Journal of Sedimentary Environments
Published by Universidade do Estado do Rio de Janeiro
4 (2): 159-173. April-June, 2019
doi: 10.12957/jse.2019.43371

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ORGANIC COMPOUNDS AS PROXIES OF THE SEDIMENTARY ENVIRONMENTAL QUALITY OF THE MARICÁ-GUARAPINA LAGOON SYSTEM (SE, BRAZIL)

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Received on 23 May 2019
Received in revised form on 7 June 2019
Accepted on 9 June 2019

Editor: Maria Antonieta da Conceição Rodrigues, Universidade do Estado do Rio de Janeiro

Citation:
Laut, L., Alves Martins, M.V., Belart, P., Lorini, M.L., Clemente, I., Nogueira, L., Juliace, A., Fontana, L.F., 2019. Organic compounds as proxies of the sedimentary environmental quality of the Maricá-Guarapina Lagoon System (SE, Brazil). Journal of Sedimentary Environments, 4 (2): 159-173.

Abstract

Bottom sediment is a natural trap for organic matter and different kinds of pollutants. The accumulation of large amount of organic matter gives rise to the eutrophication of the aquatic ecosystems. The analyses of the quantity and quality of the organic matter (biopolymers) help to determine the trophic status of coastal ecosystems. The Maricá-Guarapina Lagoon System (MGLS) is located in Rio de Janeiro and is composed by four connected lagoons: Maricá, Barra, Padre and Guarapina. It has been suffering impacts due to the intense and uncontrolled property speculation. Based on this problem, this study aimed to characterize the organic matter (OM) amount and quality in sediments and the relation with the impacted areas in this lagoon system. The collected sediment samples were analyzed for geochemical data combined with grain size and physical-chemical environmental parameters of the bottom water. Statistical results evidenced that the sedimentary environment of the MGLS is heterogenous. The organic matter supplied to the MGLS is provided from different sources but the autochthonous contribution (phytoplanktonic productivity and vegetal detritus from the mangrove fringe) prevails. The anthropogenic contribution was more evident in Padre Lagoon, where the sediments had relatively low TOC contents (0.1-0.8%). The MGLS is accumulating mainly aged organic matter. The most impacted zones were found in Guarapina, Barra and Maricá lagoons, in bottoms of fine-grained sediments, with relatively high TOC and labile biopolymeric compounds (proteins, carbohydrates and lipids) contents, which should evolve into an ever-increasing stage of eutrophication.

Keywords: Tropical Lagoons. Biopolymers. Eutrophication. Sedimentary Dynamic.

1. Introduction

The Maricá-Guarapina Lagoon System (MGLS) is located 50 km away from the Rio de Janeiro city and is composed by four connected lagoons: Maricá, Barra, Padre and Guarapina. This ecosystem are evolving to an increasing degradation, as well as other lagoons of the Rio de Janeiro State (Laut et al., 2016, 2017; Dias et al., 2017; Raposo et al., 2018). The current environmental degradation of the MGLS is being caused by the anthropic activities developed in this region, namely by the intense and disorderly property speculation. The main industrial activities in the region are the sand extraction and the mechanized exploitation of clay materials, which are

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responsible for the increasing concentrations of nutrients and suspended solids in the water column (Cruz et al., 1996).

The excessive discharge of nutrients from domestic and industrial sewage pipes as well as the surface runoff from urban and agrarian areas has created an enrichment of organic and inorganic nutrients in this transitional ecosystem (Silva et al., 2011) aggravating its eutrophication progression. This process is considered the major factor that is causing coastal environmental degradation (Meyer-Reil and Koster, 2000), as it is happening in Rio de Janeiro lagoons (Laut et al., 2016, 2017; Dias et al., 2017; Raposo et al., 2018).

Within the aquatic ecosystems, there are three main groups of organic matter compounds used as food by the biota: lipids, carbohydrates and proteins (Jones, 2001). These biopolymers are degraded by diverse bacterial groups, with different types of metabolism (Brock et al., 1994). However, when the organic matter level is higher than biodegradation capacity of the microorganisms, it tends to accumulate leading to eutrophic conditions (Marques Júnior et al., 2002).

Bottom sediment in these environments functions as a natural trap for organic matter and different kinds of pollutants (Silva et al., 2013). Therefore, the sediment becomes a great source of nutrients and/or pollutants for the water column, interfering with pelagic primary producers and, in turn, with benthic organisms (Jørgesen, 1996; Martins et al., 2016a; Laut et al., 2016).

The sediments are the temporal record of all the processes that occur in the water column and are the final destination of the produced (autochthonous) or introduced (allochthonous) organic matter in marine environments. New approaches, related to the quality of organic matter (biopolymers), are being used to determine the trophic state of coastal ecosystems (Meyers et al., 1995, Fabiano et al., 1995; Volkman et al., 1998). Dell'Anno et al. (2002), changed the classification of coastal regions classified as oligo-mesotrophic, for eutrophic environments, using biochemical compounds.

The biopolymer approaches have been applied in the Rio de Janeiro costal lagoons to identify stressed level in the sediment. The first study was carried out by Laut et al. (2016) in Itaipú Lagoon (Niteroi city). The analysis of the quantity and quality of organic matter in this lagoon allowed: to identify three distinct regions with different level of environmental stress; to deduce that the organic matter accumulation was the result of hydrodynamic conditions mostly governed by tidal currents; to infer that the quality of organic matter was mainly influenced by urban effluents, river inputs and mangroves contributions as well as by the autochthonous lagoonal biological productivity (Raposo et al., 2018).

In Saquarema Lagonal System, Dias et al. (2017) identified distinct regions such as: an inner and impacted zone where the sediments are particularly enriched in organic matter provided by contaminated effluents and by the rivers outflow; an outer-less impacted zone submitted to high hydrodynamic energy and; an intermediate area characterized by transitional conditions between the two previously mentioned.

The biopolymers analysis was evaluated in Vermelha Lagoon by Laut et al. (2017). The authors identified high concentrations of organic matter principally carbohydrates that were associated to natural microbial activity. In this study was possible to evidence hypertrophic conditions as a result of the impact of salt pans industry.

The present study is a contribution for the evaluation of the trophic state and environmental quality of sediment in Maricá-Guarapina Lagoon System (MGLS). This approach is based on the evaluation of total organic carbon (TOC), total sulfur (TS), total phosphorus (TP) and biopolymer (BPC) content and physical-chemical water parameters.

1.1 Study area

The MGLS is located on the southeast region of Brazil (22°52' - 23°00' S and 43°00' - 42°45' W) in the coastal area of Rio de Janeiro State, at Maricá city. The MGLS has a circular shape and is inserted into a kind of deformed rocky amphitheater with its boundaries determined by the Atlantic Ocean and the Itacoatiara and Negra hills (Perrin, 1984). It is composed by four lagoons comprising a total area of 43.2 km², which are currently distributed in: Maricá (20.5 km²), Barra (11.7 km²), Padre (1.6 km²) and Guarapina (9.4 km²) (Oliveira et al., 1955).

The drainage basin consists of three main sub-basins: Vigário River Basin; Ubatiba River, which mouth is located in Maricá Lagoon and; Caranguejo River Basin which mouth is located in Guarapina Lagoon (Comitê Gestor da Região Hidrográfica da Baía de Guanabara e dos Sistemas Lagunares de Maricá e Jacarepaguá, 2006).

Barbiere (1985) described the bathymetry of the MGLS: Maricá Lagoon has a maximum depth of 2.0 m and a mean depth of 1.0 m; Barra and Padre lagoons represent the shallower part of this system, since 70% of their areas have depths of ≈0.5 m; Guarapina is the deepest lagoon of this system, with depths ranging from 2.0 to 5.0 m.

The climate of the MGLS region is tropical humid to semi humid, with tropical air masses from oceanic and continental origins (Barroso-Vanacôr et al., 1994). It is characterized by mean annual temperature of ≈23 °C and the precipitation between ≈1100 and ≈1500 mm/year.

