Control of oceanic circulation on sediment distribution in the Southwestern Atlantic margin (23°S to 55°S)

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Abstract. In this study, we interpret the role played by ocean circulation in sediment distribution on the Southwestern Atlantic margin using radiogenic Nd and Pb isotopes. The latitudinal trends for Pb and Nd isotopes reflect the different current systems acting on the margin. The utilization of the sediment fingerprinting method allowed us to associate the isotopic signatures to the main oceanographic features in the area. We recognized differences between Nd and Pb sources to the Argentinean shelf (carried by the flow of Subantarctic Shelf Water) and slopes (transported by deeper flows). Sediments from Antarctica extend up to the Uruguayan margin, carried by the Upper- and Lower Circumpolar Deep Water. Our data confirm that, for shelf and intermediate areas (the upper 1,200 meters), the transfer of sediments from the Argentinean margin to the North of 35°S is limited by the Subtropical Shelf Front and the basin-wide recirculated Antarctic Intermediate Water.

On the southern Brazilian inner and middle shelf, it is possible to recognize the northward influence of the Río de la Plata sediments carried by the Plata Plume Water. Another flow responsible for sediment transport and deposition on the outer shelf and slope is the southward flow of the Brazil Current. Finally, we propose that the Brazil-Malvinas Confluence and the Santos Bifurcation act as boundaries of geochemical provinces in the area. A conceptual model of sediment sources and transport is provided for the Southwestern Atlantic margin.

30 1 Introduction

Physical and oceanographic processes, ranging from ocean current circulation, river discharge, marine fronts, wind patterns, and climate variability, have a crucial impact on sediment transport variability and fate (Storlazzi and Reid, 2010; Qiao et al., 2020). The Southwest Atlantic margin is located in a key region concerning global ocean circulation and is an excellent example of a complex interaction of physical forcings in the sediment variability. Hydrodynamics is strongly influenced by

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the Río de la Plata (RdIP) outflow, the second largest river basin in South America (discharge estimated at 23,000 m³ s -1 (Depetris and Griffin, 1968) and by the encounter of subtropical and subantarctic water masses, transported by the Brazil and Malvinas currents, worldwide known as Brazil-Malvinas Confluence (BMC) and its shelf extension, the Subtropical Shelf Front (STSF). At the BMC, water masses are transported eastwards as part of the southern limb of the basin-wide Anticyclonic Atlantic Subtropical Gyre (Boebel et al., 1997; Schmid et al., 2000). Further north, the westward flow of the gyre reaches the South American margin, where it splits into two branches, forming the Santos Bifurcation (SB; Boebel et al., 1999a; Legeais et al., 2013). The southward branch of the bifurcation feeds the Brazil Current (BC), flowing south until its eastward displacement at the BMC (Schmid et al., 2000). The northward branch flows as the Intermediate Western Boundary Current (IWBC) (Legeais et al., 2013), which is an important mechanism for the transport of the Antarctic Intermediate Water towards the northern hemisphere (Boebel et al., 1999b).

Even though the distribution of sediments, sources, and transport along the Southwestern Atlantic margin is known since the '50s (Teruggi, 1954; Etchichury and Remiro, 1960, 1963; Berkowsky, 1978; Kowsmann and Costa, 1979; Urien and Martins, 1979; Potter, 1984; Berkowsky, 1986) and recently deepened based on geochemical and mineralogical methods (Campos et al., 2008; de Mahiques et al., 2008; Razik et al., 2013; Nagai et al., 2014a) important issues regarding hydrological controls are still unresolved.

Radiogenic Nd and Pb isotopes are efficient tools in studying sediment transport in continental margins (Weldeab et al., 2002; Kessarkar et al., 2003; Maccali et al., 2012; Maccali et al., 2018; Subha Anand et al., 2019). A recent synthesis of potential sources and water masses transport in the South Atlantic was provided by Beny et al. (2020). In that study, the authors combined grain size, clay mineralogy, and Nd, Pb, and Sr isotopes to propose a deep-water mass evolution for the last ca. 30,000 years in the region. The first regional characterization of Nd and Pb isotope signatures of the upper margin surface sediments in the Southwest Atlantic was provided by de Mahiques et al. (2008), filling an extensive area without information about &Nd signatures (Jeandel et al., 2007; Blanchet, 2019). In that work the authors: 1) recognize the isotopic signatures of the sediments from the Argentinean, southern and southeastern Brazilian margins and; 2) describe potential source areas of the sediments, such as the Andes, the basalts of the Paraná basin, and the pre-Cambrian rocks of the Brazilian shield.

Notwithstanding, geographic gaps in information preclude a throughout understanding of the role of key hydrological features such as the STSF, BMC, and SB in the sediment distribution and the geochemical boundaries associated, key for paleoceanographic studies. Moreover, in another approach for sediment sources and pathways in the Southwestern Atlantic margin provided by Razik et al. (2015), the authors argued for a mixed Rio de la Plata - Andean origin for the upper slope sediments off southern Brazil.

In this paper, we extend the Nd and Pb dataset along the southwestern Atlantic margin and use the concept of sediment fingerprinting to deepen the role played by hydrodynamic forcing in sediment transport and deposition. The geographical distribution of the new samples <u>presented here</u>, covering the Argentinean margin and the Punta del Este, Pelotas, and Santos basins and covering a bathymetry range between 5 and 4066 meters, allows focusing on the role of the STSF, BMC, and BS

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in the distribution of sediments. The results are interpreted with the aid of the output of a state-of-the-art circulation model to understand the role of oceanographic boundaries in the distribution of sediments along the area.

2 Study Area

The study area comprises a Southwestern Atlantic margin sector, from the parallels 23°00'S to 54°10'S, corresponding to a linear extension of about 3,500 km (Figure 1). Syntheses of the main geological and oceanographic processes can be found in Hernandez-Molina et al. (2009), Franco-Fraguas et al. (2014), Nagai et al. (2014a), Nagai et al. (2014b), Violante et al. (2014), Hernández-Molina et al. (2015), Franco-Fraguas et al. (2016), Violante et al. (2017a), Burone et al. (2018), Piola et al. (2018), and Piola and Matano (2019), among several others.

