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https://doi.org/10.1590/2317-4889202320220083

BJGEO
Brazilian Journal of Geology

The provenance of terrigenous mud on reefs in Royal Charlotte Bank, Bahia, Brazil

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Abstract

The East Brazilian continental margin contains a large shelf sector called the Royal Charlotte Bank. It has terrigenous and carbonate sedimentation associated with coastal reefs. Studies using X-ray diffractometry (XRD) and the geochemistry of immobile elements were done in samples of these reefs to figure out the provenance of the mud that arrives there. In order to do this, samples of riverine sediment from the Jequitinhonha, Santo Antônio, João de Tiba, and Buranhém rivers as well as from the Barreiras Group's sedimentary cliffs were taken. The Jequitinhonha River was found to be the most significant mud source. This was supported by the mineral fingerprints of smectite and biotite, the concentrations of immobile trace elements such as Zr, Hf, Nb, Ta, and Th, and the distribution and ratio of rare-earth elements. These findings are supported by the prevailing north-to-south drift that occurs in the region due to the northeasterly trade winds and waves that blow throughout the region for the vast majority of the year. The findings have significant implications for hydrographic basin management and the protection of reef benthic populations.

KEYWORDS: sedimentary provenance; geochemistry; reefs; Royal Charlotte Bank

INTRODUCTION

Mud in marine environments is associated with terrigenous continental sources or carbonate factories (Hay *et al.* 1988, Scholle and Ulmer-Scholle 2003, Hanebuth *et al.* 2015). They are deposited in low-energy areas and are linked to both short- and long-term processes (Swift and Thorne 1991).

Short-term processes, such as the biogenic activities of invertebrates and cyanobacteria, as well as direct physical-chemical control and precipitation, may have controlled the carbonate factory. Carbonate is very sensitive to changes in the environment. Water temperature, light, the amount of Ca and CO_3 ions in the water, and mud suspended in the water column are the main factors that affect carbonate (Black 1933, Scholle and Ulmer-Schole 2003, Boggs 2012). Long-term processes are a dynamic balance between the relative sea level, erosional base level, the input of terrigenous sediments by rivers, and space for the accommodation of sediments (Swift and Thorne 1991, Bastos *et al.* 2015).

The southern coast of Bahia, Brazil, borders a broad sector of the Brazilian continental shelf: the Royal Charlotte Bank (Fainstein and Summerhayes 1982, Palma 1984). The bank is a typical mixed carbonate-terrigenous sedimentary shelf, similar to the Abrolhos Bank, further south (Bastos *et al.* 2015, D'Agostini *et al.* 2019). The cross-shore transition of facies is characterized by a nearshore terrigenous domain, surrounding the coastal reefs. The middle and outer shelf is composed of a carbonate domain, constituted mainly by rhodolite boundstone, beside algae packstone, and grainstone (*maerl*) (Leão *et al.* 2006, Dominguez and Bittencourt 2012, Negrão *et al.* 2021) (Fig. 1).

The most diverse coral reef communities occur as detached bank reefs, not deeper than 20 m (Leão *et al.* 2003). Other benthic communities develop in intertidal zones, on beach rock belts, or on marine abrasion surfaces (attached bank reefs), over older communities, developed under eustatic sea level rise, about 7200–5700 years before the present (Leão *et al.* 2003, Martin *et al.* 2003). Recent benthic communities are sparse and less diverse, with small colonies of hard coral species, besides green algae, echinoderms, sponges, and so on (Fig. 2) (Laborel-Deguen *et al.* 2019).

Similar to other communities in Brazil, the coral communities in Royal Charlotte Bank are made up of ancient forms with ties to the Caribbean that have adapted to turbidity in the water (Segal *et al.* 2008, Zilberberg *et al.* 2016). Nearly 60% of reef species are found in high turbidity conditions (Mies *et al.* 2020). Most terrigenous mud comes to the reef areas of the Royal Charlotte Bank during the rainy seasons, and sediment is deposited around or above corals in deeper areas (Fig. 2).

