DOI:10.21715/GB2358-2812.2018321079

Evironmental evaluation of the middle São Francisco River basin between Três Marias and Pirapora, using chemical and geophysical investigation in sediment profiles from selected marginal lagoons

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RESUMO

A ocupação industrial, agropecuária e habitacional da bacia do Alto e Médio São Francisco está se fortalecendo, resultando em um aumento do impacto ambiental. As lagoas marginais, que são um importante apoio para a vida selvagem e para o abastecimento de água, sofrem com aporte de material alóctone durante as cheias e alagamento do rio. O objetivo deste estudo foi caracterizar a distribuição granulométrica, o desenvolvimento químico e mineral mostrado nos perfis dessas lagoas marginais e correlacionar com a evolução e os impactos ambientais. Estudos granulométricos e geofísicos foram realizados utilizando peneiras combinadas, GPR (Ground Penetrating Radar), análise química total por Fluorescência de Raio-X e análises de lixiviação sequencial por Espectrometria de Emissão Ótica. Os resultados granulométricos e mineralógicos mostraram uma predominância de silte, alternando com camadas arenosas, indicando a evolução energética anual e sazonal e fonte deposicional. Investigações com GPR mostram conjuntos horizontais formados pela mudança de deposição de energia. Elementos selecionados (Ni, Zn, Co, As, Pb, Cu) mostraram um enriquecimento contínuo com maior concentração na superfície. Todas as concentrações dos elementos estão relacionadas à fração argila/silte, permitindo assim o uso dessa fração para a investigação da avaliação de impacto. O crômio está relacionado a camadas arenosas, indicando sua correlação com o conteúdo mineral. A correlação positiva entre Ni, Zn e Pb indica influência industrial (refinamento de Zn, indústria pesada) a correlação Cu, Ni, Co e As pode derivar do uso de produtos agrícolas. Os elementos Pb, Zn e Cu apresentados no topo das concentrações dos perfis acima dos limites da legislação.

Palavras-chave: Lagoas marginais, Sedimentos, Contaminação, Metais

ABSTRACT

The industrial, farming and housing occupation of the upper to middle San Francisco basin is getting stronger, resulting in an increase of the environmental impact. Marginal lagoons, which are important support for wildlife and water supply are invaded during the flooding of the river and filled with material. The aim of this study was to characterize the particle size distribution, chemical and mineral development shown in the profiles of these marginal lagoons and correlate with the environmental evolution and impacts. Granulometric and geophysical studies were carried out using combined sieves, GPR (Ground Penetrating Radar), total chemical analysis by X-Ray Fluorescence and sequential leaching analyses by *Inductively Coupled Plasma-Optical Emission Spectroscopy*. The granulometric and mineralogical results showed a predominance of silt, alternating with sandy layers, indicating the annual and seasonal energy evolution and depositional source. Investigations with GPR show horizontal sets formed by the change of energy deposition.

Selected elements (Ni, Zn, Co, As, Pb, Cu) showed a continue enrichment with higher concentration on the surface. All elements concentrations are related to the clay/silt fraction, thus allowing the use of this fraction for the investigation of the impact assessment. Chromium is related to sandy layers indicating its correlation with the mineral content. The positive correlation between Ni, Zn and Pb indicate industrial influence (refinement of Zn, heavy industry) the correlation Cu, Ni, Co and As may derive from the use of agro products. The elements Pb, Zn and Cu presented at the top of the profiles concentrations above legislation limits.
Key words: Marginal Lagoons, Sediments, Contamination, Metals

