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Southern Africa: The Missing Piece to the Dust Provenance Puzzle of East Antarctica?

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Abstract

Mineral dust is a natural tracer of atmospheric composition and climate variability. Yet, there is still much to be known about the Southern Hemisphere dust cycle during the last Pleistocene. Major efforts have attempted to solve the 'puzzle' of the origin of the potential source areas that contribute dust to the Southern Ocean and East Antarctica. Here we present a comprehensive geochemical characterization of an important potential source area, which role as a dust supplier to different environments of the SH has significantly been underestimated, that is, the Southern Africa (SAF) region. On the basis of Sr-Nd-Pb isotope ratios and rare earth element concentrations analyzed in sediments collected along the major dust-producing areas in the Namibian coast (Kuseb, Omaruru and Huab riverbeds and the Namibian sand sea region), this study demonstrates for the first time that SAF emerges as the second most important dust source to EA during interglacial times.

1. Introduction

Mineral dust is considered a sensitive natural tracer of modern and past environmental conditions and a paleoclimatic proxy that reflects the variations in the atmospheric circulation during different climatic periods and their transitions. The type and intensity of dust emissions are a consequence of different conditions at the source (sediment supply and availability) and aeolian processes (deflation, transport, deposition)(e.g., Shao et al., 2010). Once dust is emitted into the atmosphere, it remains in suspension until physical and chemical conditions favor its deposition. The deposited dust is archived in different proxy records either, over the oceans or onto continental land-masses within ice cores, deep sea sediments, loess, peat bogs, etc. For the Southern Hemisphere, reconstructions of dust buried in Antarctic ice cores are considered an excellent proxy for the paleo-record of wind patterns and its intensity. In this regard, these records indicated that dust fluxes were up to ~25 times higher during glacial periods due to dust production strengthening at their source areas, compared to warmer periods when dust concentrations were extremely low (Lambert et al., 2008).

Introduced by Delmonte et al. (2004a), the term Potential Source Areas (PSAs) denotes either a primary source of mineral dust, derived directly from the physical and/or the chemical weathering of the parent material or a secondary source for dust, where aerosol particles have already been subjected to a phase of aeolian and/or fluvial transport. For the Southern Hemisphere, Southern South America (SSA), Australia (AUS), New Zealand (NZ), Southern Africa (SAF), and the Antarctic Dry Valleys have all been identified as PSAs. The first study to trace the dust origin in Antarctica ice cores was Grousset et al. (1992). In agreement with the Sr-Nd isotope data presented in that study, all the studies since have demonstrated that the main dust contributor to the Southern Ocean (SO) and East Antarctica (EA) during glacial (MIS2-MIS20) and interglacial periods is SSA, mainly from Patagonia (e.g., Basile et al., 1997; Delmonte et al., 2004a; 2019; Gaiero, 2007; Gili et al., 2016, 2017; Vallelonga et al., 2010). Despite the agreement on the dominance of a Patagonian dust origin during glacial periods there is presently a debate regarding the Holocene dust PSAs with the following scenarios of dust provenance proposed: *i*) a weakened Patagonian source complemented by a lower latitude source region (Southern Central West Argentina, SCWA) (Gaiero, 2007; Gaiero et al., 2004; Gili et al., 2017); *ii*) a Puna-Altiplano dust source transported via the southern tropical jet stream (Gaiero, 2007; Gaiero et al., 2013; Gili et al., 2016, 2017) and, *iii*) a general contribution from Southern South America plus a secondary input from AUS (De Deckker et al., 2010; Du et al., 2018; Revel-Rolland et al., 2006) or NZ (Wegner et al., 2012). Some of these interpretations were partially supported by Atmospheric General Circulation Models (AGCM),

suggesting that while SSA dust dominates deposition over the Atlantic Southern Ocean and East Antarctica, AUS modern dust prevails over the Pacific SO and towards the western coastal regions of Antarctica (Albani et al., 2012; Li et al., 2008). Neff and Bertler (2015) concluded from back-trajectory modeling that the transportation of aeolian material from NZ to the southern high latitudes is possible. On the contrary, a recent study carried out in sediments from the major present-day dust and sediment producing regions of the South Island in NZ, has indicated this area was not a significant dust supplier to EA (Koffman et al., 2021). Using AGCMs, Li et al. (2008) showed that after SSA and AUS, Southern Africa is the third main dust supplier in the Southern Hemisphere with dust depositions mainly taking place in the South Atlantic and Indian Ocean. Even though this region is a significant dust emission source, little is still known about it being a PSA as the geochemical and modeling data available for this region is extremely sparse. Thus, it is questionable if the role of SAF as a dust contributor to the SO and EA has plausibly been underestimated. For many years, only the Makgadikgadi Pan in Botswana and the Etosha Pan in Namibia were recognized as persistent dust sources in SAF (e.g., Prospero et al., 2002; Bryan et al., 2003; Mahowald et al., 2003; Li et al., 2008), neglecting the coastal Namibian sources. In fact, it was only until recently that the Namib Desert coast has been identified as one of the largest sources of dust in Southern Africa (Von Holdt et al., 2017; Vickery et al., 2013). Using remote sensing satellite images, these authors have found that along the southwestern coast of SAF, the Kuiseb, Omaruru, and Huab Rivers beds are the three main areas of greatest dust emission located on recently deposited fluvial surfaces. Revisiting the literature, it became apparent that while most of the studies have been focused on the detailed study of SSA, AUS and most recently NZ dust sources (e.g., Gili et al., 2017; DeDeckker et al., 2019, Koffman et al., 2021), other PSAs, such as SAF, have been overlooked. Thus, there arises a compelling need for more extended geochemical studies to fill the current gap in understanding the past and present long-range transportation of mineral dust particles to the high-latitude environments.

