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ORIGINAL

Organic matter in the deltaic clinoform of the São Francisco River (Eastern Brazil)

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Abstract

The São Francisco River is one of the most important rivers in Brazil. The wave-dominated delta at its mouth includes a well-developed muddy clinoform on the adjacent shelf. This study aimed to understand sedimentary organic matter (OM) distribution and the relative contribution of terrestrial/fluvial and marine sources. A dense and evenly distributed sampling grid was used to evaluate the contribution of different OM sources and the influence of local factors. The following parameters were evaluated: grain size, total organic carbon (TOC), total nitrogen (TN), carbonates, and carbon isotopes ($\delta^{13}C$). The spatial distribution of OM properties shows similarities with the spatial arrangement of the various sedimentary provinces of the clinoform (topset, foreset, and bottomset) and the lateral contributions of sediments from neighboring reef bottoms, advected by coastal flows. TOC and TN varied respectively between 0.11 and 1.56% and between 0.02 and 0.20%. The distribution of these parameters in combination with $\%CaCO_3$ shows that the major contributors of TOC to the study area are the São Francisco River, coastal reefs located northwards, and shelf reef bottoms located northeastwards and southwestwards from the deltaic clinoform. The best indicator of river influence was provided by $\delta^{13}C$, with values ranging between − 23 and − 19‰. The spatial distribution of $\delta^{13}C$ shows a good agreement with the various sedimentary elements of the clinoform, reflecting the pattern of river plume expansion observed during the last decades, which is characterized by low discharges, compared to historical values. Our dense and evenly spaced sampling grid and its integration with local geological, geomorphological, and geophysical data allowed to better understand the sources and origin of organic matter and its spatial distribution in this deltaic setting.

Introduction

Delta clinoforms have long been recognized as natural archives of environmental changes occurring in the drainage basin and coastal sea (Bănaru et al. 2007; Bianchi and Allison 2009; Raymond and Bauer 2001; Tesi et al. 2013). Moreover, because deltas provide large contributions of terrestrial organic matter to coastal seas, they have always been considered one of the main carbon sequestering areas on continental shelves (Blair and Aller 2012; Hedges and Keil 1995; Seiter and Zabel 2004; Tesi et al. 2013).

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Carbon and nitrogen concentrations and their isotopic signatures have been widely used to characterize sedimentary organic matter (OM) and reconstruct past environmental changes (Lamb et al. 2006; Meyers 1994, 1997; Pancost and Pagani 2006; Ramaswamy et al. 2008; Zhang et al. 2007). However, organic carbon isotopic signatures vary greatly worldwide in deltaic regions such as the Rhône, the Niger, the Orinoco, the Mississippi, the Po (Bianchi et al. 2002; Boldrin et al. 2005; García-García et al. 2006; Kennicutt et al. 1987), and in those deltas located in Brazil, such as the Paraíba do Sul (Carreira et al. 2015) and the Amazon (Ruttenberg and Goñi 1997). These variations have been attributed to different causes including type of watershed vegetation, increased primary production induced by the fluvial plume, remineralization, human influences, and coastal ocean circulation (Bauer et al. 2013; Burdige 2005, 2007; Hedges et al. 1997; Tesi et al. 2007). This natural variability, resulting from intrinsic local factors, can compromise the quality of interpretations of past environmental changes using downcore variations in OM properties. Moreover, the quality of paleo-environmental reconstructions can be greatly improved by using local reference conditions assessed in situ as opposed to comparisons with similar systems elsewhere (Benito et al. 2016).

The São Francisco is one of the most important rivers in Brazil (Fig. 1). It is extremely important from a cultural, economic, and energy generation perspective for the poorer northeastern portion of the country. The São Francisco watershed is considered one of the most vulnerable in Brazil to ongoing climate changes, suffering under a prognosis of decreasing rainfall over the next decades (Marengo et al. 2011; de Jong et al. 2018). In its subaqueous portion, the São Francisco River has built a mud clinoform measuring about 35 m in thickness (Araújo et al. 2018; Rangel 2017) (Fig. 1). In order to unlock past environmental records (several decades to millennia) preserved in this clinoform, particularly information on the sources and origin of OM in these sediments, we believe that an important aspect is to identify and know in detail the present spatial distribution of isotopic signatures and C/N elemental ratios and how these relate to local intrinsic characteristics such as sedimentary provinces and coastal ocean circulation. Thus, the main objective of the present investigation was to conduct a study on the characteristics of organic matter present in bulk surface sediments of the São Francisco delta clinoform (Fig. 1).

Regional setting

The São Francisco watershed

The São Francisco River (SFR) is part of the fourth largest watershed in Brazil. The headwater is located at an altitude of 1428 m, bringing the total length of the river to 2863 km, with a drainage area of 639,219 km^2, which represents 7.5% of Brazilian territory (Knoppers et al. 2006). Until the 1980s, the SFR presented a mean flow rate of 3136 $m^3\ s^{-1}$, dropping to 1815 $m^3\ s^{-1}$ after 1986 (Fig. 1b). Also, before 1986, there was a well-defined seasonal variation associated with the rainfall regime of its watershed (Medeiros et al. 2011). During the austral summer, flow rates reached up to more than 8000 $m^3\ s^{-1}$ dropping to 1000 $m^3\ s^{-1}$ during the austral winter (Medeiros et al. 2011) (Fig. 1a). After 1986, the flow rate was kept constant year around (~ 1800 $m^3\ s^{-1}$) because of the regulatory effect of the major damns. The SFR watershed represents an important source of sediments and nutrients to the coastal/shelf region and contributes to the maintenance of primary and biological productivity in an otherwise oligotrophic region (Jennerjahn and Ittekkot 1999).

Climate in the watershed varies from a humid tropical and high-altitude temperate climate at its headwaters to a semi-arid and arid climate at its middle course, varying again towards a more humid climate in the coastal region (ANA/GEF/PNUMA/OEA 2004). Mean annual rainfall ranges between 400 mm at the middle and low courses of the river and 1500 mm at its upper course. Due to this climatic pattern, vegetation cover along the watershed includes fragments of *cerrado* (Brazilian savanna) at the upper course, *caatinga* (xeric shrubland) at the middle course, and rainforest in the coastal zone (ANA 2016). Regarding the entire extension of the watershed, the main outcropping rocks are sedimentary (sandstones and limestones) (69 %), metamorphic, metasedimentary, meta-igneous, and igneous (31%) (Kosin et al. 2004).

During the Holocene, the SFR formed a wave-dominated delta (Bittencourt et al. 1982; Dominguez et al. 1987) with a muddy deltaic clinoform on the adjacent continental shelf (Araújo et al. 2018; Rangel 2017; Summerhayes et al. 1976) (Fig. 1).

The clinoform and continental shelf

The continental shelf adjacent to the delta measures approximately 30 km in mean width and is indented by the São Francisco Canyon. The head of this canyon is located only 8 km away from the river mouth (Fig. 1a). The portion of the continental shelf facing the São Francisco delta presents a bathymetric depression associated with the canyon head, with abrupt and linear lateral limits, oriented NW-SE (southern limit) and N-S (northern limits) (Fig. 1a). A well-defined delta clinoform was built upon this topographic depression and consists of muddy sediments, extending from the shoreline until the isobath of 80 m (Fig. 1a). On the sides of this central depression, the continental shelf is considerably shallower, with mostly rocky reef bottoms (Araújo et al. 2018; Coutinho 1981) (Fig. 1a). Bioclastic sedimentation dominates in these regions, which has as its main components fragments of crustose coralline algae and rhodoliths (Carneiro et al.

