

## HIGH-RESOLUTION ACOUSTIC MAPPING OF GAS CHARGED SEDIMENTS AND LIVING BENTHIC FORAMINIFERA ASSEMBLAGES FROM THE NE REGION OF THE GUANABARA BAY (RJ, BRAZIL)

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#### Abstract

This work was performed in the NE region of the Guanabara Bay, a highly impacted Brazilian coastal system, located in Rio de Janeiro State. It aimed to: i) identify and map the areas with occurrence of gas in the sediment, as well as its acoustic signature; ii) characterize the physical properties of the sediments and; iii) document the response of microbenthic organisms (living benthic foraminifera) to changes in quantity and quality of organic matter. Seismic surveys at the frequency of 12 kHz identified a large area with about 50% gas charged sediments in the study area.

The main acoustic signatures of the shallow gas were black shadow and gas blanket. In addition, features related to gas seepages to the water column (acoustic plumes and pockmarks) and gas percolation within the sediments (intraCitation:

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sedimentary plumes, turbidity pinnacles) were also identified. The gas has a biogenic origin and results from the high acumulation rate between 0.03 to 0.9 cm.year<sup>-1</sup> and from the decomposition of large amount of organic matter (10-20%). Vertical distribution of gas ranges from few centimeters to 9 m below the water-sediments interface. These occurrences are related to both gas migration from lower sedimentary layers to Holocene muds above, and to recent generation in near-surface sediments as the area display favorable conditions for gas production. Cores ranging from 150-240 cm in length have predominantly muddy sediments and variations in the P-wave velocity followed the changes in sediment density, controlled mainly by the presence of gas in sediments, bioclasts accumulation, textural variation and percentage of organic matter.

The TOC content and Rock-Eval pyrolysis parameters evaluated in nine surface sediment samples indicate that good to excellent amount of organic matter associated with moderate to good source potential for gas production is present in the study area. In these areas living benthic foraminifera are of reduced diversity and density. The assemblages are largely dominated by *Ammonia tepida*. Statistical results evidence that areas of intense gas release

#### 1. Introduction

Gas bearing sediments are a globally phenomenon observed from coastal areas to the deep ocean but are quite common in shallow environments with a depth of less than 50 m (Judd, 2004, Vardar and Alpar, 2016). The shallow gas accumulation in the first kilometer of sediment may have biogenic or thermogenic origin (Iglesias and García-Gil, 2007; Wessels, et al., 2010).

Thermogenic hydrocarbon gasses are found in the sediment at depths where organic matter encounters high temperatures and pressure, i.e., several hundreds of meters to kilometers below the Earth's surface (Schoell, 1988) and are produced by high temperature degradation and cracking of organic compounds at considerable burial depths (Judd et al., 1997; Missiaen et al., 2002). On the other hand, the biogenic gas is formed at low temperatures by microbial methanogenesis that is the degradation of organisms such as planktonic matters, plants, fishes, and other organic material under anaerobic conditions (Kitidis et al., 2007).

The biogenic gas is isotopically depleted ( $\delta^{13}$ C <-60 ‰), and has extremely high methane content (Santos Neto, 2004). Methane (CH<sub>4</sub>) is the most common and abundant hydrocarbon gas in near-surface sediments (Judd et al., 2002). Most biogenic methane is produced by the anaerobic degradation of abundant organic matter and deposited in marine subsurface where the accumulation rate is high (Hovland and Judd, 1988).

Sediments with large quantities of organic matter, particularly with fine granulometry, are more prone to methane generation (Judd, 2004). Consequently, migration of gas or methane-rich interstitial water tends to occur by permeable migration paths (Yun et al., 1999).

Most of the shallow gas-bearing sediments, as well as gas escape features, are observed in bays, deltaic accumulations and in basins (Ivanov et al., 1998; Okyar and Ediger, 1999; Yun et al., 1999).

The gassy sediments can be easily identified by highresolution reflection seismic data (Vardar and Alpar, 2016). Depending on their seismic character, size and geometry,



affect the benthic faunas since the benthic foraminiferal assemblages are reduced in diversity and density.

Keywords: Shallow gas. Physical properties. TOC. Rock-Eval pyrolysis. Stable isotopes. Living benthic foraminifera.

various descriptive features have been used for gas charged sediments, such as acoustic blanket, black shadow, plumes, curtain, and acoustic turbidity (Taylor, 1992; Yun et al., 1999; Garcia-Gil et al., 2002; Baltzer et al., 2005).

The study of gas occurrences in sediment is important because in some cases the presence of gaseous hydrocarbons is a shallow signal of deeper and large accumulations in border basins (Okyar and Ediger, 1999). Methane is a strong greenhouse gas and is believed to contribute to rapid global climate changes depending on its concentration levels in the atmosphere (Overpeck and Cole 2006). Its global warming potential is more than 20 times higher compared to  $CO_2$ (Hovland and Judd, 1988; Hovland et al., 1993; Lowe and Walker, 1997; Judd et al., 2002).

The gas seeping can negatively affect the environment (Hovland and Judd, 1988; Hovland et al., 1993; Lowe and Walker 1997; Judd et al., 2002). However, the consequences of the gas seeping on the benthic communities of coastal ecosystems are yet poorly understood. In coastal areas, the gas escape occurs in eutrophic systems. Studies of the problems associated with eutrophication, pollution, contamination and its consequences on biodiversity, productivity of the marine ecosystem and sustainable development, seek the understanding of the physical, chemical and biological processes through indicators to prevent and tackle serious environmental problems.

Shallow water benthic foraminifera can be successfully used as proxies of environmental variables or as bioindicators of environmental status (Frontalini and Coccioni, 2008; Romano et al., 2009; Armynot du Châtelet et al., 2011; Foster et al., 2012; Schönfeld et al., 2012). They have several advantages in comparison to macrofaunal organisms (Schönfeld et al., 2012) like higher abundance and diversity, and shorter life and reproductive cycle. This means that much smaller sediment volumes are needed to obtain a high number of specimens essential to characterize the assemblages with statistical reliability. Foraminifera have in general short life cycles as compared to macrofaunal organisms and respond rapidly to environmental changes.



They have a limited ability to move and their assemblages, therefore, reflect the conditions of the environment where they live.

