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IMPACT OF EUTROPHICATION ON BENTHIC FORAMINIFERA IN SEPETIBA BAY (RIO DE JANEIRO STATE, SE BRAZIL)

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Abstract

The main objective of this work is to analyze the consequences of eutrophication in benthic meiofaunal organisms (foraminifera) in an internal area of Sepetiba Bay (State of Rio de Janeiro, SE, Brazil). This study is interesting because, at the studied site, the sediments corresponding to the period of greatest accumulation of organic matter were not contaminated by metals, although Sepetiba Bay has areas known to be polluted by this type of contaminants. Thus, in this study, it was possible to consider only the impact caused by eutrophication.

In the SP6 core, collected in the Sepetiba Bay internal area and studied in this work, textural, geochemical (concentration of chemical elements; ^{210}Pb and ^{137}Cs; radiocarbon data) and foraminifera were analyzed in a muddy section, with a few intercalations of sandy layers. The sediments of the analyzed section were deposited in the last ≈2,350 BP years. The concentrations of Al, Cd, Co, Fe, Mn, Ni, Pb, Sn, Ti, V and Zn are higher at the base of the core and decrease towards the top. The contents of TOC, P and Zr have inverse paterns. The ratios of these elements to their background value have similar patterns. However, previous studies have recognized that during the twentieth century Cd, Zn, Cr and Pb concentrations increased in several areas of Sepetiba Bay, mainly due to industrial activity. The area where this core was collected may have been dredged. Radiocarbon ages suggest a loss of ≈2000 years of sedimentary registration, marked by an unconformity at a depth of 126 cm, probably caused by dredging. A new sedimentary sequence unpolluted by metals but highly enriched in organic matter was deposited on the surface of that discontinuity. Foraminifera were quite abundants in the lower section (240-126 cm; between ≈2,400-2,090 years BP, execept in the sandy layers), corresponding to sediment deposition before dredging. After dredging, the accumulation of very fine-grained sediments rich in organic matter generated eutrophication phenomena, which caused a drastic reduction in the abundance and diversity of these organisms. This work testifies the effect of eutrophication on meiofaunal organisms. Although some coastal foraminifer species tolerate harmful effects caused by eutrophication, it is recognized that the impact has been so negative that even these species occur with reduced abundance in the study area.

Keywords: Coastal Area. Paleoenvironmental Record. Anthropic Impact. Meiofauna, Multiproxy Approach.

1. Introduction

Foraminifera are amoeboid protist organisms and have in general short life cycles and responds quickly to environmental changes caused by any kind of disturbance such as hydrodynamism, variation of physicochemical paraments, pollution by metals and organic matter (Martins et al., 2015 a; Duleba et al., 2018; Frontalini et al., 2018;

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Belart et al., 2018). After death, calcareous and many agglutinated tests are preserved in the sediment. These characteristics make the foraminifera excellent indicators of past environmental events (Ellison and Nichols, 1976).

Foraminifera in Sepetiba Bay were studied by Tinoco (1965), Zaninetti et al. (1976, 1977), Brönnimann and Beurlen (1977 a, b), Brönnimann (1978, 1979), Brönnimann et al. (1981 a, b, c), Brönnimann and Zaninetti (1984), Santos-Leal et al. (2009), Laut et al. (2006, 2012), Laut and Rodrigues (2011) among others. Only few studies used foraminifera to study the paleoenvironmental evolution of this region, e.g. Silva (2006) and Pinto et al. (2016, 2017).

Pinto et al. (2017) observed a marked reduction in foraminifera at the top of core T4 (in this work referenced as SP6) collected near the Guandú River mouth, in an inner area of Sepetiba Bay (Rio de Janeiro, SE Brazil). But the causes of such a marked reduction in meiofauna in this inner area have not been clearly explained. This study aims to clarify the possible causes for the almost disappearance of foraminifera in the recent sedimentary record of the analyzed site located in the inner area of Sepetiba Bay.

1.1 Study area

Sepetiba Bay is a semi-confined water body located in southwestern of Rio de Janeiro State between the latitudes 22° 55'S and 23° 05'S, and longitudes 43° 35'W and 44° 00'W (Fig. 1). It is an estuarine and mangrove region, and a natural breeding area for the various species of mollusks, crustaceans and fishes (Cardoso et al. 2019).

The bay has a mean depth of about 8 m in the central area and is much shallower in the inner region (Borges and Nittrouer, 2015). Microtidal semidiurnal-tides generate currents that can reach maximum speeds of 75 cm s^{-1} in the channels between islands, while mean current velocities are about 20 cm s^{-1} (DHN, 1986; Villena, 2003). Water circulation occurs clockwise (east/southwest) and is considered slow due to the tide overlapped by an almost steady flow, which is induced by different water density gradients. Ikeda et al. (1989), Creed et al. (2007) and Moura et al. (1982) divided Sepetiba Bay into three compartments based on its hydrographic and geographic characteristics: brackish (3-18, at Guandú River mouth), hyposaline (18-30, most of the bay) and hypersaline (30-40, near the islands and northwest and southwest parts of the bay).

The contributing watershed of Sepetiba Bay occupies an area of approximately 2,711 km², equivalent to almost 4.5% of the total area of the Rio de Janeiro State. It is bounded on the continent by Serra do Mar (the source of the rivers that form the Guandú drainage basin), by the isolated hills that divide the Guanabara Bay and the coastal massifs of Mendanha and Pedra Branca and an extensive area of lowland, crossed by many rivers, consisting of 22 sub-basins (SEMADS, 2001; Ferreira and Moreira, 2015). The main rivers flowing to Sepetiba Bay and its respective average discharge are Guandú (89 m^3 s^{-1}), Guarda (6.8 m^3 s^{-1}), Itá (3.3 m^3 s^{-1}), Piraquê (2.5 m^3 s^{-1}), Portinho (8.8 m^3 s^{-1}), Mazomba (0.5 m^3 s^{-1}) and Cação (1.1 m^3 s^{-1}) (Ferreira and Moreira, 2015).

The vegetal cover of the basin presents remnant areas of native vegetation and in regeneration stage, such as forests, mangroves, fields, pastures and agricultural areas (Costa, 2010). Forests are characterized by fragments of various sizes and successional stages, located on the tops and slopes of the mountains and cover about 40% of the basin area (SEMADS, 2001).

The humid tropical climate of the region and the high rainfall induce the shape of the relief, facilitating the action of rock weathering (SEMADS, 2001). Erosion is favored by geomorphological features and by human occupation in the hillside areas (Costa, 2010), contributing to the silting process of the region. Sediment accumulation in the river beds prevents free flow and causes overflowing in periods of high rainfall (Costa, 2010). However, since the seventeenth century the low river courses have been rectified, dredged, channeled, joined by valleys to avoid the constant flooding of this region due to its flat topography. The intervention works in the river basins that flow into Sepetiba Bay were began by the Jesuit priests, who lived in the Sepetiba Region between 1616 and 1759, when they were excluded by the policy of the Marquis of Pombal (SEMADS, 2001). The Guandú River mouth, near the study area, through the São Francisco Canal, in Sepetiba Bay, is occupied by mangroves and has a delta in formation (SEMADS, 2001).

The Sepetiba Bay is the target of economic, strategic and geopolitics that are reflected in a complex entanglement of mega-projects with high potential of social and environmental impact (Moreno and Kato, 2015). The so-called Sepetiba Industrial Complex covers the industrial areas around the bay, which include the Sepetiba Industrial Complex, the Itaguaí/Sepetiba Harbor backyard and the Santa Cruz Industrial District. The Sepetiba Bay includes the Southeast Port, the Itaguaí Port and the TKCSA Port (Fig. 1). According to Almeida (2004) and Sá (2008) dredging works were made aiming to deepen the main navigable channel to receive larger and faster vessels to the Itaguaí Port. Due to the industrial development and urban centers, the region has become the second main wastewater recipient of the Rio de Janeiro State, greatly affecting the diversity of aquatic organisms and generating social-environmental problems (Melges-Figueiredo, 1999). In the social and public health context, it has been repeatedly impacted with sewage discharging from domestic and industrial sources, without treatment at some points and with questionable treatment in its majority and, therefore, has high concentration of heavy metals in water, sediments and living organisms (Pellegatti, 2000; Wasserman et al., 2001; Molisani et al., 2004; Ferreira, 2009; Rocha et al., 2010; Ferreira and Moreira, 2015; Díaz Morales et al., 2019).

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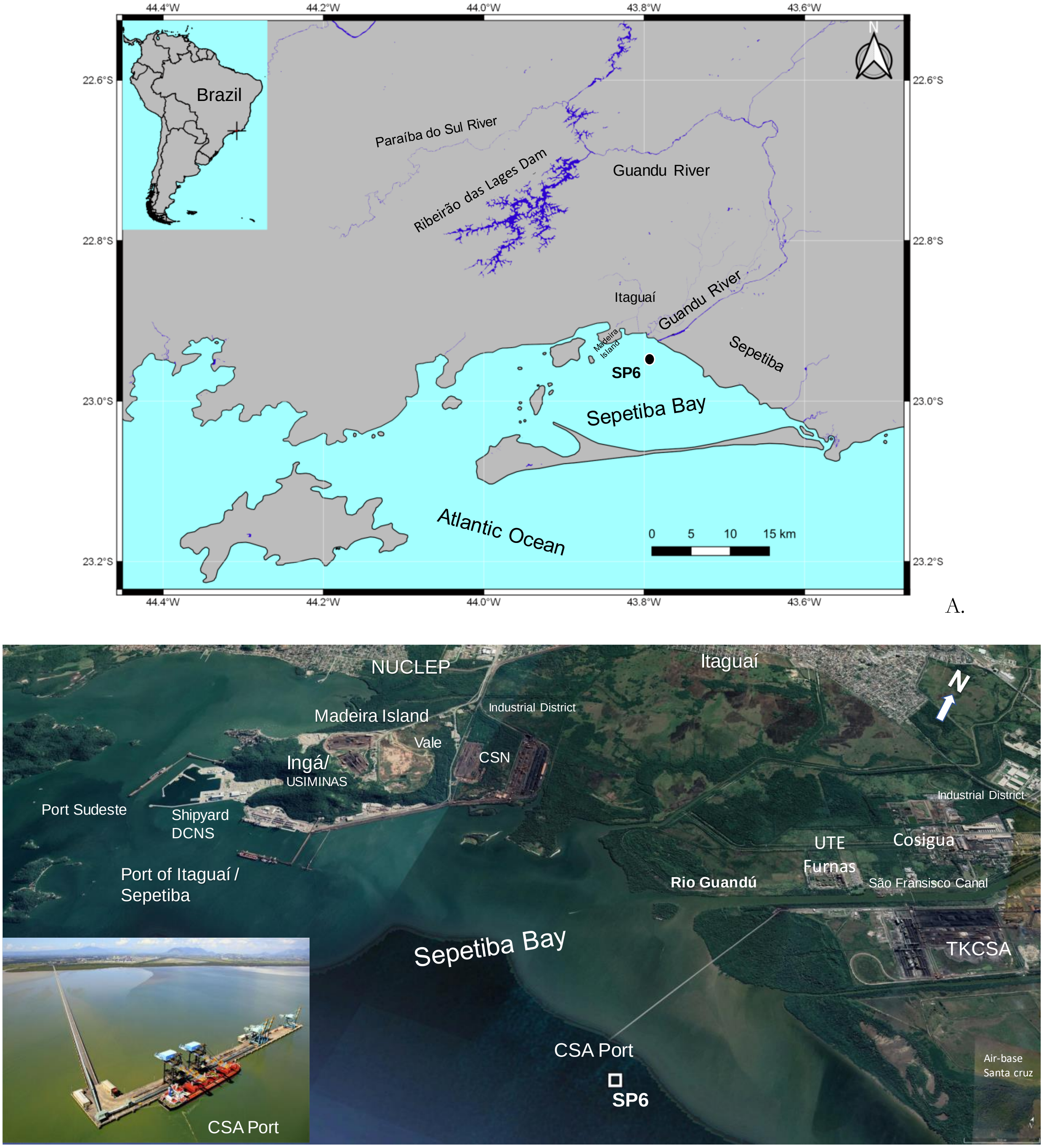

