



Late Holocene dust provenance at Siple Dome, Antarctica

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ABSTRACT

Compositions of mineral dust in ice cores serve as tracers of paleo-atmospheric circulation patterns, providing linkages between sources and sinks. Here we document the geochemical makeup of dust reaching continental West Antarctica, on late Holocene samples from the Siple Dome A ice core (spanning ~1030–1800 C.E.). The Nd–Sr isotope signature is unusual for Antarctic ice core dust samples. Siple Dome data are characterized by low Nd isotope ratios (as low as $\epsilon_{\text{Nd}} = -16.3$) along with low Sr isotope ratios (highest $^{87}\text{Sr}/^{86}\text{Sr} = 0.7102$) compared with other Antarctic dust signatures. A well-defined inverse correlation between Sr–Nd isotope ratios indicates two primary mixing sources. The low ϵ_{Nd} -values indicate involvement of ancient (Archean-to-early Proterozoic) continental crust, as either the direct source or as a precursor of the source, and the low Sr-values require low Rb/Sr ratios that often reflect high-grade metamorphism. The known Antarctic terrane with these characteristics is parts of Enderby Land, nearly at the opposite end of Antarctica. The isotopic signature of the second end-member is compatible with West Antarctic volcanoes or Patagonia in South America. The Sr–Nd isotopes and trace element abundances are also chemically compatible with mixing between volcanic material from Gaussberg, a small lamproite volcano in Kaiser Wilhelm II Land in coastal East Antarctica whose source is ancient lithospheric mantle, with dust from Patagonia or material from West Antarctic volcanoes. We assess these potential mixing scenarios and conclude that Siple Dome's unusual geochemical signature can best be explained by a mixture of Patagonian dust and a Gaussberg-like source, with additional minor contributions from old eroded Archean-to-early Proterozoic bedrock sources such as those in Enderby Land. Moreover, Siple Dome dust compositions are distinct from dust deposited on Taylor and Clark Glaciers in the McMurdo Dry Valleys of the western Ross Sea, precluding the Dry Valleys as a late Holocene dust source to this region of the eastern Ross Sea.

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1. Introduction

Knowledge of dust provenance can be a key means to inform our understanding of past climate history and atmospheric transport pathways. Dust provenance information from Antarctic ice cores has been derived primarily from high-elevation sites in East Antarctica, including Vostok, Dome C, Dronning Maud Land, and Talos Dome (Fig. 1). The application of Sr and Nd isotopic tracers at

these sites has shown Patagonia to be the dominant dust source during glacial climates (Grousset et al., 1992; Basile et al., 1997; Delmonte et al., 2004, 2008, 2010), with other sources such as Australia (Revel-Rolland et al., 2006; Delmonte et al., 2007, 2020) or the Puna-Altiplano Plateau of South America (Delmonte et al., 2008; Gili et al., 2017; Paleari et al., 2019) becoming relatively more important during interglacials. Dust can originate from both local and distant terrestrial sources, and relatively high dust fluxes, coarse grain sizes, diatom fragments, and distinct dust isotopic signatures in snow and ice samples from sites near the margin of the East Antarctic Ice Sheet, including Talos Dome, Taylor Glacier, McMurdo Sound, Camp Maudheimvidda, Roosevelt Island, Mid Point, and Berkner Island, indicate that ice-free regions of