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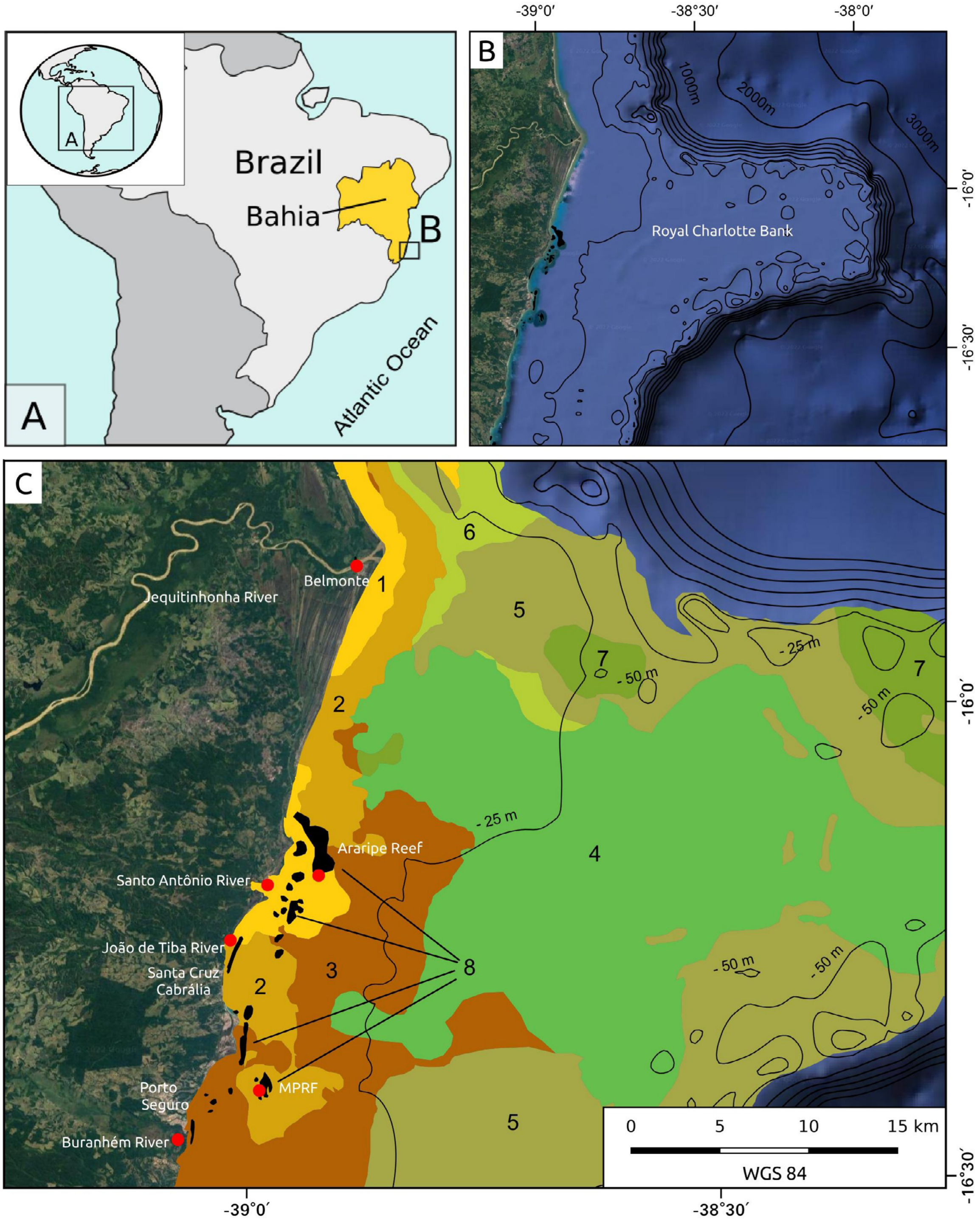
The aim is to identify the provenance of terrigenous mud in the detached coastal reef banks of Royal Charlotte Bank, such as the Recife de Fora and Araripe reefs. In the provenance method, X-ray diffractometry (XRD) and the geochemistry of some major traces of immobile elements, such as rare-earth elements (REEs), were used.

Rivers could potentially supply most of the recent muddy material for the shelf (Fig. 1). The Jequitinhonha River, further north, constitutes the largest hydrographic basin and drains areas of the São Francisco Craton, with Archaean and Paleoproterozoic rocks, besides small contributions of Neogene cover in the coastal zone of the Barreiras Group. João de Tiba, Santo Antônio, and Buranhém River drain areas with Neoproterozoic crystalline rocks of the Araçuaí Belt and Neogene covers of the Barreiras Group (Soares *et al.* 2007, Dominguez and Bittencourt 2012).

The nearshore drift is influenced mainly by NE trade winds and N-NE waves (Dominguez *et al.* 2009, Soutelino



Sources: Brazilian Navy, Directorate of Hydrography and Navigation, Dominguez and Bittencourt (2012), Negrão *et al.* (2021), and D'Agostini *et al.* (2019). 1: terrigenous sand; 2: terrigenous mud; 3: terrigenous sandy mud with carbonate traces; 4: algal sandy packstone; 5: rhodolite boundstone and algal gravel packstone; 6: mixed terrigenous and carbonate sand/gravel; 7: algal gravel packstone; 8: reefs. The geological marine bottom map was prepared from own data and third-party data.

Figure 1. Geography and surface geology of the Royal Charlotte Bank: (A) Location in South America and Brazil. (B) General aspect of the seabed morphology in the region. (C) Surface geological units and sampling sites (red dots).