1. INTRODUCTION

Marginal lagoons are lacustrine environments of periodic or permanent flooding, resulting from the overflow of the rivers, which receive contributions from direct rainfall or underground water (Junk, 1989). They are considered wetlands and play an important ecological and hydrodynamic role in the environment. In the upper to middle course of the San Francisco River Basin these Lagoons are responsible for breeding and restocking of migratory fish (Pompeu 1997, Pompeu & Godinho 2003). From a hydrodynamic point of view, these marginal lagoons are elongated depressions usually towards the river channel that, like the inundations plain, reduce the kinetic energy of the flood pulses, acting as areas of sediment deposition and retention (Wolman & Leopold 1957, Junk 1989). The particle size and mineral composition, the stratigraphic organization and the chemical composition of these sediments deposited in lagoons are important records of river hydrodynamics, climate changes and the entry of contaminants in drainage area (Godoy e*t al.* 1998a,b, Argollo 2001, Nery 2009). Förstner & Wittmann (1979) and Salomons & Förstner (1984) who studied concentrations of heavy metals and their amendments by processes of adsorption, precipitation, complexation and biological assimilation, observed that these contaminants accumulate in sediments over time. In this way, sediments are an important reservoir of metal concentration in the evaluation and diagnosis of environmental quality in a basin.

Agricultural activities and industrial processes are using a growing number of chemical compounds to enhance the quality and quantity of production. These chemical compounds contain elements of the transition group, some elements of groups 1, 2, 5 and 6 are used in various industrial processes or as micronutrients and pesticides in agriculture. Many of them have elevated toxicity when found in excess and their cumulative effects might affect the quality of natural resources and human health (Duffus 2002).

This work attempts to characterize the evolution of marginal lagoons sediments and to find their main contributors, using the particle size distribution and concentrations of selected elements, in the upper to middle São Francisco River basin in the region of Pirapora, Minas Gerais.

2. STUDY AREA

The study area is located in the surroundings of Pirapora city in the upper to middle course of São Francisco River, in the Northern part of Minas Gerais State (Fig. 1). In this area of the basin, a large number of marginal lagoons exist in the floodplain where the São Francisco River, in the periods of floods, reaches an average width of 4 km. Two of these marginal lagoons were selected for this study (Fig. 2).

The study area is totally inserted in the Sanfranciscana basin of Neoproterozoic age covered by Mesozoic and Cenozoic sediments. Neoproterozoic units are composed of sandy sediment of the Três Marias Formation of the Bambuí Group (Trindade 2010). The Mesozoic crops out as Três Barras Formation, overlaying the Neoproterozoic units and occurring on the tops of the hills. The Cenozoic is represented in the studied area by covers formed of iron oxide rich lateritic detritus with concretions and by not consolidated alluvial lateritic deposits in the valleys, forming the basement of the marginal lagoons. The marginal lagoons are located in the Sanfranciscana Depression developed by river evolution on the Sulamericana Surface. In the river plain mainly occur dystrophic alicos soils, specifically in the form of alicic or fluvic neosoils and poorly developed eutrophic and not discriminated planossolos (Baggio 2002). The following groups of vegetation can be observed in the study:

- “SEMIDECIDUOUS SEASONAL FOREST” in the form of Ciliary and gallery forests along the water courses;
- “SEMIDECIDUOUS PERMANENT FOREST" characterized as Dry or Mesophilic Forests;
- “SAVANNAS” covering the various specific formations that characterize the region of the Cerrado;
- “PIONEER FORMATIONS” included the Buritizais or veredas and lowland vegetation (varzeas);
- “ANTROPOGENIC FORESTS” which are the reforestation with pinus and eucalyptus and the agricultural systems, according to IBGE (1992).

Based on the Köppen classification, the climate is of Aw- type, tropical rainy, hot, humid with dry winter and rainy summer (Patrus *et al.* 2001). It is characterized by the average temperature of the coldest month always higher than 18ºC (CPRM 2001).

In this region the evolution of soil use and occupation was strongly influenced by the actions of the “Superintendência de Desenvolvimento do Nordeste” (SUDENE) and the “Companhia Vale do São Francisco” (CODEVASF) which granted land and tax incentives for deployment of monocultures of *pinus*, eucalyptus and fruit-farms in rural region and for heavy metal industry in the urban region of Pirapora and Várzea da Palma (Baggio, 2002). In addition, since the 1960, large industrial parks were installed.

The natural landscape has been greatly modified by the replacement of native vegetation, and the installation of industry in urban centers. From the Decade of 1970, there was a significant increase in urban population. From 1990 a diversification and intensification of the land use and occupation with the introduction of the monocultures of soy, corn and coffee in rural areas and the arrival of the textile industries in the industrial district of Pirapora was registered (Baggio 2002, Gama *et al*. 2003, Gama 2006). Currently these anthropogenic activities, historically recognized for using large amounts of heavy metals, occupying part of the inundation plain, can have strong environmental pressure on natural resources.