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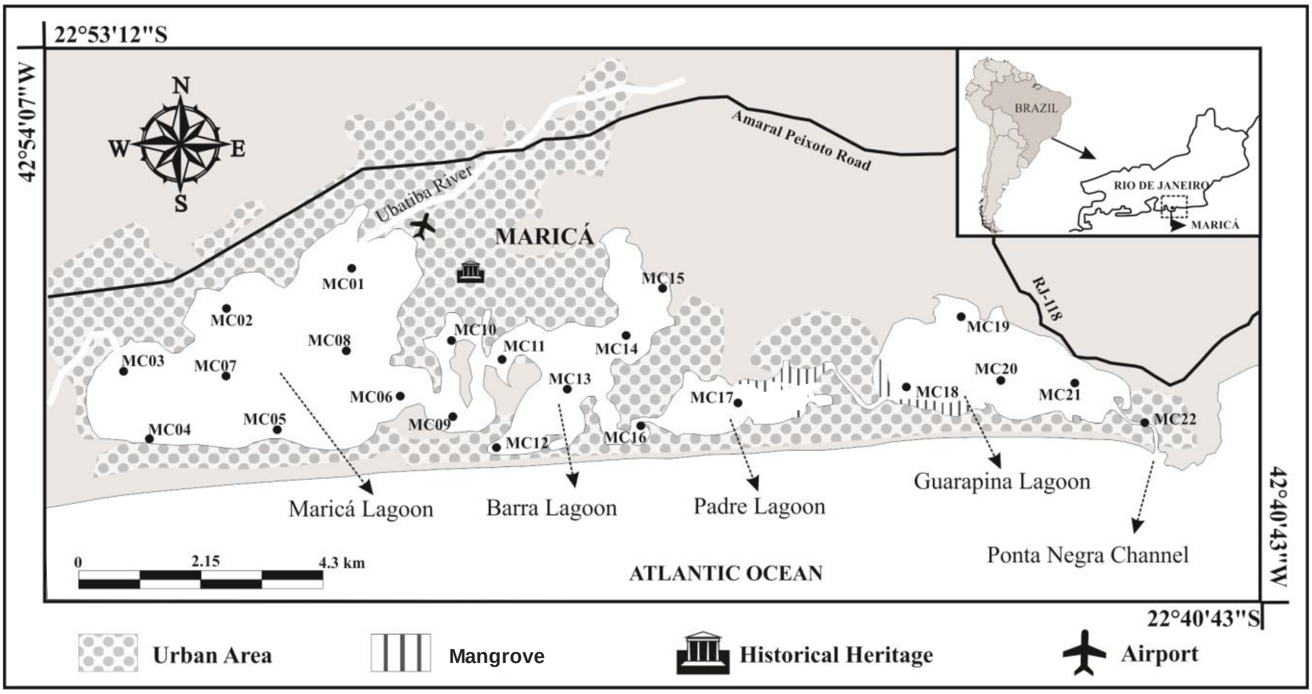

Fig. 1. The studied stations in Maricá-Guarapina Lagoon System. The urban, mangrove area, other important areas are also indicated.

Until the 50s the aperture of the sandbar to the sea was a natural process in stormy events or was carried out by fishermen when the waters reached the maximum flood in the Maricá Lagoon. In 1951, the Ponta Negra Channel was built connecting the Guarapina Lagoon to the ocean, as a result of the Governmental Sanitation Program in Rio de Janeiro State. The construction of the channel aimed to limit flooding during the rainy season and to avoid malaria focus. This work brought drastic environmental changes to the system such as reducing fishing productivity in the region and the decreasing of the water body (Amador, 1985). Currently, the channel limits the tidal dynamics and consequently the MGLS hydrodynamics has been operated by floods in rainy periods and not by tidal action (Cruz, 2010). Ponta Negra is a channel with a very limited hydraulic section due to silting (Rosman, 2007).

2. Materials and methods

2.1 Sampling

Sampling was done in March 2013 over 22 stations using a boat with short hull and a small box-corer to collect the sediment (Fig. 1). The stations were located with a GPS (Table 1). The number of samples collected in each lagoon were: 10 in Maricá Lagoon (MC1-MC10), 5 in Barra Lagoon (MC11-MC15), 2 in Padre Lagoon (MC16-MC17) and; 4 in Guarapina Lagoon (MC18-MC21), also including the Ponta Negra Channel (MC22).

During the sampling event, physical-chemical parameters, such as temperature (T), salinity (Sal), dissolved oxygen (DO), Eh and pH, were measured at the water-sediment interface using a multiparametric probe (Sanxin, model SX751) and the first centimeter was taken from the box-corer for granulometric (100 g) and biopolymeric (10 g) analyses.

2.2 Grain Size

A laser microgranulometer (Mastersizer S instrument, Malvern Instruments, Malvern, UK) was used to determine the sedimentary particle sizes (in the range from 0.05–2,000 μm) of the sediments in each sample after the removal of organic matter (with hydrogen peroxide) and carbonates (with hydrochloric acid 70%). The grain size classification system proposed by Shepard (1954) was used.

2.3 Total organic carbon (TOC) and total sulfur (TS)

Homogenized and lyophilized sediment samples (~2 g) were ground. Aliquots of ca. 0.26 g were weighed (0.1 mg precision) in a previously weighed porous porcelain crucibles. In order to eliminate the carbonate fraction, volumes of HCl (1:1 v/v) sufficient to cover the sample were added to the crucibles. Samples were treated for 24 h before filtration of solid residue, which was then washed with distilled water until complete elimination of the HCl (to pH ~6). Sample residues were dried in an oven, at 65ºC for 3 h and weighed for calculating the percentage of insoluble residue. Measurements of TOC and TS were performed with a carbon and sulphur analyser (LECO SC 144) according to a previously published method (Mendonça-Filho et al., 2003) and following the ASTM D-4239 Method (American Society for Testing and Materials, 2008).

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Tab. 1. Sedimentary Parameters of Maricá-Guarapina Lagoon System. Legend: Lat = south latitude; Long = west longitude; Sal = salinity; T = Temperature; DO = dissolved oxygen; Eh = Redox Potential; TOC = total organic carbon; TS = total of Sulphur; PTN = proteins; CHO = carbohydrates; LIP = lipids; BPC = Biopolymeric Carbon.