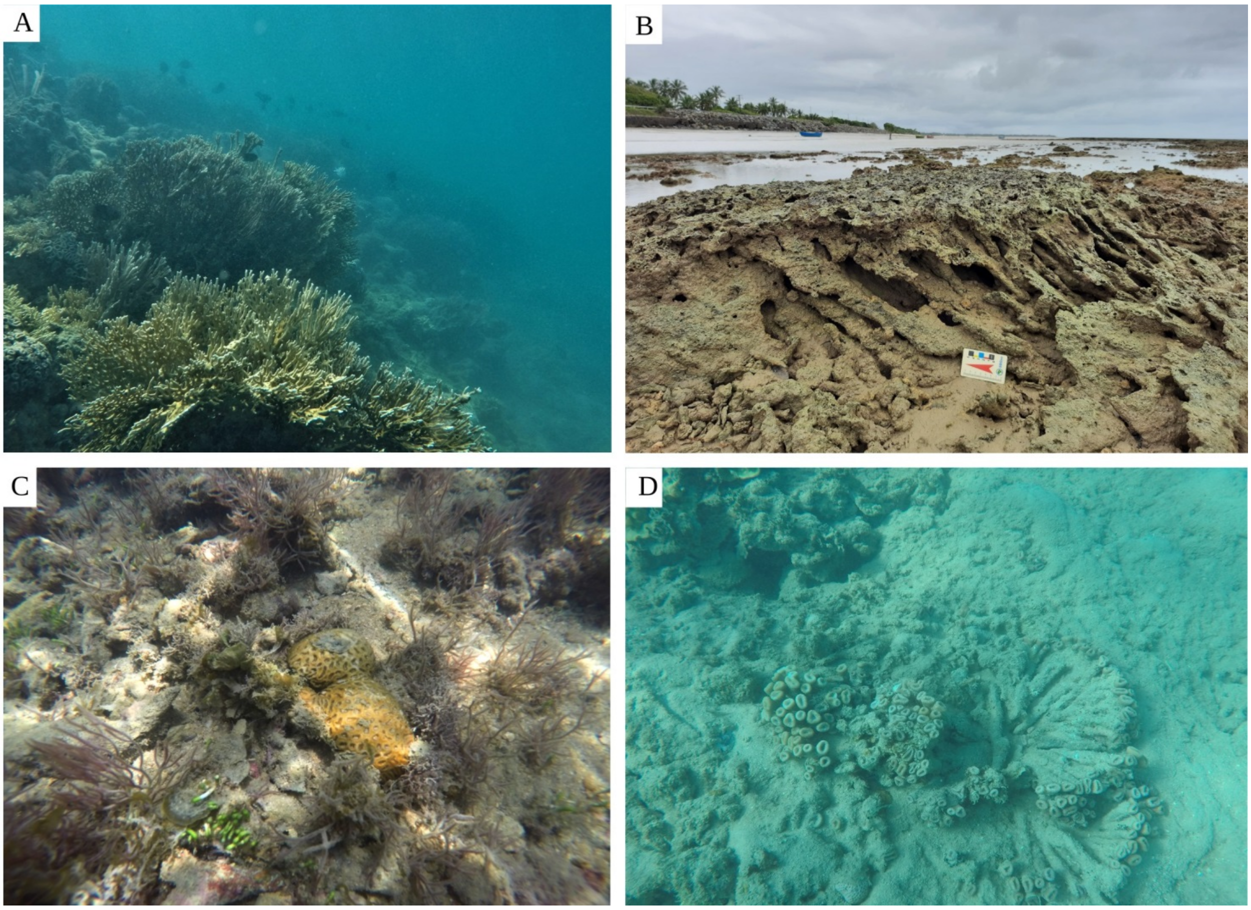


Figure 2. (A) Coral colonies in the Recife de Fora region with a high diversity of hard coral species. (B) Marine abrasion surface on older Holocene coral communities. (C) Beach rocks with sparse coral species. (D) Aspect of terrigenous mud along the reef shores.

et al. 2013). During winter, polar fronts with S and SE winds and waves influence dynamics, surface currents, and sediment drift (Dominguez *et al.* 2009).

Reef areas are under pressure and experiencing impacts from predatory tourism and uncontrolled fishing. The reefs in the region suffer from pollution from a variety of sources, including eucalyptus monoculture and other agricultural products, human sewage, and naval operations (Soares *et al.* 2021). Thus, it is necessary to improve knowledge of mud sources to assist management policies for hydrographic basins and the coastal zone.

MATERIALS AND METHODS

Two areas were chosen by the Coral-Vivo Research Network for monitoring: the Araripe Reef, further north, and the Marine Park Recife de Fora (MPRF), a full-protection conservation unit (Fig. 1).

Samples were collected in the main regional rivers in order to test possible sources, such as Jequitinhonha, João de Tiba, and Buranhém. Also, samples of the Barreiras Group were taken from coastal cliffs and escarpments on river slopes.

Marine mud samples were collected using sediment traps. It was fixed to the bed by iron bars. Sediment traps were done with PVC tubes, 75 mm in diameter by 250 mm in height (Storlazzi *et al.* 2011). Three traps were installed on each reef, not more than 100 m apart. Traps are kept in place for at least 2 months, and sediments were collected three times during 1 year, from 2019 to 2020. For each reef, the three samples made up a single composite sample. Samples washed with fresh water were dried, weighed, and sieved, and the mud fraction (< 63 μm) was separated for the analysis.

Mineralogical data were obtained by XRD. XRD measurements are performed on a diffractometer model Ultima IV (RIGAKU, Japan) with Bragg-Brentano geometry ($\theta/2\theta$), and a zirconium filter. The angular range used was 2θ 4.00 up to 70.00, with steps of 0.02, and molybdenum radiation (0.07093 nm).

X-ray data were processed using the free software PROFEX (Doebelin and Kleeberg 2015). The Rietveld refining technique is used in the software to identify the mineral phases and their relative abundances. By comparing the integrated intensities of the refined diffraction peaks for each mineral phase to a calibration standard or reference standard after refinement, it is possible to determine the fractional abundance of each mineral phase. To assess the quality of the refinement, the values of $\chi^2 \leq 5$ were considered. The observed and calculated X-ray patterns were analyzed graphically (Toby 2006).

While XRD is a powerful tool, it is important to acknowledge that there may be some limitations in differentiating between certain clay minerals. In particular, the distinction between illite and smectite can be challenging based on XRD data alone. Despite these limitations, the XRD data provide valuable information on the mineralogy of the sample and the presence of other mineral phases. It is important to recognize the potential limitations of XRD data and to interpret the results with these limitations in mind. Nonetheless, the XRD data have provided valuable insights into the mineralogy of the sample and the geological processes that have shaped it.

The geochemical approach involved using some major elements, such as Al, Ca, Na, K, Fe, and Mg, and immobile trace elements, such as Zr, Hf, Nb, Ta, and Th, as well as REEs. An amount of 20 g of each sample was milled and sent to the SGS-GEOSOL Laboratories, Brazil. Major elements were performed by inductively coupled plasma atomic emission spectrometry (ICP-AES). Inductively coupled plasma mass spectrometry (ICP-MS) was used to analyze REEs and other trace immobile elements. The major elements are expressed in weight percentage for oxides (e.g., Al_2O_3, CaO, Na_2O, and K_2O) (Table 1), while REE is expressed in parts per million (ppm) (Table 2). Iron contents are expressed as the total iron (Fe_2O_{3t}). Organic matter and volatiles were burned in muffle at 1,000°C, and their weight percentages are expressed as loss on ignition values (LOI, Table 1). Results were recalculated on an anhydrous basis.

Regarding the REE behavior in environments and surface processes such as diagenesis and weathering, it is known that their concentrations in water are extremely low and that their potential for interaction with terrigenous sediments after they have been deposited is equally negligible (Taylor and McLennan 1985, Rollinson and Pease 2021). In analyses on the provenance of terrigenous materials, REE concentrations are therefore considered to represent the sources of sediments on the continent (Rollinson and Pease 2021).