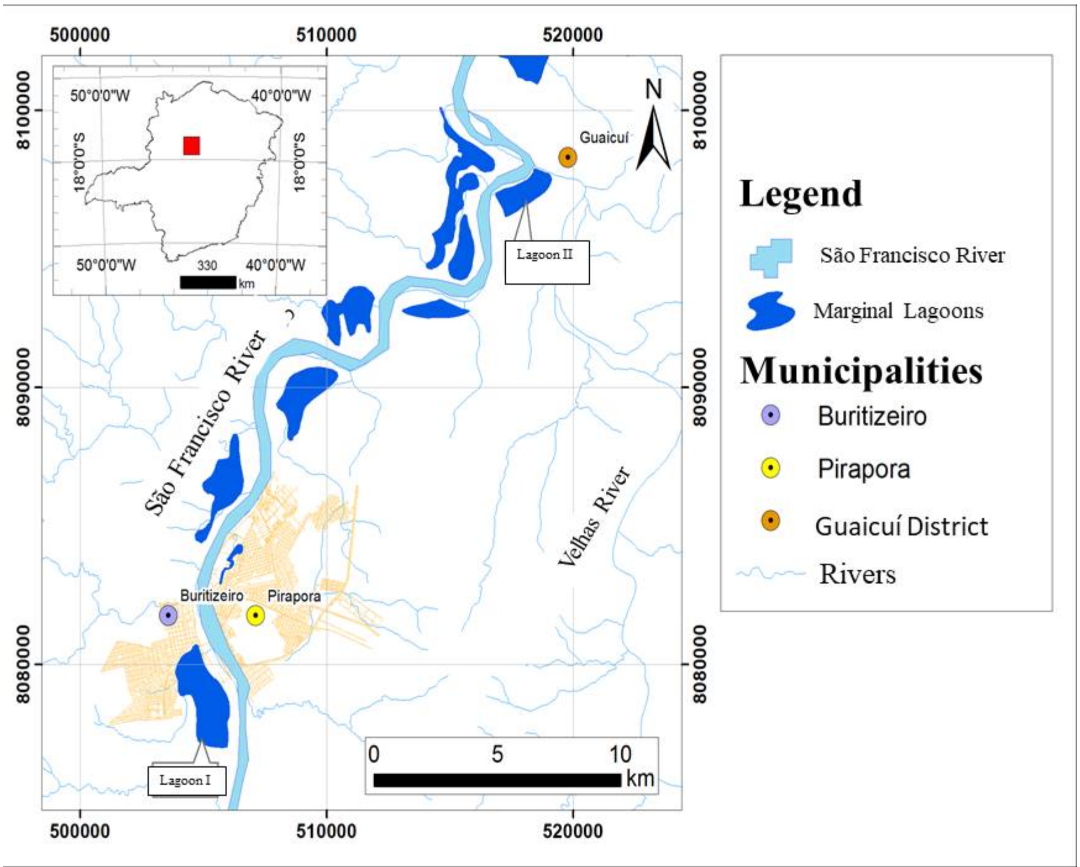

Figure 1
Location of marginal lagoons in the study area and the selected ones for sampling of sediments and GPR survey.