#### 1.1 The main goals of this study

Drawing upon the environmental, strategic and economic importance of the Guanabara Bay, this study aims to identify and map the areas with occurrence of gas in sediment, as well as its acoustic signature and the characterization of the physical properties of sediments. This work also intends to analyze how the microbenthic organisms (living benthic foraminifera) are affected by the enrichment of organic matter and gas escape in the NE region of the Guanabara Bay. To achieve this goal an integrated assessment including indicators of organic material content and quality using Rock–Eval pyrolysis parameters is applied. The results presented in this research can help in decision-making on dredging, remediation or engineering works, where proper procedures must be taken to reduce operational risks and environmental impacts.

Several works have been performed to study benthic foraminiferal distribution and ecology in the Guanabara Bay (Vilela et al., 2001 a, b, 2003, 2004, 2007, 2011; Fontana et al., 2006; Kfouri-Cardoso et al., 2006; Pereira et al., 2006; Figueira et al., 2007; Donnici et al., 2012; Eichler et al., 2013; Clemente et al., 2015) as well as in the sedimentary record to reconstruct paleoenvironmental evolution (e.g., Vilela et al., 2007; Baptista Neto et al., 2016). However, all the previous works in the Guanabara Bay have been based on total (living plus dead) benthic foraminiferal assemblages. Schönfeld et al. (2012) established as mandatory recommendation the use of living foraminifera to study the ecology of these organisms.

This work is as much as we know, the first one that uses living assemblages of benthic foraminifera in comparison of TOC content, Rock–Eval pyrolysis parameters, carbon stable isotope and sedimentological data obtained in cores and geophysical results to evaluate the influence of organic matter quantity and quality and gas releasing in the NE sector of the Guanabara Bay.

### 1.2 Study area

The Guanabara Bay is an estuarine system located in one of the most important urban areas of Brazil and presents different levels of natural and anthropogenic influence (Monteiro et al., 2012). On its margins are located the city of Rio de Janeiro and its metropolitan area including Niterói, São Gonçalo, Magé, among other cities. Approximately 75% of the population living in this great urban concentration directly discharges domestic effluents into the bay without any treatment.

The Guanabara Bay receives high loads of organic matter and pollutants and experiences a growing process of eutrophication (Amador, 1992; Baptista-Neto et al., 2000; Mendonça Filho et al., 2003; Catanzaro et al., 2004; Pereira et al., 2006). The hydrodynamic conditions in the Guanabara Bay are described and discussed by several authors that correlated them with the distribution of sediments (Amador, 1992; Quaresma, 1997; Baptista-Neto et al., 2000; Quaresma et al., 2000; Catanzaro et al., 2004).

Circulation and salinity are subject to the movements of ocean currents, tides, winds and river discharges (Kjerfve et al., 1997). The region of the Guanabara Bay connection with the sea is a very dynamic area, with strong currents in response to its asymmetrical geomorphology, in which there is a narrowing of its downstream portion and extending the upstream portion (JICA, 1994; Kjerfve et al., 1997, Guimarães et al., 2007). The bottom morphology inside the bay is also influenced by tidal currents that drain through the central channel carrying a large sediment load.

The accumulation of high amount of organic matter associated with the deposition of fine sediments has given rise to predominantly anaerobic metabolic processes (Silva et al., 2008), which has favored the formation of gas in surface and subsurface layers of sediment (Catanzaro, et al., 2004). The occurrence of seepages of gas bubbles into the water column by sediments is a well-known phenomenon that can be observed in large areas of the Guanabara Bay (Catanzaro et al., 2004).

### 2. Material and methods

### 2.1 Geophysical data and gathering of cores

Over 240 km of seismic lines were collected in the NE area of the Guanabara Bay (Brazil), (Fig. 1). These data were collected in the frequency of 12 kHz with Bathy 2010PC<sup>TM</sup> of SyQwest using the hydrographic software HYPACK® 2013 for planning and navigation and GTR-G2 TechGeo DGPS for positioning.

The physical properties, compressional wave velocity (Vp), Gamma density, magnetic susceptibility and electrical resistivity were obtained by profiling four sedimentological cores in the Multi Sensor Core Logger, combined with visual description data.

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Fig. 1. A. Geographical location of the study area in Brazil (•). B. Studied stations for surface sediments in the NE region of the Guanabara Bay (GB1-9). C. The studied seismic lines, sediment cores (T1-T4) and the positions of surface sediment samples (GB1-9).

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Two sediment cores (T2 and T4) were collected in a zone with occurrence of gas in sediments and two others (T1 and T3) without the occurrence of gas in sediments (Fig. 1). Several data were acquired in these cores: grain size, organic matter (OM), carbonates, moisture content, geotechnical properties and radiocarbon data (Delavy, 2015).

#### 2.2 Sedimentological data obtained along the core

Analysis of geotechnical properties, grain size, OM and carbonates were performed at 25-cm resolution and moisture content at 4-cm resolution along the cores. For geotechnical testing, a handheld penetrometer and Torvane shear device were used to estimate the sediments cohesion force and shear strength, respectively. The moisture content of the samples was obtained by the ratio between the weights of sediment with water and without water.

The concentrations of OM and carbonate were obtained applying a sediment calcination methodology (using 0.5 g of sediment) with the following steps: i) 1 hour at 100°C to obtain the dry sample weight; ii) 1 hour at 550°C to obtain organic matter content; and; iii) 1 hour at 1000°C to obtain carbonate content.

The grain size of core sediment was estimated using a laser granulometer model Malvern Mastersizer 2000. The data treatment was performed according to the statistical parameters of Folk and Ward (1957) through Gradistat software.

The <sup>14</sup>C analyses were performed by Accelerator Mass Spectrometry- AMS at the Radiocarbon Laboratory (LAC- UFF). Calibration was performed with OxCal software (Bronk Ramsey, 2009) using the Marine13 calibration curve (Reimer et al., 2013) with an offset of  $8 \pm 17$  years (Angulo et al., 2005).

#### 2.3 Sampling surface sediments

Nine stations were sampled for surface sediment in the NE area of the Guanabara Bay, from the Paquetá Island to the inner zone of the bay on 27.01.2016 (Fig. 1; Tab. 1). Each sampled station was georeferenced with a GPS (model GPSMAP® 78S). The physicochemical data such as salinity, temperature, dissolved oxygen and pH were obtained with a multiparameteric probe. The sediment samples were collected with a box core.

Triplicates of samples were collected in each site according to the methodology established by Schönfeld et al. (2012). The first upper centimeter of sediment was scraped with a plastic spatula and placed in referenced plastic bags and containers to get sedimentological and biotic data (foraminifera).