Fig. 1. Location of core SP6 in Sepetiba Bay. This core was collected near the TKCSA Port near the outflow of Guandú River. Other industries and industrial districts are also signaled. The study area is also close to the Santa-Cruz Airport (a military air-base). Legend: TKCSA – "Thyssenkrupp Companhia Siderúrgica do Atlântico" (Thyssenkrupp Atlantic Steel Company). NUCLEP – "Nuclebrás Equipamentos Pesados S.A" (Nuclebras Heavy Equipment S.A). Ingá - Ingá Mercantil. CSN – "Companhia Siderúrgica Nacional" (National Steel Company). Furnas – "Furnas Centrais Elétricas" (Furnas Power Stations). Cosigua – "Companhia Siderúrgica da Guanabara of Gerdau" (Guanabara of Gerdau Steel Company); DCNS – composed by Odebrecht Defense and Technology (50%) and "Direction des Constructions Navales et Services" (Directorate of Shipbuilding and Services).

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Enrichment of chemical elements such as Cd, Cu, Ni, Pb and Zn exceeding the recommended limits by the Brazilian legislation and the natural values have been found in Sepetiba Bay, as a consequence of human, harbors and industrial activities, as well as dredging, which resuspend the sediments and contribute to the reintroduction of metals in the water column (e.g. Silva-Filho et al., 1996; Wasserman et al., 2001; Patchineelam et al., 2011; Díaz Morales et al., 2019).

2. Materials and methods

The sediment core SP6 (286 cm long) was collected in 2015, in the inner area of Sepetiba Bay, at 3.0 m of water depth, in front of the Rio Guandú mouth, through the São Francisco Canal near the TKCSA Port (Fig. 1). This core was sampled every 2 cm. Samples were submitted to textural, mineralogical, geochemical and foraminiferal analyses.

In this work new geochemical (elemental concentrations and ^{210}PB and ^{137}CS results by gamma spectrometry) and mineralogical data were compared with foraminiferal and total organic carbon (TOC) and total sulfur (TS) data acquired by Pinto et al. (2017; as core T4), aiming to access the causes that lead to the almost the disappearance of foraminifers in the upper section of the analyzed site, the main aim of this work. Only the upper 240 cm of core SP6, corresponding to a fine sedimentary sequence, was studied in this work, since the base (285-240 cm) consists of coarser sediments. This option was adopted aiming to avoid the negative effect of stronger hydrodynamic conditions on foraminifera records.

2.1 Sedimentological Analyses

Grain-sizing was carried out by wet sieving. The sediment was washed with distilled water over mesh sieves of 63 µm, 125 µm, 250 µm, 500 µm, 1000 µm and 2000 µm. All the sedimentary fractions were collected and dried in an oven at about ≈60ºC. The weight and percentage of each dried granulometric fraction were determined. The sediments granulometry were classified according to Folk and Ward (1957).

For TOC, TS, carbonates and insoluble residue contents, a split of each dry sample was macerated (reduced to grain size <63µm), homogenized, decarbonated through acidification of the sample (HCl - 1mol.l $^{-1}$) and then dried in an oven (at ≈60ºC) for 12h. The decarbonation process was repeated twice in each sample. For the determination of TOC, an aliquot of 10 mg of the already processed sediment was used and the analysis was performed with a SC 634 equipment of the LECO.

Mineralogical analysis was carried out in the sediments fine fraction (<63 µm) by X-Ray Diffraction (XRD) techniques. The samples preparation and the semi-quantification were described by Martins et al. (2007).

Concentrations of Al, As, Ca, Cd, Co, Cr, Cu, Fe, La, Mg, Mn, Nb, Ni, P, Pb, Sc, Sn, Th, Ti, V, Y, Zn and Zr were estimated in sediment fine fraction by total acid digestion (with 4 acids, HNO_3-$HClO_4$-HF and HCl) followed by Inductively Coupled Plasma Mass Spectrometry (ICP-MS), and/or Inductively Coupled Plasma Emission Spectrometry (ICP-ES) at the ACME Laboratory (Analytical Services), Vancouver, Canada.

To assess the enhancement of chemical elements, in each sample, the enrichment factor (EF) was calculated for each element, according to the procedure suggested by Buat-Menard and Chesselet (1979):

$$EF = \frac{\left(\frac{Cx}{Cn}\right) Environment}{\left(\frac{Cx}{Cn}\right)\ Baseline}$$

Where Cx is the concentration of the *x* element whose enrichment is to be determined and Cn is the concentration of the *n* normalizing element assumed to be characteristic of the baseline. For the baseline, the values estimated by Pinto et al. (2019) were considered. In the present study, the Al was considered as normalizer. Since Al, is a widely used element as a geochemical normalizer to minimize possible effects of sediment granulometric and mineralogical heterogeneity (Machado et al., 2008), representing the clay minerals sedimentary component, as suggested by Soares de Almeida et al. (2019).

The variation of PTE contents was compared with the baseline value, by dividing the concentration of each element (C_{metal}) in the sediments by the respective the baseline ($C_{baseline}$) in the study area estimated by Pinto et al. (2019). Tomlinson et al. (1980) named this ratio as contamination factor (CF). In this study we considered the CF as a concentration factor:

$$CFmetal = \frac{C_{metal}}{C_{baseline}}$$

2.2 Foraminiferal Analysis

Foraminifera density (FD: the number of specimens per each species per 10 ml of sediment) and the species richness (number of species found per 10 ml of sediment in each sample) were determined in the sediment fraction 63-500 µm, separated by wet sieving from aliquots of 10 ml of total sediment collected along the core SP6. Foraminiferal tests were separated from the sediment, identified and counted under a binocular microscope (LEICA MS16F, maximum magnification 480X) and fixed in microslides.

The species identification was based on several references, such as Loeblich and Tappan (1988), Debenay et al. (1998), Semensatto-Jr. and Dias-Brito (2004), Raposo et al. (2016), Belart et al. (2017) and Alves Martins et al. (2019). The species nomenclature was updated following the World Foraminifera Database (Hayward et al., 2017).

2.3 Sediment Accumulation Rate

Sediments also were submitted to ^{210}PB, ^{226}RA and ^{137}CS analyses by gamma spectrometry in the Laboratory of Spectrometry Gamma, of the Oceanographic Institute,

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University of São Paulo (Brazil). The methodology for determining the radionuclides ^{210}Pb, ^{226}Ra and ^{137}Cs was established by that laboratory and are described in detail in Figueira et al. (2007) and Ferreira et al. (2014).

2.4 Radiocarbon dating

Three results of radiocarbon dating, performed in the Beta Analytic Laboratory (Miami, Florida), were obtained on mollusk shells recovered from 114-112 cm and on organic matter from the 135-130 cm and 215-212 cm sediment layers. The age obtained for the layers 135-130 cm and 215-212 cm was calibrated using Marine 13 database (Reimer et al., 2013) and the delta R 32 ± 44 (Alves et al., 2015).

2.5 Statistical analysis

Shannon index values were determined for samples with FD >100 specimens. Selected abiotic and biotic data were transformed by log (x + 1) and submitted to Principal Components Analysis. Pearson correlations also were estimated. Statistical analyses were carried out with the Statistica 12 software.

3. Results

Results of radiocarbon dating are presented in Table 1, indicating the following calibrated ages: for 112-114 cm, 60 years calibrated before Christ (yrs cal BP; 1955 AD; AD - *anno domini*); for 135-130 cm, 2011±30 cal BP (146 cal BC - 24 cal AD; BC – before Christ) and; 215-212- cm, 2.350 ± 30 cal BP (415-385 cal BC). Recent sediment accumulation rates (Table 2; Fig. 2) estimated by ^{210}Pb and ^{137}Cs radionuclides activity were 0.26±0.04 cm yr^{-1} and 0.35±0.03 cm yr^{-1}, respectively. Core SP6 is a muddy sedimentary sequence with intercalations of sandy sediments such as between 142-124 cm (Figs. 3, 4; Appendix 1).

3.1 Sedimentological Results

The mineralogical composition of the sediments is mostly composed by phyllosilicates, quartz, K-feldspars and plagioclase, also including, minerals, such as calcite, anatase, anhydrite and pyrite (Appendix 1). Phyllosilicates are the main abundant minerals (36.6-90.0 %; mean 73.5%). The depth plot of phyllosilicates, quartz and feldspars show that the percentage of these minerals oscillates along the core without a striking trend (Fig. 4). Pyrite is present mostly at the base and the core top (Fig. 4). The percentage of calcite is reduced (<7.6%; mean 0.3%).

The TOC values along the core SP6 were in generally high (mean = 2.0 ± 0.4%), ranging from 1.4-2.90 %. A tendency of increasing of TOC values from the base to the core top (R^2 = 0.58%) was observed (Fig. 5). The highest TOC contents were observed in the section 126-72 cm (2.0-2.9%) and the lowest at the core base between 240-168 cm (1.4-1.5%) (Fig. 5). The mean value of sulfur content was 0.90% (0.5-1.5 %; mean 0.9± 0.2%). The highest values of S were recorded between 180-100 cm and in the upper section of the core (first 50 cm) (Fig. 5). The C/S ratio varied between 1.3-3.7 (mean 2.3± 0.4). Relatively high C/S values were recorded between 124 cm and the core top (Fig. 5).