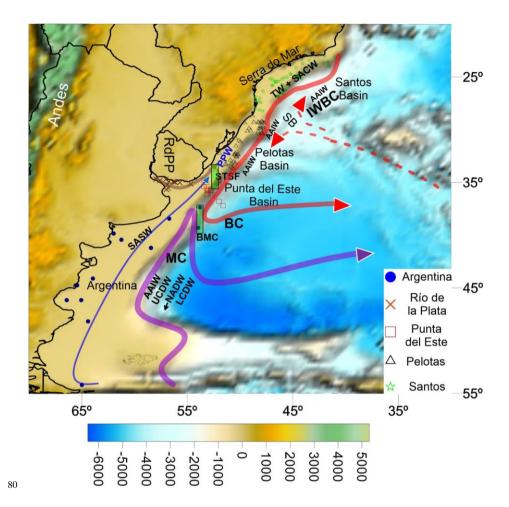


Figure 1. Location of the study area, displaying the main flows, oceanographic boundaries, and sampling stations. Violet thick line: Malvinas Current (MC); thin blue line: Subantarctic Shelf Water (SASW); thin brown line: Río de la Plata Plume (RdIPP); thick red line: Brazil Current (BC); shaded rectangles: Subtropical Shelf Front (STSF) and Brazil – Malvinas Confluence (BMC); dashed red line Santos Bifurcation (SB) and Intermediate Western Boundary Current (IWBC). Other Abbreviations: Antarctic Intermediate Water (AAIW), Upper Circumpolar Deep Water (UCDW), North Atlantic Deep Water (NADW), and Lower Circumpolar Deep Water (LCDW). Symbols: Argentina (), Punta del Este Basin (), Río de la Plata (), Pelotas Basin (), and Santos Basin (). Bottom scale: topography in meters

2.1 Morphology

The Southwestern Atlantic margin is a typical segmented volcanic-rifted margin, where several transverse basins are recognized (Bassetto et al., 2000; Moulin et al., 2010; Soto et al., 2011). Its origin and evolution are intrinsically related to the opening of the South Atlantic (Nürnberg and Müller, 1991), whose rifting processes first started in the Triassic (Lovecchio et al., 2020) but effectively occurred during the Jurassic and Cretaceous.

There is a general trend of narrowing the margin towards the North (Urien and Ewing, 1974; Zembruscki, 1979; Parker et al., 1996; Violante et al., 2017a). The shelf width varies from 850 km to the south to 70 km in its northernmost limit; the shelf-break depth ranges from 80 m, in southern Brazil, to 200 meters, in Uruguay (Zembruscki, 1979; Muñoz et al., 2010; Lantzsch et al., 2014). The shelf morphology is relatively flat, but sequences of scarps and terraces are recognized along the continental shelf at varying water depths (Corrêa, 1996; Parker et al., 1996; Baptista and Conti, 2009).

The continental slope presents a highly variable morphology, including contouritic terraces, channels, mounds, erosive surfaces, and sediment drifts all along the area (Duarte and Viana, 2007; Hernández-Molina et al., 2010; Preu et al., 2013; Hernández-Molina et al., 2015) as well as canyons (Voigt et al., 2013; Bozzano et al., 2017; Franco-Fraguas et al., 2017; Violante et al., 2017b; Warratz et al., 2019). The contouritic features and submarine canyons actively interact along the margin so that mixed contouritic-gravitational erosive and depositional features are common. Mega-slides (Reis et al., 2016; Franco-Fraguas et al., 2017) and carbonate mounds (Carranza et al., 2012; Maly et al., 2019; Steinmann et al., 2020) are also present along the margin.

2.2 Sedimentary cover

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The Southwestern Atlantic margin is dominated by a terrigenous, siliciclastic sedimentary cover, with extensive sand sheets

(Lonardi and Ewing, 1971; Frenz et al., 2003; Figueiredo and Madureira, 2004). The Argentinean and Uruguayan shelves are
capped mainly by a 5 to 15 m thick post-Late Glacial transgressive sandy sheet (with decreasing thickness towards the south)
composed of dominant medium to fine sands (sometimes muddy), with varying amounts of shells (more abundant in the
Uruguayan shelf) and gravels (more abundant in the Patagonian shelf).

Sandy and shelly sediments are mainly relicts of coastal and inner shelf environments that evolved during Pleistocene transgressive-regressive events (Kowsmann and Costa, 1979; Urien et al., 1980; Lantzsch et al., 2014). Therefore, they are considered relict and palimpsest, whereas gravelly-dominated sediments on the southern Argentinean shelf result from glacifluvial origin. More recent works emphasize the existence of mud depocenters as potential fates of modern sediments on the southern Brazilian shelf (Nagai et al., 2014a; de Mahiques et al., 2017; Lourenço et al., 2017; de Mahiques et al., 2020). In the slope and rise, there is a prevalence of very fine sands and silty sands, resulting from exclusively submarine processes occurring across- (gravitational) and along- (contouritic) slope, together with pelagic sedimentation (Violante et al., 2010;

Bozzano et al., 2011; Franco-Fraguas et al., 2016; Schattner et al., 2020). However, coarse sands and gravels occur at or near the head of submarine canyons and in contouritic channels and moats (Lonardi and Ewing, 1971; Bozzano et al., 2011; Reis et al., 2016; Franco-Fraguas et al., 2017). Razik et al. (2015) indicate increasing grain size towards coarse sands due to sediment remobilization and redistribution due to upwelling and downwelling resulting from eddies and vertical water movement generated by the slope off southern Brazil meandering Brazil Current.

2.3 Ocean Circulation

The Southwestern Atlantic margin is characterized by complex hydrography (Matano et al., 2010). It presents two main oceanographic boundaries, the Subtropical Shelf Front (STSF), as the shelf extension of the Brazil – Malvinas Confluence (BMC) (Piola et al., 2000; Severov et al., 2012), and the less-studied Santos Bifurcation (SB) (Boebel et al., 1997; Boebel et al., 1999a). The region is also influenced by the Rio de la Plata (RdlP), the second-largest hydrographic basin in South America, whose average discharge is 22,000 m³ s⁻¹ (Framiñan and Brown, 1996). This regional circulation system experiences seasonal latitudinal shifts in response to wind regimes (Schmid et al., 2000; Piola and Matano, 2001; Piola et al., 2018).

