary environmental concern for most Ceará State due to absence of proper treatment. In the most populated area of the state (Fortaleza Metropolitan Area), about 60% of waste waters is collected and transferred through an ocean disposal system (CAGECE, personal communication).

The assessment of the importance of nutrient inputs to estuarine basins from waste waters was obtained by emission factors reported by Smith et al. (1997), Howarth (1998), Bidone (2000), Bidone and Lacerda (2002), Martinelli et al. (2002). This coefficient estimates annual N and P from population parameters from latest population census available (IBGE 2006) 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. Nutrient concentrations in wastewater were those suggested by von Sperling (1996) based on local 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.

Table 6 shows the calculated N and P emission from waste water to the estuarine basins across Ceará State. Generally, N and P loads from waste waters reflect the high population density and therefore maximums loads were observed to the Cocó basin (2,116 and 596 t year^{-1}, respectively). On the contrary, minimum loads were found in Malcozinhado basin (269 and 70 t year^{-1}, respectively) which has the lower population amongst the study basins. Comparisons of the emission factors calculated for this study (4–8 and 0.6–2.9 g inhab^{-1} day^{-1}, for N and P respectively) with others from the literature showed our values in the lower range of reported factors, since emissions are expected lower than those reported for urbanized sites, due to the higher water 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).

Anthropogenic emissions: urban runoff and solid waste disposal

Such as waste waters, urban runoff and solid waste disposal are also directly associated to urbanization. In highly urbanized area, leaching of urban surfaces and solid waste disposal causes increased N and P emission to surface waters. 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

Table 6 Estimated N and P emissions from wastewaters, urban runoff, and solid wastes disposal to the fifteen estuarine basins studied (t year^{-1})

Estuário	Waste waters[a, b]		Urban runoff[c]		Solid waste disposal[d]		Total	
	N	P	N	P	N	P	N	P
Timonha	50.1	14.0	0.7	0.11	2.4	–	53.2	14.1
Acaraú	239	66.9	3.3	0.50	12.2	–	254	67.4
Coreaú	233	65.2	3.3	0.49	11.4	–	248	65.7
Aracatiaçu	56.6	15.9	0.6	0.09	3.0	–	60.2	15.9
Aracatimirim	52.5	14.7	0.8	0.11	2.8	–	56.1	14.8
Curu	98.3	27.5	1.6	0.24	4.9	–	105	27.7
Mundaú	250	70	4.2	0.63	12.8	–	267	70.6
Ceará	328	92	11.1	1.67	37.3	–	376	93.7
Cocó	2,116	596	79.3	12.0	232	–	2,427	608
Pacoti	305	85	4.4	0.65	13.6	–	323	85.7
Malcozinhado	26.9	7.5	0.4	0.06	1.4	–	28.7	7.6
Choró	115	32.1	2.0	0.30	5.3	–	122	32.4
Pirangi	76.5	21.4	1.0	0.15	3.9	–	81.4	21.5
Jaguaribe	152	42.6	2.1	0.31	7.3	–	161.	42.9
Icapuí	27.5	7.7	0.4	0.06	1.5	–	29.4	7.8

[a] Nutrient concentrations in waste waters: N=35–70 mg l^{-1}; P=5–25 mg l^{-1} (von Sperling 1996). Population data from IBGE (2006). 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)

[b] Sixty percent of waste waters from Fortaleza metropolitan area is collected and transferred through ocean disposal system (COGERH, personal communication) and therefore this load is not computed as N and P emission to Ceara and Cocó estuaries

[c] Population parameters from IBGE (2006). Annual rainfall from Hidroservice (1998). Includes leaching of solid wastes disposal sites. Solid waste production per inhabitant from ABES (1983). Average constructed housing unit of 50 m^2. Average N and P concentrations in runoff from NRC (2003) and Binner et al. (1996)

[d] Adapted from Binner et al. (1996)

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). As shown in Table 6, the importance of these sources on N and P emission is more significant in the urbanized areas such Cocó and Ceará basins. The N and P loads from urban runoff reach 79.3 and 11.1 t year^{-1}, respectively, in the urbanized Cocó basin. Emissions of N from improper urban solid waste disposal range from 232 t year^{-1} in the Cocó basin to 1.4 t year^{-1} in the Malcozinhado basin. The absence of emission factor for P from urban solid disposal impedes the calculation of P loads.