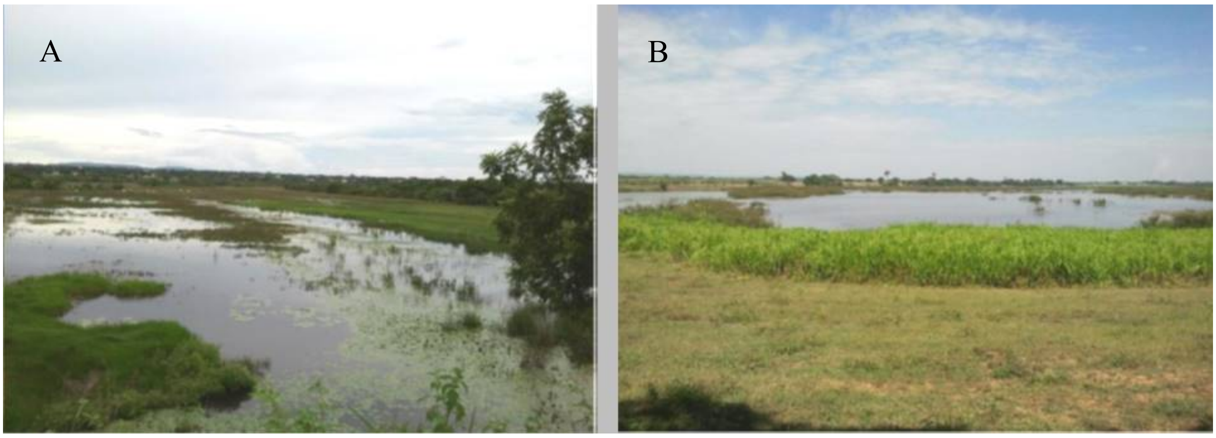

Figure 2
View of the two marginal lagoons selected for this study. The first lagoon (I) located upstream of the cities of Buritizeiro and Pirapora, occupies a total area of ~462 hectares. The second lagoon (II) occupies an area of about 190 hectares and is located downstream of the urban centers, industrial park and agricultural project (fruit production). A. Guím Lagoon (I); B. Pontal Lagoon (II).

3. MATERIALS AND METHODS

Two marginal lagoons were selected (Fig. 2), geographically separated and associated with different land uses and occupation, which are located upstream and downstream of the Pirapora city. Samples were collected in bore holes oriented by executed GPR - profiles, using equipment Mala/Ramac with antennas of 100 MHz and the following parameters: common offset, horizontal steps: 0,1m; stacks: 8; time window: 400 ns. The profiles were obtained perpendicular to the lagoon direction, indicating the best places for sampling.

Sediment samples were collected with a percussion-coring rig, which allowed the extraction of testimony with a good preservation of the sedimentary layers. In the Lagoon I, sampling reached a depth of 350 cm that was subdivided into 14 layers. In the Lagoon (II), sampling reached 690cm divided into 28 layers. The layers were identified and separated in the field, considering morphological aspects, color and texture (Fig.3). The material was sealed in plastic bags and transported in coolers under low temperature conditions and the samples were stored at 4^0C until analyzed.

Figure 3
This figure show the sediment distribution of the core samples. Abrupt contacts are indicated, showing changes in transport energy. These surfaces can be correlated with GPR indications

The GPR profiles were processed using the program Gradix (DOS version). We interpreted the radargrams based on the reflection patterns, the continuity of the reflector and interruptions of the reflector.

The granulometric fraction of each layer <0,2 mm were examined in a particle Analyzer Sympatec Laser System-Partikel-Technik H2387 model from the method of x-ray diffractometry with detection ranges between 0.2 and 2000μm and classified according to the scale of Wentworth (1922).

The samples were dried at 120°C, sieved and the particle size fraction <0.164 µm used for chemical analyses. Analytical investigation of the selected elements (Cr; Ni; Cu; Zn; Cd; Pb; Co and As) were done by using XRF (X-Ray Fluorescence) analyze, ICP OES (Optic Emission Spectroscopy with inductive coupled plasma). The bulk composition of these sediments was obtained by XRF in the fine material. The fine sediment (0.063 mm) was subjected to acid digestion in microwave MARS-CEM in accordance to the method SW-846-3051 – US EPA (US EPA, 1998). About 0.50 g of fine fraction of the sediment was digested with 10 ml of concentrated nitric acid (HNO_3) for 10 minutes (ramp time) and temperature stabilization at 180 ° C and pressure (350 psi) for 4 '30 "(hold time). Samples were then filtered in cellulose filter (0.45 µm) and analyzed in an ICP-OES (Spectroflame from Spectro Analytical Instruments).