Stations	Lat	Long	Sal	T	DO	pH	Eh	Sand	Silt	Clay	TOC	TS	TOC:TS	PTN	CHO	LIP	BPC	PTN:CHO	CHO:TOC
	S	W		°C	mg L^{-1}		mV	%	%	%	%	%		mg C g^{-1}	mg C g^{-1}	mg C g^{-1}	mg C g^{-1}		
MC01	22°55'44.54"	42°50'27.35"	0.1	33.1	6.2	9.5	-135.7	100	0.0	0.0	0.2	0.0	5.7	0.3	1.9	0.6	2.8	0.17	11.1
MC02	22°56'0.14"	42°51'44.00"	0.2	29.7	6.3	8.9	-102.1	17.4	81.2	1.4	2.6	0.4	6.2	1.6	10.6	2.4	14.6	0.15	4.1
MC03	22°56'44.73"	42°52'51.11"	0.3	30.7	6.1	8.9	-102.2	21.6	78.1	0.3	3.8	0.7	5.4	2.4	16.3	4.2	22.9	0.15	4.3
MC04	22°57'25.87"	42°52'33.92"	0.6	30.3	7.4	9.1	-108.5	48.8	51.2	0.0	0.2	0.0	20	0.4	2.2	2.7	5.2	0.17	10.9
MC05	22°57'18.12"	42°51'3.96"	0.9	28.1	8.6	8.7	-94.8	100.0	0.0	0.0	0.3	0.0	8.7	0.4	2.6	1.9	5.0	0.16	10.2
MC06	22°56'57.18"	42°49'54.45"	1.0	27.6	6.8	8.7	-88.8	100.0	0.0	0.0	2.2	0.3	6.5	1.2	8.3	5.6	15.2	0.15	3.8
MC07	22°56'52.55"	42°51'47.00"	0.5	29.9	7.1	8.7	-90.2	13.4	79.8	6.8	3.6	0.7	5.1	2.5	17.3	6.3	26.0	0.15	4.9
MC08	22°56'19.44"	42°50'33.28"	0.5	28.3	7.3	8.7	-93	2.9	87.0	10.1	4.4	0.6	7.7	2.2	15.3	5.0	22.5	0.15	3.5
MC09	22°57'10.85"	42°49'12.46"	0.5	28.9	7.1	8.6	-72.5	36.6	63.4	0.0	4.4	1.8	2.4	3.3	22.9	4.7	30.9	0.14	5.2
MC10	22°56'24.96"	42°49'17.08"	1.4	29.7	9.8	8.9	-103.2	32.1	67.9	0.0	3.8	1.0	3.9	3.0	22.2	7.1	32.3	0.13	5.9
MC11	22°56'36.77"	42°48'40.26"	1.9	29.0	4.8	8.3	-71.9	36.6	63.4	0.0	4.0	2.3	1.8	2.8	18.2	5.5	26.5	0.15	4.5
MC12	22°57'31.54"	42°48'40.88"	3.1	28.0	4.4	8.2	-64.2	100	0.0	0.0	0.2	0.0	8.6	0.4	2.1	1.3	3.8	0.17	12.4
MC13	22°57'7.35"	42°48'12.06"	1.7	29.6	9.4	8.7	-95.6	7.5	91.1	1.4	4.6	2.5	1.8	3.4	23.4	4.6	31.5	0.15	5.1
MC14	22°56'27.55"	42°47'31.45"	2.0	30.1	10.8	8.9	-106.5	4.8	91.8	3.4	4.6	2.4	2.0	3.5	24.1	5.1	32.7	0.14	5.2
MC15	22°55'52.69"	42°47'3.52"	2.7	30.5	13.0	9.2	-117.8	5.8	90.8	3.4	4.3	2.5	1.7	3.2	21.8	4.5	29.4	0.15	5.0
MC16	22°57'21.75"	42°47'4.99"	1.8	31.4	17.1	9.3	-123.8	100	0.0	0.0	0.8	0.2	4.6	2.2	15.3	5.7	23.2	0.15	18.5
MC17	22°56'41.23"	42°45'7.60"	8.0	29.0	10.1	9.3	-121.3	100	0.0	0.0	0.1	0.0	4.5	0.3	1.5	0.5	2.3	0.18	16.9
MC18	22°56'57.65"	42°44'21.49"	20.0	28.1	4.9	9.0	-109.8	13.4	86.6	0.0	2.2	0.7	3.1	2.4	16.5	5.3	24.2	0.15	7.6
MC19	22°56'3.58"S	42°43'42.04"	16.0	29.1	6.0	9.1	-114.5	79.6	20.4	0.0	5.5	2.2	2.5	4.6	32.1	5.5	42.3	0.14	5.8
MC20	22°56'44.92"	42°43'16.05"	10.0	27.8	6.3	9.2	-118.3	17.2	82.8	0.0	4.0	0.6	6.4	3.9	27.3	4.4	35.6	0.14	6.9
MC21	22°56'51.67"	42°42'25.13"	20.0	28.5	6.2	9.2	-118.5	76.4	23.6	0.0	4.2	1.4	3.0	3.9	27.1	4.3	35.3	0.14	6.4
MC22	22°57'16.50"	42°41'46.56"	20.0	28.4	5.6	9.1	-112.6	100	0.0	0.0	4.3	1.3	3.3	4.1	28.6	3.7	36.4	0.14	6.6
	Maximum		**20.0**	**33.1**	**17.1**	**9.5**	**-64.2**	**100.0**	**91.8**	**10.1**	**5.5**	**2.5**	**20.0**	**4.6**	**32.1**	**7.1**	**42.3**	**0.18**	**18.5**
	Minimum		**0.1**	**27.6**	**4.4**	**8.2**	**-135.7**	**2.9**	**0.0**	**0.0**	**0.1**	**0.0**	**1.7**	**0.3**	**1.5**	**0.5**	**2.3**	**0.13**	**3.5**
	Mean		**5.1**	**29.4**	**7.8**	**8.9**	**-103.0**	**50.6**	**48.1**	**1.2**	**2.9**	**1.0**	**5.2**	**2.4**	**16.3**	**4.1**	**22.7**	**0.15**	**7.5**

Means of the Lagoons	Sal	T	DO	pH	Eh	Sand	Silt	Clay	TOC	TS	TOC:TS	PTN	CHO	LIP	BPC	PTN:CHO	CHO:TOC
		°C	mg L^{-1}		mV	%	%	%	%	%		mg C g^{-1}	mg C g^{-1}	mg C g^{-1}	mg C g^{-1}		
Maricá	0.5	29.9	6.9	8.9	-102.4	54.1	44.1	1.9	2.2	0.5	7.3	1.5	9.9	3.4	14.8	0.16	6.9
Barra	2.3	29.4	8.5	8.7	-91.2	30.9	67.4	1.6	3.5	1.9	3.2	2.6	17.9	4.2	24.8	0.15	6.4
Padre	4.9	30.2	13.6	9.3	-122.6	100.0	0.0	0.0	0.5	0.1	4.6	1.3	8.4	3.1	12.7	0.17	17.7
Guarapina	17.2	28.4	5.8	9.1	-114.7	57.3	42.7	0.0	4.0	1.3	3.7	3.8	26.3	4.6	34.7	0.14	6.7

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2.4 Biopolymers Concentrations

The protein (PTN) content determination was carried out after extractions with NaOH (0.5 M, 4 h) and was determined according to Hartree (1972), modified by Rice (1982), to compensate for phenol interference. Concentrations are reported as albumin equivalents. Carbohydrates (CHO) were analysed according to Gerchacov and Hachter (1972) and expressed as glucose equivalents. The method is based on the same principle as the widely used method of Dubois et al. (1956), but it is specifically adapted for CHO determination in sediments. Lipids (LIP) were extracted by direct elution with chloroform and methanol and analysed according to Marsh and Weinstein (1966). Lipids concentrations are reported as tripalmitine equivalents. For each biochemical analysis, blanks were performed with the same sediment samples as previously treated in a muffle furnace (450°C, for 2 h). All analyses were carried out in 3–5 replicates. Protein, carbohydrate and lipid concentrations were converted to carbon equivalents by using the following conversion factors: 0.49, 0.40 and 0.75 μg C g^{-1}, respectively. The sum of PTN, CHO and LIP carbon was referred to as bioavailable biopolymeric carbon (BPC) (Fabiano et al., 1995).

2.5 Interpolation Maps

The maps were created with the ArcMap 10.5 software and the Spline with Barriers (SWB) tool configured with cell size 15 and 0 smooth factor. The interpolation shows the spatial of the analyzed variables. The coordinates are provided in Universal Transverse Mercator (UTM), zone 23S in the WGS84 datum

2.6 Principal Components Analysis (PCA)

This analysis was performed with software Pcord5. Before to be submitted to statistical analysis all data were standardized to the square root of 0.5. The PCA was applied to explain the variance of the main physicochemical and sedimentological (grain size and geochemical data) parameters and to describe the characteristics of the groups of stations along the MGLS.

3. Results

3.1 Water Physicochemical Parameters

Salinity (Table 1) varied between 0.1-20.0 (mean 5.1) in the MGLS. The lowest values of water salinity were recorded in Maricá Lagoon (mean of 0.5), varying from 0.1 (in station MC01) to 1.4 (in station MC10). Guarapina Lagoon had the highest mean of salinity (17.2) among the lagoons. This lagoon had the highest salinity values (20), which were recorded in the stations MC20, MC18 and MC21 (Fig. 2; Table 1), located near the lagoon mouth and margins.

The range of temperature (Table 1) was 27.6-33.1 (mean 29.4°C) in the MGLS the mean temperature was for: Maricá Lagoon, 29.9 °C; Barra Lagoon, 29.4 °C; Padre Lagoon, 30.2 °C and; Guarapina Lagoon, 28.4 °C. The station located at the river Ubatiba mouth (MC01) had the highest temperature (33.1 °C). The lowest temperature (27.6 °C) was recorded in the channel connecting the Maricá and Barra lagoons (MC06) (Fig.2).

The highest and lowest mean dissolved oxygen (DO) contents were recorded in Barra Lagoon (9.4 mg/l) and Guarapina Lagoon (4.9 mg/L), respectively (Table 1). However, the maximum (17.1 mg/L) and the minimum (4.4 mg/L) dissolved oxygen values were recorded in the channel connecting the Barra and Padre lagoons (station MC16) and in Barra Lagoon (station MC12), respectively (Fig. 2).

The pH values ranged from 8.2 (station MC12) to 9.5 (station MC01) in MGLS (Table 1; Fig. 2). The redox potential values are negative in all the analyzed stations (Table 1) and varied from -135.7 mV, near the Ubatiba River mouth (station MC01), and -64.2 mV, near of sandbar (station MC12; Fig. 2).

