



C and O Isotope Chemostratigraphy and Bulk Chemistry of Reef Limestones of the Tambaba Formation, Paraíba Basin, Northeastern Brazil

Químioestratigrafia Isotópica (C e O) e Geoquímica de Calcários Recifais da Formação Tambaba, Bacia Paraíba, Nordeste do Brasil

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Abstract

This work focuses on the behavior of the C and O isotopes and major and trace element chemistry of the carbonate rocks of the Tambaba Formation, Paraíba Basin, Northeastern Brazil. Thirteen carbonate samples collected from three vertical profiles located perpendicular to the bedding along the coast of the state of Paraíba were analyzed. The $\delta^{13}\text{C}$ values ranged from 1.6 to 2.8‰ VPDB while the $\delta^{18}\text{O}$ values ranged from -1.2 to 1.8‰ VPDB, thus suggest a restricted shallow-platform depositional environment. X-ray fluorescence analyses suggested diagenetic changes, such as the replacement of dolomite by calcite (Mn/Sr ratios from 0.6 to 28) and dolomitization, which was identified by high Mg/Ca ratios (0.5 to 0.6). The very low SiO_2 and Al_2O_3 content attested to the low terrigenous material influx. The carbon isotope values suggest that the carbonate rocks were deposited after the Paleocene-Eocene Thermal Maximum (PETM) event.

Keywords: Paraíba Basin; Tambaba Formation; isotope analysis

Resumo

Este trabalho discute o comportamento de isótopos de C e O e a química de elementos maiores e traços de rochas carbonáticas da Formação Tambaba, Bacia Paraíba, Nordeste do Brasil. Amostras de treze rochas carbonáticas de três perfis perpendiculares a estratos, localizados na costa do estado da Paraíba, foram analisadas. Os valores de $\delta^{13}\text{C}$ variam de 1.6 a 2.8‰ e os de $\delta^{18}\text{O}$, de -1.2 a 1.8‰ VPDB, sugerindo deposição em ambiente de plataforma rasa restrito. Análises por fluorescência de raios-X sugeriram alterações diagenéticas, como substituição da dolomita por calcita (Mn/Sr varia de 0.6 a 28) e dolomitização sugerido pela alta razão Mg/Ca (0.5 a 0.6). Além disso, os baixos teores de SiO_2 e Al_2O_3 corroboram o baixo influxo de materiais terrígenos. Os valores de isótopos de carbono sugerem que as rochas carbonáticas foram depositadas após o evento de máxima temperatura no Paleoceno-Eoceno.

Palavras-chave: Bacia Paraíba; Formação Tambaba; análise isotópica

1 Introduction

The Tambaba Formation is a lithostratigraphic unit of the Eocene of the Paraíba Basin. Such unit is represented by carbonate rocks deposited in a reef marine environment, and they generally present a light yellow color, bioclastic texture, and beds, with the reef constructions presenting an irregular weathered aspect due to erosion (Veras, 2017).

For a long time, this unit was considered the upper part of the Maria Farinha Formation. However, it presents specific faciological, stratigraphic and petrographic features that differ considerably from the bedding described for the typical section of the Maria Farinha Formation, which was established in the southern portion of the coastal strip of the Paraíba Basin. The Tambaba Formation occurs in the Paraíba Basin and the Alhandra and Miriri Sub-Basins, and it is mappable at 1:25.000 scale.

The age proposed by Almeida (2000) for the reef limestones of the Tambaba Formation based on the fossil content differs from that proposed for the Maria Farinha Formation, which is considered Danian (Lower Paleocene) in the Olinda Sub-Basin. Compared with the Maria Farinha Formation, which has been a target of several micropaleontological studies (e.g., Albertão, 1993; Albertão & Martins Jr., 1996; Stinnesbeck & Keller, 1996; Fauth, 2000; Gertsch et al., 2013), the ages of these reef limestones have not yet been established.

A number of chemostratigraphic studies have been performed in the Paraíba Basin due to the Cretaceous-Paleogene boundary (Sial et al., 1992; Barbosa et al., 2005; Barbosa, 2007; Veras et al., 2018). The Cretaceous-Paleogene boundary (KPB) mass extinction (~66.02 Ma) and the Paleocene-Eocene Thermal Maximum (PETM) (~55.8 Ma) are two remarkable climatic and faunal events in Earth's history that have implications for the current Anthropocene global warming and rapid diversity loss (Keller et al., 2018).

In the present work, the behavior of $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ chemostratigraphic curves and the chemistry of major and trace elements obtained for the Tambaba Formation are evaluated and compared with

chemostratigraphic curves obtained in the Global Stratotype Section and Point (GSSP) section for the PETM transition in Dababiya (55.8 Ma), Egypt (Keller et al., 2018).