4. RESULTS AND DISCUSSION

The results of particle size distribution and elemental concentrations were organized in profiles, using the STRATER 3.0 software and correlated with GPR - profiles and evaluated using STATPLUS pro 9.0 for calculating minimum, maximum, average, mean and standard deviation. Variations in the distribution of selected elements and granulometry were correlated with natural and anthropogenic events that have occurred in the basin. The data, with the exception of Co, were compared with values of sediment quality Guide (CONAMA, 344/04; 2004; Table 1). Variations in the distribution of elements and particle size can be related to natural and anthropogenic events that have occurred in the basin. In the assessment of distribution of minerals and grain size distribution in the sediment profiles, it can be indicated the variations in river transport energy (silt and clay) and also the distribution of contaminant adsorption capacity (Förstner & Wittmann 1979, Sawer *et al.* 2003). It can be seen clearly a variation among high (sand; gravel) and low energy deposition (clay, silt) and dry periods with organic material, indicating the annual and periodic evolution of the system (Fig. 4). After construction of Três Marias Lake and power plant, the sand deposition is limited to the Lagoon II (Lagoa do Pontal) at the confluence with the Das Velhas River.

Table 1 - Indicated element concentrations for level 1 and level 2 of CONAMA resolution 334/04 of 2004.

Element	As	Cd	Pb	Cu	Cr	Ni	Zn
Level 1	5.9	0.6	35	35.7	37.3	18	123
Level 2	17	3.5	91.3	197	90	35.9	315

In the Lagoon I – Guím Lagoon, granulometric distribution is relatively homogenous, typical of low-energy environments with some influx from rain flux and wind deposition. Predominant percentages of silt (45-70%; ~46%), clay fraction (~ 21%; ~20%) and sand (6%; ~34%) occurs in small amounts, indicating full. Only 6 of the 4 segments of the profile have contribution with sand.The sediments of the Lagoon II - Pontal Lagoon are predominantly siltic, with a higher dense frequency of sandy layers on the base (fig. 3) and less at the top. In 28 of the separated layers were found mean values of 45.87% of silt; 20.04% of clay; and 34.08% sand. The sediments deeper than 180cm are predominantly sandy, reaching 95.15% C10 layer. In this point, sandy layers are generally thicker showing rhythm deposition with down grading (Fig. 4). The elements show a variation in the profile, indicating different natural processes and anthropogenic influence. The total content of selected elements was compared with reference values of Bowen (1979) for geological medium shale (Tab. 2), showing that the metal concentration in our samples are much higher than in normal, not contaminated sediments. The average, maximum, minimum values are presented in table 3. All determined elements have concentrations above the reference values (CONAMA 2004) in determined profile positions. Cr, Cd and Pb are higher the reference values in all 14 profile layers of the first Lagoon. In the Lagoon II, Ni and Zn, are within the allowed range, with exception of Cu that has high concentration at the upper part of the profile. The distribution of the profiles of the two lagoons show a correlation between metals concentrations and increasing anthropogenic activities (Table 2, Figure 5). Cr and Ni concentrations increase from bottom to the top, from 76-87 mg/kg to 146 mg/kg (C5). Zinc occurs in higher concentrations at the top (124 mg/kg) and the lowest values are reported at the base (35 mg/kg). Cu, (~45 mg/kg), Pb (~36 mg/kg) and Cd (~30 mg/kg) also show a bottom-up enrichment, whereas Zn, Co and As has the highest concentrations at the base of the profile.