Responding to this need, this study aims to investigate the role of SAF as a PSA and provides the first comprehensive north-to-south geochemical characterization of riverbed sediments collected along the major dust producing river systems in the Namib desert (Huab, Omaruru and Kuiseb). Combining rare earth elements (REE) concentrations and the Sr–Nd–Pb isotope compositions, we seek to evaluate the influence of this region as a dust source to the high latitude environments of the Southern Hemisphere. We compare the geochemical signature of bulk and resuspended aerosol fractions from SAF, with the dust fingerprint reported for Antarctic ice cores (Vostok and EDC) during glacial (MIS2–MIS20) and interglacial periods

(Holocene and MIS 5.5). In addition, we also include a re-analysis of the isotopic composition of SAF samples previously reported in Delmonte et al. (2004a) and Vallelonga et al. (2010).

2. Geological and Climatic Settings

Namibia is located at the intersection of the Mesoproterozoic Namaqua and Neoproterozoic Damara orogenic belts welding the Archean Kalahari and Congo Cratons (Fig. 1a; Jacobs et al., 2008). The Namib desert extends N-S for approximately 2000 km along the western coast of southern Africa between the Kunene and Orange Rivers (Fig. 1a; Heine and Heine, 2002). The geology of the region is dominated by the lithological sequence of the Damara Orogenic system including 2.0–1.2 Ga gneissic basement overlaid by Neoproterozoic to Paleozoic metasediments intruded by 840–460 Ma granitic. Granites are the most common igneous rock type (c. 94%), while the remaining intrusions are equally subdivided between diorite and tonalite/granodiorite (Miller, 1983; 2008). The eastern border for the Namib desert is delimited by the Great Escarpment (~2000 m high) running parallel to the coast and 100–150 km inland (Fig. 1a). Along the Between the Kunene and the Orange Rivers, twelve aligned ephemeral river valleys extend sub-parallel east-west along the Atlantic Coast (Jacobson et al., 1995). Sporadic summer rainfalls of $<25 \text{ mm yr}^{-1}$ in coastal areas (Eckardt et al., 2013) sometimes generate floods of several days but otherwise, these river beds stay dry. The limited rainfall water dissipates due to infiltration and evaporation, while any fluvial transported sediments are deposited along river valley channels making them available for aeolian entrainment and transport.

In this study, sediments from three of the driest river valleys along the coast, the Huab, Omaruru, and Kuiseb are investigated (Fig.1a). These valleys cross the Damara Belt, where the main catchments are dominated by fluvial sands from the Damara Orogenic belt (Jacobsen et al., 1995). The Huab River sand composition is mainly composed of feldspars and quartz (Garzanti et al., 2014). Omaruru and Kuiseb mainly draw sediments from the Damara branch, which widely exposes highly metamorphosed rocks and granitoid intrusions (Garzanti et al., 2014; Jung and Mezger, 2003). The Kuiseb valley also runs along the northern edge of the Namib sand sea (Fig.1a), where the Orange River sediments, which are shown to be pushed northward by the dominant southerly winds and powerful along-shore currents, are known to be the predominant bed-load sediment source (Vermeesch et al., 2010; Jacobson et al., 1995; Lancaster, 1983) despite being ~1500 km apart (e.g., Compton et al., 2010).

The regional atmospheric circulation patterns and the arid climate of the Namibian coast are produced by a quasi-stationary subtropical South Atlantic anticyclone and the cold northward-

flowing Benguela current (Compton et al., 2004). For most of the year, strong onshore south-to-southwesterly trade winds over the dry river valley regions are responsible for 90 % of the sand flux, especially during winter and spring (Tlhalerwa et al., 2005; Todd and Washington, 1999)(Fig.1b). Northeasterly ‘Bergwinds’ are also another important agent of aerosol transportation and deposition off the west coast of SAF because of their high velocities and subsidence offshore (Fig.1b). Common from April to August, Bergwinds are shown to be responsible for >60% of the offshore sediment transport with dust plumes extending ≥ 100 km into the Southern Ocean (Vickery et al., 2013). In addition, with an estimated annual dust input of 4.4 tones into the South Atlantic Ocean, this wind system is in charge of supplying nutrients to the highly fertile Benguela upwelling (Tlhalerwa et al., 2005). Namibia’s offshore waters are dominated by the Benguela upwelling system, which is wind-driven by the regional alongshore southeast trade winds and recognized for being a region with the highest rates of primary production in the ocean where dust with high nutrient content deflated from the continent, it is discharged offshore fertilizing the ocean waters (Jickells et al., 214). Although minimum annual variations in wind intensities occur, the Benguela upwelling system is often separated into three zones; north, central, and south (Jury, 2012). The north and south Benguela upwelling zones are respectively bounded by the Angola and Agulhas Currents. The central zone presents the least fluctuating regime and is the most relevant zone for our research as it lies contiguous to the main dust source region investigated here between 22° and 28°S.