At the BMC, centered at 37-39°S (Maamaatuaiahutapu et al., 1992), the southward-flowing Brazil Current (BC) encounters the northward-flowing Malvinas Current (MC) (Schmid and Garzoli, 2009), transporting and mixing water masses with contrasting thermohaline characteristics. The BC is a baroclinic boundary current that concentrates its main flow in the upper 500 m water-depth upstream of 28°S, carrying the Tropical Water (TW) at the surface (Emilsson, 1961; Palma et al., 2008), and the South Atlantic Central Water (SACW) at pycnoclinic levels (Emilsson, 1961; Signorini, 1978). Near the BMC, a significant fraction of the BC transport is below 500m. The MC is a strong barotropic boundary current that advects the Subantarctic Water (SAW) near the surface (Spadone and Provost, 2009), and the Antarctic Intermediate Water (AAIW) at intermediate levels (Tomczak and Godfrey, 1994a, b).

At the BMC, water masses are transported eastwards as part of the southern limb of the basin-wide Anticyclonic Atlantic Subtropical Gyre (Boebel et al., 1997; Boebel et al., 1999a; Schmid et al., 2000; Núñez-Riboni et al., 2005; Legeais et al., 2013). At the intermediate levels of the westward flow of the gyre, the water reaches the South American margin near 28°S, where it splits into two branches, forming the Santos Bifurcation (Boebel et al., 1999a; Legeais et al., 2013). From the bifurcation, one-quarter of the transport at 40°W flows northward along the continental slope (mainly between the 800 and 1,200 m isobaths), forming the Intermediate Western Boundary Current (IWBC) (Fernandes et al., 2009; Biló et al., 2014). About three-quarters feed the BC, flowing south until its separation from the coast at the BMC (Schmid et al., 2000; Piola and Matano, 2019). This configuration leads to an overall southward flow on the outer shelf and the outer to middle slope, from 28°S up to the BMC.

150 Concerning deep circulation, the North Atlantic Deep Water (NADW) (Sverdrup et al., 1942), transported from the northern hemisphere high latitudes by the Deep Western Boundary Current, occupies the region between the 2,000 and 3,000 m isobaths. The NADW flows between two northward-flowing branches of the Circumpolar Water (i.e., Upper and Lower Circumpolar).

Deep Water). The abyssal circulation (> 3,500 m) is dominated by the Antarctic Bottom Water (AABW), which is partially trapped in the Argentine Basin (Tarakanov and Morozov, 2015).

Over the shelf, the extension of the BMC, known as the Subtropical Shelf Front (STSF), separates Subtropical Shelf Waters (STSW, formed by the mixture of the TW and SACW) and Subantarctic Shelf Waters (SASW) (Piola et al., 2000). This narrow and sharp front extends between 32°S at 50 m of water column depth and 36°S over the shelf break, and its position appears stable throughout the year (Piola et al., 2000; Berden et al., 2020). The main branch of the STSF is mixed with waters transported by the BC and exported offshore along with the BMC. A secondary branch is diluted with the PPW and TW and returns along the shelf (Berden et al., 2020).

At the surface, the low-salinity RdIP plume flows northward along the inner Uruguayan continental shelf during the austral winter. In the summer and during El Niño events, the plume remains off the RdIP mouth and extends along the entire upper continental margin (Piola et al., 2000; Piola et al., 2005; Möller et al., 2008).

3 Materials and Methods

55 3.1 Geochemical Analyses

The samples were organized in five distinct sectors in this study, corresponding to the Santos, Pelotas, and Punta del Este marginal basins, RdlP estuary, and the Argentinean margin. Due to the small number of samples, the sediments from Argentina were not divided into the corresponding sedimentary basins (Figure 1). Geographic coordinates and water depth of the samples are presented in the Supplementary Material file.

170 The results of one hundred and fifty-six sediment samples were used as a dataset, including eighty-three new samples, fifty-three samples published in de Mahiques et al. (2008), six samples published in Basile et al. (1997), eight samples published in Franco-Fraguas et al. (2016), and three coretop samples published in Lantzsch et al. (2014). The analytical methods used in those ancillary papers are described in the original references. The new samples were collected with box-corers and multiple-corers in distinct surveys onboard the research vessels Alpha Crucis (Brazilian margin), Miguel Oliver, Capitán Saldaña, and 175 Sarmiento de Gamboa (Uruguayan margin). Only the superficial samples (the upper one centimeter) of each core were used

The Nd and Pb isotopic analyses of the lithogenic fraction were conducted at the Geochronological Research Centre of the University of São Paulo, Brazil.

All chemical procedures were performed in class 10,000 cleanroom equipped with laminar flow hoods class 100. All reagents were purified before use. Water was distilled and then purified on a Milli-Q System (@Millipore Corporation) ('ultrapure' water - "Type 1"). The acids were purified in sub boiling distillers (DST-1000, @Savillex) and sub boiling stills (@Savillex) at low temperatures.

All of the samples were previously decarbonated with HCl. Sediment powder (70 mg) was dissolved with HF, HNO3, and HCl acids. Dissolution was done on a MARS-5 microwave oven. Both Pb and Nd were purified by the ion-exchange

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technique. The first stage of ion-exchange chromatography involves separating Pb from the other matrix elements using columns packed with anion exchange AG1-X8, 200-400 mesh (Biorad) resin. After Pb collection, the remaining solution is dried out, and the residue is retaken to separate the Rare Earth Elements using RE resin (EIChroM Industries Inc.) from the

bulk solution. Nd was then separated using Ln resin (EIChroM Industries Inc.).

Pb isotopic compositions were measured on a Finnigan MAT 262 Mass Spectrometer. Samples were loaded on Re filaments with H_3PO_4 and silica gel. Every single analysis consisted of 60 ratio measurements. The Pb ratios were corrected for mass fractionation of 0.13%/amu based on repeated analysis of the NBS-981 standard ($^{206}Pb/^{204}Pb = 16.893 \pm 0.003$; $^{207}Pb/^{204}Pb = 15.432 \pm 0.004$, and $^{208}Pb/^{204}Pb = 36.512 \pm 0.014$; n = 11), which yielded mass discrimination and fractionation corrections of 1.0024 ($^{206}Pb/^{204}Pb$), 1.0038 ($^{207}Pb/^{204}Pb$) and 1.0051 ($^{208}Pb/^{204}Pb$). The combination of these uncertainties and within-run uncertainties are typically 0.15%–0.48% for $^{206}Pb/^{204}Pb$, 0.13%–1.07% for $^{207}Pb/^{204}Pb$ and 0.10%–0.45% for $^{208}Pb/^{204}Pb$, all at the 2 σ (95%) confidence level. The total Pb blank contribution, <1 ng, is negligible.