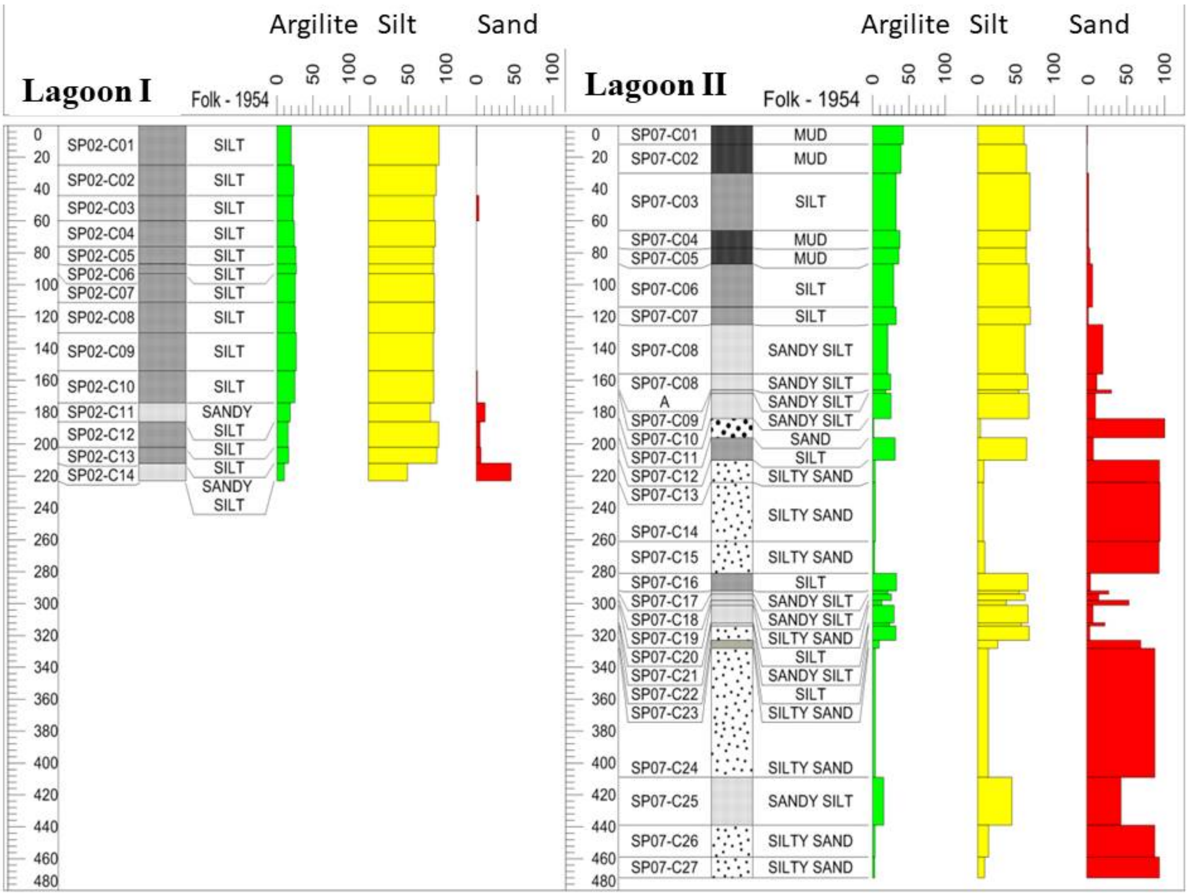

Figure 4
Granulometric profiles of the two lagoons.

Table 2 - Reference values for element evaluation after Bowen (1979)

Elements (mg/kg)	Cr	Ni	Cu	Zn	As	Cd	Pb	Co
Shales	90	68	39	120	1.8	0.22	23	19

Table 3 - Descriptive statistical data of the selected elements and particle size classes.

	Lagoon I										
	Cr	Ni	Cu	Zn	As	Cd	Pb	Co	Sand	Silt	Clay
Number of samples	14	14	14	14	14	14	14	14	14	14	14
Mean	134.1	60.2	37.4	91.3	12.1	21.1	31.2	23.6	5.2	73.8	21.0
Standard Deviation	10.6	15.4	8.1	25.7	13.7	6.3	6.5	8.5	12.1	8.8	4.9
Minimum	106.0	24.0	17.0	35.0	0.0	7.0	21.	10.0	0.0	44.8	9.7
Maximum	146.0	81.0	46.0	124.0	46.0	30.0	48.0	40.0	45.6	80.7	25.9
	Lagoon II										
	Cr	Ni	Cu	Zn	As	Cd	Pb	Co	Sand	Silt	Clay
Number of samples	28	28	28	28	28	28	28	28	28	28	28
Mean	189.7	35.1	23.3	49.8	16.5	18.0	24.9	12.6	34.1	45.9	20.0
Standard Deviation	91.4	14.3	10.7	23.6	20.3	8.5	9.0	6.9	35.4	23.6	12.5
Minimum	108.0	13.0	6.0	10.0	0.0	3.0	11.0	1.0	0.4	3.4	1.4
Maximum	440.0	61.0	42.0	100.0	93.0	40.0	41.0	25.0	95.2	67.3	40.2