3.2 Grain size analysis

The sediment grain size of most of the stations (52%) was classified as poorly sorted sandy-silt (Fig. 2). Very well sorted sand was found in the stations MC01, MC05, MC06, MC12, MC16, MC17 and MC22, located near the lagoonal margins and in the Ponta Negra Channel. Silty sediments were found at the central region of Maricá Lagoon (MC08) and in the central-northern region of Barra Lagoon (stations MC13, MC14 and MC15) (Fig. 2).

3.3 Total organic carbon (TOC) and Total sulfur (TS)

The mean values of TOC were: 2.2% at Maricá Lagoon; 3.5% at Barra Lagoon; 0.5% at Padre Lagoon; and; 4.0% at Guarapina Lagoon (Table 1). The highest (5.5%) and lowest (0.1%) TOC values were recorded in the station MC19, from Guarapina Lagoon and in the station MC17, from Padre lagoon, respectively (Fig. 3).

The TS values varied between 0.01-2.5% (mean 1.0 %) in MGLS. The highest TS values (2.3-2.5%) were recorded in Barra Lagoon (stations MC11, MC13-15; Fig. 3). The lowest (0.1 %) and highest (1.3 %) mean TS values were recorded in Padre and Guarapina lagoons, respectively (Table 1).

The TOC:TS ratio varied between 1.7 and 20.0, in the stations MC15 (near the northern margin of Barra Lagoon), and MC04 (near the southern margin of Maricá Lagoon), respectively.

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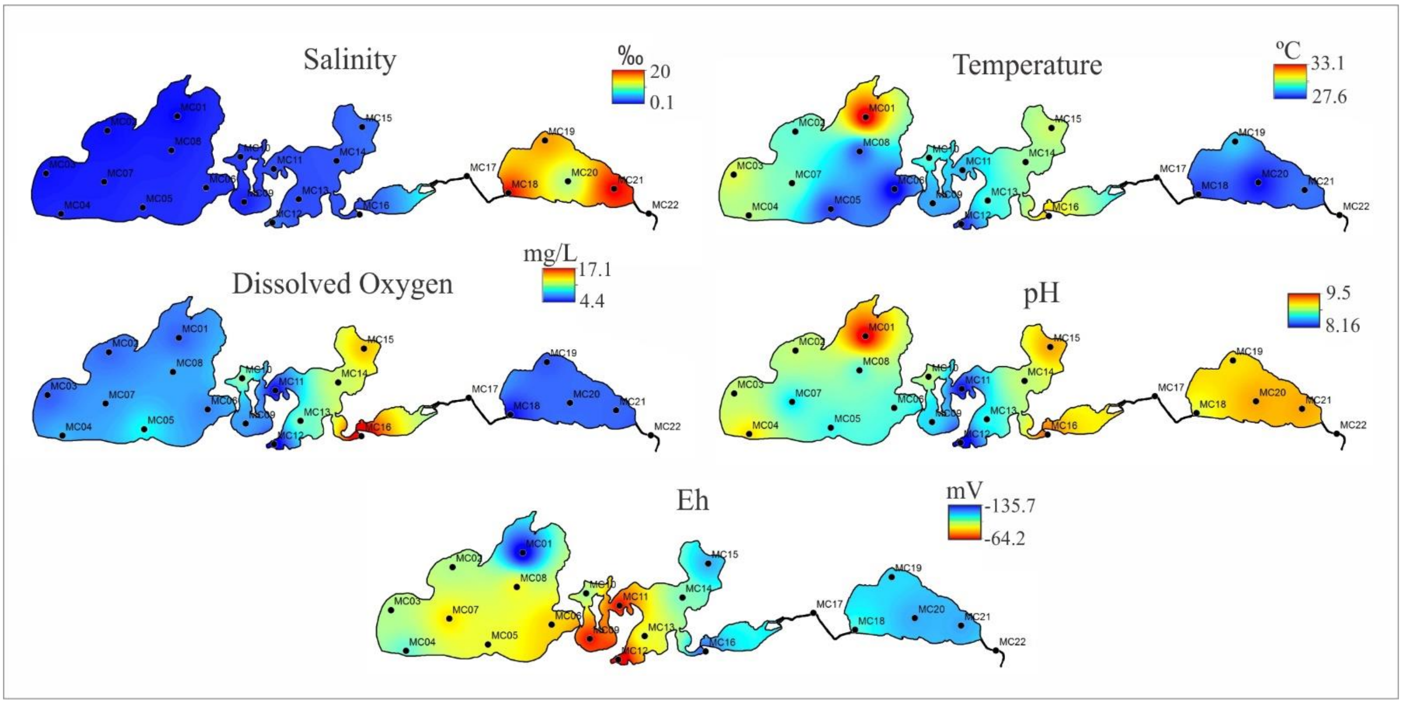

Fig. 2. Interpolation maps of water-sediment interface parameters in MGLS: salinity, dissolved oxygen (mg L^{-1}), temperature (°C), pH and Eh (mV).

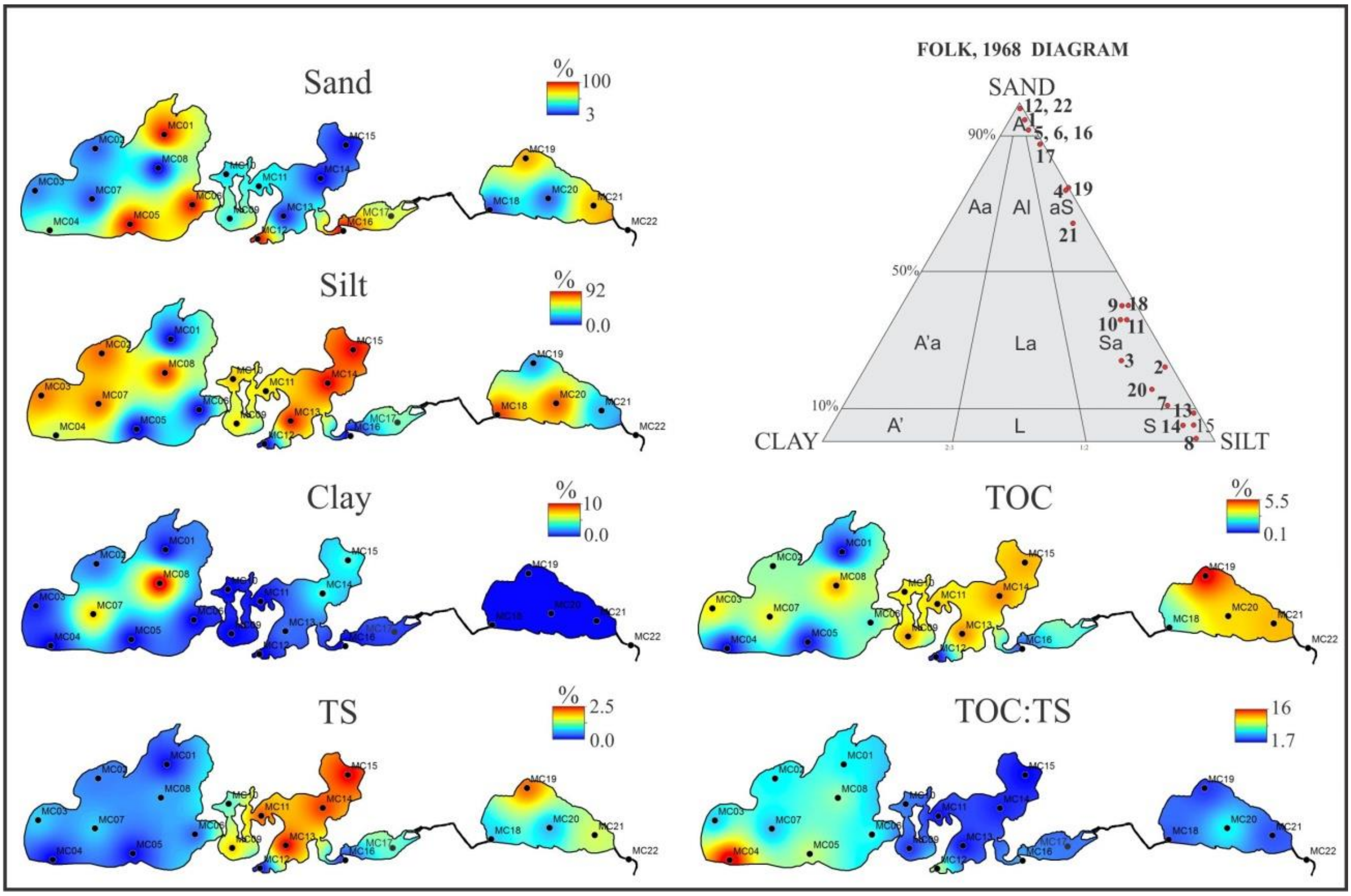

Fig. 3. Interpolation maps of sediment parameters in MGLS: Grain size (sand, silt and clay - %); total organic carbon (TOC - %); total sulfur (TS - %) and; TOC:TS ratio. The Folk (1968) diagram for sedimentological classification is also presented: A - sand, Aa – clayey sand, Al – muddy sand, As – silty sand, A'a – sand clay, La – sandy mud, Sa – sandy silt, A' - clay, L - mud, S - silt.