For the approach with REE, the normalized reference North American Shale Composite (NASC) was utilized (Gromet *et al.* 1984). To compare the concentrations of REE, ratios between light rare-earth elements (LREEs), heavy rare-earth elements (HREEs), and Eu anomalies were tested. Often, elements in the series with low atomic numbers are referred to as LREEs, whereas those with larger atomic numbers are referred to as HREEs (Rollinson and Pease 2021). REEs typically behave similarly during igneous petrogenesis, with LREEs tending to concentrate in liquid phases in systems containing both liquids and crystals. Conversely, HREEs tend to remain in mineral phases that are more refractory to melting, such as garnet and hornblende (Wilson 1989).

Eu anomalies are often measured using the geometric mean Eu_N/Eu^*, where Eu^* is $\sqrt{[(Sm_N).\ (Gd_N)]}$ (Taylor and McLennan 1985) and "N" is the normalization of the NASC standard. The formation of new diagenetic phases can lead to changes in the Gd content of marine sediments due to precipitation from interactions between seawater and porewater (Lawrence *et al.* 2006). Sm and Tb, having similar geochemical properties to Gd, are less susceptible to precipitation from seawater (Sholkovitz 1990). By using Tb values instead of Gd, the analysis can provide a more accurate and reliable assessment of the REE content in the sediment and associated geological processes. In this sense, a test was performed substituting Gd for Tb in the Eu_N/Eu^* ratio calculation to get a more accurate estimate of the REE content. According to the results, there were no appreciable differences in Eu_N/Eu^* ratios after the substitution of Gd by Tb.

The classification of geological seabed units (Fig. 1) is adapted from the proposals of Folk (1954) for terrigenous sediments and Dunham (1962) for carbonate sediments.

RESULTS

XRD analysis indicates that the predominance of kaolinite in the rivers is above 85%. Smectite appears in the Jequitinhonha River, Santo Antônio River, and João de Tiba River, ranging from 0.3 to 2.9%, while biotite in

Table 1. Major element chemical analyses (% wt) of the mud of the river, reefs, and Barreiras Group, measured by ICP-AES.

Sample	Locality	SiO_2	TiO_2	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	Na_2O	K_2O	P_2O_5	LOI
RFO 1	MPRF	31.13	0.71	17.22	5.80	0.05	3.84	39.47	0.43	1.08	0.28	31.09
RFO 2	MPRF	32.75	0.80	18.69	5.61	0.04	3.67	36.69	0.40	1.10	0.25	29.94
RFO 3	MPRF	31.33	0.73	17.71	5.95	0.05	3.85	38.63	0.37	1.09	0.28	30.97
RFO 4	MPRF	32.16	0.71	19.33	6.37	0.05	3.69	35.87	0.43	1.09	0.30	30.73
ARA 1	Araripe Reef	36.12	0.79	22.62	7.59	0.07	3.59	26.91	0.45	1.61	0.26	27.42
ARA 2	Araripe Reef	43.24	0.86	18.76	6.22	0.07	3.09	26.12	0.38	0.99	0.28	25.16
ARA 3	Araripe Reef	38.96	0.84	24.09	7.83	0.07	3.20	22.55	0.36	1.82	0.27	24.9
BUR 01	Buranhém River	57.67	1.16	21.87	8.96	0.03	1.64	4.12	2.87	1.41	0.27	21.01
BUR 02	Buranhém River	53.74	1.29	16.37	8.23	0.04	1.89	16.11	0.62	1.35	0.35	18.89
SA 01	Santo Antônio River	78.16	1.06	14.00	3.21	0.01	0.46	0.36	0.85	1.81	0.08	10.84
JEQ 01	Jequitinhonha River	63.61	0.95	21.82	7.16	0.08	1.30	1.27	0.81	2.86	0.14	10.28
JT 01	João de Tiba River	46.98	0.88	7.93	3.56	0.04	2.71	32.45	4.20	1.06	0.20	29.58
BA 06	Barreiras Group	55.44	1.32	40.21	2.44	0.01	0.06	0.09	0.05	0.14	0.24	10.19
BA 05	Barreiras Group	68.07	1.03	26.56	2.32	0.01	0.32	0.36	0.11	1.13	0.09	12.24
BA 02B-A1	Barreiras Group	58.55	1.22	38.65	0.96	0.01	0.13	0.08	0.13	0.18	0.08	13.13
BA 03-A	Barreiras Group	57.74	1.17	36.03	3.55	0.01	0.24	0.09	0.35	0.73	0.08	12.32

Table 2. Trace immobile elements and ratios of rare-earth elements (ppm) in the mud of rivers, reefs, and the Barreiras Group, measured by ICP-MS.

