

Hydrochemistry of two small estuaries: Cururupe and Acuípe Rivers (Ilhéus, BA, Brazil).

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ABSTRACT: Hydrochemistry of two small estuaries: Cururupe and Acuípe River, Ilhéus, Bahia. In this work the effect of leachate waste from the Ilhéus garbage dump on the water quality of Cururupe River estuary was focused. Sampling was carried at the estuaries of Cururupe River and Acuípe River (assumed as natural background). pH, temperature, dissolved oxygen, electrical conductivity and salinity were measured in situ. Dissolved inorganic nutrients, total alkalinity, biochemical oxygen demand and Fe²⁺ were also analyzed. The presence of dump does not affect significantly these variables. Both estuaries are dystrophic and Cururupe River and Acuípe River can be classified in class 7 and 8, respectively (CONAMA no. 20). According the new CONAMA resolution n° 357 the classification is conflicting, but Cururupe River exhibited a better water quality. This fact can be explained by depuration of leachate wastes in mangrove of Cururupe River, and the greater area covered by wetlands in Acuípe River. The BOD results also stressed the limitation of this test as an indicator of organic pollution, and suggest that correlation analysis can be used as an indirect method to evaluate environmental disturbance.

Key-words: Leachate waste, biochemical oxygen demand, dissolved inorganic nutrients, water quality, environmental regulation.

RESUMO: Hidroquímica de dois pequenos estuários: Rio Cururupe e Rio Acuípe, Ilhéus, Bahia.

Este trabalho teve como objetivo avaliar a influência da drenagem do depósito de lixo de Ilhéus sobre a qualidade da água do Rio Cururupe. Amostras foram coletadas neste estuário e no Rio Acuípe (nível de base natural). pH, temperatura, oxigênio dissolvido, condutividade elétrica e salinidade foram determinados no campo. Também foram analisados nutrientes inorgânicos dissolvidos, alcalinidade total, demanda bioquímica de oxigênio e Fe²⁺. A presença do lixo parece não influenciar significativamente estas variáveis. Os estuários foram classificados como distróficos, e suas condições atuais podem ser inseridas nas classes 7 e 8 da Resolução CONAMA n° 20, respectivamente. A aplicação da nova Resolução CONAMA n° 357 gera resultados conflitantes, porém com o Rio Cururupe apresentando melhor qualidade da água. Este fato pode ser explicado pela autodepuração do chorume no manguezal do Rio Cururupe, e pela maior área de alagados presente na bacia do Rio Acuípe. Estes resultados também enfatizam a limitação da utilização da DBO como indicador de poluição orgânica, e sugerem que a análise de correlação pode ser empregada como um método indireto de avaliação de perturbações ambientais.

Palavras-chave: Chorume, demanda bioquímica de oxigênio, nutrientes inorgânicos dissolvidos, qualidade da água, legislação ambiental.

Introduction

Clean water with quality enough to allow multiple uses is still a crucial need in several countries. In developing countries, estimates account that 80% of the diseases and about 33% of death cases are related to the use and consume of contaminated

water. The main cause of groundwater contamination is the seepage of organic compounds, heavy metals and other toxic compounds from the leachate of inadequately disposed garbage. There are estimates that the concentration of these compounds reaching the water table double every fifteen years (Banco Mundial, 1992).

Brazil produces about 20 million tons of garbage per year. According Cavinato & Rodrigues (1997), there is an average generation of 0.5 kg . day⁻¹ of solid wastes per capita. A significant amount (~ 65 %) of solid wastes produced in Brazil correspond to organic matter, which decompose and form the leachate waste (chorume; Pereira Neto, 1996; 1999). The percolation of pluvial precipitation through the waste dump containing decomposing organic matter produce this dark acid liquid, with high organic matter and heavy metal content derived from the leach of batteries and metallic packages. Microbial contaminants and micropollutents such as toluene, dichloromethane and carbon dioxide are also found in this liquid waste (Baird, 2002). Bocanegra et al. (1998) found that the biological oxygen demand range between 465 and 1,000 mg . L⁻¹, and a total leachates waste loading from 730 to 1,700 m³ . ha⁻¹ . year⁻¹ in a garbage dumping near Mar del Plata, Argentina. When these leachates reach aquatic ecosystems there is a decrease of dissolved oxygen levels and an increase of biochemical oxygen demand, resulting in environmental damages (Lima, 1991; Baird, 2002).