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4 (2): 159-173. April-June, 2019
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The mean TOC:TS value for each lagoon was: 7.3 in Maricá Lagoon; 3.2 in Barra Lagoon, 4.6 in Padre Lagoon and 3.7 in Guarapina Lagoon (Table 1).

3.4 Biopolymers

The range of total biopolymeric concentrations (BPC) in MGLS was 2.3-42.3 mg C g^{-1} (mean 22.7 mg C g^{-1}). The highest BPC contents (35.3 mg C g^{-1} - 42.3 mg C g^{-1}) were recorded in the stations MC19-MC22 located in Guarapina Lagoon (Fig. 4).

The maximum and minimum protein (PTN) contents in MGLS were recorded in the stations MC19 (4.6 mg C g^{-1}) and MC01 (0.3 mg C g^{-1}), respectively (Fig. 4). Guarapina Lagoon had the highest mean PTN content (3.8 mg C g^{-1}) and Padre Lagoon the lowest one (1.3 mg C g^{-1}; Table 1).

Carbohydrate (CHO) contents varied between 1.5-32.1 mg C g^{-1} (mean 16.3 mg C g^{-1}) in MGLS. The highest CHO contents (27.1- 28.6 mg C g^{-1}) were observed in Guarapina Lagoon (stations MC19-MC22). The highest and lowest mean CHO contents were observed in Guarapina Lagoon (26.3 mg C g^{-1}; Table 1) and Padre Lagoon (8.4 mg C g^{-1}; Table 1), respectively.

Lipid (LIP) contents varied from 0.5 mg C g^{-1} (station MC17) to 7.1 mg C g^{-1} (station MC10; Fig. 4; Table 1). The highest mean LIP content was found in Guarapina Lagoon (4.6 mg C g^{-1}) and Barra Lagoon (4.2 mg C g^{-1}).

The mean value of the PTN:CHO ratio in the MGLS was 0.15 and ranged between 0.14 and 0.17 (Table 1). The station MC17 located in the Padre Lagoon presented the highest value of this ratio (0.18). The lowest PTN:CHO value was recorded in the station MC10 in Barra Lagoon (0.13; Fig.4).

The mean value of the CHO:TOC ratio was higher in Padre Lagoon (17.7) and lower in Barra Lagoon (6.4). The stations with the highest and the lowest CHO:TOC values were MC16-MC17 (16.9-18.5), located in Padre Lagoon, and MC08 (3.5) located in Maricá Lagoon, respectively (Fig. 4).

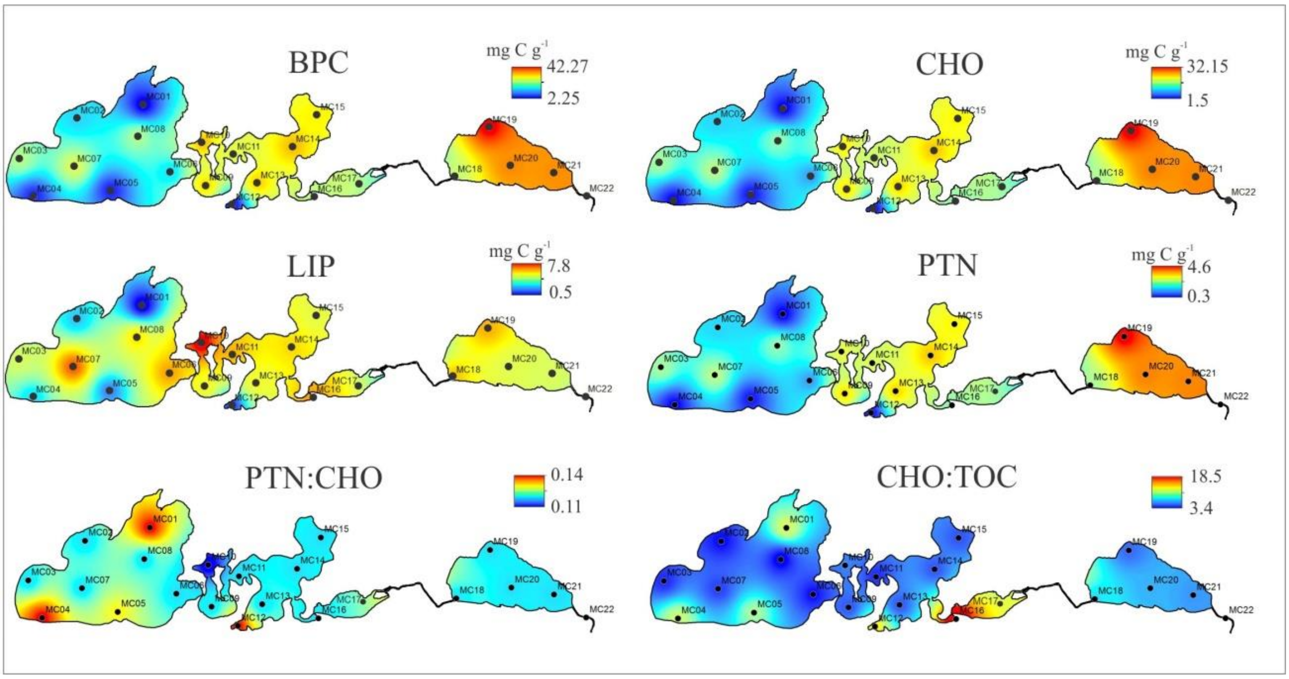

Fig. 4. Distribution maps of biopolymeric carbon (BPC; mg C g^{-1}), carbohydrate (CHO; mg C g^{-1}), lipid (LIP; mg C g^{-1}) and protein (PTN; mg C g^{-1}) concentrations, and PTN/CHO and CHO/TOC ratio values.

3.5 Statistical Analysis

The first axis of the PCA analysis explains 40% of the total variance of the samples dispersion and the second one 20% (Fig. 5). The most significant parameters related to the axis 1 are the biopolymers components (BPC, LIP, CHO, PTN), TOC and TS. Salinity, temperature, oxygen and clay are strongly related to axis 2. The arrows of silt, sand and pH make an angle of 45º in the PCA diagram (Fig. 5). According to environmental parameters distribution, five groups of stations can be considered in the PCA (Fig. 5): Group I (stations MC03, MC07, MC08 and MC13-MC15) with positive correlations with clay, oxygen and temperature; Group II (stations MC09-MC11) with positive correlations with TOC, TS and lipids; Group III (stations MC18-MC22) with positive correlations with salinity; Group IV (stations MC06 and MC16) with positive correlations with sand and pH; Group V (stations MC01, MC04, MC05, MC12 and MC17) with negative correlations with clay, silt, TOC, TS and biopolymers (Fig. 5).