Sample	Locality	Zr	Hf	Nb	Ta	Th	La	Ce	Nd	Sm	Eu	Gd	Tb	Dy	Er	Yb	Lu	Y	La/Yb	La/ Sm	Gd/ Yb	Eu/ Eu
RFO 1	MPRF	93.00	2.29	17.28	0.90	11.00	28.80	57.30	20.60	3.60	0.71	3.12	0.42	2.37	1.32	1.10	0.09	12.24	26.18	8.00	2.84	0.97
RFO 2	MPRF	108.00	2.58	17.15	1.07	12.00	30.70	60.90	22.30	3.90	0.77	3.38	0.46	2.54	1.35	1.20	0.05	12.30	25.58	7.87	2.82	0.97
RFO 3	MPRF	91.00	2.49	16.26	2.01	11.60	28.40	56.40	21.00	3.90	0.72	2.99	0.45	2.40	1.27	1.10	0.05	12.03	25.82	7.28	2.72	0.97
RFO 4	MPRF	96.00	2.50	14.75	1.24	12.00	29.90	59.40	22.00	4.10	0.84	3.34	0.47	2.60	1.35	1.20	0.05	12.17	24.92	7.29	2.78	1.04
ARA 1	Araripe Reef	105.00	2.72	17.50	1.35	14.10	35.80	70.40	26.80	5.10	1.03	4.28	0.58	3.20	1.73	1.50	0.05	15.75	23.87	7.02	2.85	1.01
ARA 2	Araripe Reef	434.00	10.85	18.29	1.34	43.40	87.10	175.20	67.00	11.30	1.08	7.43	0.92	4.41	1.91	1.80	0.18	19.86	48.39	7.71	4.13	0.54
ARA 3	Araripe Reef	114.00	3.00	15.63	1.15	15.40	38.80	77.40	28.30	5.20	1.05	4.47	0.62	3.45	1.91	1.60	0.20	16.57	24.25	7.46	2.79	1.00
BUR 01	Buranhém Riv.	987.00	25.03	23.24	1.53	87.80	164.10	340.10	129.60	19.90	1.36	12.96	1.52	7.46	3.70	3.30	0.33	35.14	49.73	8.25	3.93	0.39
BUR 02	Buranhém Riv.	2214.00	57.13	27.39	1.88	206.90	363.10	745.70	296.10	46.60	1.91	28.17	3.16	13.96	5.74	5.20	0.72	59.12	69.83	7.79	5.42	0.24
SA 01	S. Antônio Riv.	1575.00	40.24	25.53	2.31	46.80	81.00	160.50	63.40	11.10	0.98	8.65	1.27	7.18	4.03	4.40	0.54	37.45	18.41	7.30	1.97	0.46
JEQ 01	Jequitinhonha Riv.	402.00	11.00	19.60	1.84	26.10	64.30	131.00	53.00	10.30	1.75	8.49	1.27	7.16	3.80	3.70	0.56	33.91	17.38	6.24	2.29	0.86
JT 01	João de Tiba Riv.	1638.00	43.78	14.89	1.21	69.20	134.60	284.50	114.40	19.60	1.19	13.50	1.74	8.86	4.74	4.70	0.70	43.27	28.64	6.87	2.87	0.33
BA 06	Barreiras Group	1518.00	39.73	37.23	1.69	271.10	479.50	957.00	343.40	45.60	2.47	22.95	2.30	8.72	2.74	2.20	0.26	30.80	217.95	10.52	10.43	0.35
BA 05	Barreiras Group	260.00	7.25	35.61	2.81	38.60	125.10	365.60	170.70	32.30	6.72	19.76	2.46	11.85	4.27	3.40	0.40	39.44	36.79	3.87	5.81	1.22
BA 02B	Barreiras Group	913.00	24.85	23.03	1.99	102.50	190.20	358.50	120.90	18.10	1.00	12.35	1.50	7.41	3.39	3.30	0.57	32.34	57.64	10.51	3.74	0.31
BA 03	Barreiras Group	228.00	7.75	31.92	7.66	30.00	130.80	241.90	79.90	13.10	2.04	8.01	1.21	4.96	2.12	1.90	0.44	17.54	68.84	9.98	4.22	0.91
NASC	Reference standard	0.00	0.00	0.00	0.00	0.00	31.10	67.03	30.40	5.98	1.25	5.50	0.85	5.54	3.28	3.11	0.46	0.00	9.99	5.20	1.77	1.91

the Jequitinhonha River is nearly 6% (Fig. 1 and Table 1). Although illite is present in all rivers, Buranhém has the greatest concentration of about 13.5%. Barreiras Group cliffs have kaolinite up to 86%, illite up to 9.8%, and goethite around 1.5%. Regarding the mud in the Araripe Reef and MPRF, smectite is around 1–3.67%, while biotite is around 1.2%. Traces of illite and calcite are detected (Fig. 3). Reef sediments contain no goethite.

Regarding major elements, the Jequitinhonha River, Santo Antônio River, Buranhém River, and Barreiras Group are enriched in Al_2O_3, compared with reefs and João de Tiba River (Table 1). Ternary diagrams of molecular proportion (McLennan *et al.* 1993) defined two sample trends (Fig. 4). The first is related to the Barreiras Group and is close to the Al_2O_3 vertex. The other rivers shown, Jequitinhonha and Buranhém, have mud enriched in Al_2O_3 that becomes depleted toward the mud of the reef. Compared with almost all rivers, reefs contain higher amounts of alkalis, such as CaO, Na_2O, and K_2O, although it is likely that Ca makes the greatest contribution due to carbonate production in these areas.

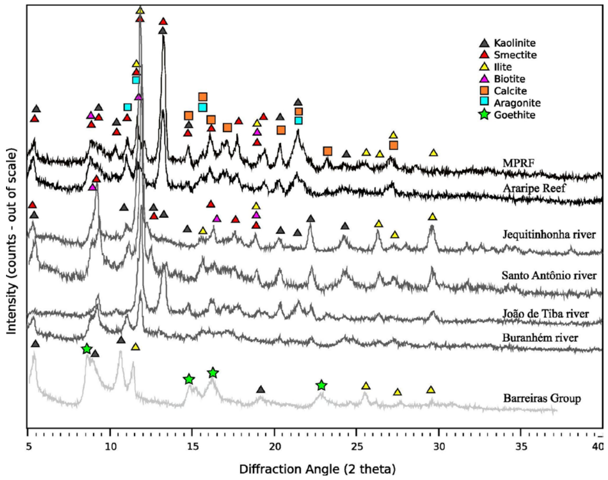


Figure 3. Diffraction diagram (MoKα, < λ > = (0.07093 nm) exhibiting the peaks corresponding to the main mineral phases present in the mud of reefs, rivers, and sedimentary cliffs of Barreiras Group.

Among the reefs, the Araripe reef, further north, is more enriched in Al_2O_3 and impoverished in alkalis than MPRF. Among rivers, João de Tiba is the most enriched in alkalis and impoverished in Al_2O_3 (Fig. 4).