The Nd analyses, here reported as ϵ Nd, values, were prepared by standard methods by the analytical procedures described by Sato et al. (1995) and Magdaleno et al. (2017), involving HF–HNO₃ dissolution plus HCl cation exchange using a Teflon Powder column to separate REEs. No visible solid residues were observed after dissolution. Samples with incomplete dissolution were discarded.

Nd determinations were performed on a Thermo Neptune Plus ICP-MS. Nd isotopic ratios (143 Nd/ 144 Nd) were normalized to the value of 146 Nd/ 144 Nd = 0.7219 (DePaolo, 1981) and 143 Nd/ 144 Nd = 0.512103 of the JNDi-1 standard (laboratory average of the last 12 months). Usually, a single analysis consisted of 60 measurements of Nd. The 143 Nd/ 144 Nd mean average of the JNDi standard during the analyses was 0.512095 ± 0.000007 (n = 3) and 0.512096 ± 0.000005 between July and November 2013 (n = 56). The daily average of 143 Nd/ 144 Nd of the JNDi-1 standard was 0.512101 ± 0.000002 (n=18). The analytical blank during the analyses varied from 51 and 53 pg.

The parameter ENd was calculated as follows:

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\epsilon Nd = ((^{143}Nd)^{144}Nd_{sample})^{143}Nd)^{144}Nd_{CHUR}) - 1) * 10^4, where ^{143}Nd)^{144}Nd_{CHUR} = 0.512638 (Jacobsen and Wasserburg, 1980).
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Reproducibility analysis was made for both Pb and Nd isotopes, using Buffalo River Sediment (NIST-RM8704) (n = 7), with the following results:

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^{143} Nd/^{144} Nd = 0.51203 \pm 0.00001 \text{ (SD)}
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$$^{206}Pb/^{204}Pb = 18.846 \pm 0.018 \text{ (SD)}$$

$$^{207}\text{Pb}/^{204}\text{Pb} = 15.646 \pm 0.005 \text{ (SD)}$$

215 208 Pb/ 204 Pb = 38.503 ± 0.016 (SD)

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Statistical analyses were performed using the software PAST (Palaeontological Statistics), version 4.05 (Hammer et al., 2001). To recognize the distinct isotopic domains over the study area, we applied the Geochemical Fingerprinting procedure, similar to the approaches proposed by Walling (2013), Miller et al. (2015), and Palazon and Navas (2017). First, a Kruskal-Wallis non-parametric analysis of variance was applied for each variable, followed by a Mann-Whitney pairwise post-hoc test to

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identify which variables presented statistically significant differences. Finally, a Discriminant Analysis with standardized values was used to determine the correct classification for the previously assigned groups.

3.2. The LLC2160 Circulation Model

To support the geochemical data distribution interpretation, we analyzed the output of the LLC2160 simulation, a global 1/24° forward run of the Massachusetts Institute of Technology General Circulation Model (MITGCM) that was spun up from Estimating from Circulation and Climate of the Ocean (ECCO). The ECCO state estimate is similar to an ocean reanalysis and assimilates millions of observations, starting in 1992. With 90 vertical levels and a horizontal resolution of about 4 km in the South American margin, the LLC2160 simulation resolves the main ocean circulation features on the continental slope and shelf of the Southwestern Atlantic. Our analysis focuses on a 12-month average spanning September 2011 through August 2012. Details of the simulation, including a description of the spin-up hierarchy and forcing, are available in Chen et al. (2018). We used annual-mean fields of LLC2160 simulation to identify two key features: the Santos Bifurcation (SB) and the Subtropical Shelf Front (STSF). The SB is recognized as the region on the continental slope where the flow within the AAIW depth range (550-1400 m) is negligible. Specifically, we search on different isobaths ranging from 500 m to 1500 m for the region where the AAIW flow is weaker than 0.01 m/s. We emphasize that the SB is not a stagnation point where the flow is zero but a shadow zone that spans nearly 100 km, wherein the intermediate flow is feeble (see the schematic SB in Figure 1). In our discussion below, we present the mean position and the latitudinal extension of the SB as a function depth. To identify the mean position of the STSF, we searched for the local maximum of the potential temperature gradient, which is a very distinct feature on the northern Argentina/southern Brazil shelf. We compute the potential temperature gradients at 40 m to avoid contamination by RdIP water (e.g., Piola et al., 2008). When applied to the LLC2160 output using seasonal averages, our method yielded frontal locations consistent with those identified by applying the isothermal criteria at 40 m proposed by Piola et al. (2008). In the yearly fields, the front follows approximately the 14 °C isotherm.

4 Results

The results of isotopic analyses are presented in the Supplementary Material and summarized in the box plots shown in Figure 2. We also present the latitudinal variation of each isotope (Figure 3).

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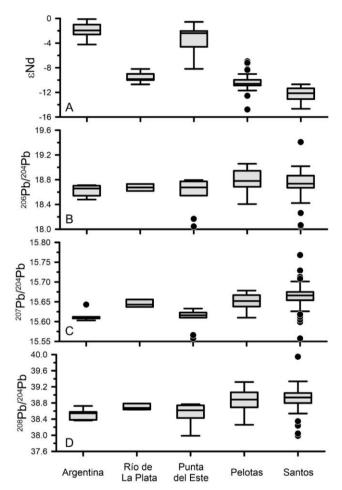


Figure 2. Box-plots of the distributions of (A) ϵ Nd, (B) 206 Pb/ 204 Pb, (C) 207 Pb/ 204 Pb, and 208 Pb/ 204 Pb. Outliers are shown as dots