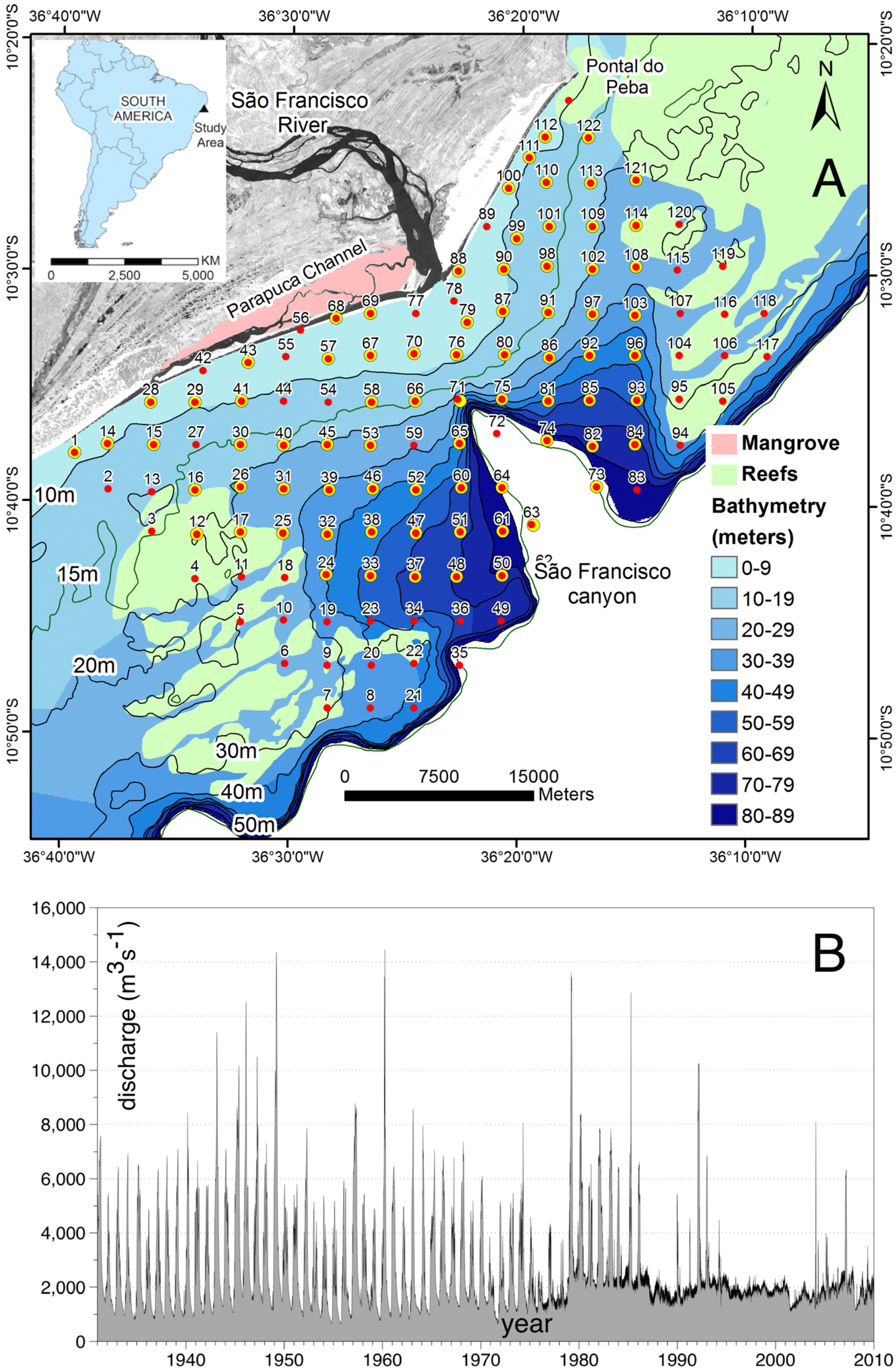


Fig. 1 **a** Location of the study area (upper left inset), sampling grid, and the São Francisco delta clinoform. Red circles with yellow outline indicate samples in which OM properties were determined. Grain size analysis and determination of %$CaCO_3$ were performed in all samples. **b** Monthly discharge for the period 1930–2010 measured 180 km from the river mouth (source: http://www.snirh.gov.br/hidroweb/publico/apresentacao.jsf)

2017; Nascimento 2017; Summerhayes et al. 1976). The shelf break in these lateral regions starts at a mean depth of 45 to 50 m which is representative for the shelf in the region (Coutinho 1981).

Figure 2 shows the major provinces of the SFR clinoform, based on Rangel (2017), who reconstructed the evolutionary history of the São Francisco clinoform using high-resolution shallow seismic records. Limits between the topset, foreset, and bottomset regions are indicated in Fig. 2. The modern clinoform advances over a maximum flooding surface that separates the highstand deposits from the transgressive ones (Fig. 2).

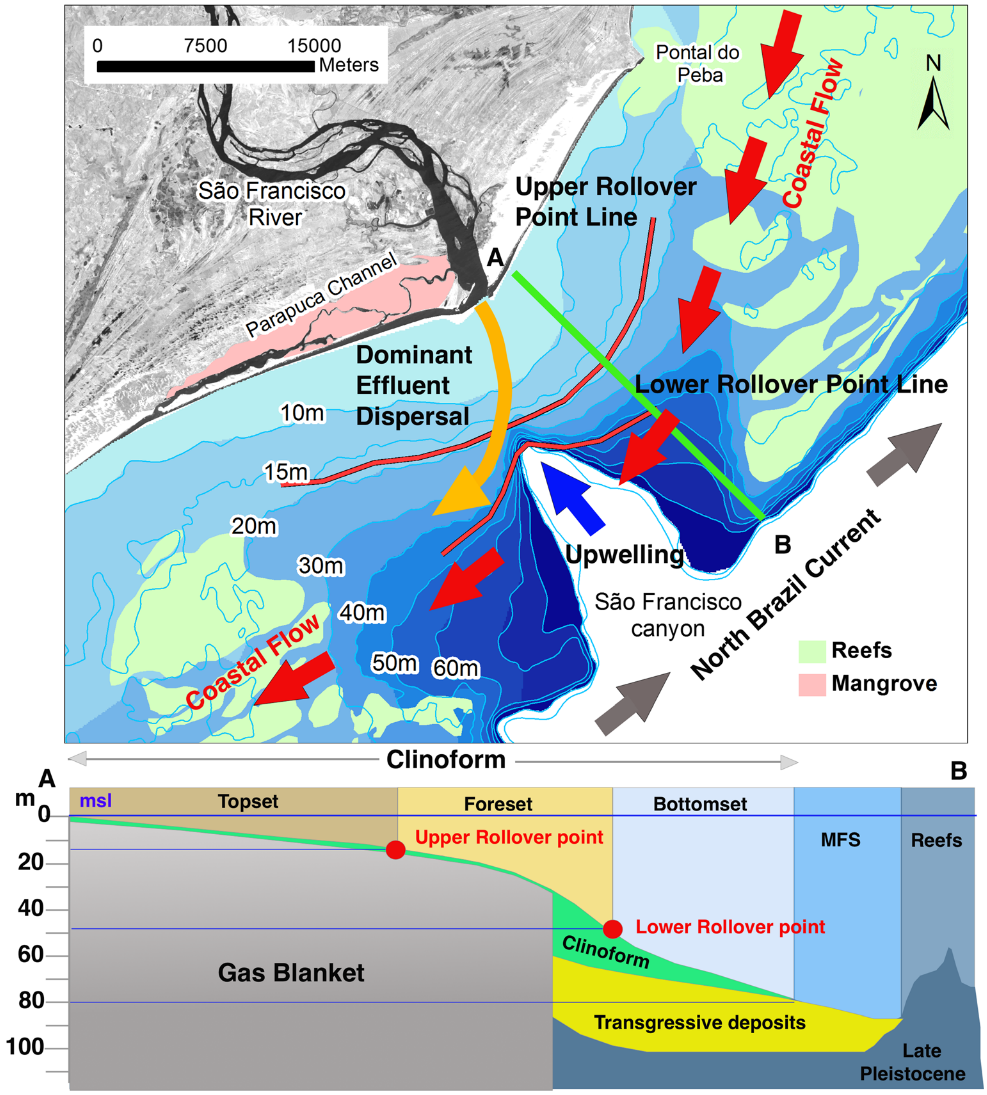


Fig. 2 Major elements of coastal circulation and sedimentary provinces of the deltaic clinoform. Cross section based on illustrations from Rangel (2017). Definitions: The rollover points are the two points of maximum curvature that separate the steeper foreset from the topset (landward) and bottomset (seaward), both with much gentler gradients (Patruno et al. 2015). MFS maximum flooding surface: depositional surface at the time the shoreline is at its maximum landward position (Posamentier and Allen 1999)

Regional oceanographic aspects

Coastal circulation at the study area is mainly controlled by trade winds that blow from east and northeast during the austral spring and summer and from east and southeast by the end of the austral autumn and winter (Bittencourt et al. 1982; Dominguez 1996; Dominguez et al. 1983). However, no direct measurements of these coastal currents are available. In order to circumvent this lack of information, we visually examined a number of satellite images (Landsat 1, 3, 5, 7, and 8—visible spectral bands) of the study area for the period 1973–2015. Sediment plumes in these images suggest the existence of a dominating southwestwards coastal flow in the study area that causes the southward deflection of the river plume, envelops the clinoform foreset, and also introduces fine-grained sediment from external regions to the clinoform (Figs. 2 and 3). Although these images provide only qualitative information on the surface flow, we believe that a similar pattern also occurs in the subsurface because this asymmetry in sediment dispersal is expressed also in the morphology of the deltaic clinoform, which presents smoother slopes southwestwards of the river mouth (Fig. 1). Thus, fine-grained sediment and fluvial plume transport occur mainly southwestwards, except during austral winter months when a northeastward transport is also present (Bittencourt et al. 2005; Dominguez 1996; Dominguez et al. 1983; de Oliveira et al. 2012). Moreover, during the austral winter, flow rates of the SFR are very much reduced (~ 1000 m^3 s^{-1}).