Concentrations range of the analyzed chemical elements (Al, As, Ca, Cd, Co, Cr, Cu, Fe, La, Mg, Mn, Nb, Ni, P, Pb, Sc, Sn, Th, Ti, V, Y and Zn and Zr) along the core SP6 are presented in Table 3 and Appendix 2. Calcium contents are low (mean: 0.29±0.04 %) and vary around the mean values.

The depth plots of Al, Fe, Ti, Ca, P, Mn and V contents presented in Fig. 6A show sudden changes in the tendency of vertical distribution of these elements around the 126 cm depth. Concentrations of Al, Fe, Ti, Mn and V (Fig. 6A) and the CF values of these elements (Fig. 6B) tend to decrease in the upper core section (126-0 cm). Zinc, Cr, Pb, Ni, Sn, Co and Cd contents also tend to decrease in this section presenting an opposite pattern to P (Fig. 7A) and Zr. The same trends are observed for the respective CF of each element (Fig. 7B). Cyclic changes around the mean were observed for As and Cu contents along the core SP6 (Supplementary Figure 1 – Fig. S1). The values of the concentration factor (As/As*) for As, also oscillate cyclically around the mean (Fig. S1). The EF values for Zn, Pb, Sn, Cd (Fig. 8) and Cu (Fig. S1) decline in the upper section of core SP6, whereas for Cr, P and Ni increase in the same section (Fig. 8).

3.2 Benthic foraminifera

Along the core SP6, 35 species/taxa of foraminifera with carbonated tests were identified (Appendix 3). Depth plots of foraminifera density (n.º specimens/10 ml), species richness (n.º species per 10 ml), and abundance of the main species/taxa (n.º specimens /10 ml), as well the *Elphidium*/*Ammonia* ratio are presented in Figure 9. These plots show that foraminifera density (FD: number of specimens/ 10 ml; <408 specimens/ 10 ml) is quite low and decrease significantly in the upper core section. The same trend is followed by species richness (SR: number of species/ 10 ml), which is quite low <16 species/10 ml per sample. The highest FD was observed between 239-130 cm, along with mollusk shells and ostracod valves.

Ordering by decreasing abundance (n.º specimens/10 ml) the following species can be listed: *Cribroelphidium excavatum* (<275), *Elphidium gunteri* (<123), *Ammonia tepida* (<53), *Elphidium oceanense* (<41), *Ammonia parkinsoniana* (<35), *Buliminella elegantissima* (<27), *Quinqueloculina seminula* (<15), *Ammonia rolshauseni* (<6), *Cibicidoides lobatulus* (<6) and *Bolivina striatula* (<6). The abundance of these species and also other miliolids (e.g. *Miliolinella circularis, Miliolinella subrotunda* and poorly preserved miliolids) and bolivinids (e.g. *Bolivina compacta, Bolivina lowmani densipunctata, Bolivina striatula* and *Bolivina variabilis)* is low or declines in the upper core section (126-0 cm depth; Fig. 9).

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Tab. 1. Radiocarbon results. Legend: pMC - Percent Modern Carbon; * 95.4 % Probability, BetaCal3.21: HPD method: SHCAL13 Calibration; ** 2 sigma calibration. Analysed Material: (1) shell; (2) Organic matter.

Depth (cm)	Beta N.°	d13C (‰)	Conventional Radiocarbon Age	Calendar Calibrated Results	Calibrated Results Before Present
114-112 cm (1)	441339	-8.6	105.4 ± 0.3 pMC	1955 AD	60 yrs cal BP
135-130 cm (2)	519822	-23.9	2090 ± 30 BP	*146 cal BC - 24 cal AD	* 2095 - 1926 yrs cal BP (2011±30 yrs cal BP)
215-212 cm (2)	441340	-22.3	2350 ± 30 BP	** 415-385 cal BC (400 cal BC)	** 2365 to 2335 yrs cal BP (2350±30 yrs cal BP)

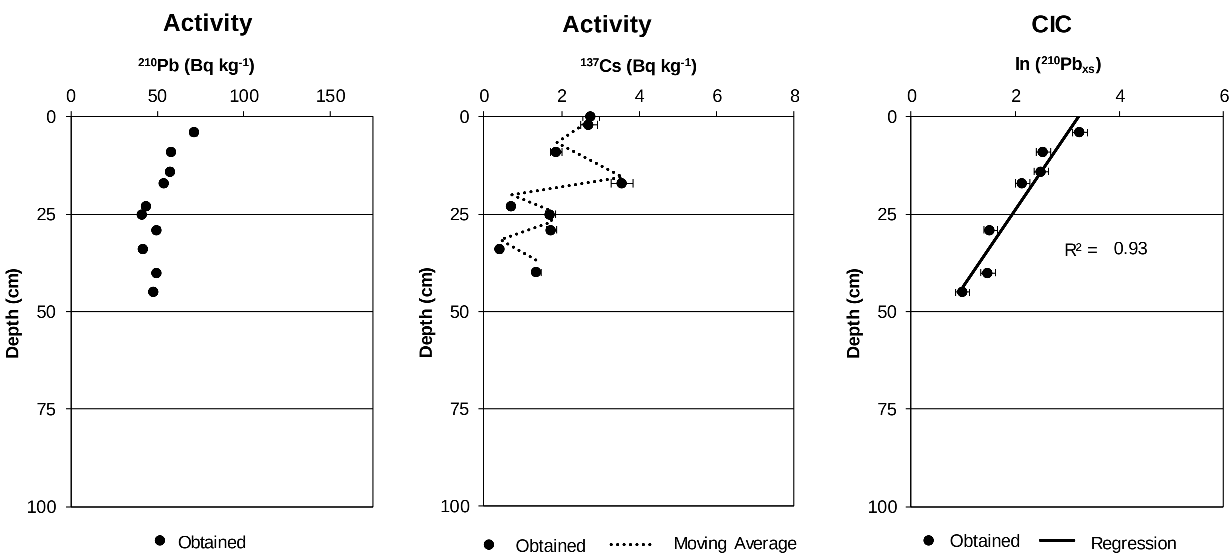

Fig. 2. Results of ^{210}Pb and ^{137}Cs radionuclides activity and the estimation of constant initial concentration (CIC).

Tab. 2. Results of ^{210}Pb (Bq kg^{-1}) and ^{137}Cs (Bq kg^{-1}) activities and the CIC values (ln ($^{210}Pb_{xs}$)) in the analysed sediment layers of the SP6 core.

	Activities				CIC	
Depth (cm)	^{210}Pb (Bq kg^{-1})		^{137}Cs (Bq kg^{-1})		ln ($^{210}Pb_{xs}$)	
	Value	Error	Value	Error	Value	Error
0			2.77	0.22		
2			2.72	0.22		
4	71.14	1.91			3.25	0.14
9	58.06	1.57	1.87	0.15	2.54	0.14
14	57.55	1.56			2.50	0.14
17	53.85	1.41	3.57	0.28	2.14	0.14
23	43.64	1.20	0.72	0.06		
25	41.10	1.13	1.72	0.14		
29	49.94	1.32	1.75	0.14	1.53	0.14
34	41.69	1.06	0.43	0.03		
40	49.74	1.35	1.37	0.11	1.48	0.14
45	48.02	1.26			0.99	0.14

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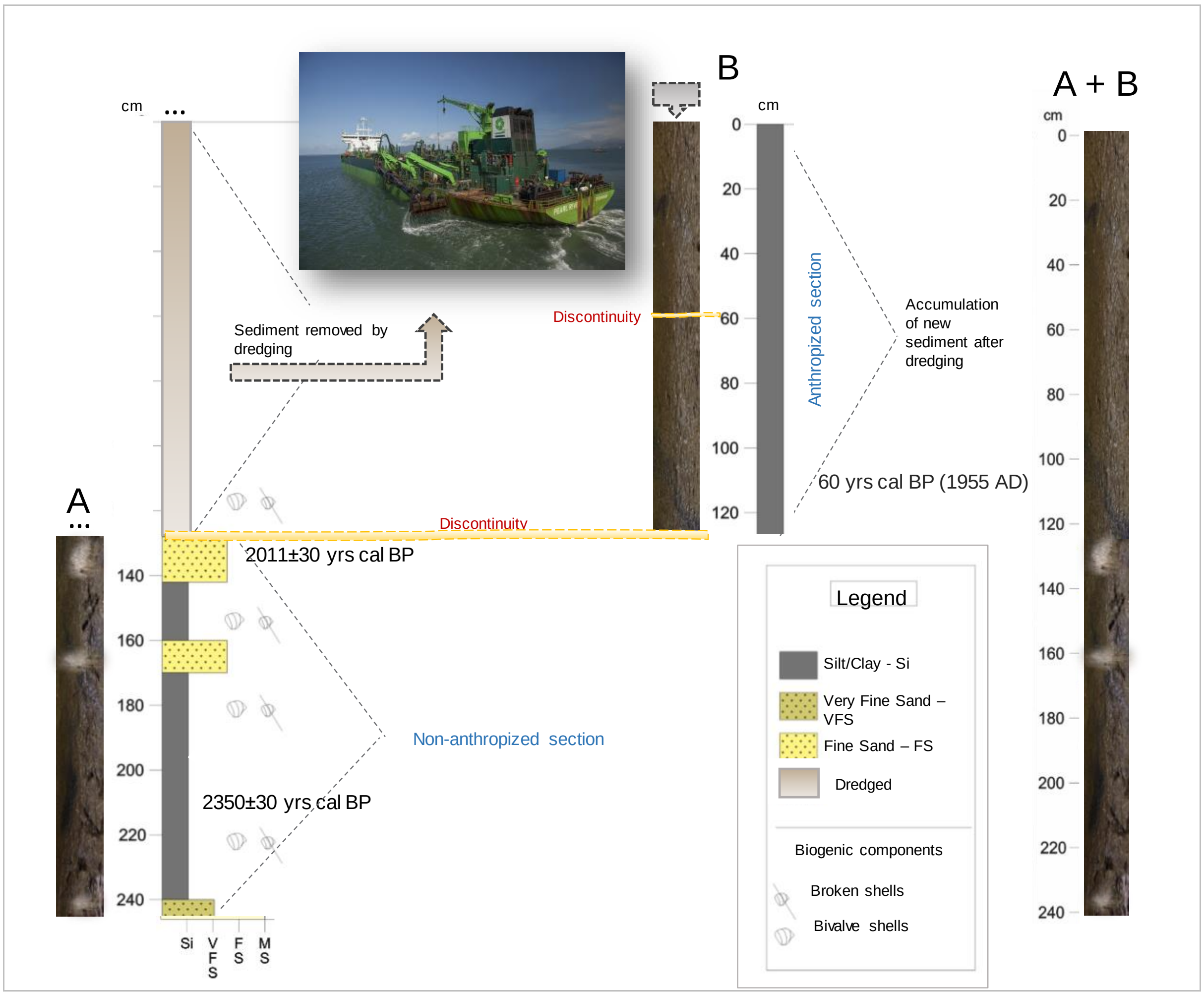

Fig. 3. Macroscopic characterization of core T4 (adapted from Pinto et al., 2017). Radiocarbon results are presented. A. Non-anthropized sedimentary section. B. Temporal gap due to dredged out (sediment removed). B. After dredging, new sediments were accumulated. Core T4 is composed by A + B sections. Below a particle size scale is presented.