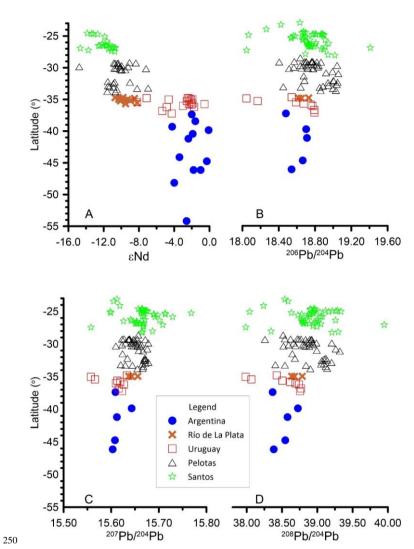


Figure 03. Latitudinal variations of (A) ϵ Nd, (B) 206 Pb/ 204 Pb, (C) 207 Pb/ 204 Pb, and 208 Pb/ 204 Pb. Symbols: Argentina ($\stackrel{\frown}{\bigcirc}$), Punta del Este Basin ($\stackrel{\frown}{\bigcirc}$), Río de la Plata ($\stackrel{\bigstar}{\nearrow}$), Pelotas Basin ($\stackrel{\frown}{\bigcirc}$), and Santos Basin ($\stackrel{\frown}{\nearrow}$).

εNd values show a northward trend to less radiogenic values, varying from -0.1 (Argentina) to -17.1 (Santos Basin) (Figure 3a). The latitudinal variation of the Pb isotopes is less clear but still visible for ²⁰⁷Pb/²⁰⁴Pb and ²⁰⁸Pb/²⁰⁴Pb (Figures 3c and 3d). On average, the Argentina sector presents the highest εNd average values (-2.1±1.3), and lowest ²⁰⁶Pb/²⁰⁴Pb, ²⁰⁷Pb/²⁰⁴Pb, and ²⁰⁸Pb/²⁰⁴Pb average values (18.620±0.104, 15.615±0.016, and 38.520±0.149, respectively). On the other hand, the Santos sector shows the lowest εNd (-12.0±1.1) and highest ²⁰⁷Pb/²⁰⁴Pb and ²⁰⁸Pb/²⁰⁴Pb average values (15.664±0.008 and 38.909±0.016, respectively). Values of ²⁰⁶Pb/²⁰⁴Pb did not show any evident latitudinal trend. For ²⁰⁶Pb/²⁰⁴Pb, the values range from 18.045, on the Punta del Este sector, to 19.409, on the Santos Basin shelf. ²⁰⁷Pb/²⁰⁴Pb values range from 15.558 to 15.768 in the same areas. Finally, ²⁰⁸Pb/²⁰⁴Pb values vary from 37.986 to 39.949, also in the same sectors. From the Kruskal-Wallis analysis, we observe that except for ²⁰⁶Pb/²⁰⁴Pb, the variables show significant differences among the compartments, thus allowing us to proceed with the Discriminant Analysis. Furthermore, the Mann-Whitney analysis allowed us to recognize the pairwise differences among the other variables (Table 1). Finally, it is to be noted that sediments from those located towards the North. On the other hand, sediments from the Rio de la Plata are statistically similar to those from the Pelotas sector for all of the variables.

270 Table 1. p values of the Mann-Whitney pairwise test. Statistically, significant differences are highlighted in blue

εNd	Argentina	Rio de la Plata	Punta del Este	Pelotas
Argentina				
Rio de la Plata	0.00			
Uruguay	0.09	<u>0.00</u>		
Pelotas	0.00	0.32	<u>0.00</u>	
Santos	0.00	<u>0.00</u>	<u>0.00</u>	0.00
²⁰⁶ Pb/ ²⁰⁴ Pb	Argentina	Rio de la Plata	Punta del Este	Pelotas
Argentina				
Rio de la Plata	0.21			
Uruguay	0.59	0.59		
Pelotas	0.02	0.31	0.08	
Santos	0.08	0.69	0.37	0.08
²⁰⁷ Pb/ ²⁰⁶ Pb	Argentina	Rio de la Plata	Punta del Este	Pelotas
Argentina				
Rio de la Plata	0.07			
Uruguay	0.42	0.29		
Pelotas	0.00	0.06	0.00	
Santos	<u>0.00</u>	0.02	<u>0.00</u>	0.01
²⁰⁸ Pb/ ²⁰⁴ Pb	Argentina	Rio de la Plata	Punta del Este	Pelotas
Argentina				
Rio de la Plata	0.04			
Uruguay	0.59	0.29		
Pelotas	0.01	0.11	0.02	
Santos	0.00	0.02	0.01	0.47

The two first axes of the Discriminant Analysis account for 99.72% (99.09% for axis 1) of the total variance considering the standardized values of ϵNd , ^{207}Pb / ^{204}Pb , and ^{208}Pb / ^{204}Pb (Figure 4). It is possible to recognize that samples from Argentina are

275 detached from the other sectors. On the other hand, samples from Pelotas Basin show a transitional character between Santos Basin, on one side, and on the other side, Río de La Plata and Punta del Este Basin.

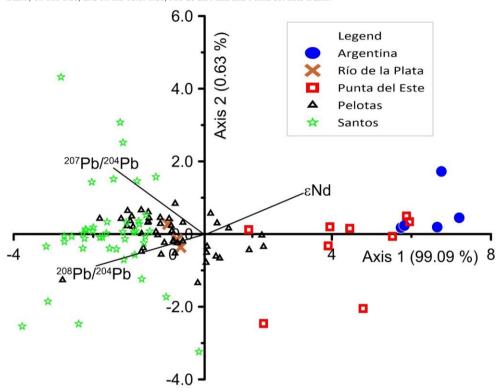
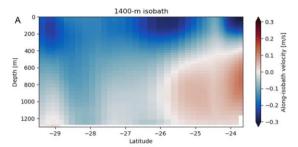


Figure 4. Scatter plot of the samples according to the two first axes generated from the Discriminant Analysis. Symbols: Argentina (\bigcirc), Punta del Este Basin (\square), Río de la Plata (\times), Pelotas Basin (\triangle), and Santos Basin ($\stackrel{\checkmark}{\bowtie}$).