Journal of Sedimentary Environments
Published by Universidade do Estado do Rio de Janeiro
4 (2): 159-173. April-June, 2019
doi: 10.12957/jse.2019.43371

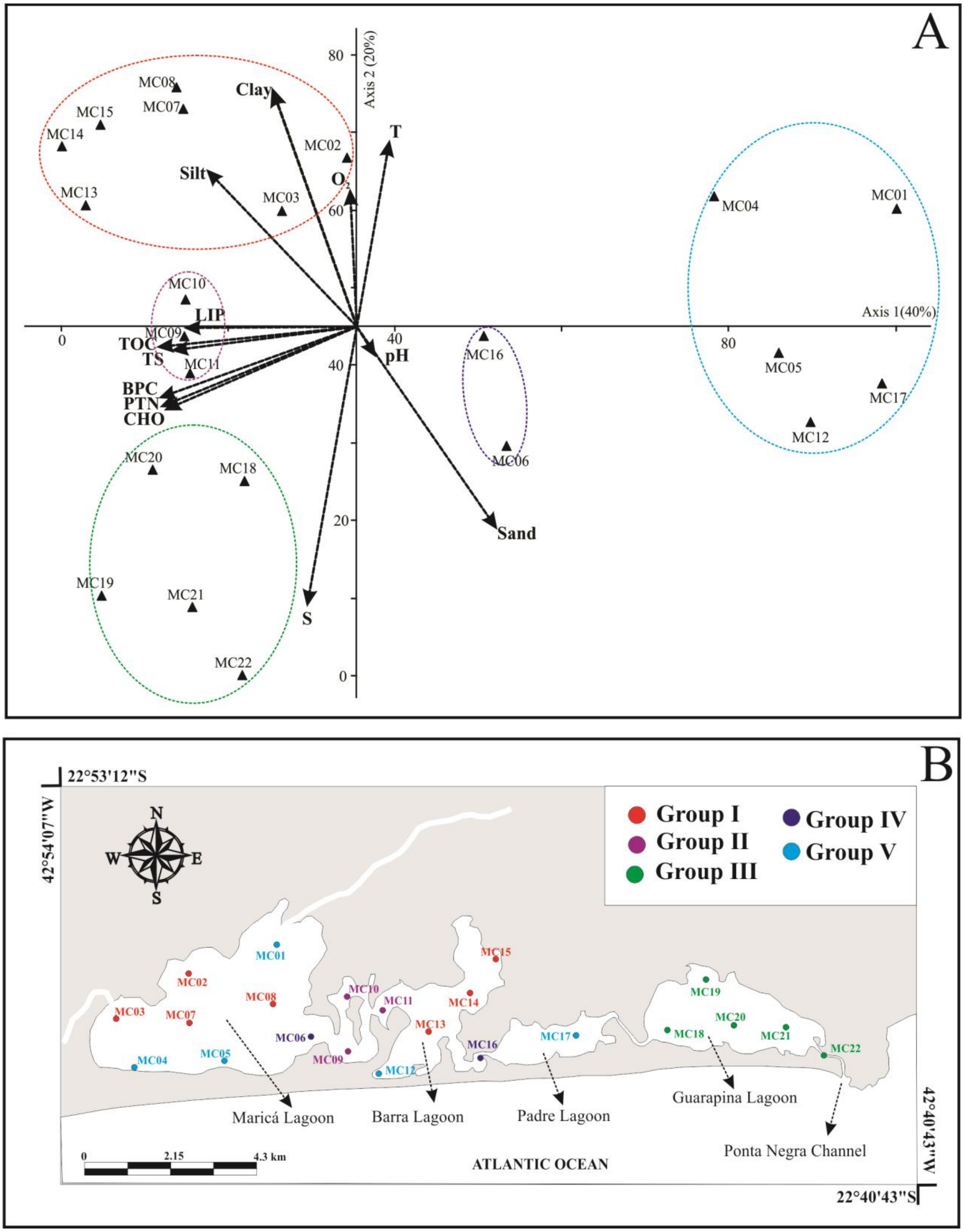

Fig. 5. The results of PCA analysis based on the physical-chemical and sedimentological parameters (grain size and geochemical data. A) PCA plot relating the studied stations and physicochemical parameters B) Distribution map of the groups of stations identified by PCA analysis. Legend: S – salinity; O_2 - dissolved oxygen (mg L^{-1}); T - temperature (°C); TOC – total organic carbon; TS – total sulfur (%); BPC - biopolymeric carbon (mg C g-1); CHO - carbohydrates (mg C g^{-1}); LIP - lipids (mg C g^{-1}) and; PTN - proteins (mg C g^{-1}).

Journal of Sedimentary Environments
Published by Universidade do Estado do Rio de Janeiro
4 (2): 159-173. April-June, 2019
doi: 10.12957/jse.2019.43371

4. Discussion

The recorded physical-chemical parameters vary gradually depending on the proximity to Ponta Negra Channel (Fig. 1), where the MGLS connects the sea, as described by several authors (Kjerfve and Knoppers, 1999; Bomfim et al., 2010; Guerra et al., 2011). The current mean salinity value recorded in Guarapina Lagoon (16.5) falls within the range documented in the literature (7.0-19.0) (Kjerfve and Knoppers, 1999; Guerra et al., 2011). In Barra Lagoon, the mean salinity value recorded in this work (2.3) is also in the range of the previously noticed values of 1.0 (Kjerfve and Knoppers, 1999) and 5.5 (Guerra et al., 2011).

Padre Lagoon presented higher mean salinity (4.9) than that found by Kjerfve and Knoppers (1999) of 3.0. The highest salinities (20.0) were recorded in the Ponta Negra Channel and close to this channel (stations MC22 and MC21, in Guarapina Lagoon) aggreging with the values recorded by Bomfim et al. (2010). These higher salinity values are the result of marine water exchange though the Ponta Negra Channel. The effect of currents movement decreases progressively in the inner region of the MGLS, especially in Maricá Lagoon, due to the presence of a bridge in the connection between this system and Padre Lagoon. This bridge acts as a barrier that reduces the flow and restricts the marine water supply to Maricá Lagoon, which is fed mainly by freshwater from rivers runoff (Cruz, 2010).

According to the Hedgpeth (1951) classification: Guarapina and Padre lagoons are strong mesohaline systems (mean salinity between 10 and 18); Barra Lagoon is a week mesohaline system (mean salinity between 1.8 – 18) and; Maricá Lagoon is a freshwater system (salinity <1.8). However, according to Oliveira et al. (1955), Maricá Lagoon was a polyhaline to mesohaline system until 1951.

The recorded temperature (27.6–33.1 ^{o}C; mean 29.5 ºC), at the sampling time, was within the mean for the region (Mello, 2007). However, in the MGLS, depending on the station, it can be found larger ranges of water temperature. Guerra et al. (2011), for instance, recorded mean temperatures 6°C lower than that found, in this work, in Guarapina Lagoon.

The range of pH (8.6-9.5; mean 8.9) and Eh (-72.5 to -135.7 mV; mean -102.4 mV) values indicate that the environment was alkaline and reducer (Stumm and Morgan, 1996) during the sampling event. The most reducing conditions were found in the station MC01 (-135.7 mV), located near the Ubatiba river mouth (Maricá Lagoon) and, in Padre Lagoon (-123.8 mV and -121.3 mV; stations MC16, MC17, respectively).

The concentration of dissolved oxygen is a very important factor for the maintenance of life in the aquatic environment and is one of the main indicators of water quality (Alabaster and Lloyd, 1982; EPA, 1976, 1986). The dissolved oxygen (DO) values (4.4-17.1 mg l^{-1}; mean 7.7 mg l^{-1}) registered in this work are higher than that previously found by Oliveira et al. (1955) and Guerra et al. (2011). The lowest mean DO values (4.4-5.6 mg l^{-1} were recorded in Barra and Guarapina lagoons differently from that reported in the literature, which mention Maricá Lagoon as having greater oxygen deficiency (Guerra et al., 2011). This variable should be quite variable, varying daily and seasonally depending on the rate of water renewal and the autotrophic and heterotrophic activity of living organisms However, the dissolved oxygen values measured in SLMG indicate that oxygen depletion should occur in some areas, since for warm-water biota, the minimum dissolved oxygen concentration is 5-6 mg l^{-1} (Alabaster and Lloyd, 1982; EPA, 1976, 1986).

The bottom sediments are silty in most of the places. Muddy sediments were found in the central areas of Guarapina and Maricá lagoons and at the northern region of Barra Lagoon. According to Fernex et al. (1992), the sandy sediments are common in shallow zones with a water column of less than 50 cm. But in our results, sandy sediments were found along the lagoonal margins and at the entrance of Guarapina and at Padre lagoon.

The coastal plain morphology of Maricá is characterized by two sand barriers that confine a small coastal plain containing a chain-likes series of isolated swampy and almost dry lagoons. The internal barrier (formed in the Pleistocene), located 5-9 m above the current mean sea level, presents a smooth and undulating relief, while the external barrier (formed in the Holocene) is located 5-7 m above the current mean sea level and, in many places, is leveled as a consequence of sand mining (Silva et al., 2012). Few dunes are still preserved in the Holocene barrier and reach 12 m high (Silva et al., 2014). The mobility of sand dunes can be the factor responsible for the presence of sand in the lagoons margins. Tiririca and Ponta Negra mountains, located near the studied region, are formed by Precambrian granites, gneisses and pegmatites which are cut by Mesozoic mafic dykes (Silva et al., 2014). The weathering of these rocks supplies medium-coarse, quartz-rich sand to this coastal plain, as well as to river mouths (e.g. station MC01). Finer grained sediments (with silt + clay fraction varying from 82.8-97.1 %) were found in the stations: MC07 and MC08, in Maricá Lagoon; MC18 and MC20, near the mangrove and in the central areas of Guarapina Lagoon and; MC13-15, in the central and northern areas of Barra Lagoon.