The wave-induced longshore drift of sandy sediments, along the shoreline, is also mainly oriented southwestwards (Dominguez et al. 1992; Guimarães 2010). This process leads to asymmetry in the distribution of deltaic plain facies, with mangrove forests found only in the sector located on the leeward side of the river mouth (Santos et al. 2014) (Figs. 1 and 2).

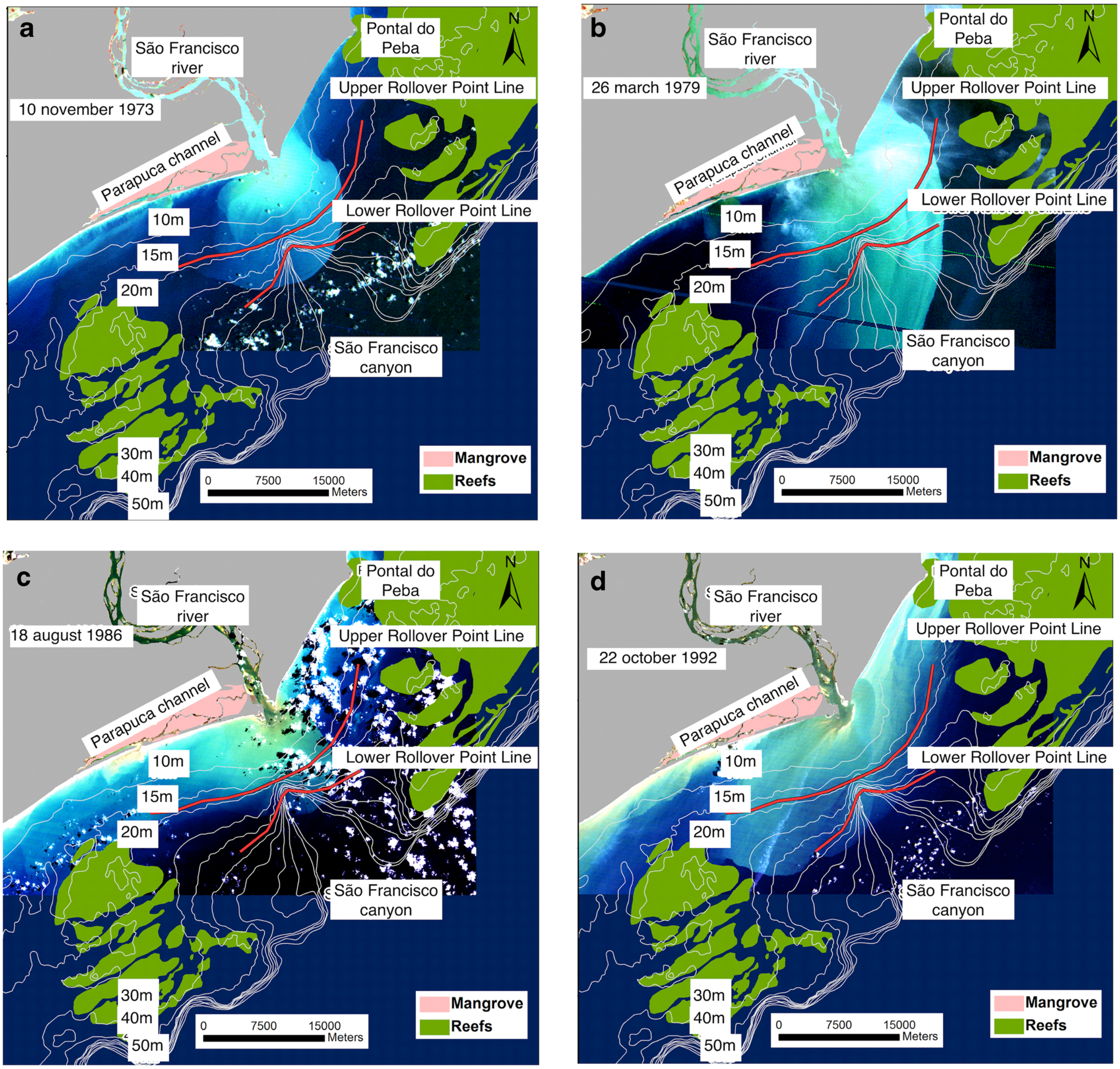


Fig. 3 Examples of satellite images of the study area for different years showing pattern of river plume expansion and its southwestwards deflection due to coastal flows. **a** November 10, 1973. **b** March 29, 1979. **c** August 18, 1986. **d** October 22, 1992. The clinoform foreset upper and lower rollover point lines are also shown in red

Circulation at the slope region is dominated by the North Brazil Current (NBC) (Fig. 2). This current is oligotrophic, with low biologic productivity (Barreto and Summerhayes 1975; Jennerjahn and Ittekkot 1999). It originates from the bifurcation of the South Equatorial Current (SEC), between latitudes 10° S and 14° S, originating the NBC and the Brazil Current (BC), the latter of which flows southwards.

Upwelling has been suggested to possibly affect the SF canyon head (Paes et al. 2007), which is a common process in these settings (Sobarzo and Djurfeldt 2004; Sobarzo et al. 2001).

Material and methods

A total of 121 surface sediment samples, representative of approximately the upper 5 cm of the sea floor, were collected between March 16 and 21, 2010 from the continental shelf adjacent to the SFR, following a sampling grid with regular spacing of 1 km (Fig. 1). Samples were stored in sterile plastic bags and cooled. Once sampling was complete, the sediment samples were frozen and maintained in this condition until laboratorial procedures.

Grain size analysis was performed in all samples of surface sediments (121). A laser diffraction particle size analyzer (Model HORIBA LA950) was used to determine the grain size of fine particles. Particles larger than 2 mm underwent dry sieving. Results were combined, and the software GRADISTAT V 5.0® (Blott and Pye 2001) was used to determine grain size parameters (sorting, median (D50), and silt, clay, sand, and gravel contents).

Of the total of 121 samples collected, only 76, texturally classified as mud (silt + clay), were analyzed for OM (Fig. 1). These 76 samples were treated with hydrochloric acid (HCl) 1 mol L^{-1} in order to remove any calcium carbonate particles present. They were then rinsed with distilled water and dried once again. Since inorganic nitrogen associated with clays cannot be eliminated (Kao et al. 2003), the nitrogen analysis included both organic and inorganic nitrogen, referred to as total nitrogen (TN).

An element analyzer (Costech) coupled to a mass spectrometer (Thermo Finnigan Delta Plus) was used to determine TOC and TN contents in OM, allowing the simultaneous determination of element composition (% of C and N) and isotopic ratio of δ^{13}C. Carbon isotopic composition was determined after eliminating carbonates. Quantification is presented in this study as the deviation (‰) from the standard Pee Dee Belemnite (PDB) isotopic composition for carbon isotopes, based on a Cretaceous marine fossil from the Peedee Formation in South Carolina, USA. The IAEA standard NBS19 (limestone, 1.95‰ vs VPDB) was acidified to produce CO_2 which was used as reference gas for the mass spectrometer calibration. The analytical error, obtained through repeated measurements, was ± 0.2‰. Total carbonate content in sediments was obtained through gravimetric analysis.

Statistical analysis of the data consisted of the application of the Spearman correlation coefficient, after Shapiro-Wilk normality tests. The inverse distance weight (IDW) algorithm was used to interpolate the values of surface sediment parameters analyzed in order to produce maps of spatial distribution. The Jenks Natural Breaks classification method was used to determine the classes represented in the maps produced, since it provided the best arrangement for the values obtained among the different classes (Jenks 1967). Graphs were produced for the variation of properties found in the sediment cores according to depth.

Because there was no available data about the spatial extension of the river plume, we used a set of 41 images from Landsat missions 1, 3, 4, 5, 7, and 8 (1973–2015) to delineate the limits of the sediment plumes on different years. Most of the images are from the austral summer, which coincides with the highest discharges of the S. Francisco river (Santos et al. 2012). Image composites of visible spectral bands were used, and limits of the fluvial plume exiting the river mouth were visually digitized on the computer screen.

Results

Grain size

Figure 4 shows the spatial distribution of median grain size (D50) values among continental shelf surface sediment samples. As a rule, predominantly fine-grained sediments were observed in the area of the deltaic clinoform, with values of D50 decreasing offshore. However, sediments tended to be slightly coarser on the topset and foreset regions of the clinoform and towards S-SW, following the same dispersion pattern of the fluvial plume. Slightly coarser sediments also occur in front of the tidal channels draining the mangrove areas located downdrift of the river mouth (Parapuca channel). In the outer portions of the shelf, beyond the deltaic clinoform, sandy and gravely bioclastic sediments dominated.