In bivariate diagrams, the concentrations of minor and trace immobile elements in the reefs, such as Hf, Nb, Ta, Th, La, and Y versus Zr, show that they are close to those found in the Jequitinhonha River (Fig. 5). In contrast, rivers south of Jequitinhonha have higher concentrations of these elements. With the exception of Nb, the Santo Antônio River and João de Tiba River have similar concentrations of immobile elements (Fig. 5). The Barreiras Group displays a scattered distribution with a high variation in immobile element concentrations (Fig. 5).

The REE was normalized to the NASC standard reference, which made it possible for the two groups to be distinguished. The first group includes the mud of MPRF, Araripe Reef, and Jequitinhonha River (Fig. 6). It has a La/Yb of 23.87–26.18, a La/Sm of 6.24–8.00, and a Gd/Yb of 2.72–2.84 (Table 2). The Eu anomalies (Eu_N/Eu^*) vary from 0.97 to 1 (Table 2). The REE pattern has LREE values ranging from 0.7 to just above 1. The HREE group exhibited a less fractionation, with values primarily in the reef muds falling below the unit. There is no negative Eu anomaly present. Certain mud samples taken from the reef exhibit significant fractionation of Lu (Fig. 6).

The second group includes mud from rivers further south of Jequitinhonha and one sample from Araripe Reef (ARA 2) (Fig. 6). The La/Yb ratios ranged from 28.64 to 69.83 and La/Sm from 6.87 to 8.25. Among the HREE, the Gd/Yb ratios ranged from 1.97 to 5.42. Eu_N/Eu^* ratios are from

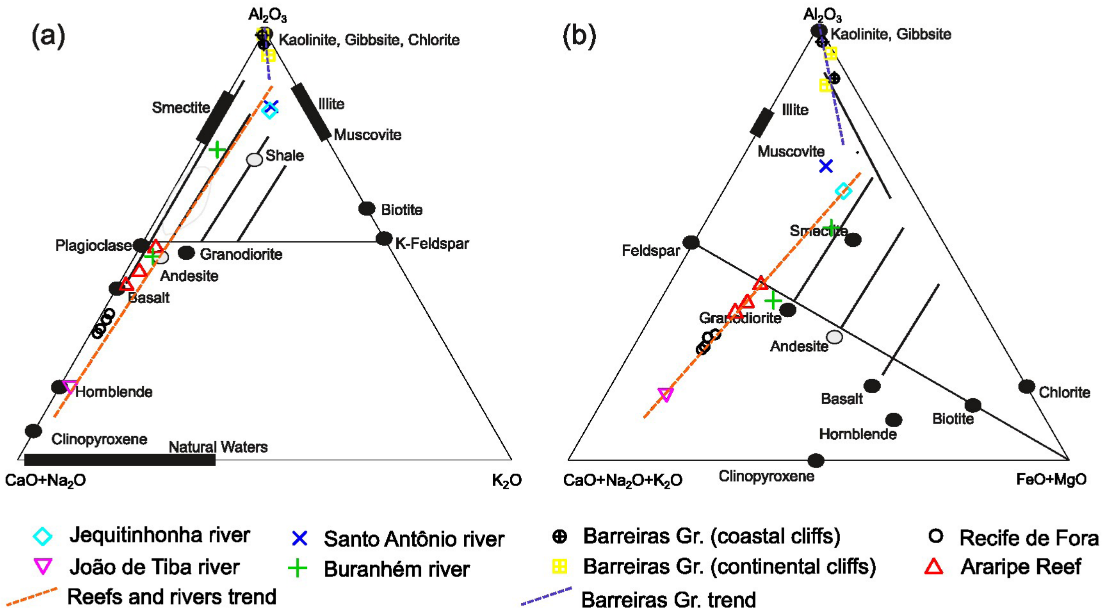


Figure 4. Ternary plots of McLennan *et al.* (1993), using immobile Al_2O_3 and mobile elements such as CaO, Na_2O, K_2O, Fe_2O_3, and MgO. Two distinct trends are observed on both diagrams. Samples from the Barreiras Group are near the Al_2O_3 vertex, while the Jequitinhonha, Santo Antônio, and Buranhém rivers form a trend that comes toward reef samples, enriched in alkalis.

0.24 to 0.54. Sample ARA 2 from Araripe Reef is closest to this group, with a La/Yb ratio of 48.39, a La/Sm of 7.71, a Gd/Yb of 4.13, and an Eu/Eu of 0.54 (Table 2). The REE concentrations and distribution of the Barreiras Group are also similar to the second group. The La/Yb ratio is 36.79–217.95, the La/Sm ratio is 3.83–10.52, and the Gd/Yb ratio is 3.74–10.43. The Eu_N/Eu^* ranges from 0.35 to 1.22 (Table 2). The REE pattern showed concentrations of LREE two times higher than NASC. Compared with the first group, the second group shows a Eu anomaly and higher fractionation of HREE in the distribution of REE normalized to NASC (Fig. 6).

DISCUSSION

The mineral assemblage found in the MPRF and Araripe Reef is similar to those observed in rivers further north, such as Jequitinhonha, Santo Antônio, and João de Tiba, characterized by the presence of kaolinite, smectite, illite, and biotite. However, the presence of a small amount of illite in the reefs suggests that the Buranhém River may have had some influence on the region, given that it has the greatest concentration of this mineral.