2 Location

The studied region is located to the south of the capital of Paraíba State, João Pessoa, in the city of Conde. The outcrops are located on the coast of Paraíba over an extension of approximately 10 km between the beaches of Jacumã and Tambaba (Figure 1). Using Recife (Pernambuco) as a reference, and the main access road is BR-101 towards João Pessoa, the northern end of the studied range can be accessed via PB-018 (approximately 113 km), and the southern end can be accessed via PB-044 (approximately 100 km).

3 Materials and Methods

3.1 Fieldwork

The fieldwork was conducted along the coastal strip of the state of Paraíba between the beaches of Tambaba and Jacumã, and three outcrops located in the municipality of Conde-PB were visited. One outcrop is located on Tambaba beach, one on Coqueirinho beach and one on Jacumã beach, and they were named DT-01, DC-01 and DJ-01, respectively. Outcrops were sampled through a vertical profile perpendicular to the strata observed at each outcrop, where a sample of each strata was taken following a bottom-up sequence (e.g., DJ-01A, DJ-01B, DJ-01C, DJ-01D – Figure 2).

UTM – DATUM WGS 1984		
OUTCROP	N (UTM)	E (UTM)
DT-01	9.185.755	301.400
DC-01	9.189.314	301.843
DJ-01	9.195.399	301.306

Table 1 Nomenclature of the outcrops and their respective UTM coordinates.

3.2 Laboratory Analysis

A total of 13 samples were collected along the coast of Paraíba at the Tambaba-Coqueirinho-Jacumã stretch, which is approximately 10 km

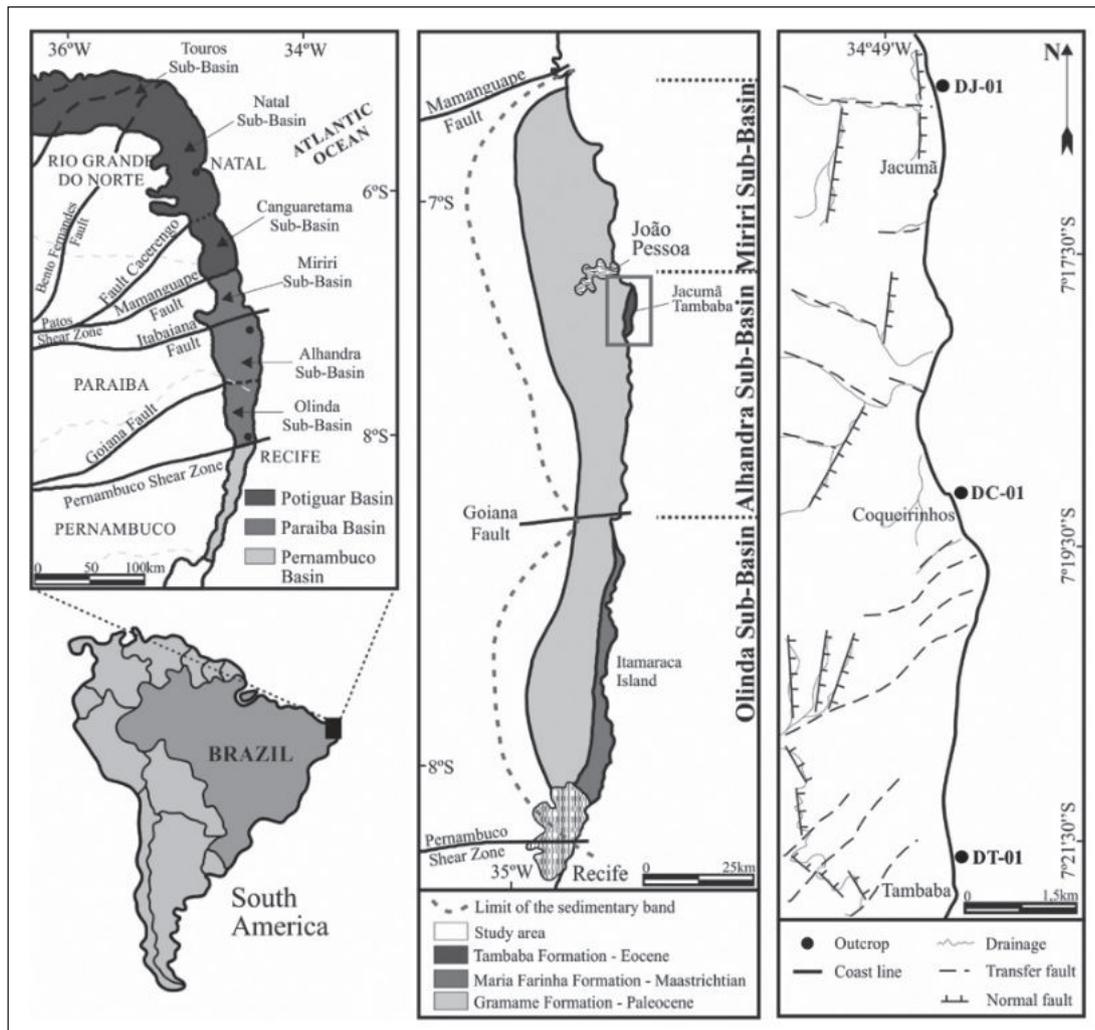


Figure 1 Location map of the occurrence area of the Tambaba Formation in the Paraíba Basin (modified from Távora *et al.*, 2017).