Calcium carbonate

Calcium carbonate contents in fine surface sediments ranged between 1.5 and 99.8% (Fig. 5). These contents progressively increased further away from the coast. In the deltaic clinoform itself, calcium carbonate contents ranged between 1.5 and 16.1%, with the lowest values found mainly on the topset area (Fig. 5). Sediment from the outer shelf, located laterally to the clinoform, was composed of almost 100% carbonate.

Total organic carbon and total nitrogen

TOC (%) and TN (%) contents are presented in Table 1S (Supporting Information), and their spatial distributions in clinoform sediments are shown in Figs. 6 and 7, respectively. TOC ranged between 0.11 and 1.56%, with mean values of 0.81 ± 0.33% (Table 1S; Fig. 6). The highest TOC contents (1.28 to 1.56%) were found at the northeastern extremity of the study area, near the reef constructions of Pontal do Peba, around the canyon head, and at the clinoform. The lowest values of TOC (< 0.6%) occurred in the southwestern portion of the study area, near the coast.

The spatial distribution of TN content was similar to the distribution of TOC (Fig. 7). Values of TN ranged between 0.02 and 0.20% (Table 1S). A significant correlation (r = 0.958; $p < 0.001$) was observed between TOC and TN

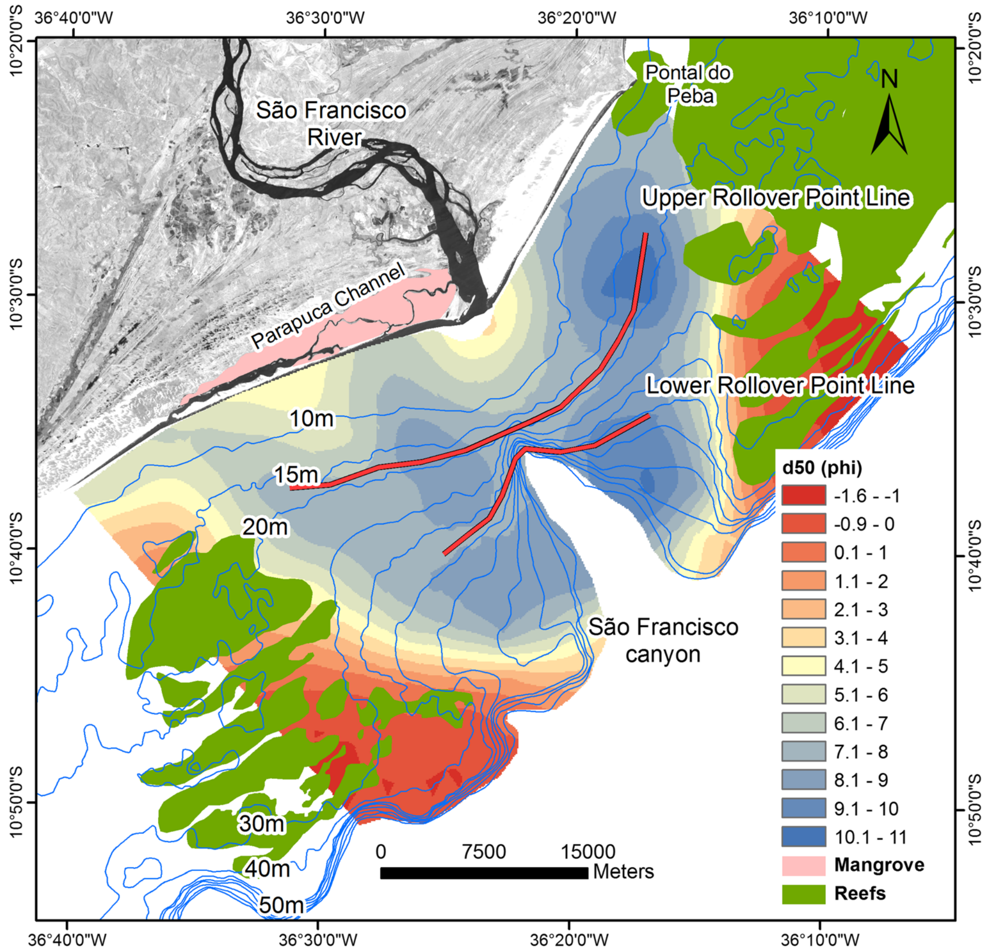


Fig. 4 Grain size distribution of sediments on the shelf and delta clinoform. The clinoform foreset upper and lower rollover point lines are also shown in red

contents (Figure 1S—Supporting Information). The regression line near the origin suggests that most of the nitrogen measured in the sediment is related to sedimentary organic carbon and, therefore, is likely to have come from an organic source (Hedges et al. 1986). Regression analyses were also conducted between TOC and the depths at which samples were collected ($r = 0.103$; $p > 0.001$) (Figure 2S—Supporting Information) and between TOC and sediment grain size ($r = 0.325$; $p > 0.001$) (Figure 3S—Supporting Information). Both analyses presented low correlation and were not significant.

Carbon stable isotopes

δ^{13}C in surface sediment presented a mean value of − 21.40 ± 0.94‰, ranging between − 23 and − 19‰ (Table 1S—Supporting Information). The spatial distribution of δ^{13}C values presented an approximately radial pattern in relation to the mouth of the SFR (Fig. 8). In general, less enriched values were found near the clinoform topset (− 23.3‰ and − 21.5‰). In the vicinities of the river mouth, values of δ^{13}C ranged between − 23.3 and − 22.0‰ along an approximately NW-SE oriented narrow belt that reaches the head of the São Francisco Canyon. Interestingly, this belt also shows a southward deflection, similar to that experienced by the river plume. Impoverishment of δ^{13}C values was also observed in the southwestern extremity of the study area, in a narrow sector parallel to the shoreline and located immediately downstream to the tidal channel (Parapuca) that drains the mangrove forests of the deltaic plain. Values of δ^{13}C increased significantly in the region beyond the clinoform, ranging between − 21 and − 19‰, possibly as a result of being nearer to the reef bottoms that predominate in the external shelf areas located NE and SW from the clinoform (Fig. 8).

Total organic carbon/total nitrogen ratio

C/N ratios in surface sediments ranged between 5.5 and 11.3 (Fig. 9), with a mean value of 8.3. The highest values coincided approximately with the limits of the deltaic clinoform (8.6 to 11.3) and decreased in the external region (9.0 to 5.5). C/N values were also higher in the southwestern portion of the investigated area, in association with the mouth of the Parapuca tidal channel, which drains mangrove areas.

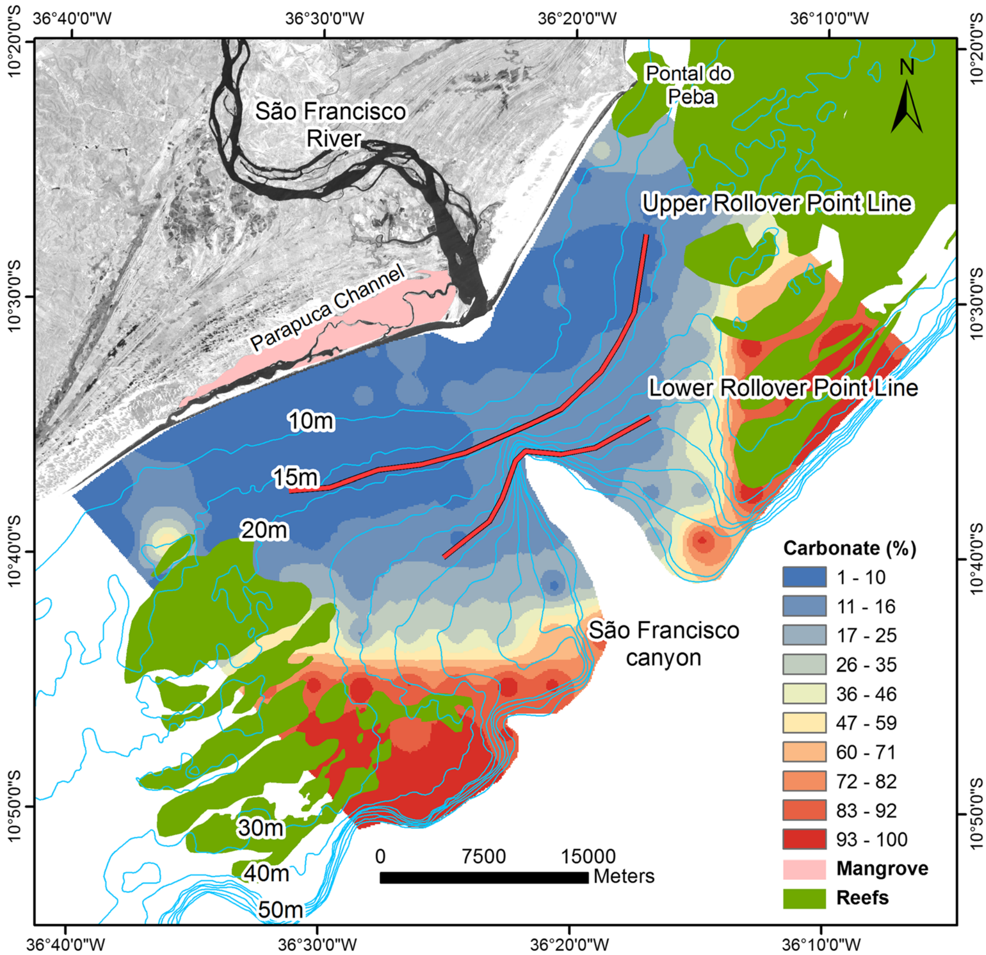


Fig. 5 Calcium carbonate content of sediments on the shelf and delta clinoform. The clinoform foreset upper and lower rollover point lines are also shown in red