The accelerated growth of population and economic activities in cities placed in the coastal zone promote an increase of the volume of produced garbage. Several of these cities solved the problem of the destination of these wastes by the dumping in mangroves. The county of Ilhéus is located in the cocoa production area, south of Bahia State. Cururupe and Acuípe Rivers comprehend two small watersheds in the coastal zone southward from the city of Ilhéus (Fig. 1). The garbage produced by the city of Ilhéus is placed near the mangrove of Cururupe River estuary, a permanent preservation area. Local inhabitants and tourists intensively use the estuarine waters as recreation. The estuarine zone of Acuípe River is not subject to any significant anthropogenic impact.

The Conselho Nacional do Meio Ambiente (CONAMA, 1986), by the Resolution no. 20 of June 18, 1986, regulated the water quality standards establishing classes according their multiple uses. These norms were substantially modified by the Resolution no. 357 of March 17, 2005 (CONAMA, 2005). This work aimed at evaluating the water quality of two estuaries, exploring the potential effects of the liquid leachate from the garbage dump near Cururupe River estuary. The estuary of Acuípe River was assumed as a natural background level for the analyzed variables. A preliminary classification of CONAMA classes and actual condition, and trophic state according dissolved nutrients and chlorophyll-a concentration were also our purpose.

Material and methods

The study site

The Cururupe and Acuípe River are two small basins at south Bahia, northern Brazil (15° 10' 14° 55' S and 39° 00'/39° 05' W; Fig. 1) draining terrains of the Tertiary (Formation Barreiras) that cover the crystalline basement. The approximate area of the watershed of Cururupe and Acuípe River is 65 and 160 km², respectively. The annual average temperature is 24.5 °C, with annual precipitation of about 2,200 mm. The typical rainy season comprise the months from May to July. Data obtained from CEPLAC reveal an anomalous pluvial distribution in 2002. Precipitation exceeded 200 mm from January to March, 100 mm in May – September and December. April, October and November presented less than 100 mm, with a minimum of 54 mm in November. Mangroves covers an area of 3.9 km² in Cururupe River (Martins, 2004), while the estuary of Acuípe River present about 9 km² of this vegetation. In addition to this greater mangrove area, large wetlands are conspicuous to Acuípe River estuary.

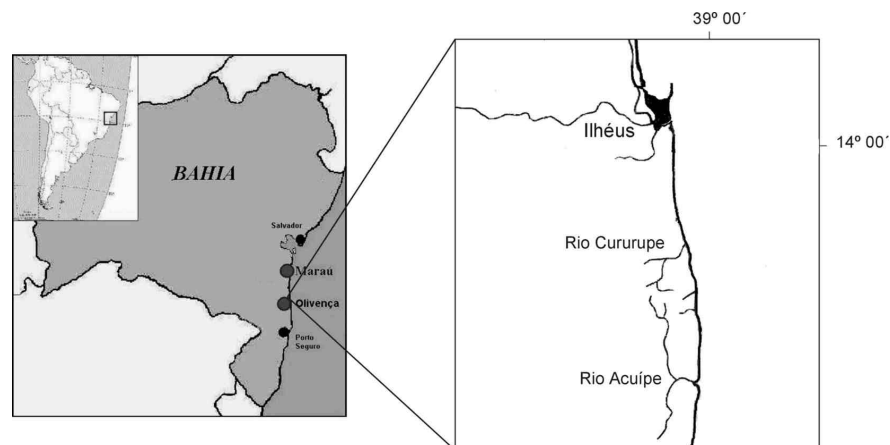


Figure 1: The study area: The Acuípe River and Cururupe River.