Figure 2 Example of the numbering of the samples collected in the field at reef limestone outcrops (outcrop DJ-01, in Jacumã). The dotted yellow lines show the boundary of the beds.

long, for a stable carbon ($\delta^{13}\text{C}$) and oxygen ($\delta^{18}\text{O}$) isotope analyses and chemical analyses of major and trace elements. These analyses were carried out at the Nucleus of Geochemical Studies-Stable Isotope Laboratory (NEG-LABISE) of the Department of Geology of the Federal University of Pernambuco.

The samples were powdered to analysis. For the stable isotope analyses, 20 mg of whole rock were used, and these samples were subjected to CO_2 gas extraction in a conventional high vacuum extraction line after reacting with 100% orthophosphoric acid at 25°C for 1 day (3 days when dolomite is present).

The liberated CO_2 gas was cryogenically purified and analyzed in a VG Micromass or Delta V (ThermoFinnigan Advantage) double-mass spectrometer, and a SIRA II triple collector (using the BSC reference gas (Borborema skarn calcite) was used to determine the isotopic ratios. The results are expressed in the notation of a per mil (‰) relative to a standard VPDB (Vienna PeeDee Belemnite). The uncertainties of the isotopic measurements are better than 0.1‰ for carbon and 0.2‰ for oxygen, based on multiple analyses of an internal LABISE standard.

Prior to the geochemical analysis, a portion (2,25g) of each sample was placed in an oven to dry at 110°C, and then it was placed in a muffle furnace at 1000°C for 2 h to determine the fire loss. Molten beads were generated using lithium tetraborate as the flux. The pearls were analyzed in a Rigaku X-ray Fluorescence Spectrometer model ZSX Primus II equipped with a Rh tube, and seven crystal analyses were performed by the calibration curve method, which was based on international reference materials. The results are displayed based on weight % (major elements) or ppm (trace elements).

4 Geology

The coastal strip of the Paraíba Basin is located between the Pernambuco Shear Zone and the Mamanguape Structural High, which is related to a branch of the Patos Shear Zone in the eastern portion of Northeast Brazil. The geological knowledge,

onshore and offshore, of this area is still deficient compared with the neighboring basins, such as the Potiguar Basin or the Alagoas Basin.

Deposits of reef-lagoon origin occur in the Alhandra and Miriri sub-basins within the sedimentary column of the Paraíba Basin between the Campanian-Danian age carbonate sequence (Itamaracá, Gramame and Maria Farinha formations) and the Miocene continental siliciclastic sequence (Barreiras Formation and Quaternary sediments) (Mabessone & Alheiros 1988, Barbosa 2004). Although few studies have evaluated the occurrence of these deposits, Almeida (2000, 2007) conducted a more systematic study of these deposits, which were named by Beurlen (1967a) as Upper Maria Farinha Formation. This last author characterized the mollusks present in these deposits and their paleoecology in addition to their important ichnofossiliferous content and suggested that these limestones were possible of Eocene age. However, an erosional/depositional gap of at least 7 million years may have occurred between the top of the Maria Farinha Formation and the reef limestones.

These deposits were always cited as belonging to the Maria Farinha Formation as suggested by Beurlen (1967a), who correlated them with Paleocene deposits that occur on the top of the Gramame Formation in the southern region of the coastal strip. However, this author observed a difference between the faciology of the classically named limestone of Maria Farinha and the deposits that occur in the northern portion of the coastal strip and referred to the latter as the “Maria Farinha Superior” Formation.

Taking into account the lithofaciological, paleontological, biochronological and depositional aspects, Correia Filho *et al.* (2015) proposed the individualization of these carbonate deposits previously named the Upper Maria Farinha Formation by Beurlen (1967a) as the Tambaba Formation. These deposits surface in the Alhandra and Miriri sub-basins north of the Paraíba Basin along a beach strip between the Tambaba and Jacumã beaches (Correia Filho *et al.*, 2015; Veras, 2017). Petrographic studies by Correia Filho *et al.* (2015), Távora *et al.* (2017), and Veras (2017) highlighted the carbonate

microfacies and diagenetic changes that occurred in these deposits.

The outcrops of these reef limestones present a rounded general morphology (“egg box pattern”), are locally layered, and exhibit a bioclastic texture and eroded appearance, with the latter due to erosion (Veras, 2017). These outcrops are between 10 and 0 m above sea level, and they vary in grain size between calcilutite and calcirudite and have molds of rhodolites, shells of bivalves and tubes of boring and burrowing organisms (Correia Filho *et al.*, 2015; Távora *et al.*, (2017), and Veras, 2017) (Figure 3).