Anthropogenic emission: aquaculture

Over the last few years there has been a steady development of the intensive shrimp farming that became the major economical activity along the studied coastal area. The area covered by shrimp farming includes about 3,279 ha of shrimp ponds with a total annual production of about 24,700 t (ABCC 2003). In the intensive aquaculture systems, N and P are two main pollutants due to high organic and nutrient loadings generated from large amount of fertilizers, feed wastage, excretion and faecal productions which are directly discharged into the coastal waters (Burford et al. 2003).

Notwithstanding the economic importance of the activity, there are few studies estimating the emission factors of nutrients from these farms (Abreu et al. 2003; Figueiredo et al. 2005). 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 1-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^- 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).

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 (Nunes 2001), the estimated average emission factor for P was about 0.05 kg ha^{-1} day^{-1} (14 kg ha^{-1} year^{-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 NO_3^-, NO_2^- and $NH_3 +$ NH_4^+ 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}. These emission factors are in the same range of those reported for Australia (0.06 and 1.08 kg ha^{-1} day^{-1}, for P and N, respectively; Jackson et al. 2003) and México (0.18–0.58 kg ha^{-1} day^{-1}; Páez-Osuna et al. 1999, 2003). Based on these emission factors, the total N and P contribution from shrimp farming to the different basins studied (Table 7) from 12 tN year^{-1} and 1.0 tP year^{-1}, for N and P, respectively, at the Mundaú basin, with only 42 ha of ponds, to 346 tN year^{-1} and 28.9 tP year^{-}1, respectively, at the Jaguaribe basin, which harbors about 1,259 ha of ponds.

Discussion

The comparison of basin area-weighted N and P emission factors showed aquaculture (0.52 t km^2 year^{-1}) and waste waters (0.47 t km^2 year^{-1}) as the most important N sources to the estuaries (Table 8). These emission factors are one order of magnitude higher than natural sources. The largest N emission from aquaculture confirms the importance of this activity, since loads are directly disposed to estuarine waters. The importance of N emission from waster waters is related to relative importance of emissions from Cocó and Ceará river basins which are located within a 2.5 million-inhabitant metropolitan area. For other basins, this source decreases in importance per unit of area as a result of low level of urbanization and low population. For P, the largest emission is related to husbandry (0.30 t km^2 year^{-1}) and waste waters (0.13 t km^2 year^{-1}) being also well-above natural loads. The Table 9 shows the N and P emission from anthropogenic sources and the relative importance (%) for each individual activity. When compared to natural emission factors (Table 3), anthropogenic sources exceed the

Table 7 Estimates for N and P emissions from shrimp farms to the studied estuaries in the Ceará State. NE Brazil (t $year^{-1}$)

Basin	Pond area (ha)[a]	N emission[b, c]	P emission[c]
Timonha	147	40	3.45
Acaraú	743	204	17.1
Coreaú	439	121	10.1
Aracatiaçu	62	17	1.4
Aracatimirim	58	16	1.3
Curu	110	30	2.5
Mundaú	42	12	1.0
Ceará	–	–	–
Cocó	–	–	–
Pacoti	–	–	–
Malcozinhado	–	–	–
Choró	–	–	–
Pirangi	90	25	2.1
Jaguaribe	1259	346	28.9
Icapuí	59	16	1.4

From emission factors of 0.47 and 0.05 kg ha^{-1} day^{-1} for N and P respectively (Abreu et al. 2003) based on pond depth 1.0 m, volume exchanged of 5×10^5 l ha^{-1} day^{-1} 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)

[a] From ABCC (2003)

[b] N estimates based on approximate speciation given in Burford et al. (2003) i.e. 70% of the total N in the effluent discharge being particulate organic N and actual concentrations of NO_3, NO_2 and NH_4 (Abreu et al. 2003)

[c] As shown in table footer a, based on concentration in effluent waters monitored during 1 year (Abreu et al. 2003)

natural emissions in all basins, being up to 78 times higher for N in the Cocó basin and 120 times higher for P in the Malcozinhado basin. Among the anthropogenic sources, agriculture and husbandry are responsible for almost all N emission to lower river basins and estuaries because of large areas utilized by these activities.