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Graphical representations of the LLC2160 output are presented for both the Santos Bifurcation (Figure 5) and Subtropical Shelf Front (Figure 6). The Santos Bifurcation is identified as the maximum horizontal velocity divergence region at the AAIW level, identified in Figure 5A close to 26°S. The visualization based on the horizontal fields is more complicated but still visible as the sector with velocities close to 0 m/s (Figure 5B). The Subtropical Shelf Front (Figure 6A) is identified as a maximum subsurface temperature gradient (Figure 6B). Vertically it is well marked below the 30 m isobath (Figure 6C).



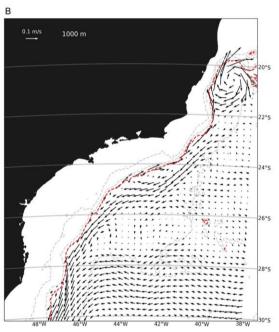


Figure 5. Graphical outputs of the LLC2160 focused on the Santos Bifurcation. 6A: the bifurcation is identified as the white zone located around 26°S between the 800 and the 1200 isobath. 6B: the bifurcation is recognized as the zone of velocities tending to 0 cm/s at 26°S – 44°W.

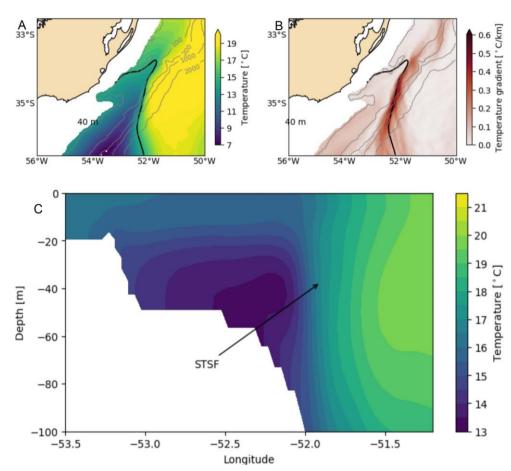


Figure 6. Graphical outputs of the LLC2160 focused on the Subtropical Shelf Front. 6A: Variations of temperature in the zone of the front. 6B: Temperature gradient; the front is recognized as the zone of maximum gradient (darker colors). 6C: Vertical transect at 35°S showing the location of the Subtropical Shelf Front (STSF) as the gradient of the Subantarctic Shelf Water (deep blue) and the Subtropical Shelf Water (light green)

300 5 Discussion

The integration of both isotopic signatures (ϵNd , $^{207}Pb/^{204}Pb$, and $^{208}Pb/^{204}Pb$) and hydrographic (water masses) and hydrodynamic (currents) information is presented in figures 7 to 9, respectively. This information is essential to infer both sediment sources and the role played by ocean circulation in the distribution of sediments in the study area.

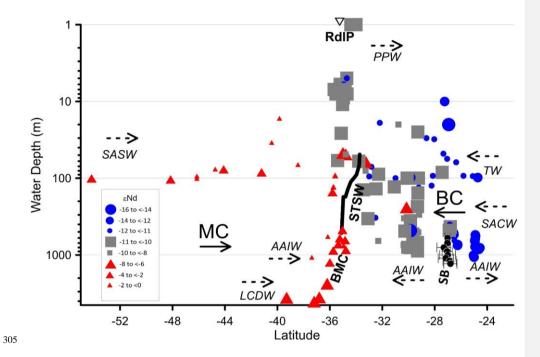


Figure 7. Latitudinal and bathymetric variability of £Nd in the sector between 23°S and 55°S. Water masses: Plata Plume Water (PPW), Subantarctic Shelf Water (SASW), Tropical Water (TW), South Atlantic Central Water (SACW), Antarctic Intermediate Water (AAIW), and Lower Circumpolar Deep Water (LCDW). Currents: Malvinas Current (MC) and Brazil Current (BC). Fronts: 310 Subtropical Shelf Front (STSW), Brazil-Malvinas Confluence (BMC), and Santos Bifurcation (SB). Vertical axis in Log(10) scale.

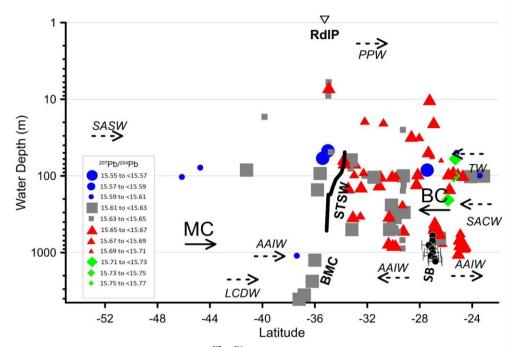


Figure 8. Latitudinal and bathymetric variability of ²⁰⁷Pb/²⁰⁴Pb in the sector between 23°S and 55°S. Water masses: Plata Plume Water (PPW), Subantarctic Shelf Water (SASW), Tropical Water (TW), South Atlantic Central Water (SACW), Antarctic Intermediate Water (AAIW), and Lower Circumpolar Deep Water (LCDW). Currents: Malvinas Current (MC) and Brazil Current (BC). Fronts: Subtropical Shelf Front (STSW), Brazil-Malvinas Confluence (BMC), and Santos Bifurcation (SB). Vertical axis in Log₍₁₀₎ scale.

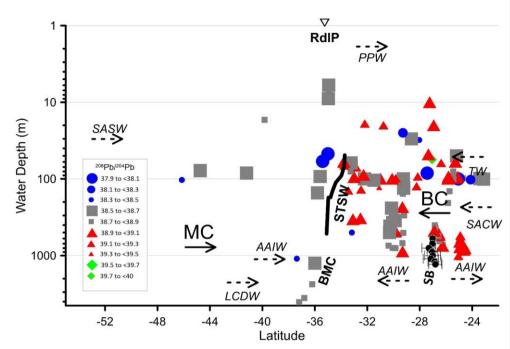


Figure 8. Latitudinal and bathymetric variability of ²⁰⁸Pb/²⁰⁴Pb in the sector between 23°S and 55°S. Water masses: Plata Plume Water (PPW), Subantarctic Shelf Water (SASW), Tropical Water (TW), South Atlantic Central Water (SACW), Antarctic Intermediate Water (AAIW), and Lower Circumpolar Deep Water (LCDW). Currents: Malvinas Current (MC) and Brazil Current (BC). Fronts: Subtropical Shelf Front (STSW), Brazil-Malvinas Confluence (BMC), and Santos Bifurcation (SB). Vertical axis in Log₍₁₀₎ scale.