Correia Filho *et al.* (2015), Távora *et al.* (2017), and Veras (2017) also studied these rocks across petrographic and catodoluminescence analyses, and observed the presence of a micritic matrix, well-formed dolomite crystals, bioclasts, framboidal pyrite, fine-grain quartz and low concentration feldspars.

In addition, the effect of dolomitization of the carbonate matrix was observed, as well as of dedolomitization reflecting evidence of diagenetic events (mineral replacement). The contribution of detrital siliciclastic components is very small, mainly represented by fine-grains quartz and feldspar, which increase the evidence of low input from the continent (Correia Filho *et al.*, 2015).

5 Results

The results obtained for all samples (Table 2) range from 1.60‰ to 2.75‰ (VPDB) for $\delta^{13}\text{C}$ and -1.18‰ to 1.76‰ (VPDB) for $\delta^{18}\text{O}$. According to Figure 4, the values obtained for the Tambaba samples vary between 1.60‰ and 2.61‰ (VPDB) for $\delta^{13}\text{C}$ and 0.66‰ and 1.76‰ (VPDB) for $\delta^{18}\text{O}$. The isotopic curves show both isotopes vary with the same trend up the penultimate point analyzed (arrows in yellow), which occur a negative curve isotope excursion. After this excursion, the last sample value (DT-01F = 1.60‰) is similar to initial profile value (DT-01A = 1.62‰).

The values obtained for the Coqueirinho samples vary between 2.05‰ and 2.53‰ (VPDB) for $\delta^{13}\text{C}$ and 0.28‰ and 0.60‰ (VPDB) for $\delta^{18}\text{O}$ (Fig-

ure 5), and do not show great variations. The curves obtained through the Jacumã outcrop do not show any abrupt variations and maintain an isotopic pattern. Their values vary between 2.32‰ and 2.75‰ (VPDB) for $\delta^{13}\text{C}$ and -1.18‰ and -0.01‰ (VPDB) for $\delta^{18}\text{O}$.

The Mn/Sr ratio is in a range from 0.59 to 27.19, thus reflecting the high predominance of the manganese concentration relative to strontium, which can reach up to 27-times higher. The Mn/Sr ratio shows strong variations with high manganese concentrations in DT-01D (red arrow – Figure 4). Compared with the Tambaba samples, the Mn/Sr ratio found in Coqueirinho reflects considerable variation but expresses a lower manganese content (DC-01A = 2.50; DC-01B = 0.86; DC-01C = 3.36). In the Jacumã section, the Mn/Sr ratio varies between 0.59 and 3.92.

The Mg/Ca ratio has a certain proportionality, where the calcium concentration for all the samples is almost twice as high as the magnesium concentration ($0.52 < \text{Mg/Ca} < 0.62$). In the Tambaba section, the Mg/Ca ratio values are within a small range without much variation, from 0.55 to 0.61. The values of the Mg/Ca ratio from Coqueirinho are between 0.53 and 0.62, while in the Jacumã section the Mg/Ca ratio varies between 0.52 and 0.59.

Despite presenting not very significant values, the Al_2O_3 values are in a range between 0.00% and 0.52%. The values of the ranges of Al_2O_3 concentrations from Tambaba, Coqueirinho and Jacumã sections are, respectively, 0.00% to 0.24%, 0.13 to 0.41, 0.21% to 0.52%.

The Figures 4, 5 and 6 show the chemostratigraphic composite logs generated from the stratigraphic sections described in the field.

6 Discussion

Attention is required when using stable C and O isotopes in carbonate chemostratigraphy for paleoenvironmental studies. Diagenetic processes may change the isotopic composition of carbonates (Hoefs, 2018). Takaki and Rodrigues (1984) assert that during diagenesis the isotopic composition of