The abundance of *Cribroelphidium excavatum, Buliminella elegantissima* and miliolids (mostly represented by *Q. seminula*) rise mainly in the lower core section (Fig. 9). The values of *Elphidium/Ammonia* ratio increase cyclically but more frequently in the lower half of the core along with small increases in the abundance of miliolides and bolivinids. FD and SR increased slightly in the upper 45 cm of the core SP6 (Fig. 9). The species percentage in the samples with density >100 specimens, presented in Figure 10 (and Appendix 3), shows that foraminiferal assemblages are largely dominated by *E. gunteri* in layers between 155-130 cm; below this section *C. excavatum* tends to be the dominant species and a greater number of species/taxa tend to be relatively frequent in the lower core section between 240-221 cm. The Shannon index values are low along the core SP6, but also tend to increase in the lower section (Fig. 10).

3.5 Statistical results

Figure 11 includes the biplot of Factor 1 against the Factor 2 of PCA based on selected biotic and abiotic data (Appendix 4). Factor 1 (46%) and the Factor 2 (14%) explain most part of data variability (60%). The Factor 1 of this PCA put in opposition: FD, SR, the abundance of *C. excavatum, E. gunteri, A. tepida, A. parkinsoniana* and *B. elegantissima* and the values of *Elphidium/Ammonia* ratio, as well as the CF of Cd, Co, Cr, Cu, Ni, Pb, Sn and Zn (I) and the CF of P and TOC contents (II). The Factor 2 of this PCA opposed FD, SR, the abundance of *C. excavatum, E. gunteri, A. tepida, A. parkinsoniana* and *B. elegantissima* and the values of *Elphidium/Ammonia* ratio (III) and the CF of Cd, Co, Pb, Sn and Zn (IV).

The correlations between TOC, Al and Sc and the analyzed chemical elements is presented in Table 4. TOC has significant negative correlations with Al, Cd, Co, Cr, Fe, Mg, Mn, La, Ni, Sc, Sn, Ti, V, Y and Zn. Aluminum and Sc also have negative correlations with these variables, except with Nb and Th. TOC has significant positive correlations with Ca, Nb, P, Th and Zr.

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Tab. 3. Range of the elemental concentrations (obtained by total digestion of the sediments and analysed by ICP-MS and ICP-ES) along the core SP6.

Elemental Concentrations		Maximum	Minimum	Mean
Al	%	12.8	7.8	10.1
As	mg kg^{-1}	14.0	8.0	11.0
Ca	%	0.4	0.2	0.3
Cd	mg kg^{-1}	5.9	1.3	2.9
Co	mg kg^{-1}	16.0	10.0	12.8
Cr	mg kg^{-1}	95.0	70.0	80.9
Cu	mg kg^{-1}	38.0	30.0	34.3
Fe	%	6.3	5.1	5.6
La	mg kg^{-1}	46.0	9.0	32.3
Mg	%	1.1	0.8	0.9
Mn	mg kg^{-1}	1006	446	578
Nb	mg kg^{-1}	30.0	24.0	27.4
Ni	mg kg^{-1}	35.0	26.0	30.1
P	%	0.2	0.1	0.1
Pb	mg kg^{-1}	90.0	33.0	52.9
Sc	mg kg^{-1}	18.0	11.0	14.1
Sn	mg kg^{-1}	22.0	8.0	12.8
Th	mg kg^{-1}	17.0	5.0	13.0
Ti	%	0.7	0.5	0.6
V	mg kg^{-1}	133	99	114
Y	mg kg^{-1}	24.0	8.0	18.6
Zn	mg kg^{-1}	1307	308	646
Zr	mg kg^{-1}	96.0	48.0	65.1

4. Discussion

4.1 Sedimentary record loss in core SP6

The sediment accumulation rate determined by C^{137} (0.35 cm yr^{-1}) is relatively higher than that determined by Pb^{210} (0.26 cm yr^{-1}), for the last hundred years, since the upper core sediments are less compacted. The sediment accumulation rate estimated in core SP6 is lower than that determined, also using ^{210}Pb geochronology, by Patchineelam et al. (2011) in Marambaia cove (0.47 cm yr^{-1}) and Borges and Nittrouer (2015, 2016), ranging from 0.37 cm yr^{-1} to 2.0 cm yr^{-1} for the last hundred years in 10 sediment cores collected in Sepetiba Bay. These results indicate large variation of sediment accumulation rates in Sepetiba Bay, probably due to hydrodynamical features and resedimenting after thunderstorms mainly in shallower waters or anthropogenic-driven changes, as also observed by Bueno et al. (2019) in several Brazilian coastal systems. The construction of channels in the drainage connecting Sepetiba Bay to the Paraíba do Sul river caused an increase of fine-grained sediment discharge (Lacerda et al., 1987; Barcellos and Lacerda, 1994; Barcellos et al., 1997; Molisani et al., 2004, 2006; Patchineelam et al., 2011; Montezuma, 2012; Borges and Nittrouer, 2015, 2016).

Tab. 4. Correlations between TOC, Al and Sc and the analyzed chemical elements. Marked in red (for negative) and blue (for positive) correlations are significant at p <0.05.

Variable	TOC	Al	Sc
TOC	…..	-0.65	-0.57
Al	-0.65	…...	0.97
As	0.00	-0.11	-0.14
Ca	0.35	-0.27	-0.25
Cd	-0.63	0.69	0.59
Co	-0.78	0.87	0.82
Cr	-0.29	0.38	0.33
Cu	-0.16	0.51	0.53
Fe	-0.51	0.87	0.85
Mg	-0.60	0.36	0.35
Mn	-0.65	0.71	0.64
La	-0.31	0.74	0.76
Nb	0.43	0.03	0.11
Ni	-0.72	0.90	0.84
P	0.89	-0.74	-0.65
Pb	-0.71	0.75	0.64
Sc	-0.57	0.97	…..
Sn	-0.65	0.72	0.61
Th	0.53	-0.25	-0.17
Ti	-0.71	0.88	0.84
V	-0.62	0.84	0.77
Y	-0.42	0.85	0.87
Zn	-0.71	0.82	0.72
Zr	0.69	-0.78	-0.71

The particle size and mineralogical composition of the core SP6 do not indicate disturbances in the sedimentary column, but the radiocarbon ages (in the layers 112-114 cm and 135-130 cm; Table 1) suggest sedimentary record loss. This assumption is also supported the sharp increase of TOC, P and Zr contents and the marked reduction of Zn, Ni, Co and Cd in the upper core section (≈126-0 cm).

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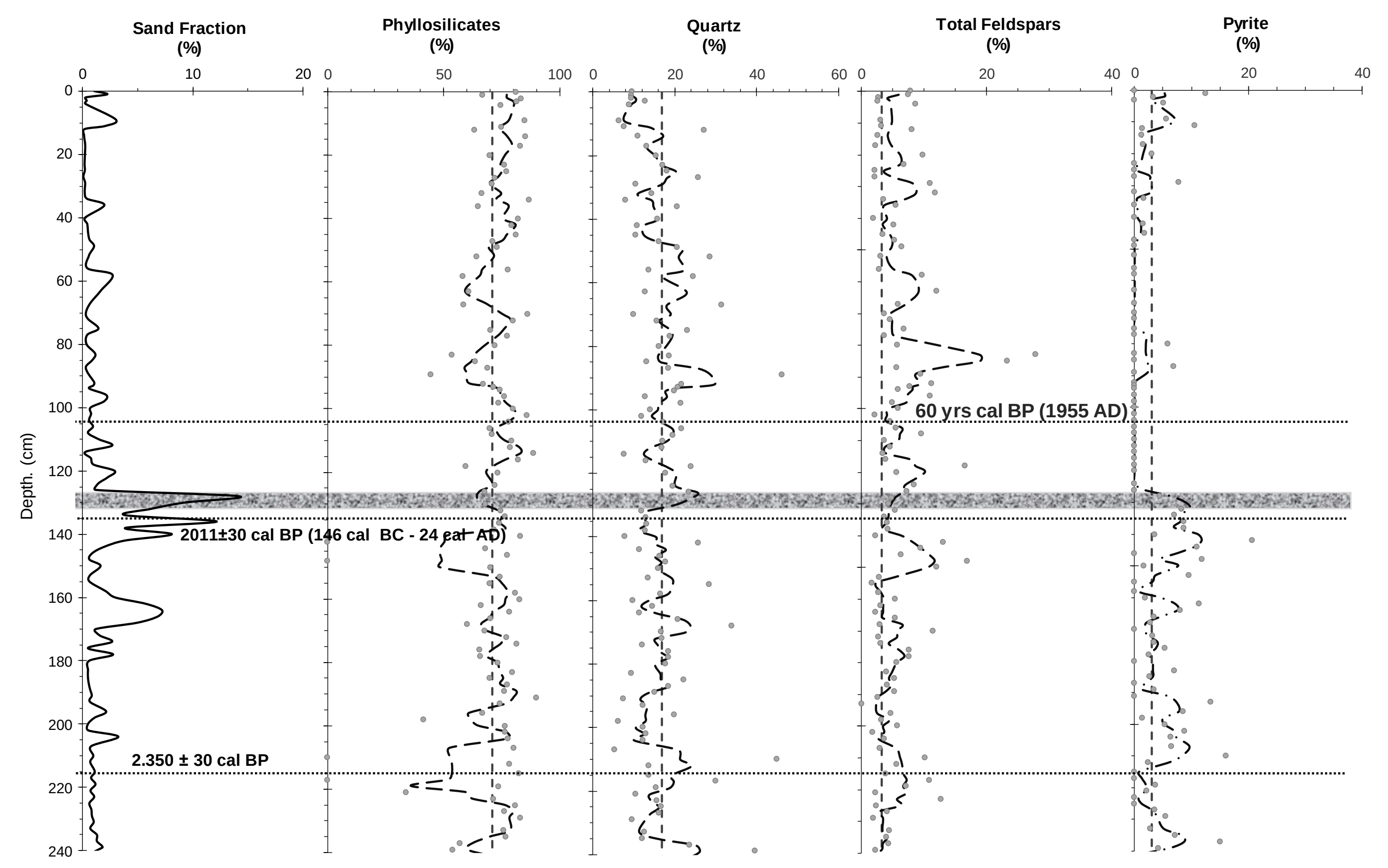

Fig. 4. Depth plots of sand fraction (<63 µm), phyllosilicates, quartz, total feldspars (k-feldspars + plagioclase) and pyrite. A temporal gap around the layer of 126 cm is gray marked. Radiocarbon data results are presented.