The surface sediments of MGLS present relatively high TS contents, which are similar to that found in other Brazilian lagoons, such as Itaipu (Laut et al. 2016; Raposo et al., 2018), Saquarema (Dias et al., 2017; Belart et al., 2018) and Vermelha lagoons (Laut et al., 2017). However, the recorded values were lower than in other polluted costal environments, such as Santos Estuary (Siqueira et. al., 2006) and Guanabara Bay (Clemente et al., 2015).

The organic enrichment at the study area was evaluated by TOC and biopolymers content. According to Peters and

Journal of Sedimentary Environments
Published by Universidade do Estado do Rio de Janeiro
4 (2): 159-173. April-June, 2019
doi: 10.12957/jse.2019.43371

Cassa (1994), TOC contents can be considered poor (<0.5%) only in a few stations (MC01, MC04, MC05, MC12 and MC17) and moderate (0.5 – 1.0 %) in station MC16. At the majority of stations relatively high TOC contents were found (2.2 - 5.5 %). The highest mean TOC value (4.0 %) was recorded in Guarapina Lagoon, despite having the communication channel to the sea. Relatively high mean TOC values were found in Barra and Maricá lagoons (3.5 % and 2.2 %, respectively), agreeing with data documented by Bruno (2013).

Comparatively, the TOC contents found in the sediments of the central area of Maricá Lagoon, central-north zone of Barra and Guarapina Lagoon were higher to that recognized in in many impacted sites elsewhere in the world. For instance, Aston and Hewitt (1977) observed sedimentary TOC concentrations ranging from 0.07% to 1.97% in Walton Backwater (Essex, England), in an impacted region due to the presence of commercial ports, drainage from agricultural land and domestic sewage disposal. In the Gulf of Izmir (Easter Aegean Sea), an intensely industrialized area, Bergin et al. (2006) measured TOC contents ranging from 0.40% to 3.12%. Carreira and Wagener (1998), found TOC concentrations ranging from 0.79% to 3.18% in an area located near the Ipanema submarine outfall (Rio de Janeiro, Brazil), which discharges on mean 8 m^3s^{-1} of untreated sewage derived from a population of ≈2 x 10^6 inhabitants (Carreira and Wagener, 1998). Cesar et al. (2007) found levels of TOC from 0.85% to 3.75% in the Santos and São Vicente estuarine systems (Brazil), a region that comprises a densely urbanized area, the biggest Brazilian industrial complex, predominantly with the petrochemical, steel, and fertilizer industries, and also the major Latin American port, Santos Port. Abessa et al. (2005) recorded 0.09–2.91% of TOC, close to the Santos submarine outfall diffusers, unloading daily 0.6–1.6 m^3s^{-1} of untreated sewage into Santos Bay (Brazil). Teodoro et al. (2010) estimated sedimentary TOC values between 0.15% to 2.27% at the influence area of the domestic sewage outfall of São Sebastian Channel in São Paulo State, Brazil. According to these authors, the high TOC values were associated with high organic matter flux and low oxygen content.

Nevertheless, the Guarapina Lagoon presented in the majority of the stations TOC values similar to that found in the most polluted regions of Guanabara Bay. Vilela et al. (2003) recorded TOC values ranging from 3.27% to 4.81% next to an oil refinery located in the most confined area of Guanabara Bay. There are no significant industrial activities in Guarapina Lagoon drainage basin, thus all the sewage flow is from domestic origin in this ecosystem.

Kjerfve and Knoppers (1999) estimated that in this lagoon the tidal range does not exceed 3 cm and the lagoon circulation is mostly driven by the wind and river flow. According to Cruz (2010), the water renewal time in Guarapina Lagoon is of ≈15 days. This relatively long time of water renewal difficult the transport of organic material and sediment out of the lagoon, facilitating its accumulation into this lagoon.

According to Mendonça Filho et al. (2003), the sedimentary TOC contents between 2.5% and 5.9% should be associated with high organic matter accumulation rate and can develop subsurface dysoxic-anoxic environments. High supply of organic matter and its breakdown can lead to dissolved oxygen depletion that ultimately might affect the proliferation and the viability of marine organisms (Borja et al., 2012). The TOC:TS ratio is used as an indicator for evaluating the oxi-reduction conditions in sediments (Morse and Berner, 2000). Ratios >3 indicate oxidizing conditions and <3 indicate reducing environments (Stein, 1991; Borrego et al., 1998). In the majority of the stations, TOC:TS indicate the presence of sediments with oxidizing conditions, except the stations located in Barra Lagoon (MC09, MC11, MC13-MC15) and in inner region of Guarapina Lagoon (MC19), where the sediments exhibited more reducing conditions. Even though, the surface sediment of MGLS is, in general, oxygenated as suggested our data, but they became low oxic or anoxic some millimeters below the sediment-water interface as indicated the subsurface sediment dark gray or black colors. As referred by Martins et al. (2016 a, b), in Bizerte Lagoon (Tunisia), shallow and well-lit waters can allow the development of benthic microalgae on the bottom. This algae mat allows the oxygenation of the surface sediment but the high accumulation of organic matter in fine-grained substrates can lead to oxygen depletion some millimeters below the interface water sediments.

The biopolymer compounds in marine and coastal environments can be used for the characterization and interpretation of the sedimentary organic matter origin (Silva et al., 2011). This methodology was used in some Brazilian transitional environments (e.g. Silva et al., 2011; Clemente et al., 2015; Laut et al., 2016, 2017; Dias et al., 2017; Raposo et al., 2018).

Carbohydrate contents (Maricá Lagoon, 9.9 mg C g^{-1}; Barra Lagoon, 17.9 mg C g^{-1}; Padre Lagoon, 8.4 mg C g^{-1} and; Guarapina Lagoon, 26.3 mg) reached higher values in Guarapina and Barra lagoons. Considering that the CHO are more related to phytoplanktonic and vegetal detrital sources (Cotano and Villate, 2006), this contribution should be more important in these both lagoons.

Protein contents also reached higher values in Guarapina and Barra lagoons (Maricá Lagoon, 1.6 mg C g^{-1}; Barra Lagoon, 2.6 mg C g^{-1}; Padre Lagoon, 1.3 mg C g^{-1} and; Guarapina Lagoon, 3.8 mg C g^{-1}). According to Dell'Anno et al. (2002) high protein concentrations are in general related to primary productivity. So, it should be higher in Guarapina Lagoon and Barra Lagoon than the other lagoons of the MGLS.

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Published by Universidade do Estado do Rio de Janeiro
4 (2): 159-173. April-June, 2019
doi: 10.12957/jse.2019.43371

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The range of PTN contents in MGLS is similar to that recorded, for instance, in Saquarema Lagoonal System (SLS), but are slightly lower than that recorded in Bizerte Lagoon (Tunisia) and Aveiro Lagoon (Portugal) and are higher than that found in Vermelha and Itaipu lagoons (Brazil). In coastal areas, the significant increase of PTN also may be associated with anthropogenic contributions of organic matter (Cotano and Villate, 2006).

Guarapina and Barra lagoons also had higher LIP concentrations (4.6 mg C g^{-1} and 4.2 mg C g^{-1}, respectively) than the other lagoons (Maricá, 3.4 mg C g^{-1}; Padre 3.1 mg C g^{-1}). The mean value of lipid contents in MGLS is similar to that recorded in other Brazilian lagoons of Rio de Janeiro State, such as Urussanga, Jardim and Boqueirão, which make part of the Saquarema Lagoonal System (SLS), Vermelha Lagoon and Itaipu Lagoon (Brazil), as well as in Bizerte Lagoon (Tunisia), but are higher than that found in Saquarema (SLS; Brazil) and Aveiro Lagoon (Portugal) (Table 2). The higher LIP values found in the MGLS should be caused by anthropogenic sources of organic matter (Cotano and Villate, 2006) which tend to accumulate in confined areas of that lagoons.