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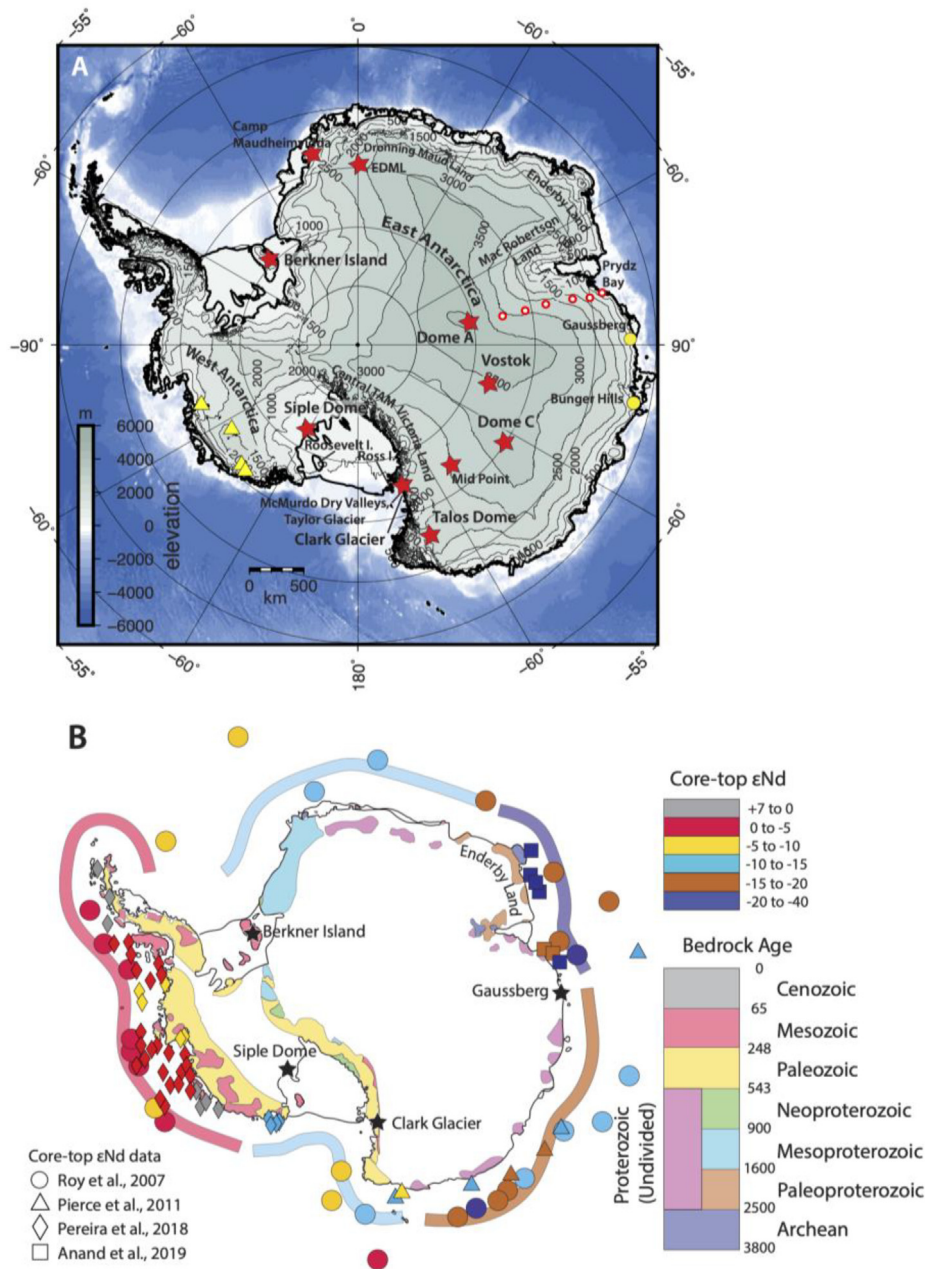


Fig. 1. Maps of Antarctica showing ice sheet elevation and sites discussed in text (A) and schematic bedrock geology and marine sediment core-top ϵ_{Nd} (B). Red-and-white symbols in (A) indicate a sampling transect from Zhongshan Station to Dome A (Du et al., 2018). EDML stands for the EPICA Dronning Maud Land ice core (EPICA: European Project for Ice Coring in Antarctica). Darker grays in (A) indicate higher elevations, except the darkest gray color near the margins, which represents ice-free landscape. Yellow triangles indicate active WAIS volcanoes: Takahe, Waesche, Berlin, Moulton. Bedrock geology in (B) based on Kirkham and Chorlton (1995) and Cook et al. (2013). Marine core-top ϵ_{Nd} data come from Roy et al. (2007), Pierce et al. (2011), Pereira et al. (2018), and Anand et al. (2019). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Antarctica also serve as important dust sources, at least during the Holocene (Delmonte et al., 2010, 2013, 2020; Bory et al., 2010; Winton et al., 2016a,b; Aarons et al., 2017).