3. Sampling and analytical methods

With the aim to evaluate the provenance and the long-range transportation of dust depositions in remote areas potentially related to Southern Africa PSAs, surface sediment samples were collected across the lower valleys of Huab, Omaruru, and Kuiseb rivers (Fig. 1a, red dots. See table S1 for more details). A total of 22 bulk samples (9 samples from Huab, 7 from Omaruru and 6 from Kuiseb) were analyzed in terms of their REE concentrations and Sr-Nd-Pb isotope compositions. Additionally, measurements of Sr-Nd-Pb isotope ratios were carried out in 5 bulk and 5 fine ($< 5 \mu\text{m}$) fraction samples from the Namib Sand Sea (Fig. 1a, blue diamonds), to produce a comparable result to the bulk samples previously analyzed by Delmonte et al. (2004) and Vallee et al. (2010). Furthermore, to assess a possible change in the geochemical fingerprint due to grain-size effects, six bulk samples from the Huab valley were selected to generate airborne aerosols dust in the fine fraction ($\sim < 11 \mu\text{m}$) using an atmospheric simulation chamber (see section 3.1 and supplementary materials for further details). The

aerosol fraction investigated in this study has also important implications for provenance studies as only certain particle sizes can be transported long distances. Hence, analyzing the right size fraction in the source region is crucial for to identify the origin of dust reaching the Southern Ocean and Antarctica.

3.1. Sample collection

Bulk surface samples from Huab, Omaruru and Kuiseb riverbeds were collected in a field camping in August 2017, using a trowel to obtain approximately 200 g of sediment from the surface. Sub-samples of ~350mg were stored in pre-cleaned centrifuges tubes and shipped to Université Libre de Bruxelles (ULB), Belgium for analysis. From the bulk of six samples from Huab, dust aerosols (fine fraction) were generated into the CESAM atmospheric simulation chamber, to mechanically reproduce the saltation and sandblasting processes responsible for the release of mineral dust in natural conditions (Di Biagio et al., 2017). Briefly, dust aerosols are generated from the bulk sediments by a mechanical shaker and the aerosols are injected into the CESAM chamber, a stainless-steel reactor with a volume of 4.2 m³ (Wang et al., 2011) where dust is dispersed, left in suspension for ~ 3 hours, and ultimately recovered on acid-cleaned 0.2 µm Teflon filters. Additional details about the dust generation in the CESAM are provided in the supplementary material. The fine fraction (<5 µm) of the Namibia Sand Sea samples was separated following the procedure explained in Delmonte et al. (2004a) using the Stoke's Law.

3.2. Geochemical analyses

Sample preparation and analyses for REE elemental and, Sr-Nd-Pb isotope compositions were carried out on bulk and fine fraction samples. Sediment samples were dissolved in an HF+HNO₃ mixture followed by an HCl bath in Savillex vials and Sr-Nd-Pb were separated through chromatography using ion-exchange resins following the procedure reported in Vanderstraeten et al. (2020). Isotopes were measured at the G-Time Lab, Université Libre de Bruxelles (Belgium) using a High-Resolution ICP-MS Plasma Nu II (Nu Instruments) in dry mode. The REE compositions were determined using a Quadrupole ICP-MS Agilent 7700. Additional details of the analytical methodology are provided in the supplementary material.

4. Results

4.1. Southern Africa sediments and their Parental Rock Sources

The REE concentrations and Sr-Nd-Pb isotopic compositions of the bulk and fine fraction of sediments from the Huab, Omaruru, Kuiseb and Namibia Sea Sand are listed in Table S1. The Southern Africa PSAs signature of the windblown samples presented in this study reflect the compositional variability of the locally outcropping rocks and sediments: a chemical and isotopic fingerprint that is the result of the mixing between the metasedimentary rocks from the Damara and the Kaoko belt and the Orange River (Garzanti et al., 2014; Goscombe et al., 2003) (Fig. S1).

4.2. Rare Earth Elements (REE) Composition

In Fig. 2, the composition of REE for the Southern Africa PSAs normalized to the upper crust (UCC, Taylor and McLennan, 1985) is plotted. This figure indicates that no significant difference in the REE composition prevails among all four areas characterized in this study and a regional differentiation is difficult. In general, samples show flat REE patterns with a minor decrease in the heavy REE abundances (Fig. 2a). Both the bulk and the fine fraction from Huab and Kuiseb exhibit $\text{Eu}/\text{Eu}^* < 1$, with Omaruru displaying slightly higher values (Fig. 2b). The Eu anomaly (Eu/Eu^*) was calculated with the equation $\text{Eu}/\text{Eu}^* = \frac{[\text{Eu}]}{([[\text{Sm}]+[\text{Gd}]]/2)}$. Figure 2b also shows that the REE composition of the bulk and the fine fraction from the Huab area are very similar, suggesting bulk samples are representative of the fine particles and that Southern Africa REE signature is independent of the grain-size fraction of sediments. For comparison purposes, soil samples from the Walvis bay in Namibia have also been included, whose source has been attributed to the local dust blown from the nearby Namibia Sea Sand (Bayon et al., 2005).