Tab. 2. Comparison of biopolymer contents in several lagoons. (1) Maricá-Guarapina Lagoon System (MGLS; Brazil), data of this work. (2) Saquarema Lagoonal System (SLS; Brazil), data reported by Dias et al. (2017) and Belart et al. (2018). (3) Vermelha Lagoon (Brazil) data reported by Laut et al. (2017). (4) Itaipu Lagoon (Brazil) data reported by Laut et al. (2016). (5) Bizerte Lagoon (Tunisia) data reported by Martins et al. (2016 b). (6) Aveiro Lagoon (Portugal), data reported by Martins et al. (2015).

Biopolymer Contents in Several Lagoons	PTN	CHO	LIP	BPC
	mg C g^{-1}	mg C g^{-1}	mg C g^{-1}	mg C g^{-1}
Maricá-Guarapina Lagoon System (MGLS; Brazil) (1)	0.3-4.6 (*2.4)	1.5-32.1 (*16.3)	0.5-7.1 (*4.1)	2.3-42.3 (*22.7)
Maricá (MGLS; Brazil)* (1)	1.5	9.9	3.4	14.8
Barra (MGLS; Brazil)* (1)	2.6	17.9	4.2	24.8
Padre (MGLS; Brazil)* (1)	1.3	8.4	3.1	12.7
Guarapina (MGLS; Brazil)* (1)	3.8	26.3	4.6	34.7
Saquarema Lagoonal System (SLS; Brazil) (2)	0.4-4.6 (*2.6)	1.1-25.9 (*16.5)	0.2-3.7 (*2.7)	1.7-31.4 (*21.8)
Urussanga (SLS; Brazil)*	2.9	15.9	4.5	23.2
Jardim (SLS; Brazil)*	3.2	19.2	5.5	27.8
Boqueirão (SLS; Brazil)*	2.6	21.7	4.5	28.8
Saquarema (SLS; Brazil)*	2.1	14.4	3.0	19.6
Vermelha Lagoon (Brazil) (3)	0.3-2.1 (*1.0)	0.2-2.5 (*1.6)	0.3-2.5 (*1.6)	0.5-3.8 (*2.0)
Itaipu Lagoon (Brazil) (4)	0.3-2.1 (*1.0)	0.2-2.1 (*0.9)	0.3-2.5 (*1.6)	0.5-3.8 (*2.0)
Bizerte Lagoon (Tunisia) (5)	2.5-3.1 (*2.8)	3.0-4.4 (*3.6)	3.5-5.3 (*4.0)	9.3-11.9 (*10.5)
Aveiro Lagoon (Portugal) (6)	0.2-5.6 (*3.0)	0.0-3.2 (*1.2)	0.1-3.6 (*1.2)	0.3-10.5 (*5.5)

The proteins versus carbohydrates (PTN:CHO) ratio have been used to indicate the organic matter age present in the sediment (Hobson, 1967; Cauwet, 1978). Danovaro et al. (1993) suggested that PTN:CHO >1 should be linked to the presence of fresh organic matter, since protein degradation is faster than the others biopolymers (Newell and Field, 1983) and for this reason only 'fresh compounds" have high values of this ratio (Cotano and Villate, 2006). In Maricá-Guarapina Lagoon System all the lagoon had PTN:CHO<1, which indicate the presence of predominantly aged organic matter.

According to Fabiano and Danovaro (1994), the composition of organic matter in labile biopolymers in coastal areas, is frequently characterized by small amounts of lipids and large quantities of PTN, frequently exceeding CHO concentrations. This trend is not observed in the present study, in which the CHO>LIP>PTN. On the other hand, in the Maricá-Guarapina Lagoon System the CHO:TOC are <20 in all the analyzed the stations (3.5-18.5; mean 7.5). As noticed by Paez-Osuna et al. (1998), CHO:TOC<20 indicate organic material of natural origin and CHO:TOC>20 indicate sewage (anthropogenic) inputs.

So, the biopolymeric analysis indicated that the quality of organic matter in most areas of Maricá-Guarapina Lagoon System results principally from autochthonous biological productivity. The same was observed in other costal lagoons of Rio de Janeiro State, such as, Saquarema Lagonal System (Belart et al., 2018) and Vermelha Lagoon (Laut et al., 2017). However, relatively high CHO:TOC values were found in MC16 and MC17, probably due to the presence of a sewage duct at Padre Lagoon, near these stations.

Based on the PCA results, it was possible to consider 5 groups of stations in MGLS related to different sedimentary environmental conditions (Fig. 5):

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Published by Universidade do Estado do Rio de Janeiro
4 (2): 159-173. April-June, 2019
doi: 10.12957/jse.2019.43371

- Group 1: stations MC02, MC03, MC07 and MC08, located in Maricá Lagoon, and stations MC13-MC15 situated in Barra Lagoon, are influenced by higher silt and clay contents, suggesting the prevalence of low bottom hydrodynamic conditions. These stations also have high TOC sedimentary contents and quite negative Eh values in bottom water, which suggests a probable tendency for the establishment of eutrophic conditions (Pelletier et al., 2011).
- Group 2: stations MC09, MC10 and MC11, located in confined channels between Maricá Lagoon and Barra Lagoon, are mostly correlated to higher BPC contents, namely PTN, CHO and LIP, as well as TOC and TS. These stations have similar characteristics to that described for the previous group (with fine-grained substrate, high TOC sedimentary contents and negative Eh values in bottom water). In addition, they have higher amount of labile organic matter, which is preferably used by aerobic organisms (Wu et al., 2018).
- Group 3: stations MC18-MC22, located in the Guarapina Lagoon and Ponta Negra Channel, were positively correlated to salinity and negatively to high temperatures. These stations are located in the region under higher marine influence and displayed the highest mean CHO content, probably due to the contributions of autochthonous primary productivity and debris of the mangrove fringe (at the margins of this lagoon), since according to Cotano and Villate (2006), high CHO contents are commonly associated with organic matter of phytoplanktonic origin and vegetal detritus.
- Group 4: stations MC06 and MC16, are positively correlated to sand contents; these stations did not have large concentrations of biopolymers nor the lowest values, which differentiate this group from the stations of group 5.
- Group 5: stations MC01, MC04, MC05, MC12 and MC17, were characterized by lower BPC values (PTN, CHO and LIP), TOC and TS. These stations also were positively correlated to the sand contents and negatively to clay and silt contents. Therefore, they can be considered the least affected by the organic matter enrichment, because they receive large terrigenous contribution from the Ubatiba river outflow (MC01) and the littoral dunes (MC04, MC05, MC12), which dilute the organic component of the sediments, or are under local active currents (MC17), which avoid the deposition of fine grained sediments and organic materials.

It can be deduced that the tendency to undergo more pronounced eutrophication are bottom with fine-grained sediments, displaying relatively high TOC and BPC contents, which were found in the stations of Groups 1, 2 and 3 of the PCA (Fig. 5), located in Guarapina, Barra and Maricá lagoons.

Conclusion

The PCA results showed that the bottom environment of MGLS is heterogenous displaying: areas with lower and stronger hydrodynamic conditions and marine influence; more or less impacted zones by organic matter and displaying different concentrations of labile biopolymeric compounds (proteins, carbohydrates and lipids).

The surface sediments of Maricá-Guarapina Lagoon System receive organic matter of mixed sources. However, the main source of organic matter should be the autochthonous productivity (phytoplanktonic and vegetal detritus from the mangrove fringe). The anthropogenic contribution of organic matter was more evident in Padre Lagoon, with relatively low TOC contents (0.1-0.8%). It has been deduced that the sediments are accumulating mainly aged organic matter.

Guarapina, Barra and Maricá are impacted lagoons by organic matter mostly in the zones with fine-grained sediments, displaying relatively high TOC and BPC contents, which should evolve into an ever-increasing stage of eutrophication.

Acknowledgment
This research was supported by National Council of Technological and Scientific Development - CNPq (Universal 445830/2014-0), Fundação de Amparo à Pesquisa do Estado do Rio de Janeiro FAPERJ (Biota RJ E26/11.399/2012). The authors would like to thank CAPES for the PhD fellowship granted to Pierre Belart (Finance Code 001) and for the postdoctoral fellowship to Iara Clemente.

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Journal of Sedimentary Environments
Published by Universidade do Estado do Rio de Janeiro
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