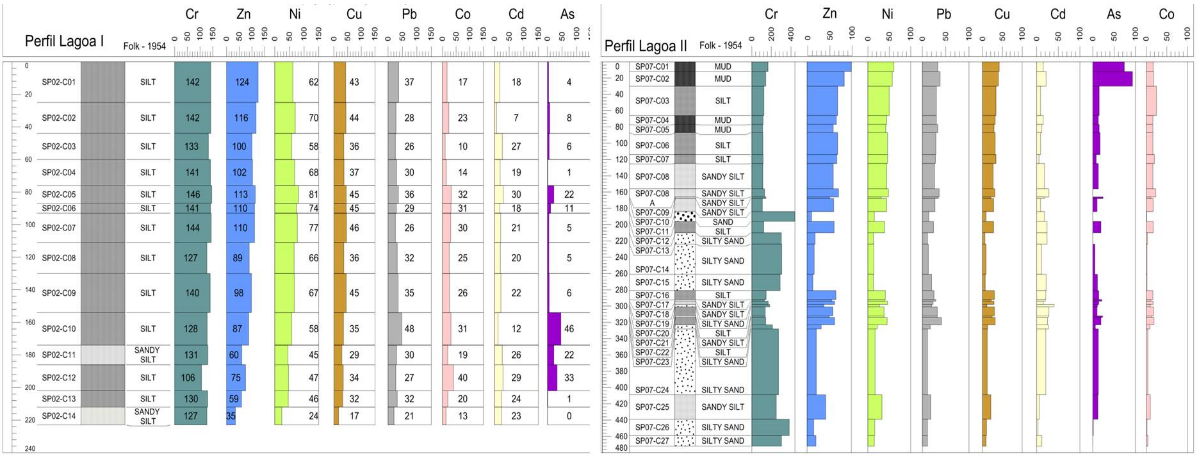

Figure 5
Element distribution of the profile from the Lagoons.

In the Lagoon II, the average concentration of Cris 126mg/kg. The distribution shows a significant hiatus at 200cm, decreasing from >400 to 180 an than an increase to 230mg/kg at the top of the profile. The As have an similar behavior, but with significant higher levels at the top. Cd decreases significantly from the top (>40cm) to the base (>450cm). A contrary behavior is shown by the elements Zn, Ni, Pb, Cu and Co with strongly increasing values to the top). Generally, the element concentration increases not homogeneously from the base to the top of the profiles with the exception of Cr, which is more concentrated in the basal part. Pearson's Correlation tests performed (Table 3) indicate a strong positive correlation between groups of elements and size classes.

Table 3 - Pearson's correlation coefficient matrix for the two lagoons.

	Clay	Silt	Sand	Cr	Ni	Cu	Zn	As	Cd	Pb	Co
Lagoon I											
Cr	0.60	0.10	-0.32	1.00							
Ni	0.92	0.62	-0.82	0.66	1.00						
Cu	0.83	0.70	-0.85	0.61	0.93	1.00					
Zn	0.78	0.64	-0.78	0.64	0.90	0.92	1.00				
As	0.08	0.21	-0.18	=0.43	-0.02	-0.01	-0.06	1.00			
Cd	-0.30	-0.11	0.20	-0.31	-0.24	-0.25	-0.34	0.01	1.00		
Pb	0.43	0.42	-0.48	0.13	0.30	0.31	0.29	0.57	-0.29	1.00	
Co	0.30	0.36	-0.38	-0.25	0.36	0.41	0.19	0.63	0.04	0.29	1.00
Lagoon II											
	Clay	Silt	Sand	Cr	Ni	Cu	Zn	As	Cd	Pb	Co
Cr	-0.83	-0.92	0.91	1.00							
Ni	0.95	0.93	-0.95	-0.82	1.00						
Cu	0.97	0.92	-0.95	-0.80	0.98	1.00					
Zn	0.95	0.92	-0.95	-0.84	0.98	0.97	1.00				
As	0.56	0.37	-0.44	-0.33	0.59	0.58	0.62	1.00			
Cd	-0.05	-0.01	0.03	-0.13	-0.07	-0.13	-0.02	0.07	1.00		
Pb	0.85	0.86	-0.87	-0.80	0.84	0.84	0.84	0.52	0.16	1.00	
Co	0.89	0.95	-0.95	-0.87	0.92	0.91	0.90	0.33	0.02	0.82	1.00