Sampling and chemical analysis

Sampling was carried out at a station near the mouth of the estuaries of Cururupe River (RC) and Acuípe River (RA) along 2002 (N = 15). The sampling station of Cururupe River is about 1 km distant from the garbage-dumping site. Water samples were collected in the morning during the low water slack tide. This tidal stage was selected to avoid the effect of dilution by mixing with seawater, and maximize the effect of the input of leachate waste.

The pH, temperature, dissolved oxygen (DO), conductivity and salinity were measured in situ with portable digital meters (Hanna HI-9143 and WTW Multiline P3). Samples were stored in polyethylene flasks and disposable syringes with no headspace (for total alkalinity determination) and maintained on ice during transportation to the Laboratório de Oceanografia Química, UESC, to further chemical analyzes. All materials used in sampling were washed in HCl 1:1 and rinsed with distilled water to avoid contamination. In laboratory the samples were filtered through fiberglass filters (GF/C; 1.2 mm), pre-combusted (450 °C; 4 h.) and weighed (precision 0.00001 g). The same type of filter was used to collect samples for chlorophyll-a determination by the spectrophotometric method in an acetone 90 % v.v. extract (Parsons et al., 1984). Dissolved inorganic nutrients (ammonia, nitrite, nitrate, phosphate and silicate) were analyzed in the filtrate by spectrophotometric methods (Grashoff et al., 1983). Biochemical oxygen demand (BOD₅) was determined in triplicate by dilution method (APHA, 1985). Total alkalinity was analyzed in unfiltered samples by titration with HCl as described in Carmouze (1994). Fe²⁺ concentration was measured using a colorimetric kit (Merck Ionoquant). The concentration of dissolved organic carbon (DOC) was estimated applying the absorbance at 254 nm to the equation proposed according Pagès & Lemoalle (1995).

Non parametric methods were used to statistical analyses, since these environmental samples rarely exhibit a normal distribution, and they are more suitable to small samples. The normality test of Shapiro-Wilk confirmed the non normal distribution. The difference between variables was assessed by the Wilcoxon matched pairs test. To determine the relation between the variables the Spearman correlation coefficient was used. A significance level of 1 % was established. All statistical analyses were performed with the SPSS (Statistical Package for Social Science, version 6.0; Norusis, 1993).

Results

The hydrochemistry of rivers Cururupe and Acuípe is summarized in Tab. 1. Average values of salinity and electrical conductivity of water were very low and the

samples were slightly acidic. The Wilcoxon test revealed that the mean of pH, dissolved oxygen, nitrate and silicate for these estuaries were significantly different ($p < 0,01$).

Table 1: Mean, range of concentration and standard deviation of the physico-chemical variables. < d.l. = below detection limit.

Variables	Açupe R.			Cururupe R.		
	mean	Range	SD	mean	Range	SD
T °C	24.9	22.1 – 26.7	1.23	24.9	22.7 – 26.8	1.21
pH	6.52	5.9 – 7.0	0.33	6.79	6.1 – 7.7	0.34
Electrical conductivity (mS . cm ⁻¹)	781.3	93 – 2830	690.9	710.8	118 – 2060	596.9
Salinity (p.s.u.)	0.81	0 – 3.1	1.14	0.85	0 – 3.7	1.02
BOD (mg . L ⁻¹)	2.43	0.8 – 7.0	1.84	2.48	0.3 – 9.3	2.59
Dissolved oxygen (mg . L ⁻¹)	5.6	4.5 – 6.6	0.69	6.2	4.9 – 7.1	0.58
Dissolved oxygen (% saturation)	68.11	52.5 – 80.8	8.79	75.8	57.6 – 83.7	7.54
Total alkalinity (mM)	253.4	4.5 – 733.9	215.8	283.1	23 – 953.9	217.8
Total suspended solids (mg . L ⁻¹)	14.14	3.5 – 59.0	15.52	7.41	3.3 – 16.5	3.65
Fe ²⁺ (mg . L ⁻¹)	0.21	0.1 – 0.6	0.15	0.25	0.1 – 0.5	0.15
N-NH ₃ /NH ₄ ⁺ (mM)	2.94	1.2 – 10.1	2.15	3.42	2.0 – 5.8	1.05
N-NO ₂ (mM)	0.29	< d.l. – 0.9	0.26	0.30	0.1 – 0.6	0.15
N-NO ₃ (mM)	5.22	2.0 – 8.8	2.01	7.70	4.6 – 13.6	2.06
P-PO ₄ ³⁻ (mM)	0.18	< d.l. – 0.3	0.10	0.18	< d.l. – 4.3	0.13
H ₃ SiO ₄ ⁻ (mM)	85.07	6.4 – 124.6	38.31	113.6.	57.6 – 135.3	23.1
Dissolved organic carbon (mM)	1.57	0.5 – 3.8	0.92	1.63	0.6 – 2.7	0.67