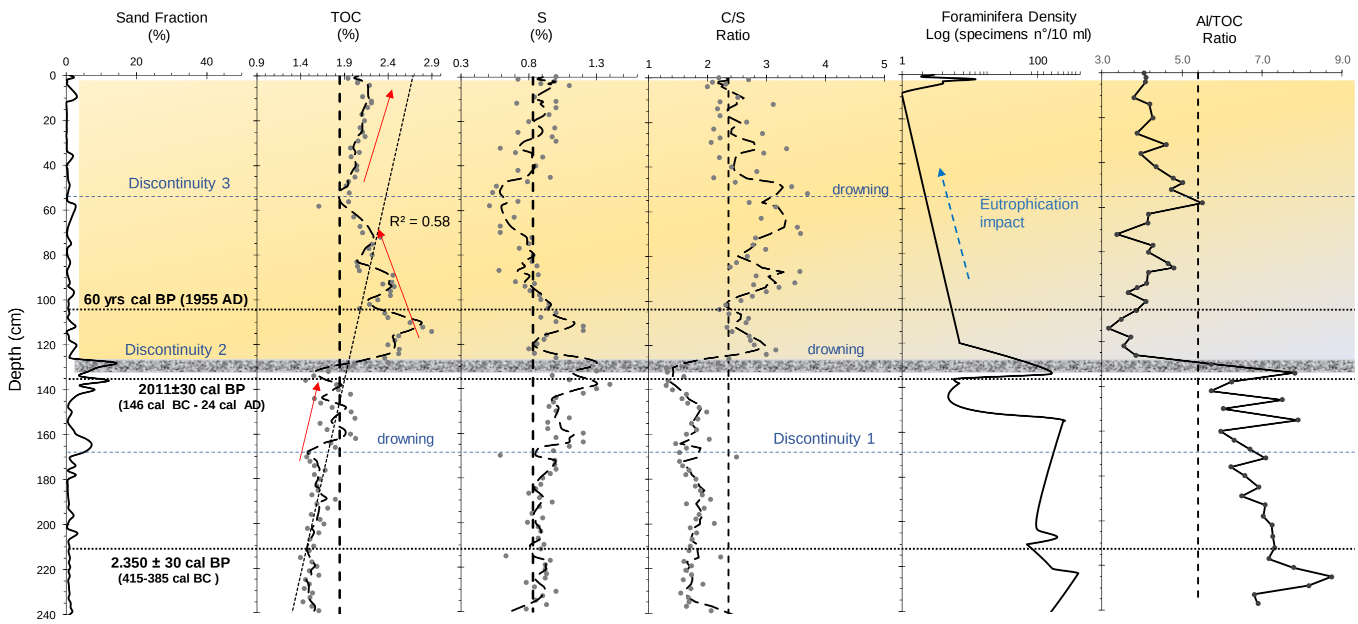

Fig. 5. Depth plots of the percentage (%) of sand fraction (<63 µm), total organic carbon (TOC) and total sulfur (S). C/S ratio and foraminifera density (logarithmic values) are also presented. The trend line and respective R^2 for TOC values and radiocarbon data are also shown, as well as indications from the interpretation of these variables. A temporal gap around the layer of 126 cm is gray marked. The upper section of the core is yellow signed.

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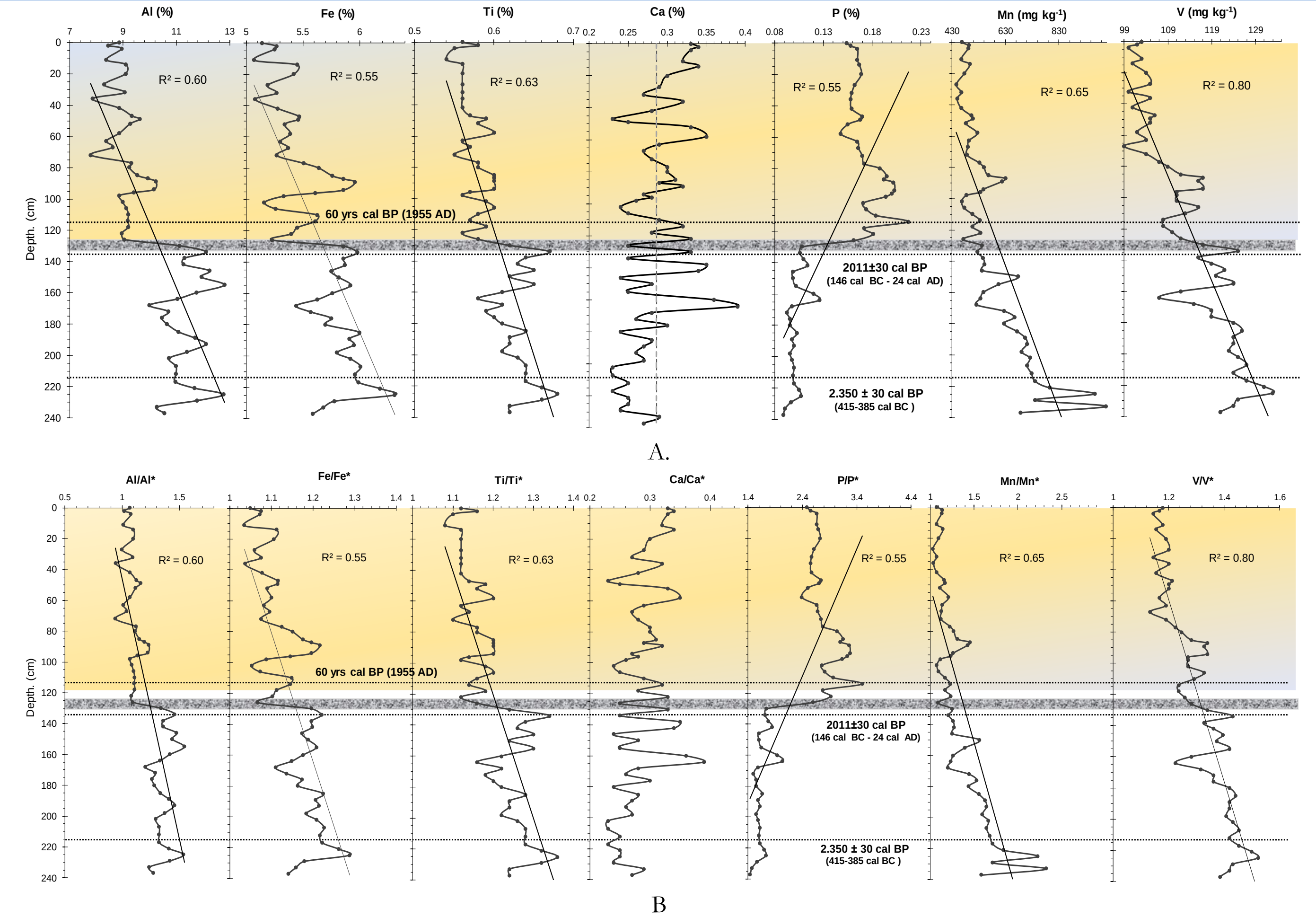

Fig. 6. Depth plots of: A. Al (%), Fe (%), Ti (%), Ca (%), P (%), Mn (mg kg^{-1}) and V (mg kg^{-1}). B. Concentration factor (CF), or ratios of each element with the respective baseline concentration (*) proposed by Pinto et al. (2019). The results of radiocarbon data are also presented. A temporal gap around the layer of 126 cm is gray marked. The upper section of the core is yellow signed.

The reduction of Zn, Ni, Co and Cd in this section (≈126-0 cm), is the opposite of what we were expecting. Several previous works revealed the occurrence of sharp increase of metals in the last century caused by the exponential population growth and industrial activity around Sepetiba Bay (Silva-Filho et al., 1996; Wasserman et al., 2001; Gomes et al., 2009; Patchineelam et al., 2011; Borges and Nittrouer, 2016; Araújo et al., 2017).

Industries established in 1966, produced Zn ingots, until the end of the 1990's. During this time, this industrial activity released significant quantities of Zn and Cd (Patchineelam et al., 2011). Copper also reaches the bay through diffuse sources (Molisani et al., 2004; Lacerda and Molisani, 2006). Enrichment of other metals such as Cd, Ni and Pb also have been observed (e.g. Silva-Filho et al., 1996; Wasserman et al., 2001; Patchineelam et al., 2011; Díaz Morales et al., 2019). However, this kind of record was not found in core SP6. Instead, the reduction of Zn, Cr, Pb, Ni, Sn, Co and Cd concentrations was documented since the seventies (section 126-0 cm; Fig. 6), as mentioned. So, we hypothesized that: i) sediments were removed from the study area; ii) three discontinuities are observed (numbered 1-3); ii) the largest discontinuity (Discontinuity 2; Figs. 3, 5) and sediment loss was recorded in core SP6 at around 126 cm; on the surface discontinuity (with over ≈2000 years), were deposited new sediments (accumulated between 126-0 cm), as suggested by the radiocarbon age obtained at the layers 112-114 cm and 135-130 cm; Table 1); iii) the two other discontinuities (Figs. 3, 5), which have lower expression can be identified in TOC depth plot and other geochemical variables: the first located around 175 cm (Discontinuity 1) and the latest around 155 cm (Discontinuity 3). So, the Discontinuity 1 should represent a transgressive process (Pinto et al. 2017). There is no evidence of section loss, only drowning, which favored the increased deposition of organic matter (TOC rise). Discontinuity 2 is an unconformity because there is erosion caused by dredging. The contact is abrupt. The increased TOC values may represent deepening and higher preservation rate of organic matter. Discontinuity 3 may address a similar process to 2, that is dredging, deepening and preservation of organic matter in the bottom.