During the fieldwork, the sediments for foraminiferal studies were stored in ethanol with Rose Bengal (2 g of Rose Bengal in 1000 ml alcohol) to differentiate living from dead specimens (Schönfeld et al., 2012).

Samples for sedimentological analysis were cool preserved and then subjected to grain size and geochemical analysis such as total organic carbon (TOC), stable isotopes and Rock-Eval pyrolysis.

Variables/ Stations	Latitude S	Longitude W	Salinity	Temperature (°C)	рН	O <sub>2</sub> (mg/l)
GB1	22° 47'42.33"	43° 5'6.29"	21.8	30.1	8.4	6.3
GB2	22° 46'16.62"	43° 4'32.85"	21.0	31.8	8.4	6.3
GB3	22° 44'56.03"	43° 4'0.82"	12.5	32	8.51	6.1
GB4	22° 42'57.70"	43° 4'31.16"	12.1	32.2	8.22	6.6
GB5	22° 43'7.02"	43° 6'6.54"	35.9	25.0	7.7	9.7
GB6	22° 45'48.52"	43° 6'22.53"	32.7	31.7	8.34	7.9
GB7	22° 48'48.27"	43° 5'38.48"	33.8	30.9	8.4	6.4
GB8	22° 49'05.2"	43° 04'57.5"	35.7	29.8	8.27	5.4
GB9	22° 48'33.00"	43° 4'27.80"	31.9	28.6	8.5	8.6

Tab. 1. Coordinates of sampling stations (GB) and physicochemical parameters of water: salinity, temperature (°C), pH and O<sub>2</sub> (ml/l).

#### 2.4 Sedimentological analysis of surface sediment samples

#### 2.4.1 Textural analyzes

The samples for particle size analysis were dried and wet sieved through a 63  $\mu$ m screen. The sediment fractions >63  $\mu$ m and <63  $\mu$ m were dried and weighted.

The sediment fraction >63  $\mu$ m was separated in several fractions using a battery of sieves (125  $\mu$ m, 250  $\mu$ m, 500  $\mu$ m, 1000  $\mu$ m and 2000  $\mu$ m).

The percentage of each sediment fraction was determined. The textural classification of sediments was based on Folk and Ward (1957) using Gradistat software.

# 2.4.2 TOC and Rock-Eval pyrolysis of surface sediment samples

About 10 mg of each sample were used for TOC analyses. After elimination of carbonates with 50% hydrochloric acid, the samples were analyzed in the Laboratory of Chemostratigraphy and Organic Geochemistry of Universidade do Estado do Rio de Janeiro (LGQM-UERJ), with a LECO SC-632 equipment, which directly provided the percentage of TOC.

As all the samples displayed TOC content >1%, sediments were further submitted to Rock-Eval pyrolysis analysis (Rock-Eval 6, Vinci Technologies), at the LGQM-UERJ. The results of TOC content were evaluated according to Peters and Cassa (1994) (Tab. 2). Rock Eval pyrolysis is used to identify the type and maturity of organic matter and to detect petroleum potential in sediments.

According to Espitalié et al. (1977), the parameter S1 is the amount of free hydrocarbon (mg HC/g sediment) liberated at 300°C. The S2 is the amount of hydrocarbon released from cracking organic compounds (mg HC/g sediment) and heavy hydrocarbons during temperature programed pyrolysis (300–600 °C) and is related to hydrocarbon source potential. The S3 represents the amount of CO<sub>2</sub> resultant from breaking carboxyl groups and other oxygen-containing compounds, obtained at 300–390 °C.

The hydrogen [HI =  $(S2/TOC) \times 100$  in mg HC/g TOC] and oxygen indices [OI =  $(S3/TOC) \times 100$  in mg CO<sub>2</sub>/g TOC] were calculated following the procedures of Espitalié et al. (1977). The HI is a parameter used to characterize the origin of organic matter. Marine organisms and algae are in general composed of lipid- and protein-rich organic matter, where the H/C ratio is higher than in the carbohydrate-rich constituents of land plants. HI typically ranges from ~100 to 600 in marine geological samples.

The OI is a parameter that correlates with the O/C ratio, and so it is related to the amount of oxygen in organic compounds (El Nady et al., 2015). The OI is high for polysaccharide-rich remains of land plants and inert organic material (residual organic matter) encountered in marine sediments. The OI values range from near 0 to ~150.

On the basis of S2, HI and Tmax values, a semiquantitative assessment of the generation potential, type and stage of thermal evolution of organic matter was performed (Espitalié et al., 1985/86).

The scales used for each case are shown in Table 2 (Espitalié et al., 1977; 1985/86). The production index (PI) values also were determined using the formula: S1/S1+S2. PI is used to characterize the evolution level of the organic matter.

# 2.4.3 Carbon stable isotopes ( $\delta^{13}C$ ) in sedimentary organic matter

The carbon stable isotopes of organic matter were analyzed at the LGQM-UERJ. After the removal of carbonates with HCl 1M hydrochloric acid, the samples were washed with Milli-Q water to removal the acid and dried in an oven and desegregated with an agate mortar. After these procedures,  $\delta^{13}$ C values were determined using Elemental analyzer coupled to Delta V Mass Spectrometer. The standard deviation of the analysis is 0.15‰.

#### 2.5 Living foraminifera analysis in surface sediment

Living benthic foraminifera analyses were performed in Laboratory of Micropaleontology of Universidade do Estado do Rio de Janeiro (LMP-UERJ). The rose Bengal treated samples collected in triplicates (about 50 ml) at each site were combined analyzed. The sediment fraction >63  $\mu$ m was wet sieved through a 63  $\mu$ m mesh screen. After washing, the samples were dried in an oven at  $\approx$ 50°C. The sediments fractions <63  $\mu$ m were discarded. The sediment fraction >63  $\mu$ m was dry sieved through a 500  $\mu$ m screen. Both dry sediment fractions, 63-500  $\mu$ m and >500  $\mu$ m, were analyzed. However, no foraminifera were found in the >500  $\mu$ m fraction. Thus foraminiferal assemblages dimension, structure and composition were evaluated in the sediment fraction 63-500  $\mu$ m.





Tab. 2. Semi-quantitative scales assessment of TOC content according to Peters and Cassa (1994), potential generator of organic matter (S<sub>2</sub>), hydrogen index (IH) and organic matter maturation – Tmax (°C), based on Espitalié et al. (1977; 1985/86).