Cururupe River presented the lower concentrations of total suspended solids (Tab. 1, Fig. 2). The higher values of total suspended solids were observed in January and September (Cururupe River) and January, March and April (Açupe River).

Dissolved oxygen concentration was significantly higher in Cururupe River along the year (Fig. 2, Tab. 1). The mean of dissolved oxygen saturation was also higher in Cururupe River (Tab. 1). The mean concentrations of BOD were similar in both estuaries. Açupe River exhibited highest concentrations of BOD in several samplings (Fig. 2).

Total Alkalinity was very low during most of the year (Fig. 2). Values above 400 mM were only observed in two initial samplings (RC and RA) and June 27 (RA). In the period from the end of March to early May concentrations of total alkalinity were between 200 – 400 mM. When pluvial precipitation decreased (from July to November) the total alkalinity was below 200 mM.

Mean concentration of ammonia was slightly greater in Cururupe River, but with no significant difference. Açupe River exhibited a higher standard deviation (Table 1, Fig. 3). Except during January and June when high concentrations were found in both rivers, Açupe River presented lower concentrations of ammonia. Samplings of June, July and December showed greater concentrations of nitrate in Cururupe River (Fig. 3). The concentrations of nitrite were low in both estuaries. Phosphate concentrations were lower than 0.5 mM (Table 1, Fig. 3). Both estuaries showed the same mean and standard deviation. Fe²⁺ concentrations showed small variation along the year in both estuaries (Fig. 3).

Chlorophyll-a determinations were not detectable by the used method. The detection limit was indeed high since it depends on the volume of filtrated water (Parsons et al., 1984), and the filters become clogged after about 0.2 L addition. According with these authors, we can assure that chlorophyll-a concentrations were lower than 2 mg . L⁻¹.

The highest concentrations of DOC were recorded in January and July, months of great pluvial input (Fig. 3). Except by samplings of July and December, with

values lower than 1.0 mM, the concentrations were high in both estuaries and reach more than 3.5 mM in June (CR).

Cururupe River showed significant correlation coefficients for eight variables (Tab. III), against 22 in Acuípe River (Tab. IV). In both rivers were found significant