4.3. Strontium, Neodymium and Lead Isotopic Compositions

Compared to REE data, the Huab, Omaruru, Kuiseb and Namib Sea Sand samples can be better discriminated based on their isotopic compositions (Fig. 3). Lead isotope compositions span within a relatively large range of values: $^{206}\text{Pb}/^{207}\text{Pb}$: 1.15–1.28, $^{208}\text{Pb}/^{207}\text{Pb}$: 2.43–2.62 defining variable Pb isotope fields (Fig. 3a). The Huab valley samples display intermediate values ($^{206}\text{Pb}/^{207}\text{Pb}$: 1.205–1.228, $^{208}\text{Pb}/^{207}\text{Pb}$: 2.482–2.518) between the Kuiseb and the Namibia Sea Sand regions which represent the highest and lowest (respectively) end-members in terms of Pb isotopic compositions. In this sense, the Kuiseb related samples have a more Th-rich signature ($^{206}\text{Pb}/^{207}\text{Pb}$: 1.23–1.277, $^{208}\text{Pb}/^{207}\text{Pb}$: 2.495–2.612) indicating derivation from sources with higher Th/U ratios. Contrarily, the Namib Sand Sea area shows comparatively the

most unradiogenic Pb isotopic values ($^{206}\text{Pb}/^{207}\text{Pb}$: 1.159–1.206, $^{208}\text{Pb}/^{207}\text{Pb}$: 2.425–2.468), with a clear distinction between the bulk and the fine fractions. Interestingly, samples associated with the Omaruru valley define two isotopic subfields: one overlapping with Huab and a second field with less-radiogenic isotopic ratios ($^{206}\text{Pb}/^{207}\text{Pb}$: 1.181–1.212, $^{208}\text{Pb}/^{207}\text{Pb}$: 2.439–2.488) showing a similar fingerprint to the fine fraction of the Namib Sea Sand (Fig. 3a).

The Sr–Nd isotopic ratios from SAF samples measured in this work (Fig. 3b) exhibit two main groups, indicating plausible dust provenance from different source end-members coherent with the geological history of this part of the continent. The isotopic variability in sediment samples derived from the Omaruru and the Kuiseb riverbeds is large ($^{87}\text{Sr}/^{86}\text{Sr}$ = 0.7208 to 0.7384 and $\epsilon\text{Nd}(0)$ = -12.80 to -10.17). In contrast, the Namibia Sand Sea samples depict an even broader range of isotopic values: $^{87}\text{Sr}/^{86}\text{Sr}$ = 0.7128 - 0.7305 and $\epsilon\text{Nd}(0)$ = -10.39 to -9.84, reflecting provenance from the granitic intrusions of the Damara belt (e.g. Miller, 2008). The Huab riverbed, which drains the Paleoproterozoic basement of the Kaoko belt (Goscombe et al., 2003), is restricted to a narrower range of isotopic values: $^{87}\text{Sr}/^{86}\text{Sr}$ = 0.725–0.728 and $\epsilon\text{Nd}(0)$ = -18 to -15. We also observe a Nd isotopic trend with values becoming progressively more radiogenic from north to south. This is, the Huab region (in the north of the Namib coastline), exhibits the less radiogenic Nd composition with $\epsilon\text{Nd}(0)$ between -18 and -15, continuing towards the Omaruru and Kuiseb valleys with intermediate Nd compositions before reaching the southernmost location of the Namibia sand sea region which exhibits ϵNd values between -10.5 and -8.

4.4. Grain-Size isotopic fractionation

Attributable to different mineral contributions, mainly clays and feldspars, that are ultimately controlled by mineral sorting processes (e.g., Garçon et al. 2014), it is observed that the fine-grained fractions show a more-radiogenic signature compared to the bulk of the same samples. In the present study, the set of samples from the Huab and Namibia sand sea regions analyzed in the bulk and the fine fractions, conforms to the longstanding observation that there are statistically significant differences in $^{87}\text{Sr}/^{86}\text{Sr}$, but much less so for $\epsilon\text{Nd}(0)$ and Pb isotopic values, when comparing the fine and bulk grain-size fractions of these sediments (Figs. 3a, b). While the Namibia Sand Sea samples show the highest Sr isotopic differences ($\Delta ^{87}\text{Sr}/^{86}\text{Sr} \sim 0.012$ units), the Huab region difference is limited to only ~ 0.002 units (Fig. 3b). Furthermore, for both areas the mean difference between $\epsilon\text{Nd}(0)$ values for the fine and bulk fractions is $<1\epsilon$ units and not significant. For Lead, these differences are also small ($\Delta ^{206}\text{Pb}/^{207}\text{Pb}$ and

$^{208}\text{Pb}/^{207}\text{Pb}$ ~ 0.022 units for Namib Sand Sea samples and, even smaller Δ for the Huab samples: ~ 0.01 units) meaning that the sorting by size, although not a zero effect, does not play a significant role that could complicate attempts to relate the Pb isotope data in remote locations to PSAs. Overall, our results agree with the observed small differences between clay and silt for grain-size fractions in river sediments (e.g. Bayon et al., 2015). The new data presented here corresponding to the finer fraction of Southern Africa sediments agrees with the previous Pb isotopic ratios ($n=2$) reported by Vallelonga et al. (2010) for the same region. Surprisingly, the strontium isotope ratios previously presented in Delmonte et al. (2004a) for this area, are slightly less radiogenic in $^{87}\text{Sr}/^{86}\text{Sr}$ compared with the new values obtained here for the same samples (Fig. 3b). Despite the disparity, this does not affect our interpretations and any difference in the Sr isotopic data between the bulk and the fine grain-size fractions of sediments are small compared to the variability of the whole Southern Africa region, reinforcing the hypothesis that the isotopic signatures can be used to fingerprint provenance.