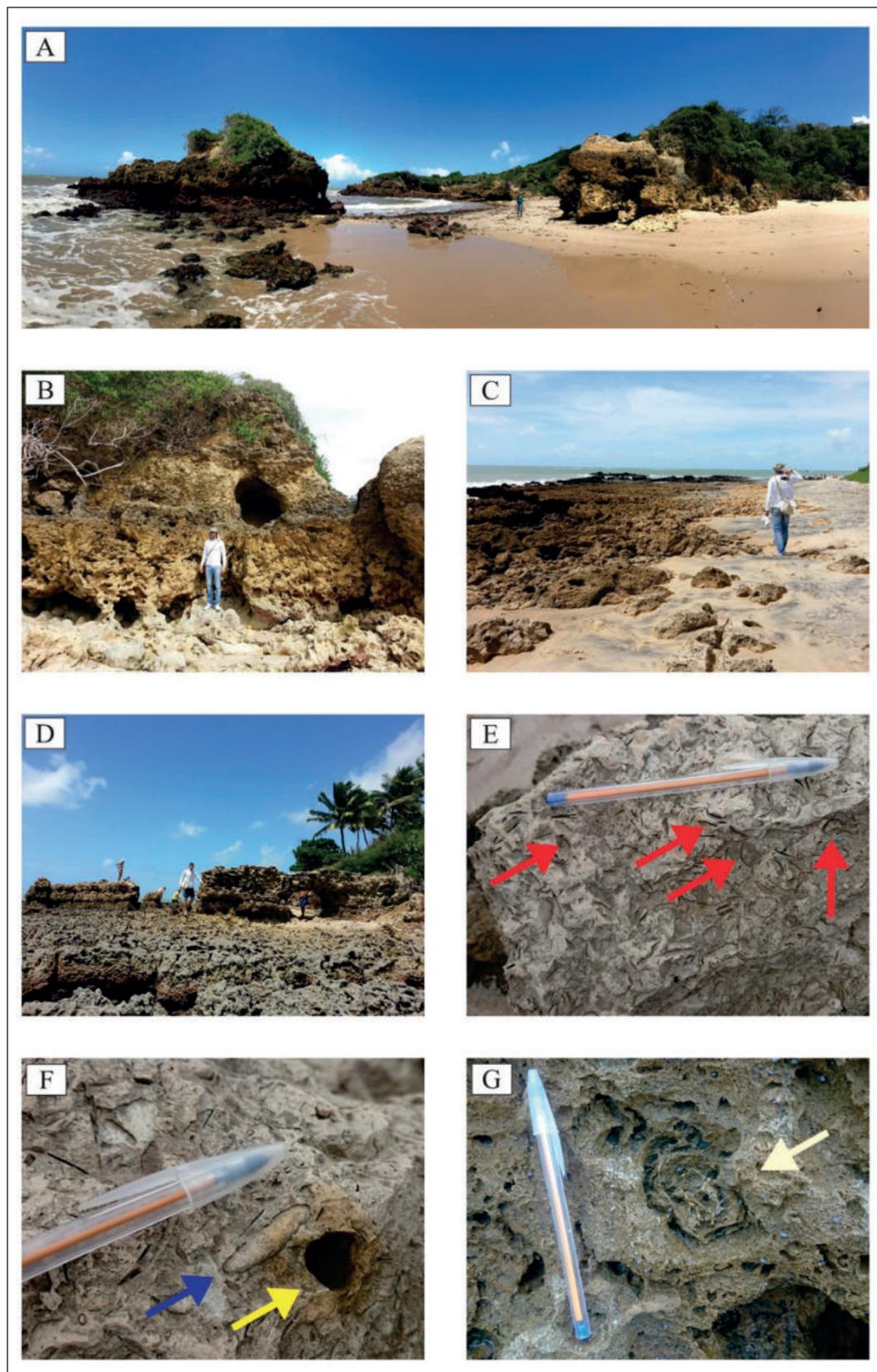


Figure 3 Outcrops of reef limestones. A) General appearance of limestones; B) Tambaba outcrop (DT-01); C) Coqueirinho outcrop (DC-01); D) Jacumã outcrop (DJ-01); E) bivalves molds (red arrows); F) bioturbation molds (blue and yellow arrows); and G) mold of incrusting organisms, i.e., algae (rhodolites – white arrow).