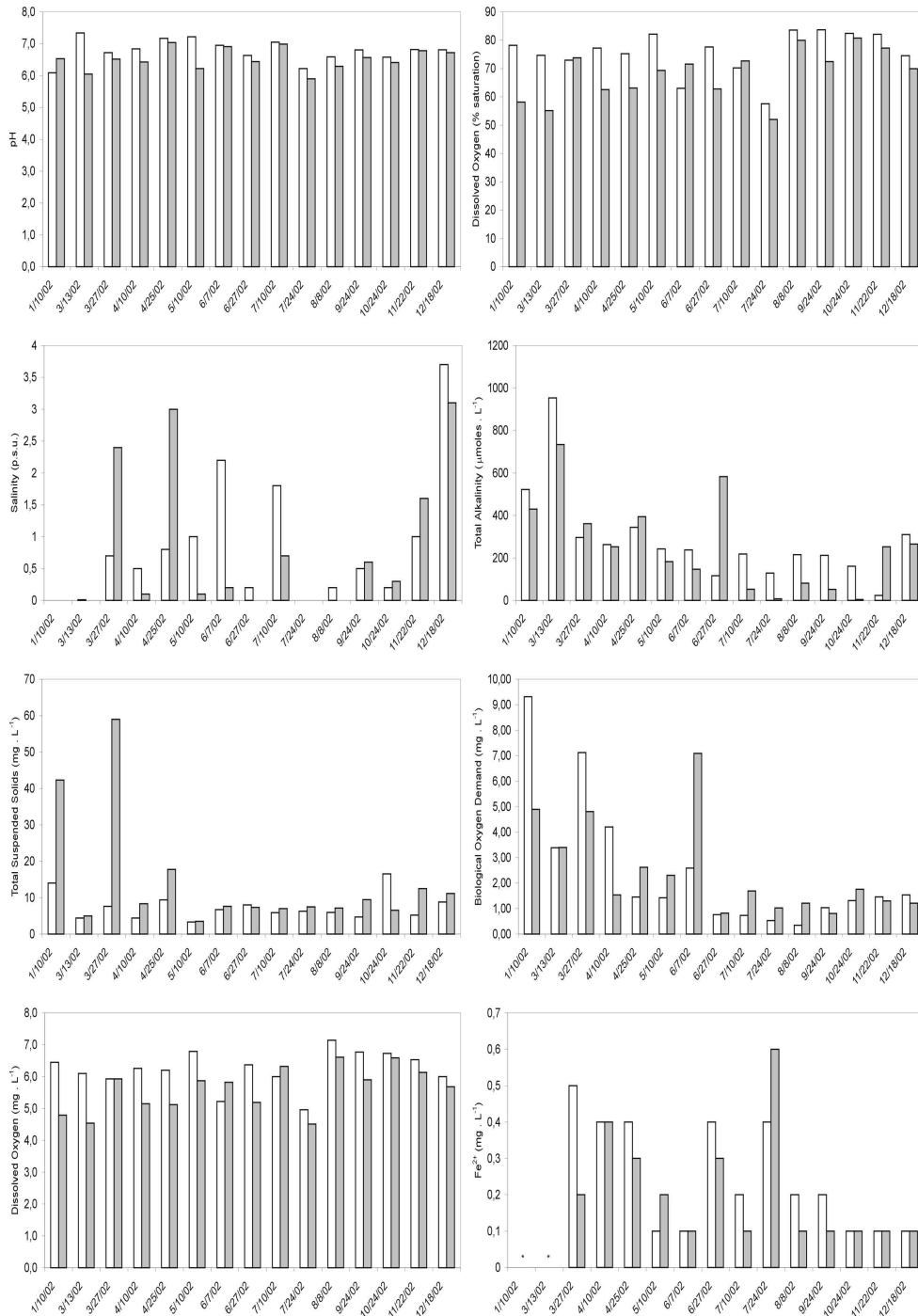


Figure 2: pH, salinity and concentrations of total suspended solids, dissolved oxygen, total alkalinity, biological oxygen demand and ferrous iron. Blank columns = Acuípe River; Gray columns = Cururupe River.

Table II: Correlation matrix for variables in Cururupe River. $P < 0.01$ in bold.

Variable	T°C	pH	pH μScm^{-1}	S	TSS	DO mg/L	DO %	Fe ²⁺	BOD	Total _{alk.}	N-NH ₃	N-NO ₂	N-NO ₃	PO ₄ ³⁻	SiO ₂ H ₃	DOC
T°C	1.000															
pH		1.000														
αScm^{-1}			1.000													
S				1.000												
TSS					1.000											
DO mg/L						1.000										
DO %							1.000									
Fe ²⁺								1.000								
BOD									1.000							
Total _{alk.}										1.000						
N-NH ₃ ⁺											1.000					
N-NO ₂												1.000				
N-NO ₃													1.000			
PO ₄ ³⁻														1.000		
SiO ₂ H ₃															1.000	
DOC																1.000

Table III: Correlation matrix for variables in Açupe River. $P < 0.01$ in bold.