5. Discussion

During the late Quaternary, an enhance in continental windblown dust production triggered by the expansion of outwash plains and exposed land areas in general, led to higher dust fluxes during glacial periods. For over three decades, Sr-Nd-Pb isotopes in combination with chemical, physical, and modeled studies have focused on the provenance of dust recovered from East Antarctica ice cores. In this sense, compelling evidence has attributed Southern South America as the primary source of dust to Antarctica during both, cold and warmer periods (e.g. Basile et al., 1997; Delmonte et al., 2004a; Gaiero et al., 2007; Gili et al., 2016, 2017; Albani et al., 2021; Li et al., 2008). Nevertheless, the contribution from other areas in the Southern Hemisphere is still unclear due to the inability to discriminate each individual PSAs because the significant overlap between the different geochemical fingerprints. This matter has kept most of the attention on determining whether if possible to distinguish these different dust signatures and possibly overlooked other important secondary contributions, as is the case of Southern Africa. Even still there is no consensus on whether these PSAs (SSA, AUS, and NZ) should be considered and/or excluded as the dust supplier to EA during the Holocene; here we adhere to the observations of previous studies suggesting that neither AUS or NZ are relevant sources to explain the signature of dust found in East Antarctica (Gili et al., 2016, 2017; Koffman et al., 2021), and therefore have not been taken into account in the present

discussion. In contrast, we provide compiling evidence in favor of SAF as an important PSA contributing dust to the high-latitude atmosphere and ocean.

5.1. Southern Africa (SAF) and Southern South America (SSA) fingerprints

The SAF and SSA signatures can be partially discriminated by combining trace elements (REE) with isotope data (Fig. 4). In general, SAF dust sources evidence low radiogenic Nd (with ϵNd values lower than -10), and more radiogenic values of $^{206}\text{Pb}/^{204}\text{Pb}$, reflecting the signature of the Damara basement (Figs. 4a, b). In terms of REE, Huab and Kuiseb sediments exhibit an average $\text{Eu}/\text{Eu}^* < 1$, while Omaruru appears divided in two groups of samples; one following the same trend with $\text{Eu}/\text{Eu}^* < 1$ and another group with scattered data, showing higher values of Eu/Eu^* (Fig.4a). Contrarily, SSA exhibits a fingerprint that varies from more “mafic-like” compositions from Patagonia, reflected by Eu/Eu^* values > 1 , higher $^{143}\text{Nd}/^{144}\text{Nd}$ and low $^{206}\text{Pb}/^{204}\text{Pb}$, in contrast with more “crustal-like” compositions of material from the Puna-Altiplano Plateau area (Gili et al., 2017; 2016) (Figs.4a, b). For the Namibia sand sea region, only one data point for a bulk sample is available in terms of REE and its composition is similar to the Huab area (Figs. 2a, b). In terms of $^{143}\text{Nd}/^{144}\text{Nd}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ isotope ratios, the Namibia sand sea reveals a similarity with some areas of SSA, overlapping with the Southern Altiplano and Northern Puna isotopic composition field (Fig.4b).

5.2. Southern Africa fingerprint in High-Latitude Environments

Based on Sr-Nd-Pb isotopes, only two earlier studies (i.e., Delmonte et al. (2004a) and Vallelonga et al. (2010)) suggested the possibility of SAF as a PSA to the polar Antarctic regions. However, due to the limited number of sediment samples and their different signature with respect to EA, they concluded isotopic data was not enough to support a dust contribution from SAF, excluding this region from the provenance puzzle. Contrarily, the new and expanded dataset of Southern Africa Sr-Nd-Pb isotopic ratios and REE concentrations presented in this study indicate that, SAF emerges as PSA that together with high-latitudes sources in Southern South America (that is, the Puna-Altiplano Plateau) can explain the interglacial dust signature found in East Antarctica ice cores, as well as the signature of marine sediments from the South Atlantic sector of the Southern Ocean (Fig. 5).

In general, the $^{87}\text{Sr}/^{86}\text{Sr}$ and $\epsilon\text{Nd}(0)$ values are more diagnostic than the $^{206}, ^{208}\text{Pb}/^{207}\text{Pb}$ for identifying the glacial/interglacial dust provenance in paleo-archives. This means that even though the Pb isotopic data is usually less indicative because of the Pb input related to local (Antarctic) volcanoes in comparison to the dust related Pb (Vallelonga et al., 2010), the Pb data

in the present study add critical support to our interpretations. This is, the new Pb isotope data of SAF samples define a field that can account for the entire range of Pb isotopic compositions observed in the Antarctic dust from Vostok and EDC ($^{208}\text{Pb}/^{207}\text{Pb}$ values from 2.44-2.50) (Fig. 5a). It should also be noticed that the very radiogenic Pb data similar to Kuiseb field are also those of the Antarctic local volcanoes (Mt. Erebus); contribution that has been recognized in previous studies (e.g., Hinkley, 2007; Vallelonga et al., 2010; Gili et al., 2016) and cannot be denied. Our results show that EA dust samples show two main compositional trends (orange dashed lines in Figure 5a): *i*) the Namibia Sea Sand in combination with a more radiogenic end-member, that is, either Kuiseb PSA or the local Antarctic volcanism and, *ii*) as suggested by Gili et al. (2016), Patagonian dust plus a McMurdo-Erebus volcanic contribution. In conclusion, Antarctic Pb dust data fall on a compositional line where a mix of SAF and SSA with a local volcanic end-member could explain the EA dust fingerprint.