Discussion

Sources of organic matter to the study area

TOC values in bulk surface sediments at our study area are similar to those found in other deltaic regions in Brazil, such as the Paraíba do Sul (Carreira et al. 2015) and Amazon (Ruttenberg and Goñi 1997; Siqueira et al. 2013) rivers and elsewhere in the world, such as in the Rhône, Niger, Orinoco, Mississippi, and Po rivers (Kennicutt et al. 1987; Tesi et al. 2013; García-García et al. 2006; Bianchi et al. 2002; Shields et al. 2019).

No relationship was found between TOC and sediment grain size or water depth (Figures 2S and 3S—Supporting Information). However, as expected, a good correlation existed between TOC and TN, since these are the two major components of organic matter (Mayer 1994).

If the São Francisco River was either the major or sole OM contributor to the area, such gradients (TOC vs grain size vs depth) would normally be expected. We interpret this lack of correlation and of any onshore-offshore gradients in the spatial distribution of TOC and TN as a result of there being different organic carbon production areas. As already noted, the deltaic clinoform was built in a low-lying area of the continental shelf bordered laterally by reef bottoms, where carbonate sedimentation is dominant, and with the presence of the São Francisco canyon head in the offshore region.

The spatial distribution of $CaCO_3$ (Fig. 5) clearly suggests that besides the fluvial input of sediments, characterized by low $CaCO_3$ values, lateral contributions of sediments also occur from the Pontal do Peba reefs located in the extreme northern portion of the study area and from the lateral reef bottoms located northeastwards and southwestwards from the clinoform (Figs. 2, 3, and 5). These lateral contributions, expressed by higher $CaCO_3$ values, are more significant on the bottomset region of the clinoform and at the maximum flooding surface over which it progrades (Figs. 2, 3, and 5). These different sources of sediment are also expressed into different sources of OM, which also help us understand the spatial distributions of TOC and TN in the study area (Figs. 6 and 7). Interestingly, no significant external lateral contribution of $CaCO_3$ and OM is found associated with the western extremity of the clinoform (Parapuca channel). Although the seafloor in this region

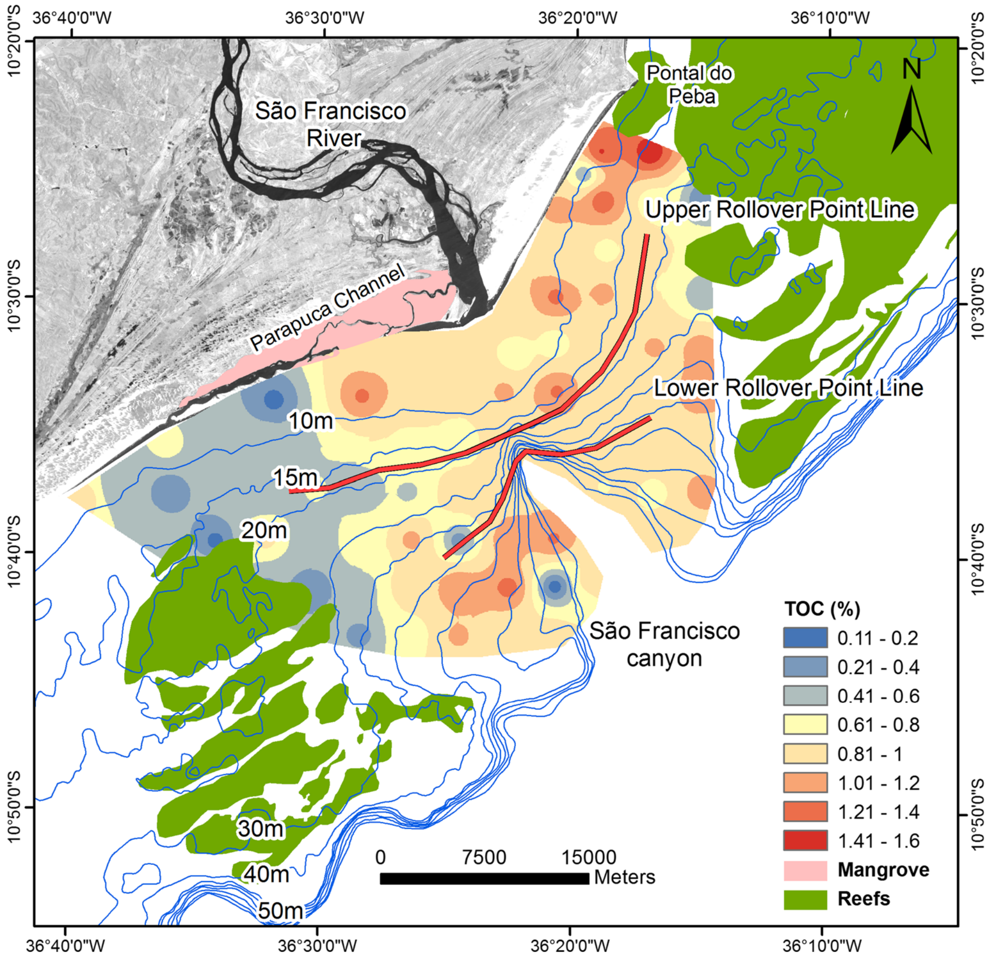


Fig. 6 TOC (percent) content of sediments on the shelf and delta clinoform. The clinoform foreset upper and lower rollover point lines are also shown in red

is also characterized by rocky outcrops, carbonate production and associated OM are inhibited due to the influence of the river plume, which flows predominantly southwestwards (Figs. 2, 3, 5, 6, and 7). This is corroborated by the fact that $CaCO_3$ content in the sediment is reduced. Moreover, older sedimentary rocks outcropping offshore in this southwestern region still exhibit its original bedding, which is clearly visible in satellite images, implying the absence of biogenic reef constructions.

Finally, upwelling at the canyon head, which enhances primary productivity, is also thought to be a possible source of OM to the region.

Organic matter origin

The spatial distribution of $\delta^{13}C$ and C/N ratios offers corroborating evidence to what we have hypothesized above, i.e., that the spatial distributions of TOC, TN, and $CaCO_3$ contents are linked to the existence of three to four major sources of organic matter production for the study area.

$\delta^{13}C$ and C/N ratios have been widely used to distinguish sources of organic matter and carbon flux in coastal sediments (Carreira et al. 2015; García-García et al. 2006; Meyers 1994; Meyers 1997; Lamb et al. 2006; Pancost and Pagani 2006; Ramaswamy et al. 2008; Usui et al. 2006). In shelf areas, these values reflect the contribution of terrestrial organic matter and nutrients, local increase in primary production associated with the fluvial plume, and lateral contributions of organic matter from other shelf areas (Bianchi et al. 2002; De Haas et al. 2002).

Usually, C/N ratio values between 4 and 8 are indicative of a predominantly marine origin (algae), whereas values greater than 15 are suggestive of a terrestrial origin (vascular plants) (Gao et al. 2012; Lamb et al. 2006; Meyers 1997). Values in between these two extremes would indicate a mixture of these sources (Meyers 1997). In our study area, C/N ratios lower than 8 and therefore indicative of a marine origin occurred in areas that also presented evidence of lateral contribution of sediments (higher $CaCO_3$ values) and OM, as discussed above (Pontal do Peba reefs and reef bottom areas neighboring the canyon head).