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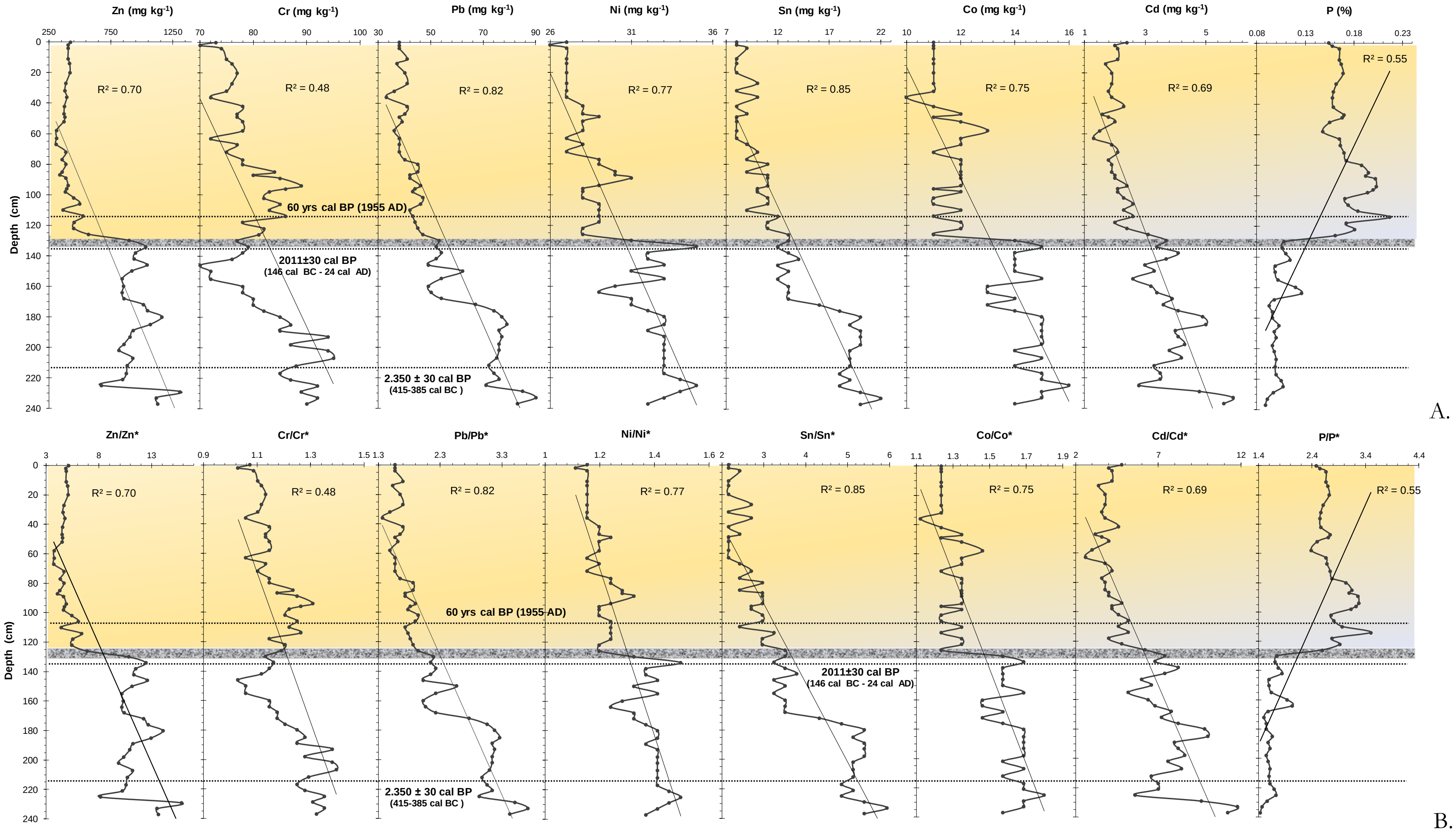

Fig. 7. Depth plots of concentrations of: A. Zn (mg kg-1), Cr (mg kg-1), Pb (mg kg-1), Ni (mg kg-1), Sn (mg kg-1), Co (mg kg-1), Cd (mg kg-1) and P (%). B. Concentration factor (CF), or ratios of each element with the respective baseline concentration (*) proposed by Pinto et al. (2019). The results of radiocarbon data are also presented. A temporal gap around the layer of 126 cm is gray marked. The upper section of the core is yellow signed.

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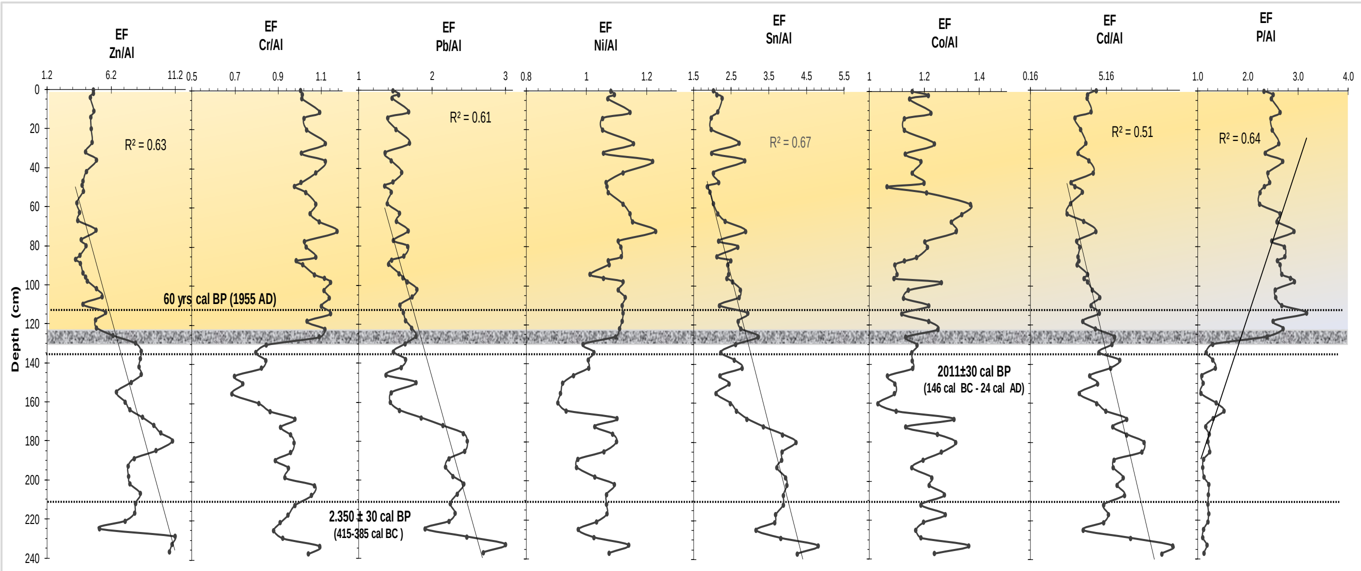

Fig. 8. Depth plots of the values of enrichment factors (EF) for Zn, Cr, Pb, Ni, Sn, Co, Cd and P. The results of radiocarbon data are also presented as well as the trend line and respective R^2 for the EF of Zn, Pb, Sn, Cd and P. A temporal gap around the layer of 126 cm is gray marked. The upper section of the core is yellow signed.

Between 1935 and 1941, engineering works were carried out in practically all the river sections of Sepetiba lowlands, including the Guandú, river basin (SEMADS, 2001). Construction of 270 kilometers of canals, 620 kilometers of ditches and 50 kilometers of dikes were completed in that period. In environmental terms, these works have eliminated or drastically reduced the floods.

We assume that the discontinuity 2 observed in core SP6, at 126 cm depth, may eventually be related to works carried out during this period, regarding to settlement of the Guandú River flow. These activities removed materials accumulated during ≈2000 years (sedimentary record lost), on the discontinuity surface were accumulated new sediments. This discontinuity had a higher impact on benthic foraminifera as analyzed in the next sections.

In 1973, the government of Guanabara State promoted feasibility studies for the implementation of a maritime terminal in the Santa Cruz region, intended primarily to serve the industrial complex that would be deployed in that area. With the merger of the states of Guanabara and Rio de Janeiro, the implementation of the port was the responsibility of Companhia Docas do Rio de Janeiro - CDRJ. The construction of the pier began in 1976, followed in 1977 by dredging, rockfill and landfill. The port was opened on May 7, 1982, with the start-up of the Coal Terminal, leased since July 10, 1997, by Companhia Siderúrgica Nacional – CSN (Fig. 1).

The sedimentary discontinuity 3, observed in several variables such as TOC, P and Co in around 60 cm depth of core SP6, was possibly caused by these works and had some influence on the recent sedimentation of this area.

4.2 Characteristics of the sediments deposited before and after the sedimentary discontinuity 2

The new sediments accumulated between 126-0 cm, in core SP6, are fine-grained and have general mineralogical characteristics similar to those previously deposited (between 240-126 cm; Fig. 3). However, the distribution of, for instance, Al, Fe and Ti (Fig. 6) as well as Zr and La, suggests slight mineralogical differences between these sections probably related to changes in the sedimentary dynamics and mechanical and chemical weathering, as well. These changes should be related to the anthropogenic interventions in the Sepetiba hydrographical basin (Smoak and Patchineelam, 1999; Molisani et al., 2004, 2006; Marques et al., 2006) and also climatic conditions influencing river discharge. TOC has significant negative correlations with Al, Fe, Mg, Mn, La, Nb, Sc, Ti, V and Y (Table 4). The main sources of these elements should be outside the basin. They are introduced in the basin through mineralogical components of the sediments transported mostly by the rivers. This significant negative correlation should indicate that this mineralogical component decreased, relative to the accumulation rate of organic material. This assumption is also supported by the values presented in the depth plot of Al/TOC (Fig. 5). Aluminum concentrations are mainly associated with phyllosilicates (Martins et al., 2015b, 2017), which are the main mineralogical component of the sediments of core SP6. This core was collected in a marginal area of the Guandú River delta front, probably in a protected zone, out of the high continental sediment load. On the other hand, as it is located in a shallow area, sediments can be disturbed by the local hydrodynamics during violent storm events.