Agriculture contributes with the largest fraction of the N load to the rivers Timonha (52%), Aracatimirim (48%), Curú (59%), Icapuí (47%) whereas agriculture contributes to highest P emissions to Acaraú (57%), Aracatimirim (68%) and Icapuí (76%). Husbandry is the principal source of N and P to Coreaú, Aracatiaçú, Mundaú, Pacoti, Malcozinhado, Choró and Pirangi basins. Waste waters are the dominant source of nutrients to urbanized watersheds such as Cocó basin. Aquaculture is the most important source of N to the Jaguaribe (43%) where the largest pound surface is observed. 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 order to validate our estimates, the emission factors were compared to available N and P concentrations for the studied estuaries. These concentrations were obtained at upper estuarine section where influence from watershed is predominating (salinity less than 0.1‰) which decreases dilution by seawater. If emission factors were corrected, the basin receiving largest emission per unit of area and lower dilution capacity expressed by area and volume ratio should present highest nutrient concentrations in estuarine waters. Among the available data for N and P concentration in studied estuaries (Molisani 2005), nitrogen species (NH_4 and NO_3) were highest in Cocó estuary (1.6 and 3.9 mg l^{-1}, respectively) where was observed highest N emission per unit of area (5.69 t km^{-2} $year^{-1}$). On the other hand, Aracatiaçú and Ceará estuaries have the highest inorganic phosphorous concentrations (5.8 and 5.2 mg l^{-1}, respectively) in their waters although emission factors indicated relatively smaller P yield (0.18 and 0.31 t km^{-2} $year^{-1}$) compared to other estuaries. Since relatively smaller P load entering these estuaries, the low dilution capacity, mainly for the Ceará estuary which presented highest area to volume ration among the studied estuaries (Table 1), imply the potential for more susceptibility to nutrient-related impacts. Thus, the emission factor seems to be a useful tool to scaling and quantifying the N and P sources to coastal areas from northeastern Brazil, where available data

Table 8 Range and average of N and P emission from the different anthropogenic sources standardized to t km^{-2} $year^{-1}$, for the specific conditions existing in the fifteen estuarine basins studied along the coast of Ceará State, NE Brazil

Source	N	P
Natural sources	0.008–0.07 (0.04)	0.001–0.07 (0.02)
Waste water	0.04–5.04 (0.47)	0.01–1.41 (0.13)
Husbandry	0.08–1.8 (0.38)	0.05–1.49 (0.30)
Agriculture	0.01–0.87 (0.22)	0.005–0.32 (0.12)
Urban runoff	0.0004–0.18 (0.015)	0.0001–0.03 (0.002)
Aquaculture	0.06–1.95 (0.52)	0.005–0.17 (0.05)

Table 9 Average estimates of N and P emissions (t year^{-1}) from anthropogenic sources in the studied basins from the coast of Ceará State