This is also in part supported by Sr and Nd isotopic data (Figs. 5b, c), where the new SAF defines a compositional field that turns out to be a key end-member for describing the composition of dust deposited in the South Atlantic and EA. In general, during the last glacial climatic periods of the last Quaternary, EA data is very uniform converging to an unequivocal signature from Patagonia with additional inputs from other lower latitude areas in SSA, such as, the Southern Central Western Argentina and Southern Puna (Fig. 5b) (Gili et al. (2017) and references therein). On the other hand, the EA interglacial dust fingerprint, as well as the glacial/interglacial signature of marine sediments from the South Atlantic are marked by a variable mix of sources (Fig. 5b). An important finding of this study is that a tri-mixing hyperbola with common end-members can be drawn and help to decipher this variability. This is, the best candidates to elucidate the origin of dust during the Holocene and likely during the last 15 ka are a more mafic-like end-member represented by Patagonia and/or SCWA, in addition to a second crustal-like end-member, defined by the Southern Altiplano, plus a third end-member, which can either be the northern Puna (black solid line) or Southern Africa (dotted black line) and/or a mix of these two (Fig. 5c, Table S2). This is also in agreement with the isotopic composition of present-day dust deposited on the Berkner Island in the Atlantic sector of coastal Antarctica, which can clearly be ascribed to a mixing isotopic composition from PAP (Bory et al., 2010; Gili et al., 2017) and SAF PSAs (Fig. 5c). In this sense, Bory et al. (2010) pointed out that neither of the PSAs in SSA could account for the entire Sr-Nd isotopic range of the dust deposited on Berkner and the contribution of another secondary source with more negative ϵNd (0) was missing. The Sr and Nd signature of the SAF samples presented here can account for that ‘missing’ end-member. Alike, Noble et al. (2012), indicated

the signature identified in ocean sediments from the South Atlantic is associated with material derived from SAF, but also with compositions similar to the northern-Puna (Fig.5b, c). Aeolian dust blown from the Namibian region by the SE trade winds is considered one of the main sources of detritus to the South Atlantic Ocean (e.g., Dieckmann et al., 1996). In this sense, interglacial detrital fractions recovered from marine cores from the SE Atlantic region display $\epsilon\text{Nd} (0)$ values between -10 and -22 that have been attributed to a Southern Africa origin (Bayon et al., 2003), which is also in accordance with the SAF Nd isotopic data presented here. As presented by the evidences above and as seen in Fig. 5, the composition of interglacial and present-day dust deposited in East Antarctica and the South Atlantic ocean can be explained by the mixing between Patagonia, the Puna-Altiplano Plateau and Southern Africa, although the distinction and relative contribution from the different PAP and SAF source areas remains puzzling. It should also be noted that the SAF source becomes more important in the SO and marginal sites of EA where the SSA signature is not enough to justify the data, although local Antarctic sources may also play a role. The similar geochemical composition exhibited by these two areas (i.e. SAF and the PAP) could be related to the comparable climatological aspects that the Puna-Altiplano Plateau and Southwestern Africa share from their tropical to sub-tropical location (between about 17° and 30°S latitude) (Tyson, 1986). Both regions are influenced by atmospheric circulation dominated by subsidence air zones (SE Atlantic-Namibia- and SE Pacific -PAP-), promoting extreme arid conditions and producing the world's desert lands. Reinforcing the aridity in these areas is the presence of an upwelling, north-flowing, cold oceanic current; the Humboldt along the west coast of South America and the Benguela along western South Africa. Precipitation is the result of tropical convection and rainfall patterns are controlled by the South American summer monsoon (SASM) and East African summer monsoon (EASM), respectively. Both areas have shown important climatic variability during the late Quaternary, indicating altering cycles of wet/dry conditions, however interpretations about the main factors determining these changes is still controversial. Some authors have pointed to a precessional orbital control after finding a correlation between periods of Southern Hemisphere maximum insolation and enhancement of moist air on land, increasing precipitation delivered by the SASM and EASM (Baker et al., 2001; Fritz et al., 2004; Partridge et al., 1997). Others have suggested that southwestern Africa responds to variations in phase with the Northern Hemisphere summer insolation (e.g., Chase et al., 2009). Despite the debate of these complex mechanisms, there is persuasive evidence supporting the important role of both the PAP and SAF regions as dust suppliers during the late Quaternary. The arid and dry environmental conditions in Namibia have been similar to what it is today for the last 50 Ky,

with the late Holocene being the driest period (Chase et al. (2019) and references therein). Based on a pollen marine record, Shi et al. (2000) indicated two arid peak periods (14-12 Ky and 11-9 Ky) for the early Holocene in this part of the Southern Africa continent. Authors suggested a northward migration (from its present position) of the intertropical convergence zone (ITZC) resulted in a reduction of the water vapor supply in southern subtropical Africa during the early Holocene. A marine grain-size record (Stuut et al., 2002) and a collection of terrestrial records (Chase and Meadows, 2007) also suggested arid conditions in southern Africa during interglacial stages due to meridional shifts in the atmospheric circulation system. These authors indicated intensified trade-winds during glacials compared to interglacials. Depleted isotopes in precipitation have also pointed out to dry conditions during warm periods (Collins et al., 2014). In line with this, a more recent pollen record from the southern Namib Desert obtained from rock hyrax middens have shown the Holocene was a time where high temperatures and evapotranspiration resulted in increased aridity and an expansion of the desert vegetation (Lim et al., 2016). These environmental conditions of dryness during interglacial times are also indicated for the PAP area (see details in Gili et al., 2017), especially during the Holocene (Fornace et al., 2014) and MIS 5.5 (Baker and Fritz, 2015). In addition, a combined grain-size study of two marine cores taken off the coast of southwestern Africa (~20S) and western South America (~27S), observed both sites showed arid conditions for at least the last 35,000 years (Stuut and Lamy, 2004).