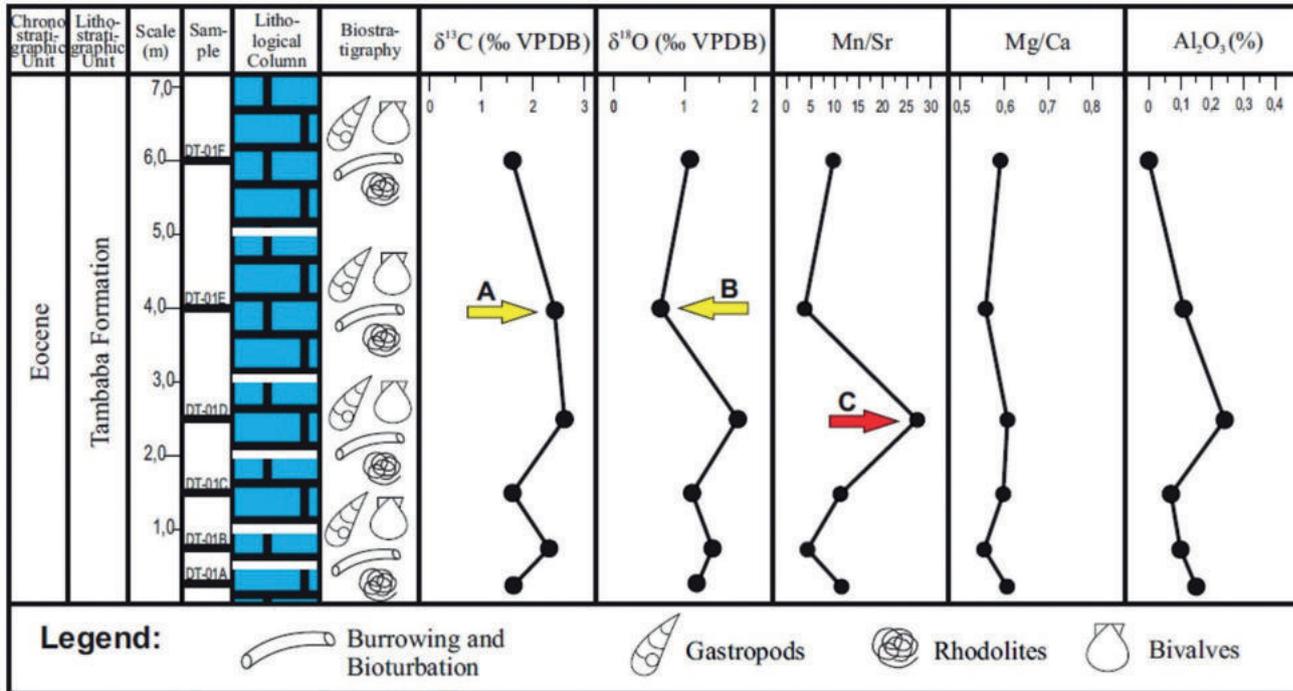


Figure 4 Chemostratigraphic profile of the section located on Tambaba beach. Both isotopes vary with the same trend up the penultimate point analyzed (yellow arrows); High concentration of manganese in DT-01D (red arrow).

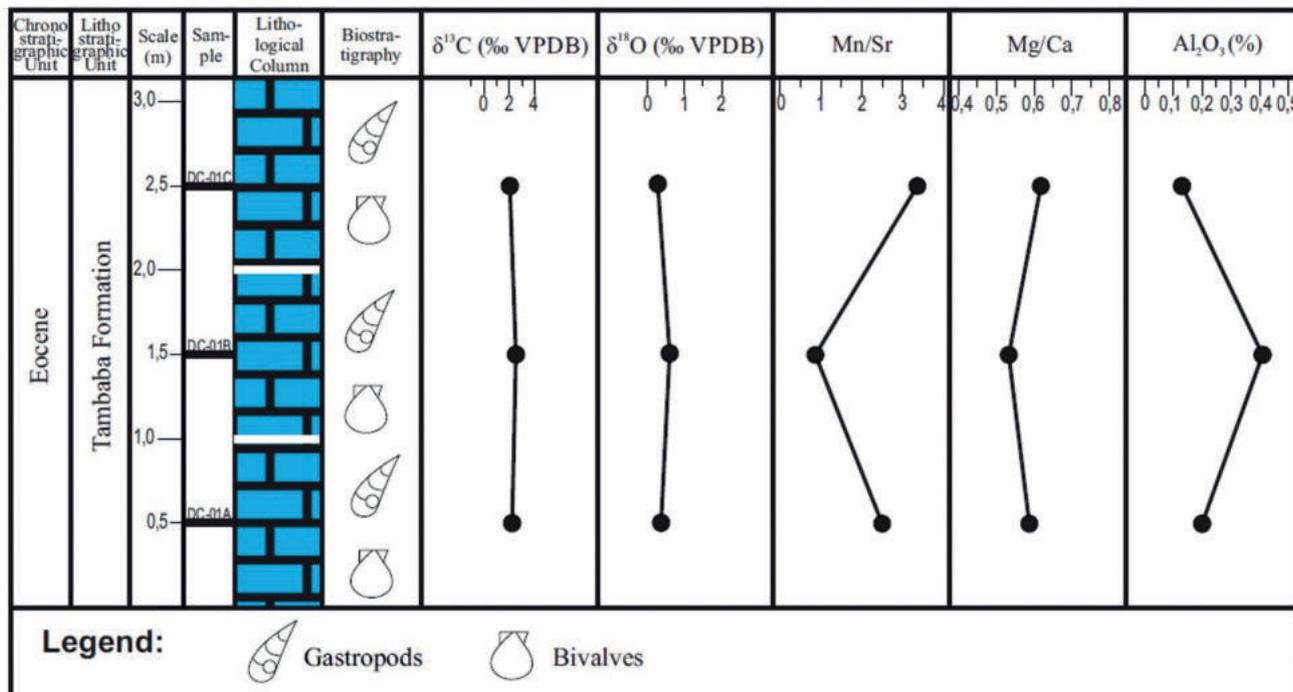


Figure 5 Chemostratigraphic profile of the section located on Coqueirinho beach.