Ternary diagrams of molecular proportion (McLennan *et al.* 1993) reinforce the provenance possibilities observed in the mineralogy (Fig. 4), considering the drift trends

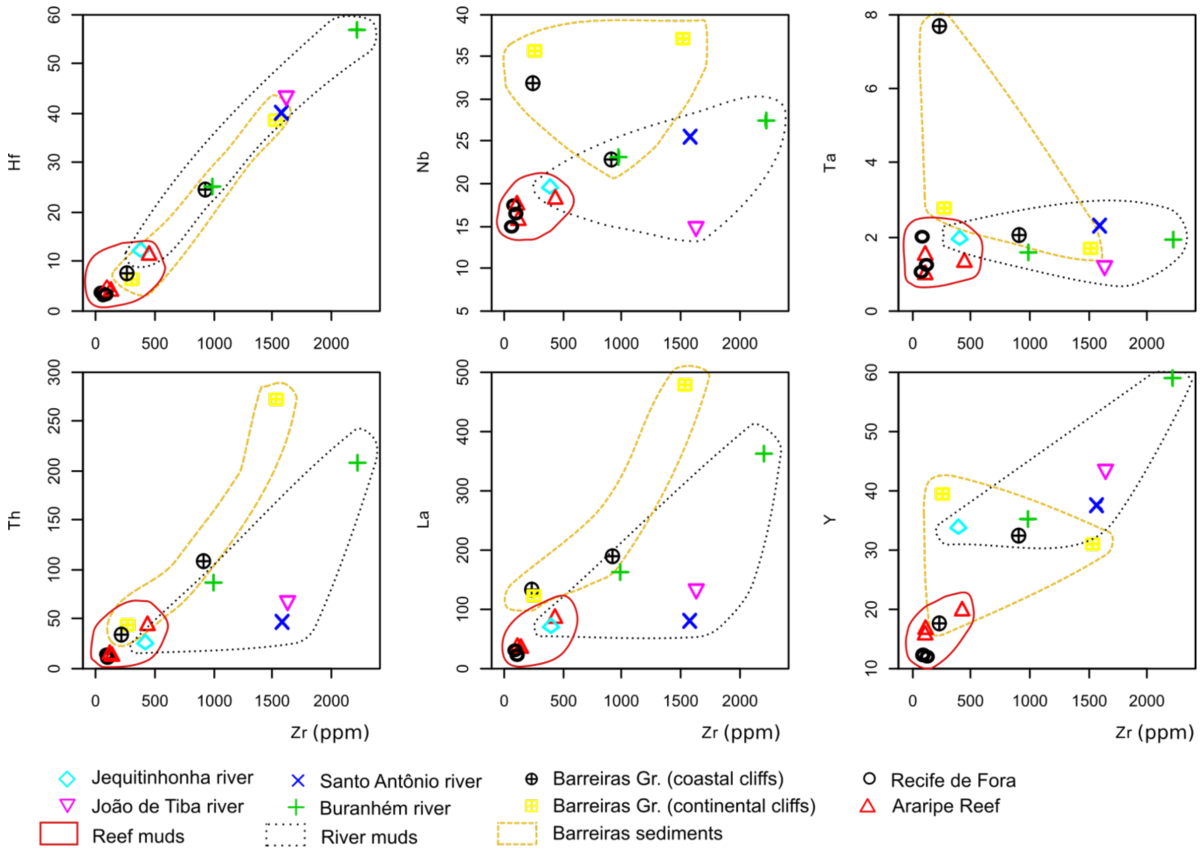


Figure 5. Bivariate correlation diagrams of Zr (ppm) versus other immobile elements in ppm. In many cases, the reef samples form a cluster, close to the sample from the Jequitinhonha River (except for the Y). In contrast, samples from the Barreiras Group and other rivers are scattered.

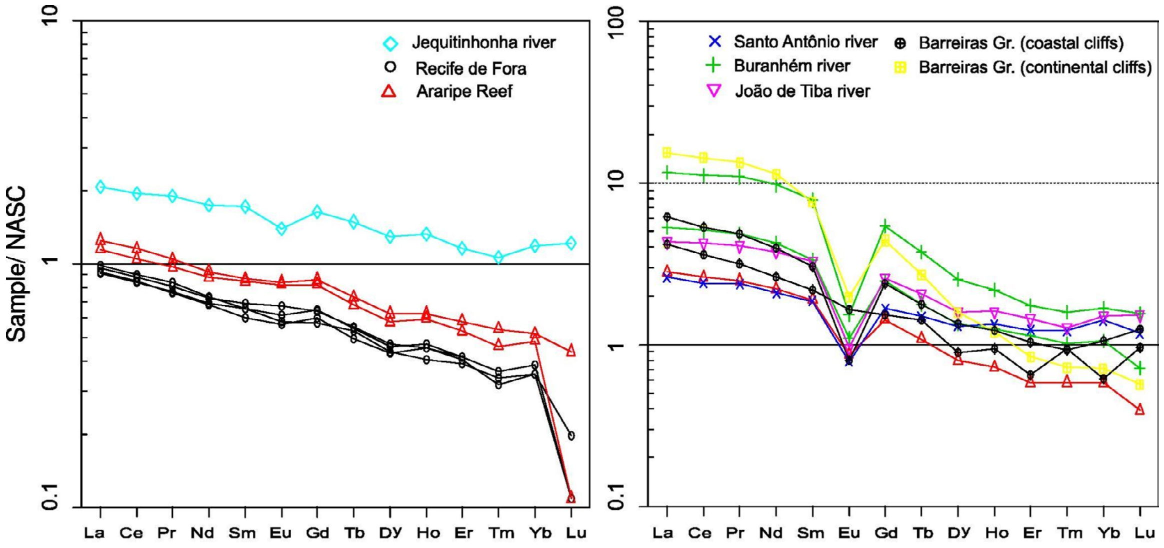


Figure 6. Distribution of REEs normalized to NASC (Gromet *et al.* 1984).

influenced by waves and winds in fair weather (N-NE) and polar fronts (S-SE). Immobile elements are not soluble by fluids in alteration zones and surface processes (MacLean and Barrett 1993), reflecting their presence and proportions in the source area. Aluminum is not soluble in surface waters, except when the pH is high, and it usually accumulates in clays on top of weathering mantles. When present in sediments, they reflect the transport of clays from the source area (McLennan *et al.* 1993). Alkalis are highly mobile and are rapidly removed from aqueous systems during the weathering process (Albarède 2009). In mud, they are present mainly in clay minerals, micas, and carbonate.