In contrast to East Antarctica, dust provenance in West Antarctica has been less well studied. Considering its lower elevation, the dominant atmospheric circulation patterns around the Antarctic continent (Reijmer et al., 2002; Nicolas and Bromwich, 2011), and the geographic distribution of mid-latitude continental dust sources, we expect that sites in West Antarctica receive dust from different sources, or mixtures of sources, than those that supply East Antarctica. Identifying the provenance of dust reaching

West Antarctica has two-fold significance. First, it provides critical observational constraints on atmospheric transport pathways and their variability around the entire Antarctic continent, especially related to ice core sites and associated efforts to reconstruct past circulation and climate changes (e.g., Dixon et al., 2012; Koffman et al., 2014). Second, knowledge of dust sources and transport pathways can facilitate better estimates of the potential biogeochemical impact of iron and other dust-derived nutrients reaching the Southern Ocean (e.g., Martínez-García et al., 2011; Shoenfelt et al., 2018). In both cases, observational data are fundamental to assess model-based estimates of dust transport and deposition.

Moreover, modeling efforts to date have included primarily mid-latitude sources of dust, namely, Australia, New Zealand, southern South America, and southern Africa (Li et al., 2008; Albani et al., 2011; Neff and Bertler, 2015), and so do not assess potential contributions from Antarctic dust sources.

Here we present new dust provenance data from Siple Dome, located near the margin of the West Antarctic Ice Sheet (WAIS) and eastern shore of the Ross Sea. We also present new data from Clark Glacier in the McMurdo Dry Valleys, which allow us to compare dust transport to the eastern and western sides of the Ross Sea embayment. We characterize the ice core dust samples using Sr and Nd isotope ratios, which reflect the ages and compositions of their bedrock sources, as well as rare earth element (REE) patterns. Both approaches have been widely used for ice core dust source attribution since the pioneering work for the EAIS (e.g., Grousset et al., 1992; Basile et al., 1997; Delmonte et al., 2004; Grousset and Biscaye, 2005; and others listed in the supplement).

2. Site description and sample collection

Siple Dome (81.6667° S, 148.8167° W, 621 m. a.s.l.) is a coastal ice dome located on the eastern side of the Ross Sea embayment in West Antarctica (Fig. 1). Annual snow accumulation is ~12 cm water equivalent (Kreutz et al., 1999). The Siple Dome A deep ice core was drilled during the late 1990s, and samples for Sr–Nd isotopic analyses were collected from late Holocene ice by P. Biscaye. Dust was concentrated from melted ice samples using a Sharples™ continuous-flow supercentrifuge, which operated at ~30,000 RPM. Dust particles were deposited on mylar sheets that lined the centrifuge bowl, and were rinsed subsequently into sample vessels. This system was deployed successfully in Greenland, where observations suggest complete recovery of dust samples (Bory et al., 2002). Samples were stored in acid-cleaned Teflon beakers under clean conditions until chemical processing for this study, about two decades later. The 24 ice core samples analyzed for this study span two intervals within the past millennium: ~1030–1200 C.E. and ~1650–1800 C.E. There are no meaningful differences between the data from the two periods, and the samples are considered in aggregate here. Interpretation of the time series is beyond the scope of this paper, and will be the topic of a separate study.

Clark Glacier (77.4054° S, 162.3656° E, 816 m. a.s.l.) is located in the Olympus Range, between Wright Valley and Victoria Valley in the largely ice-free McMurdo Dry Valleys (Fig. 1). The annual accumulation rate is ~2.5 cm water equivalent (range: 1–7 cm/y; Williamson et al., 2007). An ice core was collected in 2005 and samples were kept frozen in lay-flat bags until analysis. The samples analyzed for this study span the late Holocene, and ages are not well-constrained. Cores were melted in large clean plastic bins at room temperature, and meltwater was filtered onto Whatman 42 paper filters (2.25 µm pore size) using vacuum filtration. Given the extremely local dust sources and large, visible dust grains in the melted samples, we assume this pore size captured the bulk of the lithogenic material in the samples. However, finer dust from the Dry Valleys might have higher Sr isotope ratios than those we measured in these samples (e.g., Biscaye and Dasch, 1971). Dust was removed from the filters using ultrapure water (Millipore™ Milli-Q) and ultra-sonication, and collected in acid-cleaned Teflon beakers. Remaining water was evaporated from the samples prior to acid digestion.

Ice core dust samples were digested using HF–HNO₃ following standard procedures, and elements for isotopic analysis were separated using column chromatography, with isotopic analysis via MC-ICP-MS. Full analytical details are in the Supplementary Information.