5.1. Sediment sources

Recognizing the role of circulation on the deposition of sediments requires an association of the sedimentary provinces with potential source areas. Indeed, radiogenic isotopes are considered good sediment source fingerprints (Owens et al., 2016). Two seminal papers, by Goldstein et al. (1984) and Bayon et al. (2015), used Nd isotopes and other proxies from the world's rivers and provided the basis for comprehending distribution detrital Nd in the world's oceans. Beny et al. (2020) provided the summary of Nd, Pb, and Sr signatures in the South Atlantic, looking for the potential sources and circulation in the area. More recently, a work by Höppner et al. (2021) provided new data on the isotopic signatures of the river sediments that feed the RdlP basin. Worth noting that the RdlP-Paraná-Uruguay riverine system drains several types of terranes, such as pre-Cambrian rocks of the Brazilian shield, Paleozoic sediments, and tholeiitic basalts from the Paraná Basin, and Cenozoic Andean rocks.

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Table 2 provides a list of Nd and Pb isotopic signatures of potential continental materials (rocks and sediments) for the study area. It is possible to recognize a trend of decreasing values of ϵ Nd towards the north, as already observed in our samples. Concerning Pb isotopes, the small number of data hampers the recognition of a trend.

Table 2. Nd and Pb isotopic values of distinct continental materials from Antarctica and Southeastern South America

Material	εNd	²⁰⁷ Pb/ ²⁰⁴ Pb	²⁰⁸ Pb/ ²⁰⁴ Pb	Source
Sediments from the Antarctic Peninsula and Western Antarctica	-4.3±1.0	n.a.	n.a.	Roy et al. (2007)
River bed sediments from Patagonia	-2.0±0.7	n.a.	n.a.	Gaiero et al. (2007)
Topsoils and river bed sediments from Southern Patagonia	-1.7±1.6	15.63±0.01	38.61±0.12	Khondoker et al. (2018)
Clay and silt fractions from the Chubut River (Patagonia)	-0.4 -1.6	n.a.	n.a.	Bayon et al. (2015)
Bulk sediment from Paraná River	-10.3	n.a.	n.a.	Goldstein et al. (1984)
Suspended sediments from Paraná River	-10.8±0.5	n.a.	n.a.	Henry et al. (1996)
Low-Ti basalts from the southern Paraná Igneous Province	-8.0±1.2	15.68±0.02	38.97±0.24	Barreto et al. (2016); Melankholina and Sushchevskaya (2018)
Ribeira River sediments. The river drains Proterozoic low- to middle- rank metamorphic rocks from southeastern Brazil (river mouth located at 24.68°S – 047.42°W)	n.a.	15.55±0.02	37.67±0.4	Moraes et al. (2004)
Neoarchean and Proterozoic metasediments from the coastal region of southeastern Brazil	-21.5±5.6	n.a.	n.a.	Ragatky et al. (2000)
Proterozoic granites from the coastal region of southeastern Brazil	-18.7±1.4	n.a.	n.a.	Mendes et al. (2011)
Paraíba do Sul River sediments. The river drains middle-rank to high- rank metamorphic rocks from southeastern Brazil (river mouth located at 21.61°S – 41.02°W)	-19.3±1.4	n.a.	n.a.	Roig et al. (2005)
Proterozoic metasediments from southeastern Brazil	-8.8±1.9	n.a.	n.a.	Mantovanelli et al. (2018)
Proterozoic granites from southeastern Brazil	-18.2±1.2	n.a.	n.a.	Mantovanelli et al. (2018)

345 Isotopic distinctions and similarities among the sectors are recognized from the interpretation of the results of the Discriminant Analysis (Figure 4). Sediments from Argentina and part of the Punta del Este Basin present isotopic signatures similar to the values obtained for Patagonia (Gaiero et al., 2007; Bayon et al., 2015; Khondoker et al., 2018). The deepest samples of the dataset, located in the Punta del Este basin, at water depths between 3579 and 4066 meters, present εNd values of -5.33 and -4.26, respectively. These values are consistent with those from the Antarctic Peninsula and West Antarctica (Roy et al., 2007).

4.26, respectively. These values are consistent with those from the Antarctic Peninsula and West Antarctica (Roy et al., 2007).
 They can indicate a provenance of sediments via the flow of the Upper- and Lower- Circumpolar Deep-water masses (UCDW and LCDW, respectively) (Beny et al., 2020) or even from ice-rafted debris (Bozzano et al., 2021). Another sample, located at 2378 meters, presents an εNd value of -4.80 but is presently under the influence of the NADW southward flow (Figure 7).
 The distinct character of these samples also resides in the fact that they are lower 207Pb/204Pb and higher 208Pb/204Pb radiogenic than those located in shallower areas (Figures 8 and 9). The remainder of the Punta del Este basin samples, situated on the shelf, might represent a mixture of Patagonian and Río de la Plata sediments.

Most of the samples from the Pelotas Basin are under the influence of the RdlP. Apart from Nd and Pb isotope values, other independent proxies confirm the Plata Plume Water (PPW) as a source of the sediments to the southern Brazilian margin (Pelotas Basin) and part of the southeastern margin (Santos Basin). Campos et al. (2008) and Nagai et al. (2014a) used clay mineralogy to indicate sediments from the Río de la Plata to the North. Also, the maps presented by Govin et al. (2012) show similarities in the ln(Ti/Al) and ln(Fe/K) between the Uruguayan and southern Brazilian upper margin. Finally, Mathias et al. (2014) used magnetic properties of sediments in a core located at the latitude of 25°30'S to recognize the influence of the Río de la Plata on the southern Brazilian shelf since 2 cal kyr BP. Finally, in a study that included the analysis of potential source rocks from the continent, Mantovanelli et al. (2018) confirmed the contribution of the Paraná Basin basalts along the Holocene off the southern Brazilian shelf (27°S). The authors observed a remarkable change to less radiogenic Nd in the sedimentary column further North (23°S).