Variable	T°C	pH	pH μScm^{-1}	S	TSS	DO mg/L	DO %	Fe ²⁺	BOD	Total _{alk.}	N-NH ₃	N-NO ₂	N-NO ₃	PO ₄ ³⁻	SiO ₂ H ₃	DOC
T°C	1.000															
pH		1.000														
αScm^{-1}			1.000													
S				1.000												
TSS					1.000											
DO mg/L						1.000										
DO %							1.000									
Fe ²⁺								1.000								
BOD									1.000							
Total _{alk.}										1.000						
N-NH ₃ ⁺											1.000					
N-NO ₂												1.000				
N-NO ₃													1.000			
PO ₄ ³⁻														1.000		
SiO ₂ H ₃															1.000	
DOC																1.000

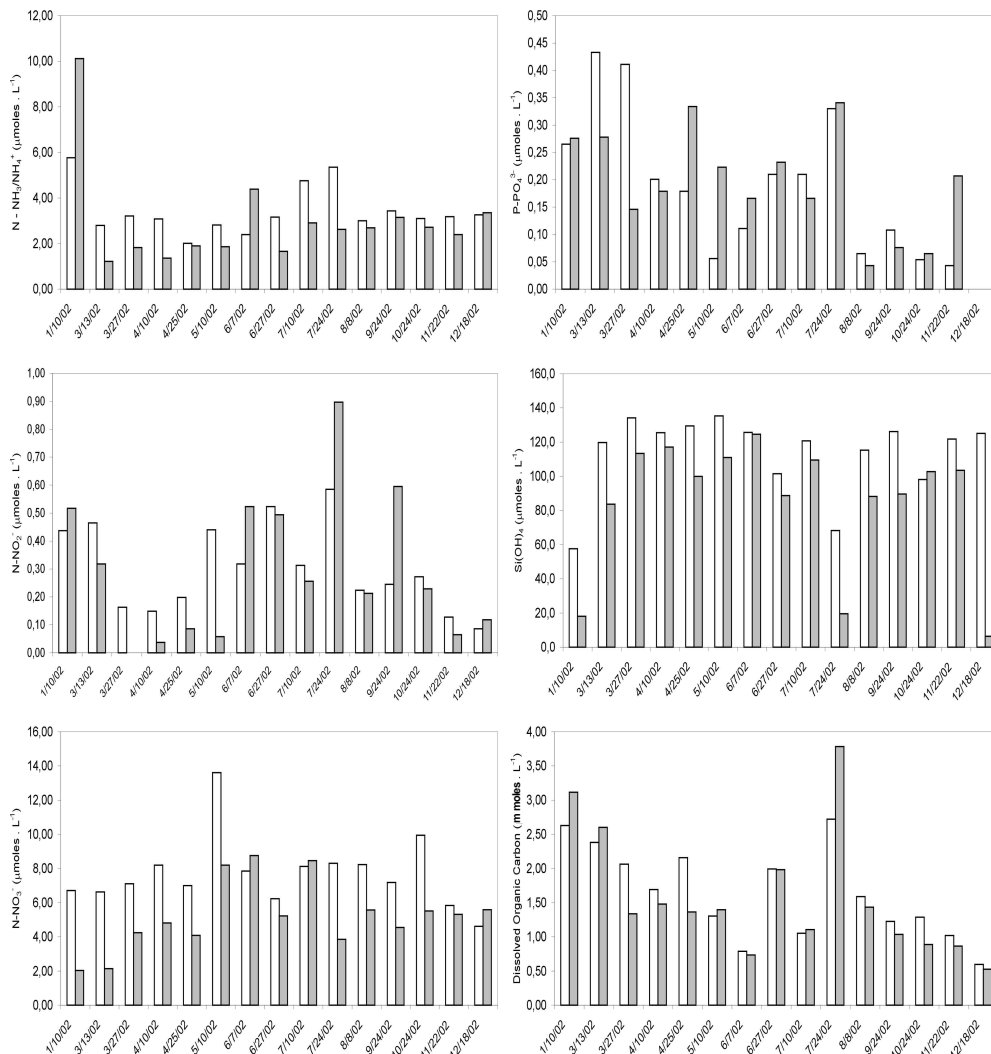


Figure 3: Concentrations of ammonium, nitrite, nitrate, phosphate, silicate and dissolved organic carbon. Blank columns = Acuípe River; Gray columns = Cururupe River.

correlation for $\text{Fe}^{2+} \times \text{PO}_4^{3-}$, $\text{Fe}^{2+} \times \text{DOC}$, salinity $\times \text{DOC}$ and $\text{PO}_4^{3-} \times \text{DOC}$. It was observed an inverse correlation of DO concentration and saturation and Fe^{2+} . Nitrate and DOC were negatively correlated in Acuípe River.

Discussion

The salinity values allow the classification of these water samples as brackish (class 7) according the earlier CONAMA resolution no. 20. Most samples were of fresh water according Resolution no. 357, with brackish water being recorded at both estuaries in only five occasions. Brackish water was observed in one and two occasions, for Acuípe and Cururupe River, respectively. In the initial samplings the high concentrations of total suspended solids seems to be related with rain fall. In situ observations of Acuípe River samples showed that there was a great amount of colloidal aggregates in the water.

The concentrations of the DOC were high, typical of tropical black waters (Drever, 1997). The results of DOC were comparable to those found in larger estuarine systems

draining great extents of salt marshes (Cai & Wang, 1998) and mangroves (Pagès & Lemoalle, 1995). There was no significant difference between the concentrations observed in Acuípe River and Cururupe, which could be attributed to the chorume leakage in the later.