3. Siple dome dust provenance

We obtained 23 Sr and 27 Nd isotope ratio data on dust from a late Holocene section (~1030–1800 C.E.) of the Siple Dome deep ice core (SDMA; Table S1). The data form a tight array in Sr–Nd isotope space, with ⁸⁷Sr/⁸⁶Sr ranging from ~0.7087 to 0.7102 and εNd ranging from –16.3 to –7.3 (Fig. 2). εNd is the deviation in parts per 10⁴ of a sample's ¹⁴³Nd/¹⁴⁴Nd from estimates of the average chondritic value; this study uses 0.512638 (Jacobsen and Wasserburg, 1980) in order to be consistent with most literature data. The ⁸⁷Sr/⁸⁶Sr and εNd-values are strongly inversely correlated (Pearson's $r = -0.72$; $P < 0.001$), indicating the dust primarily reflects mixing of two distinct end-member compositions. Using isotope mixing plots, we are able to exclude seawater (e.g., salts or carbonates) as a potential Sr end-member (explained in Supplementary Text and Fig. S3). The Sr–Nd isotopic compositions of the Siple Dome dust are unusual among known Antarctic ice cores. The low εNd values coupled with low ⁸⁷Sr/⁸⁶Sr values occupy a distinct low-Nd/low-Sr isotope field on the Sr–Nd isotope diagram (Fig. 2).

Dust in Antarctic ice cores can be sourced from both local (e.g., ice-free areas of Antarctica) and remote (e.g., midlatitude) regions. In general, lower-elevation snow and ice core sites close to the Antarctic ice sheet margin receive significant contributions of dust from nearby exposed surface sediments, including dunes, regolith, and drift (Bertler et al., 2005; Ayling and McGowan, 2006; Dunbar et al., 2009; Delmonte et al., 2010; Aarons et al., 2016, 2017; Diaz et al., 2020). This local input is reflected by geochemical compositions, high dust fluxes, and coarse grain size distributions (e.g., Delmonte et al., 2010; Aarons et al., 2017). At higher elevations and farther inland on the ice sheet, dust comes primarily from mid-latitude sources such as deserts and glacial systems (e.g., Delmonte et al., 2004, 2013, 2020). These ice core sites are characterized by low dust fluxes, fine particle size distributions (e.g. volumetric mode diameter of ~2 µm), and geochemical compositions that differ from most potential Antarctic sources (e.g., Delmonte et al., 2004). The combination of grain size and isotopic compositions

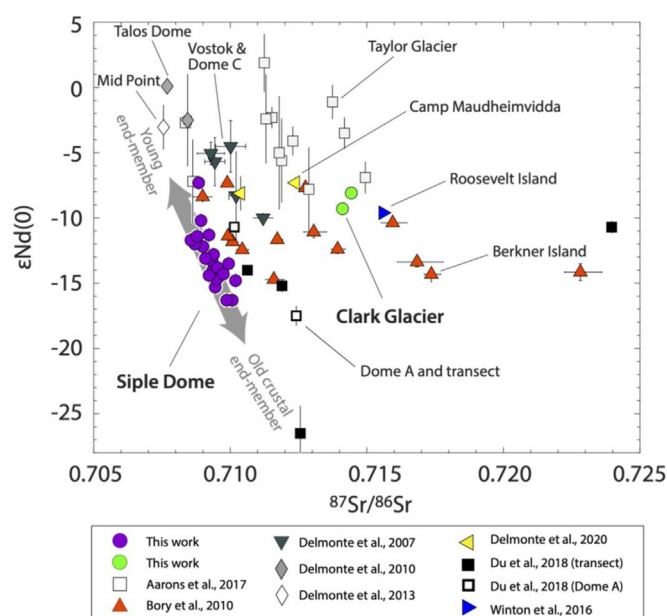


Fig. 2. Intergracial Sr–Nd data from Antarctic ice cores and snow pits, including Holocene and MIS 5.5 aged samples. Siple Dome data form a linear array, indicating two primary end-members. Clark Glacier data, representing local Dry Valleys sources, have significantly higher ⁸⁷Sr/⁸⁶Sr than Siple Dome, indicating the Dry Valleys are not a significant supplier of dust to Siple Dome. References are listed in the Supplement.

can be particularly diagnostic in determining likely local vs. remote dust sources. Unfortunately, grain size measurements have not been made on Siple Dome dust. However, during sample processing, we observed some visible dust grains, consistent with the presence of at least some local (Antarctic) dust.