The samples from Santos Basin present lower radiogenic Nd and higher radiogenic Pb values, thus indicating a Pre-Cambrian source, as Mantovanelli et al. (2018) stated. Nevertheless, the values obtained for Pb isotopes differ significantly from those reported by the literature for the Precambrian metasediments and granites of the southeastern Brazilian coast (Ragatky et al., 2000; Moraes et al., 2004; Mendes et al., 2011). A possible explanation for this discrepancy is that the input of sediments from the adjacent coast is hampered by the Serra do Mar mountain chain, limiting the development of expressive drainage basins in the area (Riccomini et al., 2010; Cogné et al., 2011). In this sense, we cannot rule out the possibility that a significant part of

sediments that presently cover the shelf and upper slope of the Santos basin is originated further north and transported by the Brazil Current and derived shelf dynamics (Castro and Miranda, 1998; Silveira et al., 2017).

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5.2. Isotope fingerprinting and ocean circulation

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The geochemical fingerprinting approach confirmed the suitability of using Nd and Pb isotopes (except ²⁰⁶Pb/²⁰⁴Pb) as reliable proxies for the discrimination among the distinct sectors of the Southwestern Atlantic margin. Indeed, the first axis provided more than 99% of explained variance (Figure 4). The distribution of the samples and, together with the recognition of the potential sources (Table 2), allows tracing a correlation among isotopic signatures, sediment sources, and ocean circulation. Figures 7 to 9 present the bathymetrical variations of the isotopes and positions of the STSF (continuous line) and SB (continuous line with horizontal bars) identified in the LLC2160 output. The positions of both oceanographic features in the LLC2160 simulation are broadly consistent with previous studies (e.g., Boebel et al., 1999a; Boebel et al., 1999b; Piola et al., 2008). The LLC2160 output, together with the isotopic values, allows us to present the bathymetrical variations of those features. As observed, there are clear distinctions in the signature corresponding to both fronts. The STSF presents only minor seasonal variations, and its control is probably related to the interaction between the RdIP plume and the subsurface water masses distribution. During austral summer, the strong stratification (Möller et al., 2008) inhibits RdlP sedimentation southward of the STSF. During austral winter, the northeastward RdlP plume promotes the offshore displacement of subtropical waters (Möller et al., 2008), enabling the deposition of fine sediments on the shelf north of the STSF. On the upper and middle slope, the southward displacement of the thickened Brazil Current, carrying the recirculated AAIW, likely limits RdlP sedimentation (Schmid et al., 2000). There are apparent differences in the Nd and Pb signatures in the intermediate zone, at about 34°S – 35°S; this boundary might represent the northernmost limit of the BMC (Benthien and Müller, 2000; Pezzi et al., 2009).

This integrated analysis suggests no transport of sediments from the Argentinean sector to the southern Brazilian margin. On the other hand, based on the same analysis, we can confirm that sediments from the Rio de la Plata reach, at least partially, the Santos sector, i.e., to the north of 28°S. Concerning the SB, there is a clear distinction in isotopic signatures below the 500 m isobath, less radiogenic Nd prevailing to the north of the bifurcation. We thus argue that both STSF and SB also separate distinct geochemical provinces on the Southwestern Atlantic margin.

The Argentinean and part of the Uruguayan upper margins are covered by Andean-Patagonian sediments, redistributed by the shelf circulation and Malvinas Currents. The STSF and BMC block the transport of these sediments to the north. This finding corroborates Hernández-Molina et al. (2016) and Franco-Fraguas et al. (2016), who defined the northernmost limit of a megacontouritic feature on the Uruguayan slope. Sediments located more profound than the 2,000 m isobath present an Antarctic signature, transported either by the bottom circulation (UCDW and LCDW) or ice-rafted debris.

Sediments from the Río de la Plata estuary advance along the inner shelf towards southern Brazil and a mixture of Pelotas and Santos signatures are observed between 28°S and 30°S. This mixture is visible in the scatter plot presented in Figure 4, in which sediments of Pelotas Basin constitute a mixture of distinct populations, i.e., Santos Basin and Río de la Plata. It is essential to highlight that, on interannual time scales, the influence of El Niño-Southern Oscillation (ENSO) in the precipitation regime determines changes in the freshwater outflow of the RdlP. Cold and warm episodes of ENSO cause, drought and

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abundant rainfall in Uruguay, southern Brazil, and north-eastern Argentina (Pisciottano et al., 1994; Cazes-Boezio et al., 2003).

In addition, changes in the wind patterns during the warm phase of ENSO determine the influence of the PPW towards higher latitudes. In conjunction with the Coriolis force, this phenomenon determines that the PPW follows a NE direction close to the shelf break, explaining the distribution up to 28°S and, in anomalous years, 25°S (Piola et al., 2005).

Finally, sediments located northward of 27°S originate from the Precambrian rocks that dominate the coastal domains off SE and E Brazil, being mainly transported by the intense flow of the BC on the outer shelf and upper slope. Limited input comes

425 from the small rivers that drain the mountainous areas of the Serra do Mar, as previously reported by Lourenço et al. (2017) and de Mahiques et al. (2017)

430 Conclusions

In this paper, we use Nd and Pb radiogenic isotopes to recognize the role of ocean circulation in the sediment distribution of the Southwestern Atlantic margin.

Andean and continental Patagonian sediments are the primary source for the deposits of the Argentinean and Uruguayan shelves, while the lower slope is more influenced by more distant sources, such as the Antarctic Peninsula. Nevertheless,

435 sediments on the shelf and upper slope are carried by the flows of the SASW and AAIW, while the UCDW and LCDW transport sediments from the lower slope.

The Río de la Plata is the primary influencer of the sediments off southern Brazil up to the 27°S parallel. The sediments are transported northwards by the PPW, which is transported by a wind-driven current. A mixture of sediments from the PPW and the north is transported towards the slope between 34°S and 28°S.

Finally, Pre-Cambrian terrains are the primary sources of the sediments deposited further north. They are originated from rivers located northward of the area of study and, on a smaller scale, by the small drainages that face the ocean in the Serra do Mar region.

We propose that the main oceanographic boundaries of the southwestern South Atlantic margin, i.e., the Subtropical Shelf Front and the Santos Bifurcation, separate distinct geochemical provinces.

445 Data availability

All of the data used in this paper is presented as Supplementary Material

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The authors declare that they have no known competing financial interests or personal relationships that could have <u>influenced</u> the work reported in this paper.

800 Data availability

All of the data used are available as a Supplementary file

Excluído: appeared to influence