Basin	Source						
	Waste Waters	Solid wastes	Urban runoff	Agriculture	Husbandry	Aquaculture	Total
Timonha							
Nitrogen	50.1 (13)	2.4 (0.6)	0.7 (0.07)	191 (52)	81 (22)	40 (11)	365
Phosphorus	14.0 (13)	–	0.11 (0.01)	39 (37)	49 (46)	3.4 (3.2)	106
Acaraú							
Nitrogen	238 (15.6)	12.2 (0.08)	3.3 (0.2)	471 (31)	595 (39)	204 (13.3)	1,524
Phosphorus	66.9 (6.3)	–	0.50 (0.04)	611 (57.4)	368 (34.6)	17.1 (1.6)	1,063
Coreaú							
Nitrogen	233 (22.6)	11.4 (1.1)	3.3 (0.3)	133 (12.9)	530 (51.4)	121 (11)	1,031
Phosphorus	65.2 (13)	–	0.49 (0.1)	101 (20.1)	324 (64.7)	10.1 (2.0)	501
Aracatiaçú							
Nitrogen	56.6 (19.2)	3.0 (1)	0.6 (0.2)	71 (24.1)	146 (49.6)	17 (5.8)	294
Phosphorus	15.9 (8.7)	–	0.09 (0.05)	72 (39.5)	92 (50.5)	1.4 (0.8)	182
Aracatimirim							
Nitrogen	52.5 (16)	2.8 (0.8)	0.8 (0.2)	159 (48.4)	97 (29.5)	16 (4.9)	328
Phosphorus	14.7 (6.2)	–	0.11 (0.05)	161 (68)	60 (25)	1.3 (0.5)	237
Curú							
Nitrogen	98.3 (11.3)	4.9 (0.6)	1.6 (0.2)	520 (59)	215 (24.7)	30 (3.4)	870
Phosphorus	27.5 (9.0)	–	0.24 (0.1)	115 (37.8)	159 (52)	2.5 (0.8)	304
Mundaú							
Nitrogen	250 (18)	12.8 (0.9)	4.2 (0.3)	411 (29.5)	701 (50.3)	12 (0.9)	1,391
Phosphorus	70 (7.2)	–	0.63 (0.06)	373 (38.4)	526 (54.1)	1.0 (0.1)	971
Ceará							
Nitrogen	328 (40)	37.3 (4.6)	11.1 (1.4)	71 (8.7)	368 (45)	–	815
Phosphorus	92 (23)	–	1.67 (0.4)	25 (6.4)	274 (70)	–	393
Cocó							
Nitrogen	2,116 (82)	232 (8.9)	79.3 (3.1)	4.1 (0.1)	157 (6.1)	–	2,588
Phosphorus	592 (80)	–	12.0 (1.6)	1.9 (0.2)	132 (18)	–	738
Pacoti							
Nitrogen	305 (27.8)	13.6 (1.2)	4.4 (0.4)	84 (7.6)	690 (63)	–	1,097
Phosphorus	85 (12.1)	–	0.65 (0.1)	27 (3.8)	584 (83.7)	–	697
Malcozinhado							
Nitrogen	26.9 (11.8)	1.4 (0.6)	0.4 (0.2)	62 (27.3)	136 (60)	–	227
Phosphorus	7.5 (5.7)	–	0.06 (0.04)	11 (8.3)	113 (85.6)	–	132
Choró							
Nitrogen	114 (21.8)	5.3 (1.0)	2.0 (0.4)	81 (15.5)	318 (61)	–	521
Phosphorus	32.1 (9.2)	–	0.30 (0.1)	52 (15.0)	262 (75.7)	–	346
Pirangi							
Nitrogen	76.5 (13.1)	3.9 (0.6)	1.0 (0.2)	159 (27.1)	320 (54.7)	25 (4.2)	585
Phosphorus	21.4 (4.0)	–	0.15 (0.01)	251 (47.2)	257 (48.3)	2.1 (0.4)	532
Jaguaribe							
Nitrogen	152 (19.5)	7.3 (1.0)	2.1 (0.3)	146 (18.3)	145 (18.2)	346 (43.3)	798
Phosphorus	42.6 (12.9)	–	0.31 (0.1)	168 (51)	90 (27.2)	28.9 (8.8)	330
Icapuí							
Nitrogen	27.5 (14.6)	1.5 (0.8)	0.4 (0.2)	88 (46.8)	55 (29.2)	16 (8.5)	188
Phosphorus	7.7 (4.5)	–	0.06 (0.03)	131 (76.2)	32 (18.6)	1.4 (0.8)	172

The relative contribution (%) of each individual source appears in parenthesis from N and P emission is scarce still water quality is a matter of increasingly urgent concern.

Acknowledgements This work was supported by Superintendência Estadual de Meio Ambiente do Estado do Ceará (SEMACE) and Rede RECARCINE-FINEP. The author MM Molisani is grateful to CNPq/FUNCAP-CE for concession of DCR grant. This work is part of "Instituto do Milenio" (CNPq/420050/2005-1).

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