Total Organic Carbon TOC (%; Peters and Cassa, 1994)							
<0.5	Poor						
0.5 - 1.0	Moderate						
1.0 - 2.0	Good						
2.0 - 4.0	Very Good						
>4.0	Excellent						
Potential ge	nerator of organic matter $S_2$ (mg HC/g sediment)						
<2.0	Low potential generator						
2.0 - 5.0	Moderate source potential						
5.0 - 10	- 10 Good source potential						
>10	Excellent source potential						
Hydro	gen Index –HI [(mg HC/g TOC) x100],						
IH < 200	Gas Potential						
$200 < \mathrm{IH} < 300$	Gas and Condensate Potential						
IH > 300	Oil Potential						
Maturation – Tmax (°C)							
<440	Immature						
440 - 470	Mature						
>470	Senile						

Foraminiferal density (FD) as the number of living specimens (stained) per gram of sediment was calculated. The assemblages of living foraminifera were characterized by counting at least 300 specimens of rose Bengal stained foraminifera per sample.

The generic taxonomical classification of Loeblich and Tappan (1987), and specific concepts of Cushman and Brönniman (1948 a, b), Boltovskoy et al. (1980), Debenay et al. (1998; 2001), Martins and Gomes (2004) and Laut et al. (2012a) were followed. After identification, the species names were checked in World Register of Marine Species (WoRMS: http://www.marinespecies.org/index.php; accessed on 01.09.2016). The specimens were stored in micropaleontological slides (as suggested by Schönfeld et al., 2012).

The percentage of each species per sample was determined. Several assemblages' parameters were also calculated. The number of species found in each sample, named richness of species (S) was determined. The Shannon index (H') was used as measure of diversity: H' =  $\Sigma$ pi Ln pi, where pi represents the portion of i- species in the sample and Ln is the natural (base e) logarithm. Equitability or evenness (J') was obtained through J'=H'/Ln (S), where S is the richness of species, H' is the Shannon diversity index and Ln the natural (base e) logarithm. These biotic parameters were determined with the software Primer 6.

#### 2.6 Statistical Analysis

As a very limited number of living specimens was found at station GB8, this sample was not considered in statistical analysis. The most frequent foraminiferal species (with a relative abundance  $\geq 3\%$  in at least one site) were further used in the statistical analysis. Before the statistical analysis the biotic and abiotic data were logarithmically transformed log (X+1). Principal components analysis was carried out in

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Statistica 12.0. Maps were performed with Arc Gis 9.2 B using coordinates according to WGS84 (UTM zone 23 S) datum.

#### 3. Results

#### 3.1 Acoustic signatures and core sediment properties

The analysis of seismic profiles revealed a large area, of about 50% of the total analyzed area, affecting signal penetration due to gas charged sediments. Following the classifications of Taylor (1992) and Baltzer et al. (2005), it was possible to identify acoustic signatures of black shadow and acoustic blanket. Features related to percolation of gas as intra sedimentary plumes, turbidity pinnacle, and structures related to seepages to the water column, as acoustic plumes and pockmarks were also identified.

The black shadow (BS) is a very dark signal commonly with sharp lateral limits and multiple distinct reflections to the end of the profile (Fig. 2). It differs from the acoustic blanket because it is not possible to detect the signal penetration into the substrate, all the signal energy appears to be reflected by the surface sediments (Baltzer et al., 2005). When the acoustic blanket (AB-I) is a few centimeters deep and up to 1.3 m below the bottom surface it generates diffuse and more attenuated sequences of surface multiple due to less reflected energy than the black shadow. When the acoustic blanket (AB-II) is lying at depths between 1.3 m and 9 m below the bottom surface there is no seismic signal below its occurrence (Fig. 2). Turbidity pinnacles and intrasedimentary plumes appear as gas percolation seepages into the sediment originated from the acoustic blanket - II (Fig. 2).

Turbidity pinnacles develop towards the surface and range between 1 and 2 m in height, while the intrasedimentary plumes can reach up to 7.5 m toward the surface. Acoustic plumes appear in several dimensions. The largest one is 3 m in height and 12 m width located in deeper water near Paquetá Island. The smallest one, with 0.5 m height and 2.5 m width, is located in shallower areas.

The cores (with 150-240 cm in length) are composed by muddy sediments, passing from fluid mud at the top to compacted mud in the base. Bioturbation structures and many bioclasts were observed along the cores. Cores are composed by the following sediment fractions: silt (83-93%), very fine sand (0.1-17.2%) and clay (3-12.6%) (Fig. 3). Regarding OM and carbonate contents, the percentages range from 11-22% and from 3.8-11.6%, respectively (Fig. 3). The OM percentage gradually decreases with increasing depth, while the carbonate content is variable along the cores. The geotechnical properties of shear strength (SS) and cohesive force (CF) is lower at the top, where the water mass is 3.85 times greater than the dry solid mass. From 100 cm deep to the base, the values of SS (0.26-0.72 kg.cm<sup>-2</sup>) and CF (0.04-0.11 kg.cm<sup>-2</sup>) increase and water mass content decrease (1.27-2.26). The range of P-wave velocity (Vp), Gamma density, magnetic susceptibility and electrical resistivity measured along the cores T1-T4 are shown in Table 3.

The depth plots of these variables along the cores T1 and T2 are presented in Figure 3. In gas free sediments, the density values ranging from 1.08 to 1.48 g·cm<sup>-3</sup> and the Vp ranged from 1441 to 1541 m·s<sup>-1</sup>. In gas charged sediments, density varied between 1.10 and 1.46 g·cm<sup>-3</sup> and Vp between 1372 and 1505 m·s<sup>-1</sup>. All cores show a general trend of increasing in density values from the top to the base. However, punctual decreases of these values were observed along the cores (Fig. 3).

The radiocarbon dating showed that sediment accumulation rates are smaller in core T1 and higher in cores T2 and T4. Core T1 has small accumulation increments from 0.03 to 0.09 cm.year<sup>-1</sup> in the last 4,000 BP, while cores T2 and T4 showed a significant increase in accumulation rates from 0.16 to 0.90 cm.year<sup>-1</sup> in the last 200 years.