5.3 Atmospheric circulation pattern and dust deposition over Antarctica

While the geochemical evidences presented here, demonstrate that SAF sediments have a dust signature comparable with the interglacial and modern dust deposited in different environments of the SH, the concept about how the transport occurs from the Namibian coastal regions to Antarctica is still not well known. The deposition of dust around Antarctica can happen by either a scenario whereby it is deposited in the interior high elevation sites or in the periphery low elevation sites of the Antarctic plateau (e.g., Delmonte et al., 2008). Mineral dust reaching the EA plateau is windborne long-range transported from the continental land-masses carried through the mid-to high troposphere. For dust sources that are not located at high latitudes, such is the case of SAF, there is need of strong convective uplift of particles up to 200-400 mbar to reach the interior plateau and the presence of SAF particles has mostly been reported in the lower troposphere (Chazette et al., 2019). Another possibility is that the source itself is already elevated in altitude allowing the injection of dust particles directly in the high

441 troposphere. This is the case of the Puna-Altiplano Plateau in SSA, where the subtropical jet
442 stream acts as main mechanism mobilizing large amounts of dust that ultimately, get deposited
443 onto the polar plateau (Gili et al., 2017; Gaiero, 2007; Gaiero et al., 2013) and whose signal
444 has also been found in peripheric areas of Antarctica (Bory et al., 2010). Although a significant
445 dust emission source, modeling data has suggested that material from SAF only contributes
446 with ~5% of the total dust burden over Antarctica, mostly because the emissions are localized
447 near the source, and a southward transport to the inner part of the Antarctic continent is unlike
448 (Albani et al., 2012; Li et al., 2008). However, modeled dust emission fluxes from the different
449 PSAs of the SH and their subsequent deposition driven by the local trade winds, can vary by
450 as much as a factor of four or more (Neff and Bettler, 2015), which could mislead to
451 controversial interpretations. In this sense, comparatively with the study of Neff and Butler
452 (2015), who indicated that even in small amounts SAF dust could be found in East Antarctica,
453 a more recent global model simulation indicated that together with SSA, that accounts for
454 almost the bulk of the deposited dust, Southern Africa represents a secondary source that
455 contributes to the total dust deposition flux to Antarctica (Kok et al., 2021). Similar
456 observations have also been highlighted for the Southern Ocean. Using HYSPLIT, a trajectory
457 model, Bhattachan et al. (2012) showed that dust emissions from the Southern Kalahari
458 commonly reaches the Southern Ocean, with most of the trajectories traveling beyond 40°S
459 during the dry period (April-September) when conditions favor the dust production and
460 transportation (Bryant et al., 2007).

461 We note however, that most of the regional to global dust models regarding Namibian sources
462 are solely based on the atmospheric circulation of the trade winds to emit dust. While in reality,
463 it is the Berg winds, a local southwestern Africa topographically enhanced thermal wind that
464 represents a more important mechanism responsible of dust mobilization and deposition over
465 the ocean (Vickery and Eckardt, 2013; Todd and Washington, 1999). These hot and dry winds
466 are also more common in winter and they are the result of adiabatic heating; a process that
467 occurs when air coming from the escarpment and plateaus descending towards the Namibian
468 coastline, where it is compressed and heated. Multiple dust streams and dust storms originated
469 in this part of the African continent evidencing the substantial transport of material to the
470 Southern Ocean as a result of Berg winds have been spotted by different satellite platforms
471 (Von Holdt et al., 2017; Vickery and Eckardt, 2013). This has important implications for the
472 transportation of material to the marginal and coastal sites of Antarctica, which can receive an
473 unquantified amount of SAF dust. On this matter, more recent studies have showed strong
474 support that highlights Southern Africa as a secondary and significant dust supplier to East

Antarctica. Gili et al. (2019), indicated the Sr, Nd and Pb isotope ratios of Antarctic modern dust recovered from snow samples collected along coastal sites of the Dronning-Maud Land area (DML-East Antarctica), which showed similar composition to the Southern Africa sediments. This data is consistent with the results observed by Vanderstraeten et al. (2019), in a back trajectory FLEXPART particle dispersion transport model where a mix of modern Southern Africa and Southern South America sources was clearly spotted in the coastal area of East Antarctica. Likewise, using a statistical analyses Vanderstraeten et al. (2021) revisited the only EDML ice core rare earth elements dataset available from Wegner et al. (2012), and observed that the Southern Africa fingerprint appears at ~15kyr as an important mixing end-member that together with SSA, can explain the East Antarctica interglacial dust signature. Ultimately, another study carried out in a core from the Southern Indian Ocean, has also proposed a mixing model where SSA and SAF have been found to be the main distal dust sources through most of the Holocene (Li et al., 2020). In resume, we have presented different evidences indicating the transport of SAF material reaches the South Atlantic sector of the Southern Ocean and the marginal sites of East Antarctica. Thus, one could infer that the same material could also be transported farther distances and ultimately get deposited in the interior of the polar plateau. However, more modeling studies are necessary to fully prove this type of transportation.