Given the Sr–Nd compositions of dust, the strong Sr–Nd correlation, and compositions of potential source areas, we infer that Siple Dome dust comes mainly from two sources of very different crustal ages. The younger high- ϵ Nd/low-Sr isotope end-member is likely to be from one of two sources. The first is reworked, remobilized detritus from volcanic eruptions within West Antarctica (e.g. on Ross Island, or in the McMurdo Sound region more generally (Figs. 1 and 3) (Panter et al., 1997, 2000; Blakowski et al., 2016; Winton et al., 2016a)). These volcanoes are Mesozoic to Cenozoic in age, and activity has continued through the Quaternary (e.g. Wilch et al., 1999). The second potential young end-member is dust from Patagonia, which is the only known mid-latitude source region with sufficiently low $^{87}\text{Sr}/^{86}\text{Sr}$ ratios to serve as a source (Fig. 3) (Delmonte et al., 2004; Gaiero et al., 2007; Gili et al., 2017). That is, Australia, New Zealand, southern Africa, and other regions of South America are incompatible as young crustal-age sources; whereas the ϵ Nd-values are similar to those found in the new Siple Dome data, their $^{87}\text{Sr}/^{86}\text{Sr}$ values are too high (Delmonte et al., 2004; Gili et al., 2017; De Deckker, 2019; Koffman et al., 2021). Additional ‘young’ sources could include reworked volcanic material from sub-Antarctic islands or the Antarctic Peninsula; however, we favor one of these two sources (West Antarctic volcanism or Patagonia) based on areal extent and known dust emissions activity.

The other ‘older’ end-member must have a lower ϵ Nd-value than the lowest dust sample (–16.3), along with low $^{87}\text{Sr}/^{86}\text{Sr}$ compared to all other known Antarctic dust samples (Fig. 2) and potential dust sources (Fig. 3). In general, ϵ Nd-values closely reflect the ‘crustal residence age’ of a sample, that is, the average amount of time the Nd in the sample has been in the continental crust; this is because many upper crustal compositions have similar Sm/Nd ratios of 0.190, equivalent to $^{147}\text{Sm}/^{144}\text{Nd} = 0.115$ (Goldstein et al., 1984). For example, using the parameters of Goldstein et al. (1984), the minimum crustal residence age of the dust is 2.1 Ga. The low ϵ Nd-values thus indicate involvement of Archean-to-early Proterozoic continental crust in the formation of the dust source (Fig. 3B). As explained below, such ancient continental crust might be the direct dust source as generally assumed in provenance studies, while in the case of Siple Dome dust the direct source may be recent unusual volcanism that samples the lithospheric mantle.

In contrast to the Sm/Nd isotopic system, Rb/Sr ratios are highly susceptible to alteration during metamorphism and chemical weathering, and vary greatly in different lithologies. For the Siple Dome dust samples, the low $^{87}\text{Sr}/^{86}\text{Sr}$ ratios require low Rb/Sr ratios for much of the source’s history. Such a combination of low Sm/Nd and low Rb/Sr is sometimes fulfilled by granulite terranes (e.g. Rudnick and Presper, 1990). During deep crustal (high T-high P) granulite-facies metamorphism, Rb is often preferentially lost, effectively resulting in low Rb/Sr that supports only small increases of $^{87}\text{Sr}/^{86}\text{Sr}$ over time (Heier, 1973; Cuney and Barbey, 2014). Shallow crustal weathering has the opposite effect, as Sr is more soluble than Rb and therefore is preferentially lost, causing weathered sediments to have high Rb/Sr ratios (e.g., Biscaye and Dasch, 1971) that develop correspondingly high $^{87}\text{Sr}/^{86}\text{Sr}$ over time. Using the median Rb/Sr ratio of Archean granulite (~0.12, corresponding to $^{87}\text{Rb}/^{86}\text{Sr} \sim 0.348$; Rudnick and Presper, 1990), we find that the calculated $^{87}\text{Sr}/^{86}\text{Sr}$ of granulite rocks that are generated from an upper mantle-like source at 3.0 Ga, that also experienced early, high-grade metamorphism, would likely have $^{87}\text{Sr}/^{86}\text{Sr}$ lower than 0.720 today (Fig. 3B; more information is given in the Supplement). Using the median Sm/Nd value of Archean granulite

(Rudnick and Presper, 1990), the corresponding present-day ϵ Nd-value would be ~ -33 . Thus an Archean or early Proterozoic granulite terrane is a viable source for the older low- ϵ Nd Siple Dome mixing end-member.