# 3.2 Abiotic variables related to the event for sampling surface sediment

During surface sedimentsampling, the water temperature varied between 25°C and 32.2°C (mean 30.2°C), salinity from 12.1 to 35.9 (mean 24.4; Fig. 4A) and pH from 7.7 to 8.5 (mean 8.3) (Tab. 1). The DO in the water column ranged from 5.4–9.7 mg/l (mean 7.0 mg/l). Temperatures were relatively elevated agreeing with the summer season. The lowest values of salinity were recorded near the rivers out flow of the NE region of Guanabara Bay (GB) and of pH in station GB5, located near the Surui River mouth.

The estimated sediment mean grain size varied between 17-124  $\mu$ m (mean 41.9  $\mu$ m) with fine fraction ranging from 27% to 94.6% (mean 72.9%; Fig. 4B; Tab. 4). In most of the analyzed sites mud and sandy mud compose the substrate.

In the studied surface sediment samples the following (Tab. 4) variations were observed: TOC between 1.5-5.8 % (mean 3.7 %; Fig. 4C);  $\delta^{13}$ C between -21.6 ‰ to -24.6 ‰ (mean -23.1 ‰; Fig. 4D), S1 between 0.5-1.8 mg HC/g (1.0 mean mg HC/g; Fig. 5A); S2 between 2.7-10.5 mg HC/g (mean 6.2 mg HC/g; Fig. 5B) and; S3 between 2.2-7.6 mg CO<sub>2</sub>/g (mean 5.5 mg CO<sub>2</sub>/g; Fig. 5C).





Fig. 2. Profile 1 displaying the identified acoustic signatures.

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Fig. 3. Depth plots of grain size, P-wave velocity (Vp), density, water content, organic matter (OM) and carbonate parameters in the cores T1 (gas free sediments) and T2 (gas charged sediments).



Core	Sediment	Length (cm)	Vp (m.s <sup>-1</sup> )		Der (g.c	nsity cm <sup>-3</sup> )	E (Ω	CR .m)	MS (x10-5SI)	
			Min.	Max.	Min.	Max.	Min.	Max.	Min.	Max.
T1	free gas	240	1450	1541	1.08	1.33	0.54	1.98	-3.6	5.0
T3		180	1441	1487	1.24	1.48	0.71	1.37	0	7.2
T2	– charged gas	150	1372	1505	1.10	1.26	0.51	0.87	-0.19	6.4
T4		200	1439	1486	1.31	1.46	0.65	0.91	2.77	8

Tab. 3. Range of P-wave velocity (Vp), Gamma density, electrical resistivity (ER) and magnetic susceptibility (MS) at cores T1-T4.



Fig. 4. Maps of distribution: A. Salinity; B. Fine fraction (Fines; %); C. TOC (%); and D. δ<sup>13</sup>C (‰).

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**Fig. 5.** Maps of distribution of Rock-Eval pyrolysis parameters, namely: A.S1 (mg HC/g sediment);B. S2 (mg HC/g sediment);C. S3 mg CO<sub>2</sub>/g sediment; D. Hydrogen Index [HI; (mg HC/g TOC) x100]; E. Oxygen Index [OI;(mg CO<sub>2</sub>/g TOC) x100] and;F. Production Index (PI).

The hydrogen index (HI) ranged from 127.7 to 210.1 mg HC/g TOC (mean 168.7 mg HC/g TOC; Fig. 5D) and oxygen index (OI) from 131.2 mg CO<sub>2</sub>/g TOC to 195.5 mg CO<sub>2</sub>/g TOC (mean 150.4 mg CO<sub>2</sub>/g TOC; Fig. 5E). The Tmax values are <440 °C. The production index (PI) values (S1/S1+S2) oscillated between 0.12-0.16 (Fig. 5F). The most positive  $\delta^{13}$ C values were recorded in the stations located nearer the Guanabara Bay connection with the ocean as well as HI index values.

#### 3.3 Living benthic foraminifera in surface sediment

Data of foraminifera are presented in Appendix 1. Living FD is <3487 ind/g of bulk sediment (Fig. 6A), H' is <1.74 (Fig. 6B) and J' is <0.57. These values are indicative of low abundance, diversity and equitability of living foraminifera assemblages in the studied stations. In these stations, 44 species of living foraminifera were identified. The most abundant species by decreasing order are: *Ammonia tepida* (48-95%), *Cribroelphidium excavatum* (0.3-32%%), *Ammotium* 

salsum (<15%), Bolivina striatula (<12%), Gavelinopsis praegeri (<9%), Buliminella elegantissima (<8%), Discorbis parkeri (<7 %), Reophax nana (<6%), Elphidium gerthi (<5%), Bolivina compacta (<3%) and Rosalina floridana (<3%).

#### 3.4 Principal Components Analysis (PCA)

Factor 1 and Factor 2 of the PCA presented in Figure 7 explain together most part of data variability (63%, contributing with 43% and 20%, respectively). The PCA separated four groups of variables.

Group 1 is composed by *B. striatula*, *G. praegeri*, *D. parkeri*, *E. gerthi*, *B. compacta* and R. *floridana*, H', J' and S. These species are more common in the outer sector of the study area. The variables of Group 1 are negatively related to the Group II, which includes TOC, S1, S2, S3 and fine fraction. Group 3 contains *A. tepida*, *B. elegantissima*, PI and FD and is in opposition to Group 4 that is composed by *C. excavatum* and *A. salsum*.



Fig. 6. Maps of distribution of: A) for aminiferal density (FD; n.º/g) and; B) Shannon Index (H').

### 4. Discussion

#### 4.1 Surface sediments

The grain size of surface samples indicates that in most part of the studied sites the bottom currents are weak and favorable to the deposition of fine grained sediments enriched in organic matter (mean TOC values 3.7%) on the bottom of the Guanabara Bay. This is also supported by the significant positive correlations between TOC and sedimentary fine fraction. These results of surface sediments well agree with data obtained from sediment cores presented in this work and also that performed by Figueiredo Jr. et al. (2014).

In costal environments, carbon origins consist of complex mixtures from autochthonous and allochthonous sources. The isotopic composition of organic matter ( $\delta^{13}$ C and  $\delta^{15}$ N) obtained by Carreira et al. (2002) in surface sediments (vertical cores collected at eight locations) allowed to verify that in the Guanabara Bay, a mixture of marine, land



and estuarine sources of organic matter is present. The  $\delta^{13}$ C values of organic matter produced from atmospheric CO<sub>2</sub> by C3 terrestrial plants range from -23 ‰ to -34 ‰, while C4 plants have  $\delta^{13}$ C values between -9 ‰ and -17% (Schubert and Calvert, 2001).