Dissolved oxygen in Cururupe River was in the lower range observed in the small unpolluted estuary of Picinguaba River (Sartori & Nogueira, 1998) and a non polluted course of Cachoeira River (Pinho, 2001). The average concentration of dissolved oxygen in Acuípe River is similar to those found in stretches of Cachoeira River which receives high organic loading (Pinho, 2001). Despite the lack of important sources of pollution, there were three samples of Acuípe River in which dissolved oxygen values were below the late CONAMA no. 20 class 7 standards. According the new CONAMA resolution no. 357, six samples of Acuípe River were in the range of freshwater class 2 and 3 samples in class 3. Only one sample of Cururupe River was not included in the freshwater class 1 (5.0 mg/L; upper limit of class 3). All brackish water samples were in the range established for class 1. The concentration and saturation of dissolved oxygen are evidence that Acuípe River receives high natural inputs of organic carbon.

The values of BOD were much lower than the observed in the polluted Cachoeira River (Pinho, 2001). There are some concerns about these results, since BOD is a key variable to assess water quality in the environmental legislation. In fact its determination is subject to sample manipulation and artificial conditions which imply that it can not express what actually happens in nature. Other point is that the natural inputs of organic matter from wetlands and mangroves can result in high values not related to anthropogenic impacts, as seems to be the case of Acuípe River. Most freshwater samples exhibited BOD below the limit of class 1 of CONAMA no. 357 in the two estuaries. Acuípe and Cururupe River presented two samples in class 2 and only one in class 3. This new legislation conveniently substituted the BOD variable for total organic carbon (TOC) in brackish and saline waters. All brackish water samples exceeded the limit of class 3 only considering with the concentrations of dissolved organic carbon (DOC). This classification differs from that based in DO in brackish water samples, resulting in all samples included in class 1. These estuaries present great concentrations of organic matter, but this is in a great extent refractory to oxidative degradation.