6. Conclusions

Because wind-borne minerals are strongly linked to atmospheric dynamics, the characterization of present-day Southern African dust and its potential sources is useful for the interpretation of paleo-records. Improving our knowledge about the dust cycle in the Southern Hemisphere can provide important information about atmospheric circulation patterns during the last climatic cycles. This study represents a step forward in the documentation of aeolian dust sources in SAF. We have combined chemical and isotopic data to characterize for the first time the main desert areas along the Namibian coast, a potential dust source area, whose contribution to the high-latitude environments of the SH has been underestimated, mostly due to the lack of SAF data from suitable samples. Here, we have compared the rare earth elements concentrations and Sr–Nd–Pb isotopes of SAF PSAs with those indicated as the main dust sources to the Southern Ocean and Antarctica during glacial and interglacial periods. Our results show that the Huab, Omaruru, Kuiseb, and the Namibia sand sea regions in SAF, denote a dust signature very similar to the one exhibit by the PAP in SSA. Moreover, both SSA and

SAF fingerprints match very well with the composition of interglacial sediments deposited in the SO and EA. However, a distinction of which areas are more dominant remains challenging, which can be related to the common position of these regions in subsidence air zones within the SE Pacific and SE Atlantic anticyclones. This characteristic is also responsible for the extreme aridity conditions that characterize both regions in the present climate and that has also been observed during warm times, especially the Holocene.

In summary, this study agrees with previous findings suggesting that SSA is the best candidate to explain the dust signature recorded in the SO and EA during cold and warm stages. For interglacial periods, this work proposes a scheme where the typical isotopic variability observed in interglacial and present-day dust sediments deposited in the South Atlantic Ocean and East Antarctica can be related to the contributions of two main PSAs: Southern South America, through westerlies dominating the atmospheric circulation over Patagonia and SCWA and, the jet stream over PAP, in combination with Southern Africa in response to the plausible transport by strong Berg winds. More modeling studies focusing on the atmospheric mechanisms responsible for the combined mix of air masses depositing dust over these high-latitude environments are immensely needed. Westerly winds and subtropical jet stream atmospheric currents have been extensively indicated as the responsible for the Patagonia/SCWA and PAP signatures, respectively (Gili et al. (2017) and references therein). For the Namibia region, while Berg winds account for the dust load deposited in the South Atlantic (Shannon and Anderson, 1982), the extent of this transport to higher latitude areas, such as, the East Antarctic plateau is still unknown. Likewise, more mineralogical and geochemical studies focusing on the PAP and SAF PSAs will allow to gain a better understanding of the similarity between the regions and ultimately help to improve our knowledge about major atmospheric circulation patterns that occurred during the last glacial-interglacial cycles in the Southern Hemisphere.

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Figures

Figure 1: Left panel (A): Southern Africa Potential dust Potential Source Areas characterized in this study: Huab, Omaruru, Kuiseb (red dots) and the Namibia Sea Sand (blue diamonds). A satellite image of dust plumes originated off the coast in SAF detected by MODIS on 29 July 2015 are also showed and available at NASA Worldview. The hydrographic Huab, Omaruru and Kuiseb systems are indicated by the blue lines. Right panel (B): map of southwestern Africa showing the major features of the regional climate system. Main oceanic currents are indicated in bold green arrows and atmospheric circulations in red (south easterly trade and westerlies winds) and blue (Berg winds). BC: Benguela Current; ACC: Antarctic Circumpolar Current; AC: Agulhas Current.

Figure 2: Rare Earth Elements (REE) data of surface sediments representing PSAs of SAF: (a) Mean REE patterns, (b) Relationships between Eu^*/Eu and La/Yb_N . REE data here are based on normalizing to upper continental crust (UCC, Taylor and McLennan, 1985).

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Figure 3: Sr-Nd-Pb isotopic compositions of SAF sediments. (a) $^{208}\text{Pb}/^{207}\text{Pb}$ vs. $^{206}\text{Pb}/^{207}\text{Pb}$ (b) $^{87}\text{Sr}/^{86}\text{Sr}$ vs. $\epsilon\text{Nd}(0)$. Grain size effects on Sr-Nd-Pb isotopic compositions for the Huab and Namibia sand sea are also shown and indicated by symbols in gray (fine) and open symbol (bulk).

Figure 5: Comparison of SAF and SSA isotopic signatures with the glacial/interglacial fingerprint of dust deposited in East Antarctica and the South Atlantic sector of the Southern Ocean. (a) $^{208}\text{Pb}/^{207}\text{Pb}$ vs. $^{206}\text{Pb}/^{207}\text{Pb}$, the orange dotted lines represent compositional trend lines necessary to explain EA dust compositions. (b, c) $^{87}\text{Sr}/^{86}\text{Sr}$ and $\epsilon\text{Nd}(0)$, the black lines represent the calculated tri-mixing hyperbola between different PSAs of SAF (dotted line) and SSA (fill line). End-member values used to calculate Sr–Pb mixing curves are in Table S2. Local Antarctic Volcanoes data in plot (a) is from Vallelonga et al. (2010).

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