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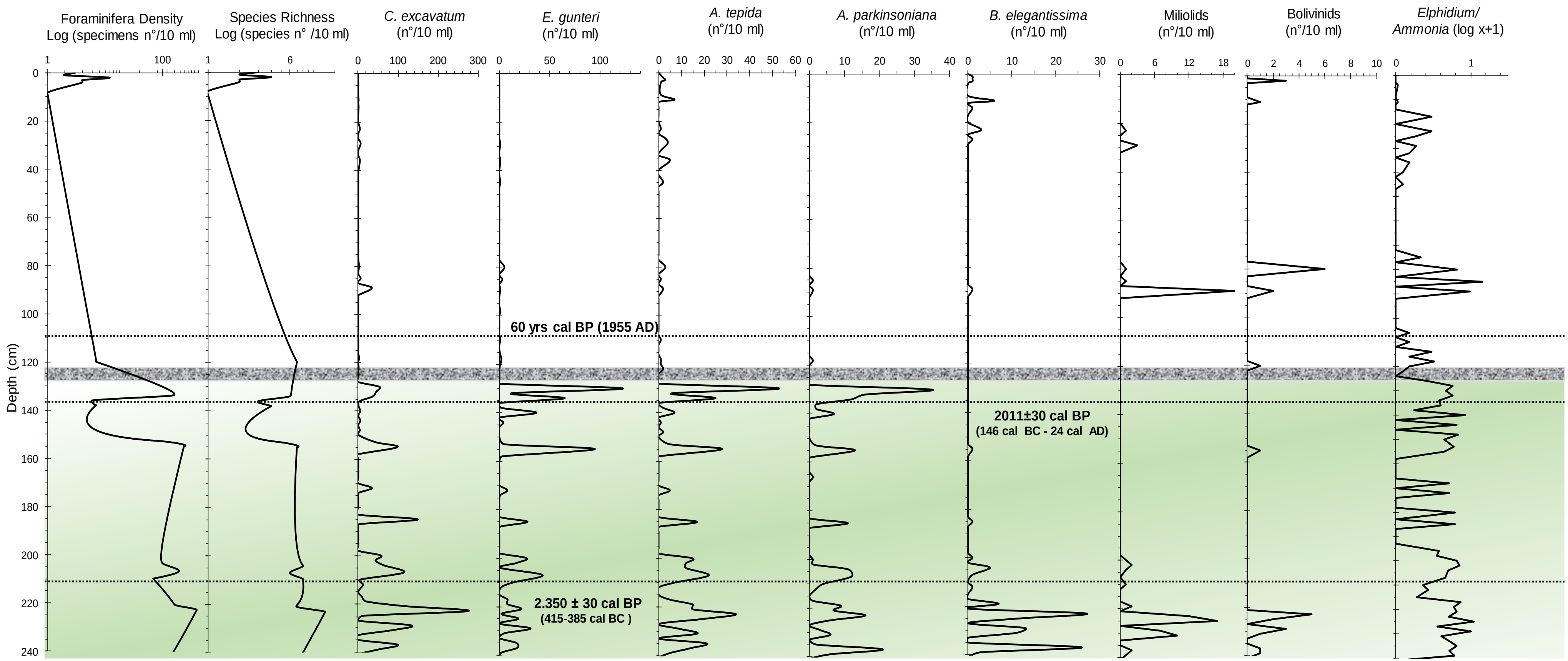

Fig. 9. Depth plots of foraminifera density (n.° specimens per 10 ml), Species richness (n.° species per 10 ml), and abundance of the main species/taxa (n.° specimens per 10 ml). The results of log (x+1) of *Elphidium*/*Ammonia* values and radiocarbon data are also presented. A temporal gap around the layer of 126 cm is gray marked.

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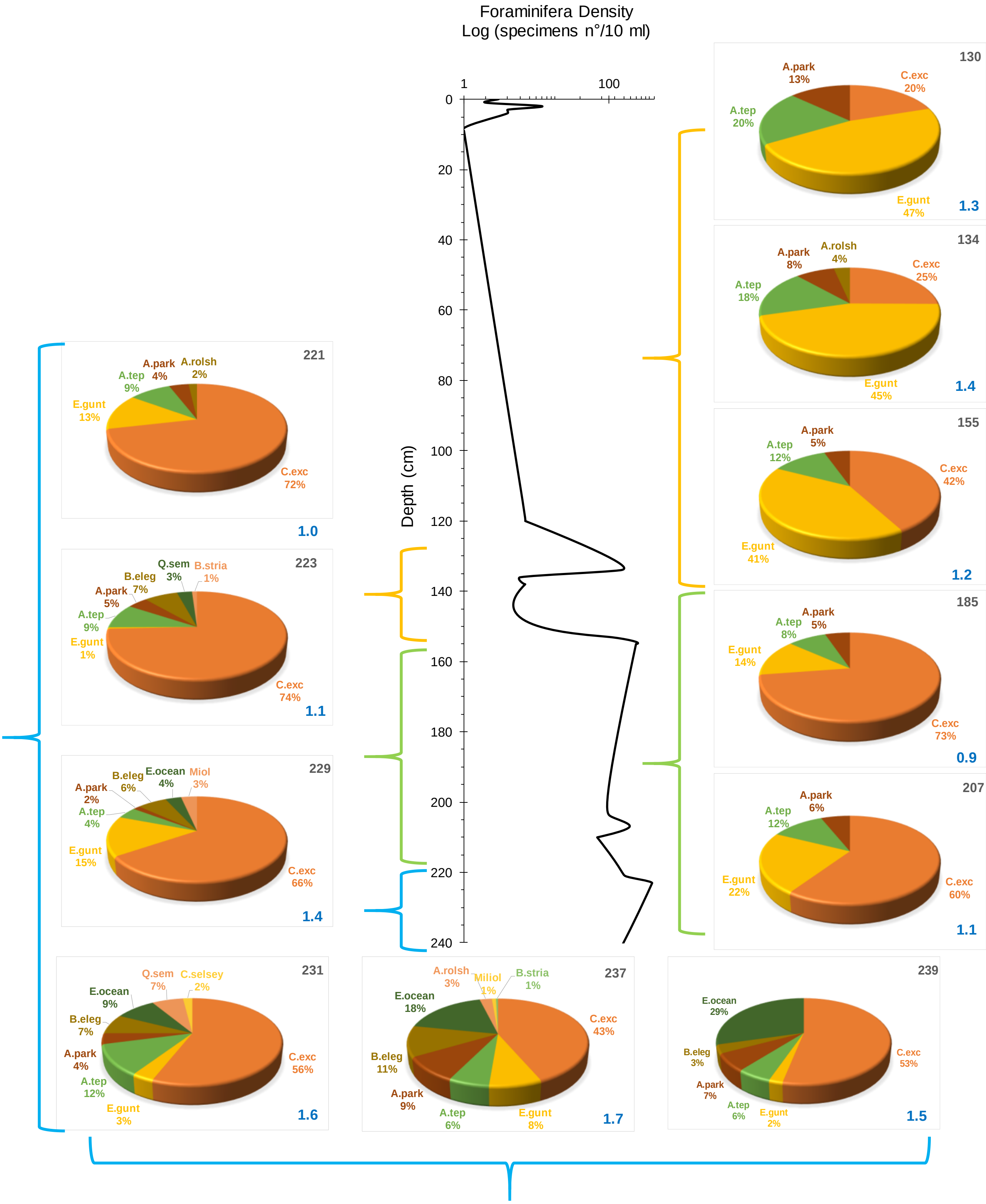

Fig. 10. Percentage of the main species/taxa in selected layers (depth in black at the top right corner) with relatively high foraminifera density (n ° specimens per 10 ml) along the core SP6. The blue values in the lower left corner represent the Shannon index

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The new sediments accumulated between 126-0 cm have relatively low PTE concentrations, such as Zn, Cr, Pb, Ni, Sn, Co and Cd. The highest concentrations of these chemical elements were recorded between 240-114 cm. The significant positive correlations (Table 4) of Al and Sc (tracing mineralogical contributions) with Cd, Co, Cr, Cu, Ni, Pb, Sn and Zn (PTEs), as well as with Fe, Mg, Mn, La, Li, Th, Ti, V, Y and Zr (lithogenic elements), indicate that the relatively high PTE concentrations in the lower section of core SP6 are related to lithogenic sources. So, the new sediments supplied to the study area, are not polluted by metals as also indicated the CF and EF values of PTEs (Fig. 7B and 8) but are impacted by organic matter (Fig. 5).

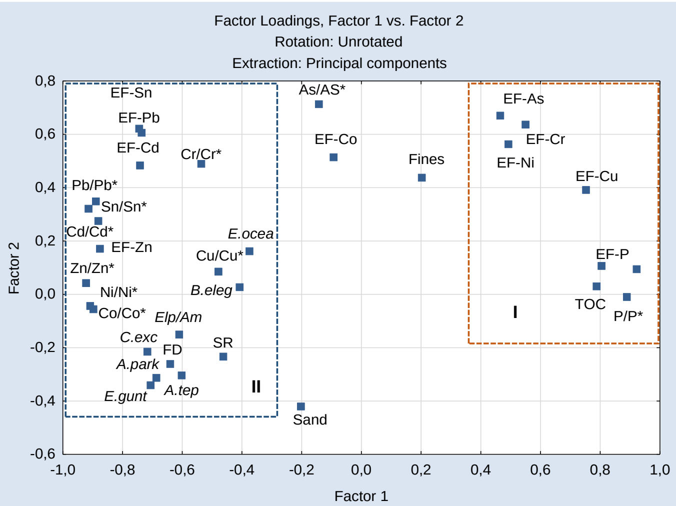

Fig. 11. Biplot of Factor 1 against the Factor 2 of PCA based on selected biotic data and abiotic data (such as TOC, C/S, sand and fine fractions, CF (or ratios of each element with the respective baseline concentration (*)) for As, Cd, Co, Cr, Cu, Ni, Sn, P, Pb and Zn. Legend: Sand – sand fraction (%; <63 μm); Fines – fine fraction; FD – foraminifera density (n.º specimens/ 10 ml); SR – species richness (n.º species/ 10 ml); *C.exc.* - *Cribroelphidium excavatum* (n.º specimens/ 10 ml); *E.gunt* - *Elphidium gunteri* (n.º specimens/ 10 ml)*;* A.tep - *Ammonia tepida* (n.º specimens/ 10 ml)*; E.ocea* - *Elphidium oceanense* (n.º specimens/ 10 ml)*; A.park* - *Ammonia parkinsoniana* (n.º specimens/ 10 ml)*; B.eleg* - *Buliminella elegantissima* (n.º specimens/ 10 ml)*; Elp/Am* – *Elphidium/Ammonia ratio.*

These results can be explained by two possibilities: core SP6 is located in a protected area, out of the direct influence of metals pollution (e.g. Lacerda et al., 2004; Patchineelam et al., 2011; Díaz Morales et al., 2019) or; if the new sediments are being introduced in Sepetiba Bay by the Guandú River, we can deduce that this river is not introducing significant PTE-polluted sediment into this basin when compared to that supplied from the Ingá area. Guandú River is the most important supplier of the Sepetiba Bay watershed and is responsible for supplying water to several cities, being the main source of Rio de Janeiro city (Cunha et al., 2016). However, the catchment area is located upstream of the industrialized and most populated zone. Guandú River large watershed, crosses highly vegetated areas (SEMADS, 2001) and introduces large amount of sediment and organic matter into Sepetiba Bay (Smoak and Patchineelam, 1999; Molisani et al., 2004, 2006; Marques et al., 2006). Circulation within Sepetiba Bay and weak hydrodynamic conditions favor the sediment accumulation in its inner region, facilitating the accumulation of fines (Borges and Nittrouer, 2016) and contributing for its delta formation (SEMADS, 2001).