At the deltaic clinoform itself, C/N ratios were dominantly in the range of 8 to 9 but reaching values of up

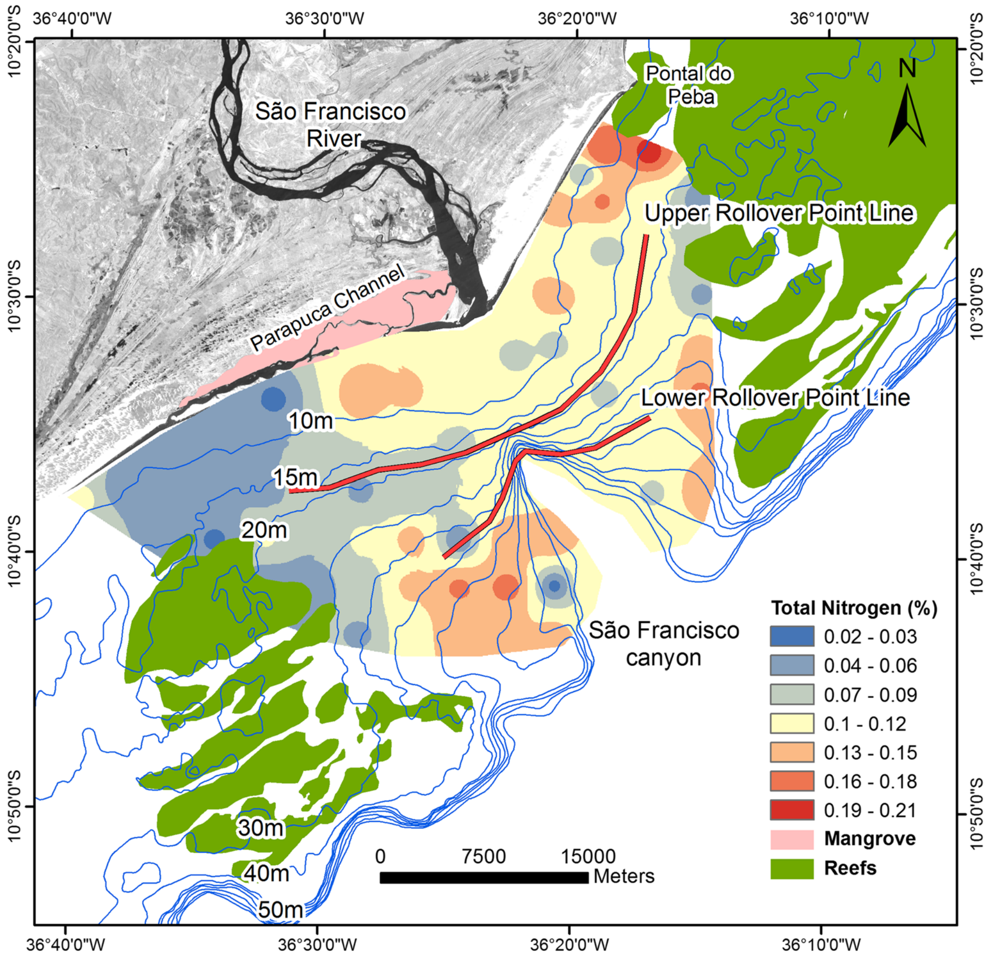


Fig. 7 TN (percent) content of sediments on the shelf and delta clinoform. The clinoform foreset upper and lower rollover point lines are also shown in red

to 12 in front of the river mouth and at the western limit of the region downdrift to the Parapuca river channel. According to Meyers (1997), vascular C3 plants have C/N ratios around 12, whereas C4 plants show values of around 30. Our results showed that, at the clinoform, C/N ratios were well below those expected for an exclusively continental origin, suggesting a mixture of sources. Low C/N ratios have also been found in other tropical shelf areas located near river mouths and were attributed to anthropogenic influences and/or natural causes (Ramaswamy et al. 2008; Ruttenberg and Goñi 1997). Moreover, preferential sorption of inorganic N by minerals and preferential remineralization of N has also been observed (Hu et al. 2006; Ruttenberg and Goñi 1997)

δ^{13}C values for terrestrial plants, characterized by the C3 photosynthetic pathway, typically vary between − 33 and − 22‰, whereas C4 plants such as grasses have values around − 14 to − 9‰ (Meyers 1997; Pancost and Boot 2004). Typical isotopic ratios of marine OM produced by phytoplankton show values between − 17 and − 22‰ (Gearing et al. 1984; Hedges et al. 1997; Kao et al. 2003). Until finally being deposited in the bottom sediment, this organic matter experiences several decomposition transformations. This process is controlled by several factors such as chemical composition and amount of organic matter, nutrient and oxygen availability and temperature, local fauna, and microorganisms found in the soils of the drainage basin and in the marine environment (Leithold et al. 2016; Sanderman and Amundson 2016). Thus, δ^{13}C in bulk marine sediments will not necessarily preserve the original signatures of organic matter sources. In addition, the overlap of δ^{13}C from different sources can complicate the attribution of an origin to OM (Mayer 1994). In this sense, the simultaneous use of δ^{13}C and C/N ratios can provide more reliable interpretations (Lamb et al. 2006).

The distribution of δ^{13}C values in our study area exhibited a clear gradient from the river mouth to offshore areas (Fig. 8). Values from − 22.2 to − 23‰, and from − 21 to − 22.2‰ characterize respectively the topset and foreset regions of the clinoform. δ^{13}C values greater than − 21‰ characterize the clinoform bottomset and the maximum inundation surface regions. Comparisons between this distribution and the areal

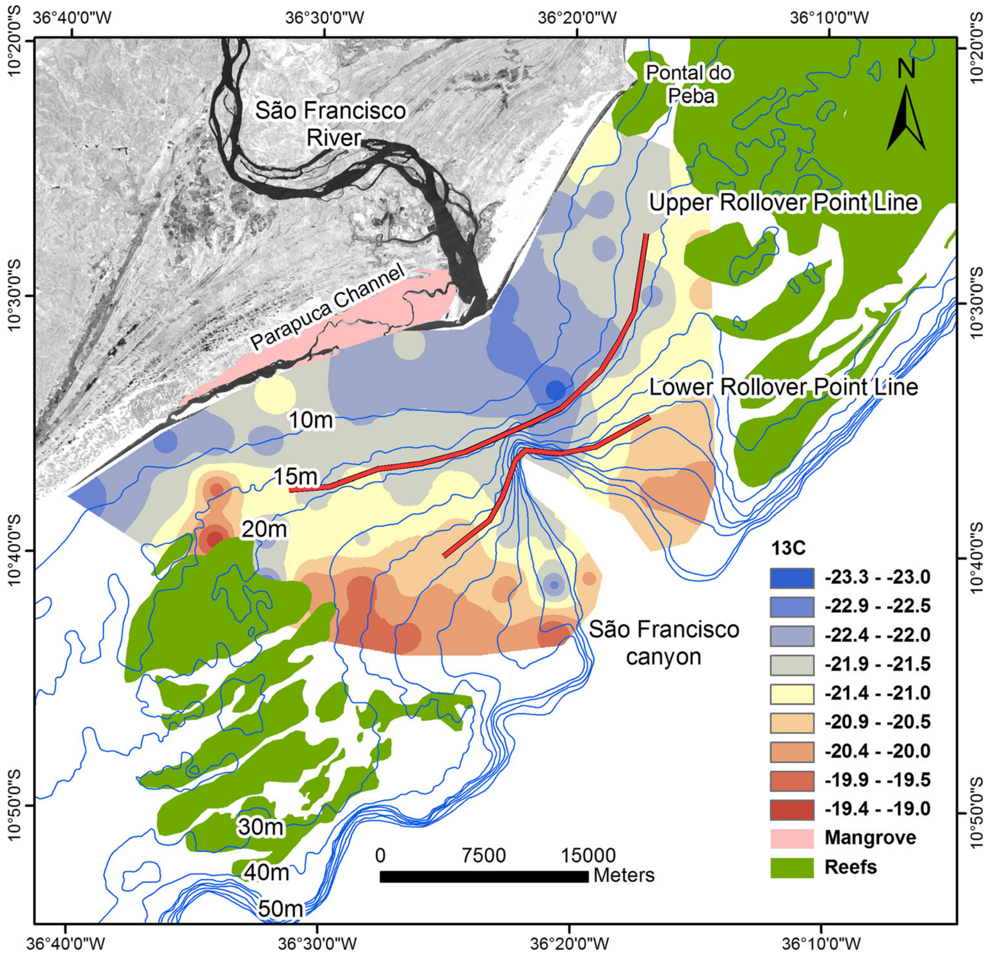


Fig. 8 δ13C in sediments on the shelf and delta clinoform. The clinoform foreset upper and lower rollover point lines are also shown in red

extent of the river plume, which was visually mapped from satellite images over the last 30 years, show a good agreement between the lower $\delta^{13}C$ values found (− 22.2 to − 23.3‰) and the extension of the plume for this period (Fig. 10a). This range of values was also found downdrift from the Parapuca channel, which drains the mangrove forests of the subaerial delta.