Mud from the Jequitinhonha and Buranhém rivers exhibits higher amounts of Al_2O_3 when compared with the mud in the reefs. Increased Al_2O_3 concentrations may be related to riverine clay, which may indicate the drift process. In this sense, trade winds would be responsible for the drift of clays from north to south during the summer, transferring the Jequitinhonha River's mud to the reefs. In contrast, the greater concentrations of Al_2O_3 in the Buranhém River, compared with the reefs, may suggest its role in the sedimentary drift. In this way, clays from the Buranhém River might be supplied to reefs during the winter by polar fronts. Regarding the alkalis, the reef samples contained a significant amount of CaO, which may be connected to the carbonate production and the rise in total alkalis in the ternary diagram. The content of Al_2O_3 and alkalis in the João de Tiba River could represent local conditions, with no contribution to the mud on reefs. The ternary diagram shows a clear trend in the amounts of Al_2O_3 and alkalis present in the samples from the Barreiras Group, suggesting that it did not contribute directly to the mud supply on the marine platform.

Bivariate diagrams of immobile elements indicated concentrations in MPRF and Araripe Reef very close to those in the Jequitinhonha River, lower than the rivers south of Jequitinhonha. Also, the samples from the rivers south of Jequitinhonha and the Barreiras Group show that all of the other immobile elements, except for Hf, are spread out. A linear or polynomial relationship would be expected if sediments were carried to the reef and deposited there by any of these rivers. In this regard, the proximity of the samples from the reefs and the sample from the Jequitinhonha River indicates the sedimentary origin of the mud.

The ratios of REE and their concentrations, normalized to the NASC pattern, support previous data and divide the terrigenous muds into two groups. The first group includes the mud of the Jequitinhonha River, the mud of the MPRF, and almost all samples of the Araripe Reef. The second group includes the muds of the other rivers further south (Santo Antônio, João de Tiba, and Buranhém), one sample from Araripe Reef (ARA 2), and the Barreiras Group. The sample from Araripe Reef in this group reinforces the possibility of south-north drift forced by winter fronts, carrying mud from the Buranhém River to the reefs.

REEs have low mobility during weathering and in the sediment cycle, and their concentrations in river and sea waters are typically very low (Rollinson and Pease 2021). This means that, once weathered rocks are on the surface, they will transfer their REE signatures to soils and sediments, primarily in the clay fraction (Fleet 1984, Cullers *et al.* 1987, McLennan 1989).

As a result, the REE distribution in fine terrigenous sediments could reflect the average composition of the regional upper crust (Taylor and McLennan 1985).

A negative Eu anomaly in post-Neoarchean source rocks indicates that minerals such as plagioclase and clinopyroxene did not melt during rock formation. This anomaly suggests that the rocks were created by melting the sublithospheric mantle in subduction zones, which resulted in the retention of plagioclase in the source (Wilson 1989, Martin *et al.* 2005). In contrast, the Eoarchean to Mesoachean continental crust was formed from magma originating from the melting of the oceanic lithosphere during subduction or from its metamorphosed form, such as eclogites. In this case, plagioclase does melt, resulting in higher Eu concentrations than Sm and Gd, which reduces the negative anomaly in the distribution of normalized REEs. Currently, this type of melting only occurs in very specific situations, such as when adakites are formed through the subduction of young and highly hot oceanic lithosphere (Wilson 1989, Rollinson and Pease 2021).

Differences in REE patterns between the two groups of muds, with or without slight Eu anomalies, could be due to variations in the composition of the sources of continental sediments. For example, rivers south of Jequitinhonha drain areas with rocks from Neoproterozoic to Phanerozoic. In contrast, the Jequitinhonha River exhibits a distinct REE pattern, probably due to its passage over rocks from the São Francisco Craton. This distinct pattern can be attributed to the abundant presence of Archean rocks in the region, which brings the concentrations of Eu closer to those of Sm and Gd, resulting in a diminished negative anomaly pattern when compared with other rivers.

Regarding heavy REEs, the more pronounced fractionation pattern in the reefs compared with the rivers provides evidence for the preferential retention of heavy minerals, particularly zircon, in the silt fractions (Sholkovitz 1990).

CONCLUSION

The Jequitinhonha River is proposed as a major source of most of the terrigenous muds in the Araripe and MPRF reefs, according to the provenance method based on XRD mineralogy and mainly by the geochemistry of immobile elements. The Buranhém River may function as a seasonal source of muds when winter fronts are active.

The results may support the management of the main hydrographic basins that contribute to marine sedimentation and the protection of benthic communities in the region.

ACKNOWLEDGMENTS

We thank the Coral Vivo Institute, Petrobras (through the Petrobras Environmental Program), the Arraial d'Ajuda Eco Parque, and the Directorate of Hydrography and Navigation (DHN) of the Brazilian Navy, for the data provided.

ARTICLE INFORMATION
Manuscript ID: 20220083. Received on: 02 NOV 2022. Approved on: 25 JULY 2023.

How to cite: Turbay C.V.G., Orlando M.T.D., Lacerda C.H.F., Duarte E.B. 2023. *Brazilian Journal of Geology*, **53**(3):e20220083. https://doi.org/10.1590/2317-4889202320220083

C.V.G.T.: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Software, Supervision, Validation, Visualization, Writing – original draft, Writing - review & editing. M.T.D.O.: Data curation, Funding acquisition, Investigation, Methodology, Review & editing. C.H.F.L.: Funding acquisition, Data curation, Formal analysis, Investigation, Methodology, Review & editing. E.B.D.: Formal analysis, Investigation, Methodology, Review & editing.

Conflict of interests: The authors declare no conflict of interests.

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