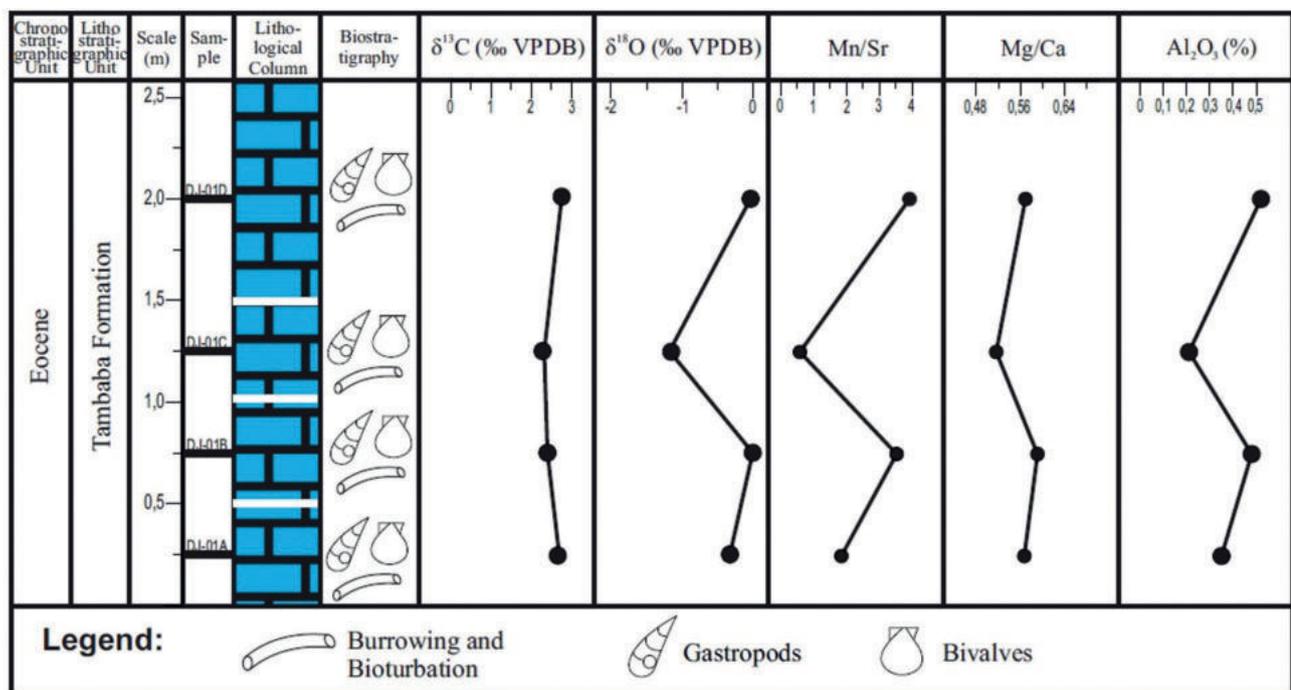


Figure 6 Chemostratigraphic profile of the section located on Jacumã beach.

Sample	$\delta^{13}\text{C}_{\text{VPDB}}\text{‰}$	$\delta^{18}\text{O}_{\text{VPDB}}\text{‰}$	Mn/Sr	Mg/Ca	Al_2O_3
DT-01A	1.62	1.18	11.36	0.61	0.15
DT-01B	2.31	1.40	4.25	0.55	0.10
DT-01C	1.60	1.11	11.13	0.60	0.07
DT-01D	2.61	1.76	27.19	0.61	0.24
DT-01E	2.41	0.66	3.64	0.56	0.11
DT-01F	1.60	1.06	9.62	0.59	0.00
DC-01A	2.23	0.37	2.50	0.59	0.20
DC-01B	2.53	0.60	0.86	0.53	0.41
DC-01C	2.05	0.28	3.36	0.62	0.13
DJ-01A	2.69	-0.33	1.85	0.57	0.35
DJ-01B	2.40	-0.01	3.53	0.59	0.48
DJ-01C	2.32	-1.18	0.59	0.52	0.21
DJ-01D	2.75	-0.04	3.92	0.57	0.52

Table 2 Results of isotopic and lithochemical chemostratigraphy for the described reef limestones.

carbon in relation to the total rock would not change much, because the carbonate volume in carbonates is much higher than in the formation water.

The isotopic curves in DT-01 (Figure 4) show that both isotopes vary with the same trend up to the penultimate sample, where $\delta^{13}\text{C}$ undergoes a nega-

tive excursion (arrow A) and $\delta^{18}\text{O}$ undergoes a positive excursion (arrow B), which may indicate: a fall in productivity, during transgressions there was an anormal proliferation of organisms that extract the $\delta^{13}\text{C}$ from seawater and cause an enrichment of $\delta^{13}\text{C}$ in seawater and precipitated carbonates. This dropping productivity may also be linked to a regression event; and a cooling of the temperature, according to studies by Barbosa (2007) on the Cretaceous-Paleogene (K-Pg) boundary of the Paraíba Basin carbonate platform, where the behavior of the $\delta^{18}\text{O}$ curve shows more negative values close to the K-Pg boundary. These values corroborate the heating and subsequent cooling of a period of environmental stress (Li & Keller, 1998; Keller, 2001; Keller *et al.*, 2002; Keller *et al.*, 2003; Keller *et al.*, 2004a). According Barbosa (2007), the enrichment in Mn is associated to the replacement of dolomite by calcite during diagenetic events (dedolomitization), where this replacement occurred after the dolomitization process that is evidenced by the Mg/Ca ratio.