The supply, accumulation and preservation of organic matter were high in the last ≈3,500 yrs cal BP, in the study area. But the TOC values increased significantly in the first 126 cm of the sedimentary column, as did the P contents. The very sharp increase in P contents may be associated with additional contributions of agricultural and urban effluents. Phosphorus should also be released from anoxic layers and retained in oxic sediments (Almroth-Rosell et al., 2015). Increasing P levels in the water column may have enhanced algal blooms within the bay which, once accumulated in the bottom, in addition to continental organic matter, were responsible for the large increase of TOC contents and for the oxygen drop in sedimentary pore-water. This reduction is signed by the C/S ratio values <3 (Stein, 1991; Borrego et al., 1998; Morse and Berner, 2000) in most layers of the core SP6 (Fig. 5).

The most pronounced oxygen scarcity, and the establishment of anoxia, at least in micro environments, may have occurred for example in the first 60 cm of the sedimentary column (new sediments) and in the section 240-126 cm depth, leading to the production of biogenic pyrite (Fig. 4), as also observed by Araújo et al. (2017). In the lower core section, the establishment of anoxic conditions should have not been contemporary of the sediment deposition. It should have been established at a later stage in subsurface sedimentary layers not producing high impact on benthic communities as suggest the large abundance of benthic foraminifera.

4.2 Influence of Eutrophication on Benthic Foraminifera

Foraminifera assemblages of core SP6 (Fig. 9, 10; appendix 3) are commonly found in living assemblages of Brazilian coastal shallow waters (e.g. Delavy et al., 2016; Martins et al., 2016 a, b; Raposo et al., 2016, 2018; Belart et al., 2017, 2018, 2019; Duleba et al., 2018). However, in the lower sedimentary layers (240-229 cm), in addition to a relatively high foraminiferal density and slight increase of diversity, the presence of *Ammonia parkinsoniana*, bolivinids and buliminids species (Figs. 9, 10 and Appendix 3) suggest the occurrence of more favorable environmental conditions even though associated to high organic matter flux and low oxic conditions (Martins et al., 2016 a, b; Belart et al., 2018).

In the next upper section (220-155 cm), species diversity is decreasing and increasing dominance, first of *C.*

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excavatum (up to 185 cm) and later of *E. gunteri* in addition to the rise of *Ammonia* spp. (up to 125 cm; Fig. 9). It is difficult to attribute an environmental cause to explain the exchange of *C. excavatum* dominance by *E. gunteri* since the ecological niche of these species is not yet well known.

However, as observed by Belart et al. (2018), in Saquarema lagoon (Saquarema Lagoon System, Brazil), *A. parkinsoniana*, *C. excavatum* and *E. gunteri* occur in areas with the highest marine influence and the last two species reach higher abundance under relatively low temperatures, TOC, protein and lipid values and relatively high salinities and carbohydrate contents. In core SP6, the abundance of *A. parkinsoniana* increases in the same layers where *C. excavatum* and *E. gunteri* are more abundant, mostly in the lower section, which probably reveals food preference for sources of carbohydrates.

Belart et al. (2018) also observed that *A. tepida* is related to confined areas and regions with higher temperature, TOC, protein and lipid values but to relatively low biopolymers content. These results are confirmed by works performed in other regions (Martins et al., 2016a, b; Duleba et al., 2018). They also agree with that obtained by Pregnolato et al. (2018) that used the *Ammonia-Elphidium* Index to assess the oxygenation levels in the Petrobras Polo Atalaia Production complex area (Sergipe, Brazil), since the genus *Ammonia* (but mainly *A. tepida*) has a greater resistance than the genus *Elphidium* to low oxic conditions, and both are abundant in the coastal zones, which makes possible the use of this index to access the impact caused by organic matter. So, this index can be seen as a confinement proxy.

Based on this assumption, the *Elphidium-Ammonia* Index (based on *Cribroelphidium excavatum* plus *Elphidium gunteri* versus *A. tepida*) was used as a proxy to access higher/lower marine influence in this work. The *Elphidium-Ammonia* Index along the core SP6 (Fig. 9), suggest, higher marine influence in the lower core section and larger environmental instability in the upper core section.

The higher CF and EF values of Cd, Co, Cr, Ni, Pb, Sn and Zn (Figs. 7, 8), in the period between ≈2400-2000 yrs BP (section 240- 126 cm of not anthropized sediment; absence of catchment system and regulation of rivers flow and industrial activity), should have had natural causes. These results suggest a landscape in which stormy events induced large sediment transport from continental source areas. These metals were introduced in the bay through the mineralogical component transported mostly by rivers. The increased EF of Cd, Co, Cr, Ni, Pb, Sn and Zn (Fig. 8) seems to be mainly associated with the clayey component and especially to the presence of clay minerals in the area (Fig. 6A: highest Al contents in this section; Fig. 4: relatively high phyllosilicate contents). Several authors observed the presence of clay minerals with high metal fixation capacity in Ribeira Bay (SE of the study area) (Bidone and Silva Filho, 1988; Corrêa et al., 1996). The highest concentration of metals in the lower core section was associated with immobile sedimentary phases. Sulfide formation should have also trapped metals in relatively immobile sedimentary phases (Burdige, 1993; Martins et al., 2015a).

In the PCA results (Fig. 11), the Factor 2, put in opposition FD, SR, the abundance of *C. excavatum, E. gunteri, A. tepida, A. parkinsoniana* and *B. elegantissima* and the values of *Elphidium*/*Ammonia* ratio (III) and the CF of Cd, Co, Pb, Sn and Zn (IV). These results indicate that, to some extent, the higher concentrations of metals in the lower section of core SP6 may have had a negative effect on benthic meiofaunal organisms. Metal adsorption to clay minerals is one of the most mobile sedimentary phases, with the possibility of damaging meiofaunal organisms (Martins et al., 2015a). Organic matter is another possible sedimentary phase capable of concentrating metals (Martins et al. 2015a) whose flow and accumulation have been high in the study area, in the last 2400 yrs. BP.

Foraminifera density was quite reduced in the upper core section between 126-0 cm. Even in the lower core section, the large variation of foraminifera abundance indicates great environmental instability, probably caused by sediment disturbance during storm events when the wind and ocean circulation is more active.

Factor 1 of PCA suggests that the main cause (considering the analyzed variables) for the reducing abundance of foraminifera should have been the high sedimentary organic matter contents (Figs. 5, 11), which were probably induced not only by natural causes but also by anthropogenic influences. The new sediment deposited after dredging seems to be not polluted by metals. However, the study area, is an environment of high environmental stress caused by silting and increasing eutrophication, as indicated by benthic foraminifera record.

5. Conclusion

The benthic foraminifera abundance and assemblages' composition found along the core SP6 indicate that the environment was a quite instable shallow marine setting in the last ≈3,500 yrs cal BP. The natural evolution of the area was probably interrupted by dredging. The new sediment deposited after dredging was not polluted by metals but enriched in organic matter. As the main sediment supplier for this area should be the Guandú River, the obtained data in core SP6, suggest that this river is not being an important source of metal pollution for Sepetiba Bay. However, the study area, is an environment of high environmental stress caused by silting and increasing eutrophication, as indicated by benthic foraminifera record.

Foraminiferal repopulation after dredging, seems not to have been successful. The cause of this failure should have been associated with the increasing degree of silting and eutrophication in the study area.

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Appendices 1-4 are attached as supplementary material (SM1-SM4) in:
http://www.e-publicacoes.uerj.br/index.php/jse/article/view/47327

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Supplementary Figure

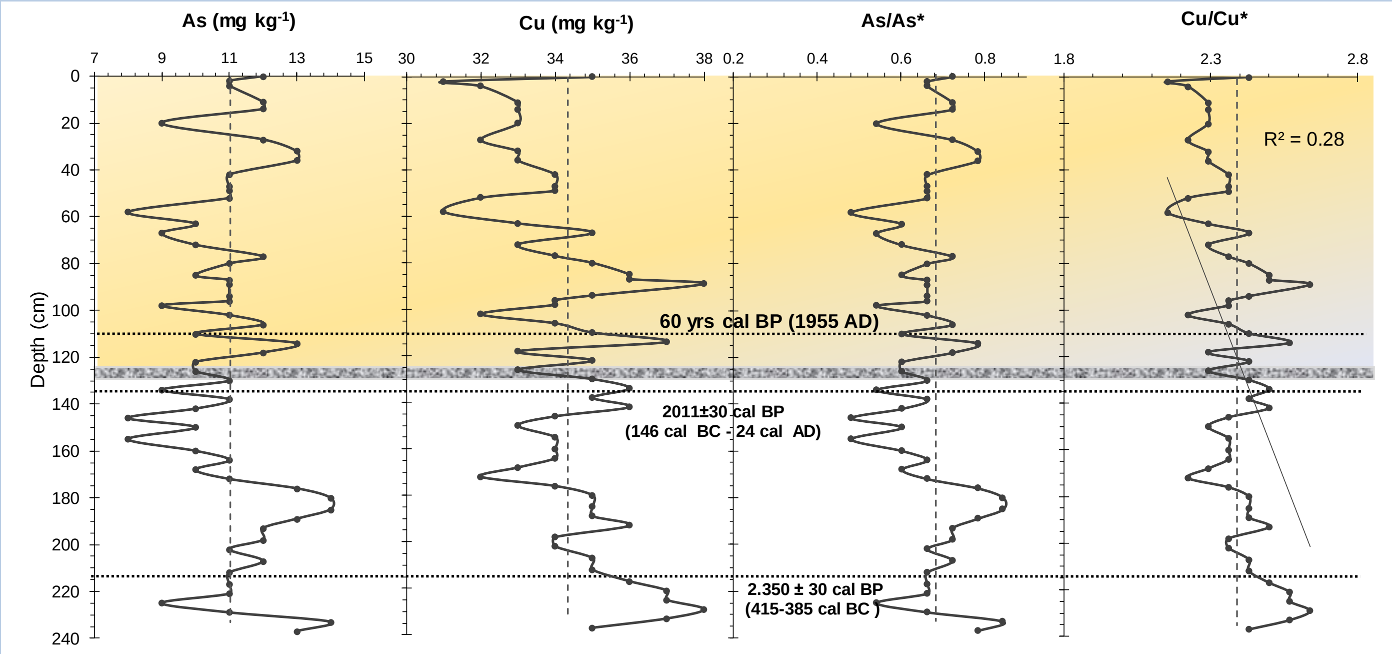

Fig. S1. Depth plots of As and Cu (mg kg^{-1}) contents and the respective CF values (As/As*, Cu/Cu*). The results of radiocarbon data are also presented. A temporal gap around the layer of 126 cm is gray marked. The trend line and respective R^2 was presented for Cu/Cu*. The upper section of the core is yellow signed and the mean values are marked with a gray dashed line.