Santos et al. (2013) described $\delta^{13}C$ values for vegetation occurring in the estuarine region of the São Francisco: *Elodea* sp. (grass) − 16‰, *Montrichardia linifera* (aquatic macrophyte) − 27‰, and *Rhizophora mangle* (mangrove tree) (− 28‰). In the same study, these authors showed that $\delta^{13}C$ bulk sediment values progressively became enriched along the river channel, with C/N ratios also decreasing along the same gradient. Along the Parapuca channel, $\delta^{13}C$ values were more typical of C3 plants because of the dominance of mangrove forests. The lowest $\delta^{13}C$ values (− 23‰) found by Santos et al. (2013) were from sediments collected very close to the river mouth (both inside and outside of the estuarine area) and at the outlet of the Parapuca channel. C/N values close to 12 from these same samples suggest that OM in bulk river-borne sediments have significant contributions from vascular C3 plants. Sedimentary OM exiting the fluvial system presented an isotopic signature of about − 23‰ (Santos et al. 2013). This value rapidly increases away from the river mouth due to dilution from lateral sediment sources (reef bottoms) and possibly also from nutrient recycling by phytoplankton.

Usually, clinoform topsets are characterized by high energy levels from waves and currents, resulting in low accumulation rates (Cattaneo et al. 2007; Pirmez et al. 1998; Swenson et al. 2005). This reworking will probably cause changes in the $\delta^{13}C$ signature of offshore bulk sediment. Close to the upper rollover point line and at the clinoform foreset region, $\delta^{13}C$ values already reach − 21‰ probably as a result of mixing between terrigenous and locally produced marine OM and of the contribution of laterally advected sediments and marine OM from the Pontal do Peba and shelf reefs. Because the foreset portion of a clinoform has the highest accumulation rates (Patruno et al. 2015), its isotopic signature will dominate the bulk of clinoform sediments. At the bottomset

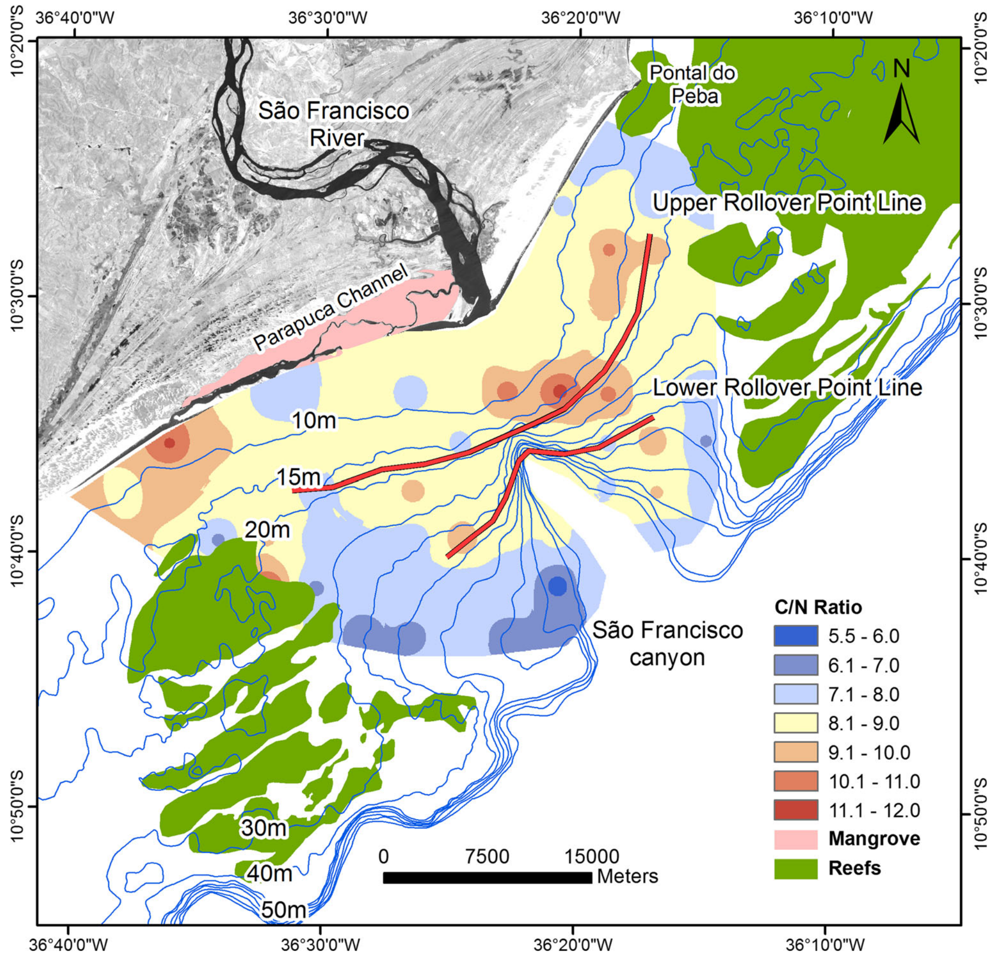


Fig. 9 Total organic carbon/total nitrogen ratio (C/N) in sediments on the shelf and delta clinoform. The clinoform foreset upper and lower rollover point lines are also shown in red

and maximum flooding surface regions, the $\delta^{13}C$ isotopic signal is typically marine. Higher marine productivity induced by upwelling may have also played a role in the signatures at and around the canyon head.

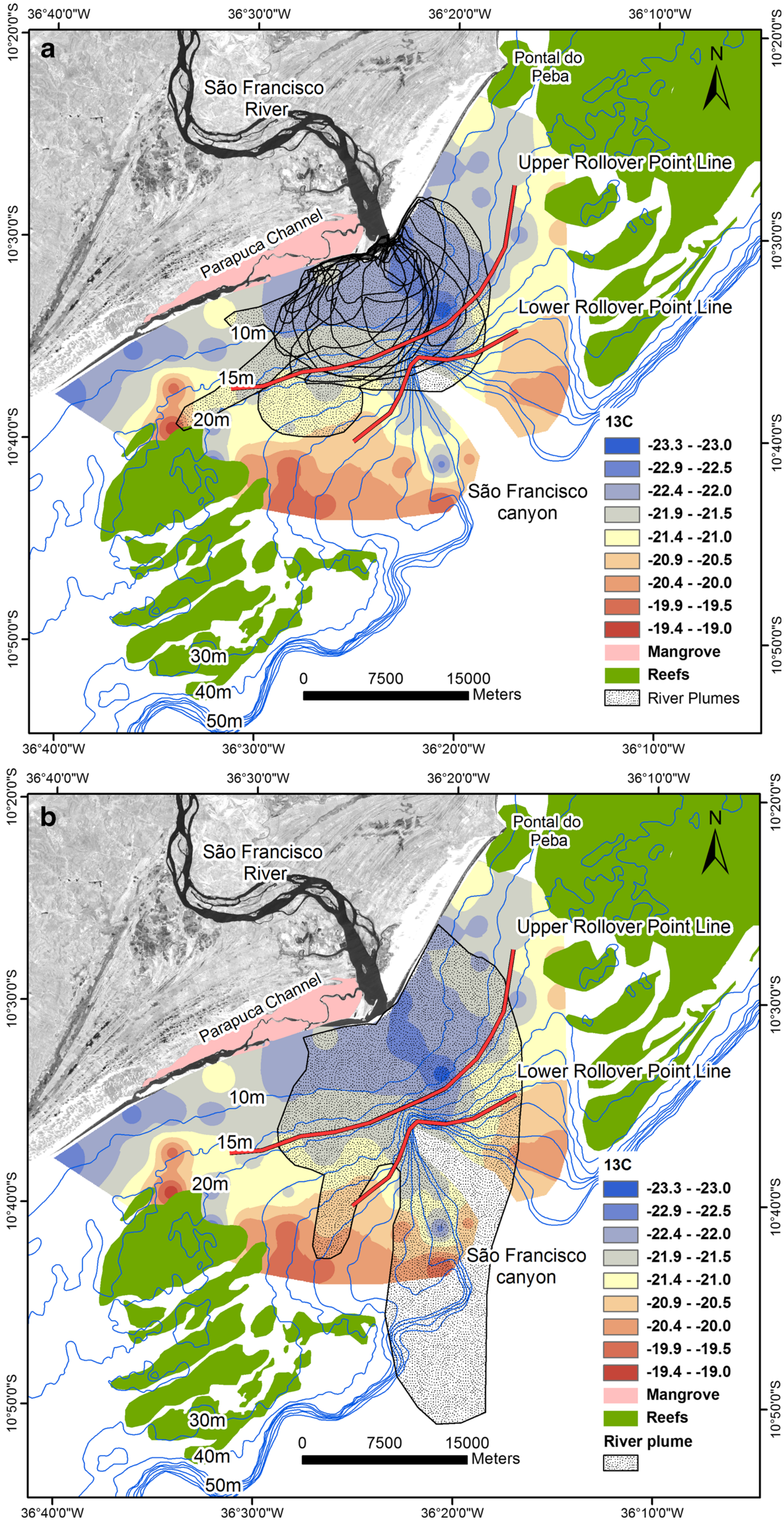
Our results, considering overall average values, are similar to other documented fluvially influenced shelf areas in Brazil and elsewhere (Bianchi et al. 2002; Carreira et al. 2015; Ruttenberg and Goñi 1997). In a number of these regions, relatively high $\delta^{13}C$ values have been attributed to a dominance of C4 plants in large portions of the catchment area. This is also the case of the São Francisco River because of the dominance of a semi-arid climate in the catchment area.