Marine organic matter has  $\delta^{13}$ C values ranging between - 20 ‰ and -22 ‰ (Meyers, 1994). The  $\delta^{13}$ C values found in the studied samples (ranging from -21.60 ‰ to -24.56 ‰; mean -23.1±1.5 ‰) can suggest that the NE part of Guanabara Bay receives predominately organic matter from higher plants.

Carreira et al. (2002) reported an increase of inputs of terrestrial organic matter in recent years in the bay. They also verified that although high respiration rates occurring in the water column, rapid sedimentation rates therein result in the transfer of a significant proportion of carbon to the anoxic sediments. In the stations where surface samples were collected sediments displayed dark gray to black color that might suggest reducing conditions just some millimeters below the water-sediment interface.



Fig. 7. Results of PCA for the first two components, based on the percentage of the main species of benthic foraminifera, foraminiferal density (FD; n°/g), specific richness (S), Shannon index (H') and equitability (J'). Some abiotic variables also were used: salinity (sal); sedimentary fine fraction content (Fines; %);total organic carbon (TOC; %);  $\delta^{13}$ C (d13C; ‰) and Rock-Eval pyrolysis parameters, namely S1 (mg HC/g sediment),S2 (mg HC/g sediment),S3(mg CO<sub>2</sub>/g sediment), hydrogen index [HI; (mg HC/g TOC) x100]; oxygen index [OI; (mg CO<sub>2</sub>/g TOC) x100] and production index (PI).

The values of TOC, fine fraction, S1, S2 and S3 present a similar pattern of distribution as indicated by the results of PCA (Fig. 7). Accordingly, the highest TOC content results in a higher source generation potential (indicated by S2 values) and free hydrocarbon content (indicated by S1), and can give place to a higher production of  $CO_2$  (indicated by S3 values) from organic matter degradation. Peters (1986) and Espitalié et al. (1970, 1980, 1985/86) reported that oil generation from source rocks began at  $Tmax = 435-465 \text{ }^{\circ}\text{C}$ , and production index "PI" between 0.2 and 0.4. The organic matters are in an immature stage when "Tmax" has a value <435 °C, and "PI" <0.1 and the gas generation from source rocks began at "Tmax" 470 °C, and production index "PI" >0.4. Thus, the Tmax values (<440 °C) in the studied samples suggest that the organic matter in surface sediments are thermally immature, which is in agreement with the geological context. Being the organic matter immature, it can be expected that the PI was <0.10 (Espitalié et al., 1985/86). However, the estimated PI values are >0.10, suggesting that the samples are contaminated by oil released probably by boats and by the activities of the oil refineries such as the REDUC, Duque de Caxias. Moreover, as the organic matter is terminally immature, the TOC content and the S2 (related to the source potential) and HI values can be considered originals, signifying a presence of good to excellent amount of organic matter associated with moderate to good source potential for gas production. Plotting the analyzed samples on a diagram of Van Krevelen type (van Krevelen, 1950; Fig. 8), the predominance of organic matter content of Type III can be observed. This kind of organic matter, when subjected to anaerobic bacterial degradation or to high temperatures, can give place to gas production.

#### 4.2 Distribution of gas and core sediment properties

Geophysical results presented in this work evidences that gas charged sediments are distributed from the Paquetá Island until the innermost area of the Guanabara Bay, and mainly towards the São Gonçalo margin (Fig. 9). The distribution of gas acoustic signatures when correlated with bathymetry, it is associated with higher topographic gradient area (E/NE). This margin receives the influence of most significant drainage from the basin of the Guanabara Bay (Figueiredo Jr. et al., 2014). The bottom sediments at the gas occurrence area are classified as coarse silt to very coarse silt and in this zone the organic matter content can reach 10-20% (Galvão, 2014). Black shadow occurs in 59.3% and acoustic blanket occurs in 40.7% of the total gas charged sediments. The shallow gas vertical distribution ranges from surface sediments to 9 m below the interface water-sediment.



The spatial distribution of the different types of gas signatures depends to some extent on the overlying lithology, but can be also associated with the amount of gas trapped in sediment. Thus a high concentration of gas might favor its release (Missiaen et al., 2002). Garcia-Gil et al. (2002) showed that presence of gas in sediments is closely linked to Holocene mud because its ability in trapping underlying gas reservoir. Such connection can also be true for the gas charged sediments in the studied bay because deposition of muddy fluvial and marine Holocene sediments over Pleistocene sediments according to the evolution of the bay (Amador, 1980 a-b; Galvão, 2014). The origin of gas in Guanabara Bay is biogenic, since the geology of the area does not suggest possible thermogenic sources. Therefore, gas generation is a result of the high sediment accumulation rate, from 0.03 to 0.90 cm.yr<sup>-1</sup>, and intense input of organic matter in the area.



Fig. 8. Diagram of Van Krevelen type (van Krevelen, 1950).

The black shadow (BS) is associated with the recent generation of gas in the surface sediments and is typically related to the input of organic matter (Baltzer et al., 2005). Furthermore, in this study the BS signatures are related to both the migration from the lower layers and the recent generation in near-surface sediments (Figs. 9 and 10) as the area presents favorable conditions for gas production, as indicated by geochemical data. The density decrease along the cores agrees with the reduction in Vp. According to Ayres Neto (1998), the geological factors that control the Vp



in marine sediments are porosity, density and confinement pressure and, in general, there is a trend of increase in Vp, with decreased porosity and an increase in density. Decreases in Vp and density in gas free sediments are linked with the reduction of fine sand percentage and increase in the percentage of clay, water content and carbonate shells (Fig. 3). On the other hand, in gas charged sediments, besides the already mentioned factors, the reduction in density values are mainly associated with the presence of the gas and the large percentage of OM (13% to 20%).

Core T2, for example, shows that the intervals with lower Vp velocities,  $1372-1400 \text{ m} \cdot \text{s}^{-1}$ , recorded at 45 cm to 70 cm (Fig. 3), are associated with a layer of dark mud, where

circular structures caused possibly by bubbles marks were observed in the sediment. The existence of trapped gas bubbles in sediment layers produced by biochemical degradation of organic matter, scatter and attenuate sound (Hamilton, 1972) since volumes of 0.1% can affect the geoacoustic behavior of sediment (Ayres Neto, 1998). All cores showed very low magnetic susceptibility (maximum 7.98x10<sup>-5</sup> SI; minimum -3.6x10<sup>-5</sup>SI). This can be attributed to significant concentrations of organic matter and biogenic materials and sometimes to presence of diamagnetic minerals, which weaken the magnetic field (Ellwood et al., 2006).