In the Coqueirinho section there is not much variation between the values obtained (from 2.05‰ to 2.53‰ for $\delta^{13}\text{C}$, and $\delta^{18}\text{O}$ from 0.28‰ to 0.60‰ - figure 5). C- and O-isotope studies of carbonate

rocks from the Paraíba Basin (Nascimento-Silva *et al.*, 2011) reveal that the behavior of $\delta^{13}\text{C}$ e $\delta^{18}\text{O}$ provides information on productivity, temperature and paleoenvironment. Our samples show a carbon and oxygen variation with the same trend are observed with no abrupt variation, indicating a period of environmental stability, without major changes in productivity, temperature or eustatic variations. It is also possible to observe that there is still a predominance of manganese over strontium, especially the reasons found are lower than those in the Tambaba section (<4), and may imply that the intensity of diagenetic events was lower in the DC samples when compared to the DT.

As in Tambaba, Jacumã also presents variation in the behavior $\delta^{18}\text{O}$ curve, indicating again a change in the environmental conditions during the deposition period, diagenetic changes (dolomitization and replacement of dolomite by calcite) and little contribution of siliciclastics. The ratio Mn/Sr in figure 6 presents a variation similar to that observed in Coqueirinho.

The values presented from the mineral chemistry (table 2) indicate that in all three outcrops: i) replacement of dolomite by calcite (dedolomitization) due to the high concentration of manganese on strontium, establishing an order of diagenetic intensity of Tambaba to Jacumã (DT > DC \geq DJ); (ii) Reef limestones are classified as dolomitic limestones, since they have MgO contents above 12%, however, considering that the CaO > MgO contents and the occurrence of diagenesis previously mentioned, it is possible to conclude that there was initially precipitation of the calcite, then a dolomitization causing the occurrence of dolomites, and finally, a phase of replacement where the dolomites were dedolomitized and returned to the calcite mineral phase; iii) all Al_2O_3 values are below 0.50%, indicating a low terrigenous influx over the entire occurrence area of the Tambaba Formation.

Variations in the $^{13}\text{C}/^{12}\text{C}$ and $^{18}\text{O}/^{16}\text{O}$ value in the world's oceans through time have been documented through chemostratigraphic studies of marine carbonate rocks ($^{13}\text{C}_{\text{carb}}$). This variation has been used to date and correlate sediments. Saltzman and

Thomas (2012) analyzed isotope curves between different authors and geological time periods and built a $\delta^{13}\text{C}$ curve from Archean until Pleistocene explaining the isotope behavior.

Keller *et al.* (2018) studied environmental changes in two remarkable climatic and faunal events in Earth's history: The Cretaceous-Paleogene Mass Extinction (KPB) and Paleocene-Eocene Thermal Maximum (PETM). For this study They chose the globally recognized most complete sections for the KPB and PETM events. For the KPB these are the Global Stratotype Section and Point (GSSP) at El Kef, Tunisia, and for the PETM, the global GSSP at the Dababiya quarry in Egypt. They analyzed the carbon isotope behavior across chemostratigraphic sections before, during and after the main events (KPB and PETM), which show excursions (positives and negatives), environmental changes and isotope signal recovery.

Comparing the isotope curves from the reef limestones of the Tambaba Formation (Eocene) with the PETM event (Keller *et al.*, 2018) and the curves built by Saltzman and Thomas (2012), it is possible that the reef limestones are fit well after the PETM event when there is a recovery of the $\delta^{13}\text{C}$ values. The curves built by these authors are in a range from 0‰ to 2‰, as such as the reef limestones here studied.

7 Conclusions

The carbon isotope ($\delta^{13}\text{C}$) behavior in the analyzed samples is generally constant, and the values vary from 1.6 to 2.8‰ and are fit well with the previous values of PETM event. However, the PETM event produced a strong negative excursion that reached values up to -4‰ for $\delta^{13}\text{C}$. This finding added to the previous data from Tambaba Formation corroborates that the carbonate deposits were formed after the PETM event since the $\delta^{13}\text{C}$ values had stabilized by approximately 2‰. The field data were correlated with the chemostratigraphic profiles of the studied outcrops and the bibliographic data, and they favor the hypothesis of a shallow and restricted platform environment with a low sedimentation rate that formed along with the coastal systems. There-

fore, the isotopic variations suggest the occurrence of such a paleoenvironment during a transgression event of smaller magnitude, which allowed for variations in the water blade and generated the deposits studied here.

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