Finally, it should be noted that our results are probably representative of low discharge periods, which have dominated the last 30 years in the SFR catchment area (de Jong et al. 2018) (Fig. 1b). This decrease in rainfall was greatly amplified by the regulatory effect of the large Sobradinho dam, located 748 km upstream from the river mouth. This is corroborated by the good agreement found between the spatial extent of river plumes during this period and the lower $\delta^{13}C$ values found in the topset region (Fig. 10a). Larger discharges would cause an expansion of the bottom area impacted by the river plume, as exemplified by the last great flood event that affected the region in 1979 (Figs. 3b and 10b).

Conclusions

The São Francisco clinoform provinces (topset, foreset, and bottomset regions) and neighboring areas (maximum flooding surface, canyon head, and reef bottoms) provide a mosaic of bottom types and dynamical provinces that leave distinct OM signatures imprinted in bulk sediments and their spatial distribution patterns (Fig. 11). Untangling the causes of these patterns was made possible through integration of the different datasets, with local geological, geomorphological, and geophysical data discussed herein.

Fig. 10 **a** Areal extension of the river plume, visually mapped from satellite images of the last 30 years, plotted on top of bulk sediment δ13C values. The clinoform foreset upper and lower rollover point lines are also shown in red. **b** Areal extension of the river plume during the 1979 high discharge event, visually mapped from satellite images, plotted on top of bulk sediment δ13C values. The clinoform foreset upper and lower rollover point lines are also shown in red ▶


a
São Francisco River
Parapuca Channel
Pontal do Peba
N
Upper Rollover Point Line
Lower Rollover Point Line
10m
15m
20m
30m
40m
50m
São Francisco canyon
13C
-23.3 - -23.0
-22.9 - -22.5
-22.4 - -22.0
-21.9 - -21.5
-21.4 - -21.0
-20.9 - -20.5
-20.4 - -20.0
-19.9 - -19.5
-19.4 - -19.0
Mangrove
Reefs
River Plumes
0 7500 15000 Meters
36°40'0"W 36°30'0"W 36°20'0"W 36°10'0"W
10°20'0"S 10°30'0"S 10°40'0"S 10°50'0"S
b
São Francisco River
Parapuca Channel
Pontal do Peba
N
Upper Rollover Point Line
Lower Rollover Point Line
10m
15m
20m
30m
40m
50m
São Francisco canyon
13C
-23.3 - -23.0
-22.9 - -22.5
-22.4 - -22.0
-21.9 - -21.5
-21.4 - -21.0
-20.9 - -20.5
-20.4 - -20.0
-19.9 - -19.5
-19.4 - -19.0
Mangrove
Reefs
River plume
0 7500 15000 Meters
36°40'0"W 36°30'0"W 36°20'0"W 36°10'0"W
10°20'0"S 10°30'0"S 10°40'0"S 10°50'0"S

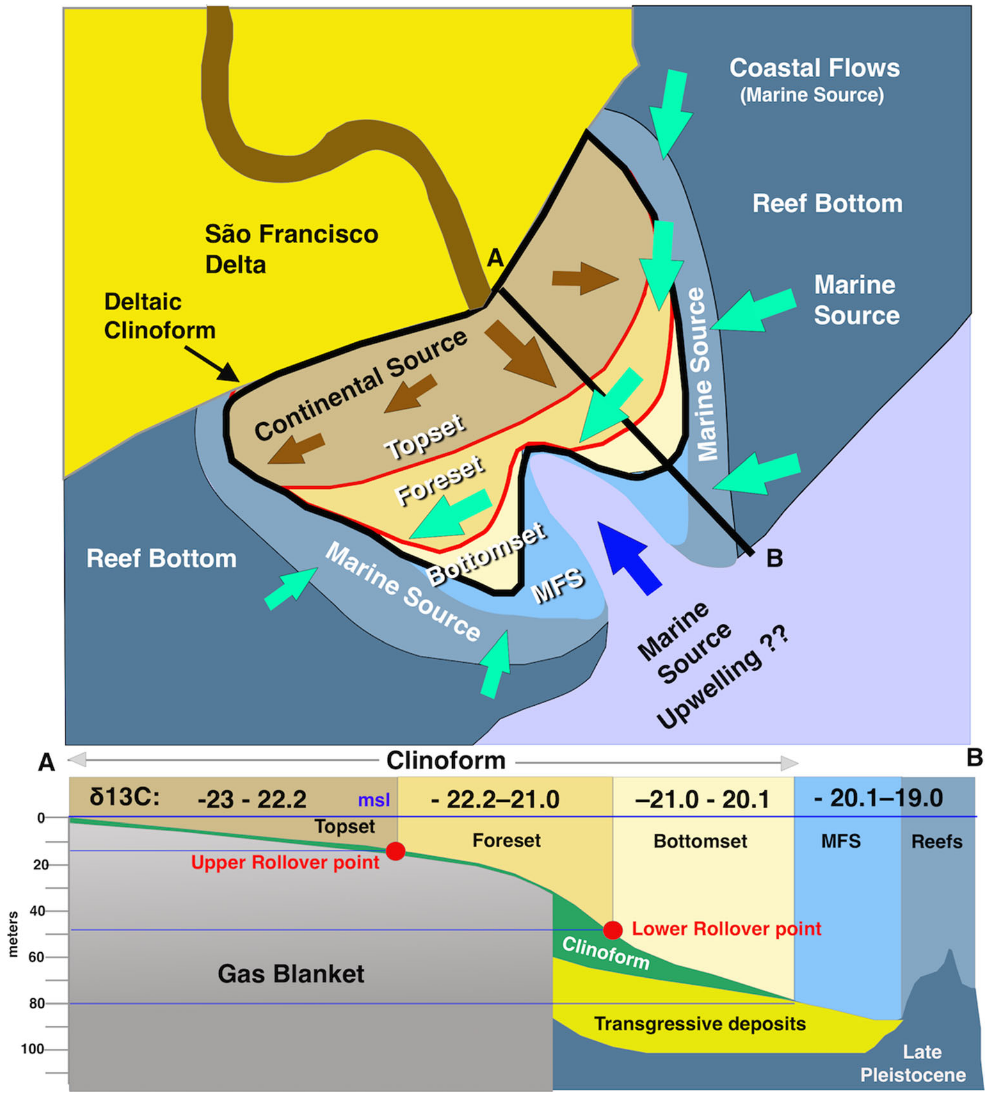


Fig. 11 Conceptual model showing sources of sediments and OM in the deltaic clinoform of the São Francisco and neighboring areas. Major sedimentary provinces and average δ13C values in bulk sediment are also depicted. Cross section based on illustrations from Rangel (2017)

Sources of OM to the São Francisco clinoform are river-borne combined with lateral contributions from reef areas and possibly in situ phytoplankton production related to local increase of nutrients brought by the river and upwelling at the canyon head region.

The topset region of the clinoform under the direct influence of the river plume presents $\delta^{13}C$ bulk sediment values more akin of C3 vascular plants, in association with the lowest $CaCO_3$ content found in the data. $\delta^{13}C$ values progressively increase seaward, as a result of lateral contributions from reef areas located either close to the shoreline or on the shelf, and brought into the clinoform foreset region by the southwards directed coastal flows. Typical marine $\delta^{13}C$ and C/N ratios are found in the distal clinoform bottomset and in the maximum flooding surface that outcrops around the canyon head. These two regions also exhibit high TOC contents.

We hope this study will provide a template to improve quality of future paleo-environmental investigations at the São Francisco River clinoform using downcore analysis of OM properties either isolated or in combination with other parameters such as foraminifera, ^{14}C, sediment characteristics, biomarkers, and metal ratios, among others (Carlin and Dellapenna 2014; Mendes et al. 2015; Zhou et al. 2016).

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