It can be noted a decrease of ammonia concentrations while nitrate concentrations increase. This can be the result of nitrification, especially in Cururupe River. Nitrite concentration was also low, as expected to well oxygenated waters with an efficient nitrification/denitrification coupling (Carmouze, 1994). The temporal change observed in phosphate concentrations seems to be influenced by precipitation. The temporal distribution of silicate was similar in both estuaries, and also related to rain. There was an oscillation from high values in the driest months to low concentrations ($< 20 \text{ mM}$) in the rainy period. High concentrations of Fe^{2+} were found in June and July, when precipitation exceeded 150 mm. The forms of dissolved inorganic nitrogen (ammonia, nitrite and nitrate) and chlorophyll-a were included in the new legislation. The concentrations observed were much lower than the limits proposed in the CONAMA n. 357. The new resolution still does not establish values for chlorophyll-a in brackish/saline waters, despite the serious problems of eutrophication in the coastal ecosystems. The nutrient and chlorophyll-a concentrations suggest that these humic waters were dystrophic.

Cururupe River showed a lower number of significant correlation coefficients than Acuípe River. Though this kind of interpretation was not found in the literature, this can suggest that the expected estuarine processes involving these variables are being disturbed, possibly by the leachate waste seeping. The inverse correlation between salinity and DOC can be explained by the input from wetlands, including mangrove and in Cururupe River, the chorume. Fe^{2+} concentration, phosphate and DOC were well correlated. These variables enter in the estuaries mainly by the drainage of low oxygen water of adjacent wetlands, rich in organic matter, dissolved nutrients

and Fe^{2+} . The iron ions influence the phosphorus dynamic in aquatic ecosystems, depending on dissolved oxygen concentration, its oxidation state and pH. The iron (II) compounds can be easily oxidized to Fe^{3+} ions, reducing the Fe^{2+} concentration while oxygen can be replaced by physico-chemical and biological processes (Kleerekoper, 1990; Lee, 1996). Ferric monohydrogen phosphate (FeHPO_4)⁺ is one of the soluble forms of phosphate (Esteves, 1988). The neutral and oxic condition of the water favors the formation of ferric ions, especially oxyhydroxides which precipitate carrying phosphate ions adsorbed (Schäfer, 1985; Esteves, 1988).

The CONAMA no. 357 class 1 limit to dissolved iron was exceeded in both Acuípe (2) and Cururupe River (3). In the brackish samples only Cururupe River presented two values higher than the proposed to class 1. Despite the estuarine processes resulting in iron removal (Liss, 1976) this variable strongly reflects the lithological influence (Drever, 1997), rather than the effects of the leachate waste in Cururupe River.

The pH show strong direct correlation with salinity and inverse with DOC in Acuípe River, as a result of the freshwater input, mineralization of organic matter and increased buffering effect of seawater. The inverse correlation exhibited by DO and DOC reflect the oxidative degradation of these organic compounds. The negative correlation between nitrate and DOC suggests that the former compound was being used as acceptor of electrons in the oxidative degradation of organic matter at oxic/anoxic interfaces.

Based in these results we conclude that the presence of the municipal waste dump do not affect significantly the variables measured in these dystrophic estuaries. The variable that are significantly different (pH, DO and nitrate) were lower in Acuípe River, and this can not be related to the garbage deposit. Cururupe River conditions can be inserted in the early CONAMA no. 20 class 7 while Acuípe River, that exhibited three samples in which DO do not agree with this standard, can be classified in class 8. The DO concentrations in six samples of Acuípe River were below the limit established by class 2 and three samples below class 3 of the new CONAMA's resolution. BOD concentrations exceeding the limit established by CONAMA resolution were observed in both estuaries. The depuration of leachate within wetlands and mangrove of Cururupe River, and the greater area covered by mangroves and brackish/freshwater wetlands in Acuípe River can explain this paradoxical conclusion. These results also stressed the limitation of DO and BOD as indicators of organic pollution, and suggest that a comparative correlation analysis can be used as an indirect method to evaluate environmental disturbance. It would be also important to assess microbiological and heavy metal contamination, since Cururupe River waters are widely used to primary contact (e.g., recreation) and fisheries.

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