Fig. 9. Gas acoustic signature distribution in Guanabara Bay superimposed on bathymetric contours.





Fig. 10. Profile 2 displays percolation of gas within the sediments.

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Contrary to expectations, since the gas has resistive properties, the profiles with the highest values of resistivity were found in the gas free sediments (0.54 to 1.98  $\Omega$ ·m). These findings may be related to the presence of high percentage of clay in the gas charged sediments. According to Becegato and Ferreira (2005), clays have the property to facilitate the passage of electrical current due to the increased amount of interstitial fluid.

# 4.3 The benthic foraminiferal response to TOC enrichment and gas release

Living benthic foraminiferal assemblages of the NE sector of the Guanabara Bay include species that have been reported in other works previously performed in this ecosystem (e.g. Kfouri-Cardoso et al., 2006; Laut et al., 2012; Pereira et al., 2006; Figueira et al., 2007; Vilela et al., 2001 a, b, 2003, 2004, 2007, 2011; Donnici et al., 2012; Eichler et al., 2013; Clemente et al., 2015; Baptista Neto et al., 2016) as well as in other Brazilian (e.g., Brönnimann et al., 1981; Bonetti and Eichler, 1997; Raposo et al., 2016), and worldwide (Murray, 1991, 2006; Laut et al., 2014, 2016 a; b, c; Martins et al., 2015 a, b, 2016 a, b) transitional coastal environments. Laut et al. (2012b) based on published works gave an overview of the main species that occur in the Guanabara Bay.

Living benthic foraminiferal assemblages of the studied area include species that can tolerate changes in physicochemical parameters such as temperature, salinity, pH and oxygen, which change seasonally depending on the season and meteorological conditions and daily depending on tidal phases. They show, in general, a reduced density and diversity and are largely dominated by *A. tepida* and locally by *C. excavatum*.

However, some variability was observed depending on some abiotic variables as suggested by the PCA (Fig. 7). These statistical results indicate that living benthic foraminifera diversity (indicated by S and H') and equitability (J') and some species, such as *B. striatula*, *G. praegeri*, *D. parkeri*, *E. gerthi*, *B. compacta* and *R. floridana* are negatively correlated to TOC, S1, S2 and S3 but positively correlated with  $\delta^{13}$ C and OI values. This group of species reaches the highest relative abundance in the stations located nearest the Guanabara Bay entrance and in waters with relatively high salinity. All these relationships indicate that this group of species is sensitive to an excessive increase of organic matter, prefer high quality of OM and are favored by aerated waters with mainly marine salinity. Ammonia tepida, B. elegantissima and FD are positively correlated with PI suggesting their probably tolerance to contamination by oil and to the exhalation of gas from the sediments. In particular, *A. tepida* has been recognized to be one of the most tolerant species to pollution (e.g., Frontalini and Coccioni, 2008; Romano et al., 2009; Armynot du Châtelet et al., 2011; Foster et al., 2012).

All the other species have negative or weak correlations with PI. In opposition to the increase of PI there are *C. excavatum* and *A. salsum*. Both *C. excavatum* and *A. salsum* are detritivorous species (Boltovskoy, 1965; Murray, 1991, 2006). Results of this work indicate that both species, which are also known to be tolerant to large environmental variability caused for instance by oscillation of temperature, salinity and oxygen content (Murray, 1991, 2006), are probably sensitive to organic matter contaminated by oil.

#### 5. Conclusion

Results of this work evidence that significant accumulation of gas occurs in the NE part of the Guanabara Bay. The main drainage that flows into the bay and the currents circulation pattern favor the occurrence of high sediment accumulation rates and the deposition of fine grained sediments enriched in organic matter. Geochemical data suggest that organic matter present in surface sediments can potentially generate gas.

The highest TOC content results in a higher source generation potential and free hydrocarbon content and can give place to a higher production of  $CO_2$  from organic matter degradation.

Geophysical results allowed the identification of high concentration of shallow gas from the water-sediment interface until 9 m below this interface. These occurrences of shallow gas are related to the migration from the lower layers, possibly related to the capping of those layers by Holocene muds, and a recent gas generation in near-surface sediments is also possible by favorable conditions (high OM content, high sedimentation rate and fine grained sediment); gas escape to the water column are occurring in several places of the NE sector of the Guanabara Bay.

The impact on benthic organisms of the high accumulation of organic matter and releasing of gas from the sediment is yet not well known in the Guanabara Bay. However, the living assemblages of benthic foraminifera are characterized in general by low density, diversity and equitability and high dominance, which are indicative of high environmental stress. In the stations affected by the highest accumulation of organic matter, located in zones where gas



seeping takes place, foraminiferal assembles displayed the lowest density, diversity and equitability. Foraminiferal density is slightly higher where TOC content decreases but the quality of organic matter increases due to a relatively high supply of materials resultant of autochthonous biological productivity and smaller deposition of organic materials provided from continental sources.

Thus, it can be speculated that the excessive accumulation of degraded organic matter (low food quality and generation of redox conditions) and the gas escape represent adverse factors for benthic communities. Some foraminiferal species however seems to have a higher tolerance to these adverse environments. *Ammonia tepida* seems to be the most tolerant species to stress caused by reducing conditions, high amount of organic matter with low quality and gas seeping.