In the lagoon I, Ni, Cu, Zn showed strong correlation, Cr, Ni, Cu and Zn and Pb, Co, Zn, a weak one (Table 3). Ni, Co and Zn are well correlated with the silt and argillite fraction and Co and As weak. For the lagoon II were observed strong coefficient of Pb, Ni, Cu, Zn and Co, The moderate, Ni, Cu, Zn, Pb and Co and weak between Pb and Cd (Table 3). Here Ni, Cu, Zn, Co and Pb was Strong correlated with silte and argillite and Cr strong with sand. The results obtained in this study show clearly a correlation between natural climatic factors and the interfering of anthropogenic activities. The granulometric evolution show a correlation between rainy and dry periods in the form of deposit evolution and sediment surface conditions. The installation of the Três Marias

Lake and power plant regulated the water flux to medium quantities, limiting sand deposits in the marginal lagoon to minimum events. Element distribution is clearly related to overall concentration of fine fractions in the sediments showing an evolution with incase of anthropogenic activities, like observed by Lima *et al.* (2011), Sabaris (2011), Nery (2007), Argollo (2001) and Godoy *et al.* (1998a,b) in other environments.

The different distribution in the two Lagoons show clearly a time related reaction to upstream industrial and agriculture activities and the influence of peculiar industrial activities in the Pirapora region This explains also the concentration variation between the elements in the profiles from the two lagoons. The high concentrations of Cr, Zn, Ni e Cu (samples between 76 to 79 cm; Fig. 5) are probably indicating changes in sediment sources, probably due to anthropogenic caused mass movements (lake construction; soil movements for agriculture). The regulatory effect of the lake is shown by the predominance of fine sediments over coarse-grained causing a reduction of energy potential (Collinson & Thompson 1982). The enrichment of the analyzed elements observed at the top of the profiles of the both lagoons (I and II), and its environmental available forms exceeds the values established in the Quality Guide of Sediments (CONAMA 344/04). Cu goes beyond the level II at the top (35mg/kg) while Ni shows high values (18 mg/kg) at whole profiles culminating at the profile of lagoon I (> 88 mg/kg) observed also by Trindade (2010) in current upstream sediments, indicating contamination by agricultural activities.

Zn show similar distribution in the upper parts of the two lagoons, but with higher concentration (>Level II) in the second lagoon, showing the lower influence of Zn industry from Três Marias and the contributions of agriculture and industry, observed in samples from the river (Ribeiro *et al.* 2010). Cr (>level II) e Pb (between level I and II) show higher values in the lower part, indicating a reduction of the use of these elements in traffic, industry and agriculture. Baggio (2008) and Trindade (2007) connected this to agricultural and industrial evolution, too.

5. CONCLUSIONS

The marginal lagoons of the São Francisco River without any doubt are of great importance for biodiversity conservation. However, the data presented in this work demonstrate, even preliminarily, that these environments act as sediment reservoirs that can provide important information about the flow of contaminants in the floodplain.

The application of XRF and LPSA (laser particle size analysis) in vertical profiles of the marginal lagoons proved their fast and effective use in characterization of sediments. It also has shown the occurrences of mineral areas of heavy metals in depth and the existence of distinct from deep-water depositional patterns.

The results showed that the highest concentrations of heavy metals in the marginal lagoons are strongly associated with fine sediments typical of low-energy environments, demonstrating the importance of these systems for spatial-temporal evaluation of environmental quality.

The use of geochronological tracer may support the time distribution of the contamination and help to follow contamination profile in time and space.

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