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Appendix 1. Biotic data. Ecological parameters and percentage of living benthic foraminifera species per sample. N - number of living specimens identified and counted in each sample; S specific richness; H' – Shannon index; J' – equitability; FD - Foraminifera density  $(n.^{\circ}/g)$ 

Percentage of Species in each Station	GB1	GB2	GB3	GB4	GB5	GB6	GB7	GB8	GB9
Ν	1041	761	607	345	635	641	945	1	349
S	22	11	18	6	10	26	30	1	8
J'	0.28	0.27	0.35	0.14	0.34	0.42	0.52	nd	0.57
Н'	0.88	0.64	1.01	0.25	0.78	1.37	1.75	nd	1.19
FD	302	213	49	42	3487	129	625	0	10
Ammonia parkinsoniana (d' Orbigny, 1839)	0.0	0.0	0.0	0.3	0.0	0.3	0.0	nd	0.0
Ammonia tepida (Cushman, 1926)	80.2	87.0	75.8	95.1	81.4	67.9	57.6	nd	48.4
Ammotium salsum (Cushman & Brönnimann, 1948)	0.2	0.0	0.0	0.0	0.0	0.0	0.8	nd	15.2
Asterigerinata mamilla (Williamson, 1858)	0.0	0.0	0.0	0.0	0.0	0.0	0.1	nd	0.0
Bolivina compacta Sidebottom, 1905	0.2	0.0	0.0	0.0	0.0	0.6	2.5	nd	0.0
Bolivina lowmani Phleger & Parker, 1951	0.5	1.3	0.7	0.0	0.6	0.0	0.0	nd	0.0
Bolivina striatula Cushman, 1922	0.8	0.8	0.3	0.0	3.5	12.3	1.9	nd	0.0
Bulimina aculeata d' Orbigny, 1826	0.1	0.1	0.2	0.0	0.2	0.9	0.2	nd	0.0
Bulimina gibba Fornasini, 1900	0.5	0.0	0.3	0.0	0.5	0.2	0.0	nd	0.0
Bulimina marginata d' Orbibny, 1826	0.0	0.0	0.0	0.0	0.0	0.0	0.1	nd	0.0
Buliminella elegantissima (d' Orbigny, 1839)	8.3	2.9	8.1	0.9	5.8	2.2	4.9	nd	0.0
Glocassidulina crassa (d' Orbigny, 1839)	0.2	0.0	0.5	0.0	0.2	0.0	0.0	nd	0.0
Cornuspira involvens (Reuss, 1850)	0.0	0.0	0.0	0.0	0.0	0.0	0.1	nd	0.0
Cribroelphidium excavatum (Terquem, 1875)	5.4	3.5	5.1	0.3	1.7	1.7	2.1	nd	32.1
Cribroelphidium poeyanum (d'Orbigny, 1826)	0.0	0.0	0.0	0.0	0.0	0.2	0.2	nd	0.0
Discorbis parkeri Natland, 1950	0.0	0.0	0.0	0.0	0.0	0.0	6.6	nd	0.0
<i>Elphidium gerthi</i> van Voorthuysen, 1957	1.0	0.5	0.0	0.0	0.0	2.3	4.4	nd	0.3
Elphidium gunteri Cole, 1931	0.3	0.0	0.0	0.0	0.0	0.0	0.3	nd	0.0
Elphidium incertum (Williamson 1858)	0.2	0.0	0.0	0.0	0.0	0.0	0.0	nd	0.0
Fissurina lucida (Williamson, 1848)	0.0	0.0	0.0	0.0	0.0	0.5	0.2	nd	0.0
Fursenkoina conspiqua (Pishvanova, 1960)	0.0	0.0	0.2	0.0	0.0	0.0	0.0	nd	0.3
Fursenkoina pontoni (Cushman, 1932)	0.0	0.0	0.0	0.0	0.0	0.0	0.0	nd	0.3
Gavelinopsis praegeri Heron-Allen and Earland, 1913	0.4	0.9	0.7	0.3	0.2	0.8	9.2	nd	0.0
Haplophragmoides wilberti Andersen, 1953	0.0	0.0	0.0	0.0	0.0	1.1	0.2	nd	0.0
Hopkinsina pacifica Cushman, 1933	0.3	0.0	0.2	0.0	0.0	0.2	0.3	nd	0.9
Lagena striata var. (d'Orbigny, 1839)	0.0	0.0	0.2	0.0	0.0	0.0	0.1	nd	0.0
Lepidodeuterammina ochracea (Williamson, 1858)	0.3	0.4	0.5	0.0	0.0	0.2	0.1	nd	0.0
Miliolinella lutea (d'Orbigny, 1839)	0.1	0.3	0.2	0.0	0.0	0.3	0.0	nd	0.0

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Percentage of Species in each Station	GB1	GB2	GB3	GB4	GB5	GB6	GB7	GB8	GB9
Neoconorbina terquemi (Rzehak, 1888)	0.0	0.0	0.0	0.0	0.0	0.5	0.0	nd	0.0
Nonionella atlantica Cushman, 1936	0.0	0.0	0.0	0.0	0.0	0.0	0.1	nd	0.0
Ophthalmidium balkwilli Macfadyen, 1939	0.1	0.0	0.0	0.0	0.0	0.0	2.1	nd	0.0
Pseudoclavulina curta Cushman & Bronnimann, 1948	0.1	0.0	0.0	0.0	0.0	0.2	0.0	nd	0.0
Quinqueloculina lamarckiana, s. l. d'Orbigny, 1839	0.0	0.0	0.0	0.0	0.0	0.2	0.0	nd	0.0
Quinqueloculina seminula (Linné, 1758)	0.0	0.0	0.0	0.0	0.0	0.2	0.0	nd	0.0
Reophax curtus Cushman, 1920	0.0	0.0	0.0	0.0	0.0	0.0	0.5	nd	0.0
Reophax nana Rhumbler, 1913	0.7	2.2	6.1	3.2	6.0	3.7	3.1	nd	2.6
Rosalina bradyi (Cushman, 1915)	0.0	0.0	0.2	0.0	0.0	0.2	0.1	nd	0.0
Rosalina floridana (Cushman, 1922)	0.0	0.0	0.0	0.0	0.0	2.5	0.4	nd	0.0
Rosalina globularis d'Orbigny, 1826	0.0	0.0	0.3	0.0	0.0	0.0	0.2	nd	0.0
Sagrina primitiva (Cushman, 1920)	0.0	0.0	0.0	0.0	0.0	0.0	0.2	nd	0.0
Siphonina reticulata (Czjzek, 1848)	0.0	0.0	0.0	0.0	0.0	0.0	0.1	nd	0.0
Spirobolivina curta (Cushman, 1923)	0.0	0.0	0.0	0.0	0.0	0.3	0.0	nd	0.0
Textularia earlandi Parker, 1952	0.1	0.0	0.0	0.0	0.0	0.0	0.0	nd	0.0
Trochammina inflata (Montagu, 1808)	0.3	0.0	0.7	0.0	0.0	0.0	1.1	nd	0.0

Appendix 1 (cont.).Biotic data. Ecological parameters and percentage of living benthic foraminifera species per sample.