The identity of the older mixing end-member is uncertain. The narrow, low $^{87}\text{Sr}/^{86}\text{Sr}$ range, combined with low ϵ Nd, and seemingly binary mixing trend of the Siple Dome data, limit the possibilities. Although there are sediments derived from Archean and early Proterozoic bedrock in Australia (primarily in Western Australia and Northern Territory), the material is weathered with high Rb/Sr (and $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.715–0.775) and therefore not a plausible source (Revel-Rolland et al., 2006; De Deckker, 2019). There are sediments in southern Australia with similar Nd isotope ratios as observed in this study, and a few samples have similar Sr isotope ratios as well, but the mid-latitude source area as a whole is characterized by much higher $^{87}\text{Sr}/^{86}\text{Sr}$ and the data plot far to the right of the Siple Dome data (green highlighted region in Fig. 3B; De Deckker, 2019; Koffman et al., 2021). If this region were a source, we would expect to see a wider range of Sr isotope values in the ice core data. Likewise, while much of East Antarctica is underlain by Archean and Proterozoic bedrock, as evidenced by extremely low marine core-top ϵ Nd values (Fig. 1B), the high Sr isotope compositions of many outcrops exclude their serving as the older source to Siple Dome (Black et al., 1986; Collerson and Sheraton, 1986; Young et al., 1997; Peucat et al., 1999, 2002; Mikhalsky et al., 2010). This includes the Bunge Hills ice-free area (Fig. 1A), which is characterized by ϵ Nd values of ~ -24 to -20 and high $^{87}\text{Sr}/^{86}\text{Sr}$ of ~ 0.72 – 0.76 in fine-fraction sediments (Delmonte et al., 2020). Furthermore, existing bedrock and till data from within the central and southern Transantarctic Mountains (e.g., Victoria Land) preclude this region from being the older end-member, as Sr isotope ratios and/or ϵ Nd values are too high (Fig. 3B) (Delmonte et al., 2010, 2013; Blakowski et al., 2016; Farmer and Licht, 2016). It is worth noting that these tills would contain some contribution of material from beneath the EAIS, including potentially unknown lithologies; therefore, existing till compositions indicate that the older end-member to Siple Dome is unlikely to be located in a region draining toward the Transantarctic Mountains.

Instead, we have found two possible sources that are geochemically compatible as the older (low ϵ Nd) end-member to Siple Dome. The first is material eroded from highly metamorphosed Precambrian bedrock underlying the EAIS and outcropping as moraine or drift, where it is subsequently wind-eroded. Coastal Enderby Land and Mac Robertson Land near Prydz Bay (Fig. 1) contain Archean to Proterozoic granulite-facies bedrock outcrops that are geochemically compatible as this end-member (Fig. 3B) (McCulloch and Black, 1984; DePaolo et al., 1982; Asami et al., 2002; Suzuki et al., 2006). Further, this region is characterized by some of the highest erosion and sediment flux rates in East Antarctica (Jamieson et al., 2010; Colledge et al., 2013). The second possible ‘older’ end-member is volcanic dust derived from Gaussberg or a similar but unknown source. Gaussberg is a 370 m tall lamproitic (leucite basalt) Quaternary volcano in coastal East Antarctica (Murphy et al., 2002; Sushchevskaya et al., 2014). Lamproites are ultra-potassic volcanic rocks from the mantle with high Mg and K/Al; low Ca, Al, and Na; and high incompatible trace element abundances (Foley et al., 1987). While this source reflects recent volcanism, its unusual chemistry has been interpreted as reflecting late Archean continent-derived sediment that was subducted beneath the Antarctic craton and isolated for 2–3 Ga (Murphy et al., 2002). Gaussberg’s last known eruption was 56 ± 5 ka (Tingey et al., 1983), and unlike Precambrian bedrock in East Antarctica, it is very limited in areal extent. Presumably the ice-free flanks of the volcano could serve as dust deflation areas; or alternatively, material with this composition could be eroded by the ice

sheet and deflated at a downstream site such as a blue ice area. In addition to Gaussberg, there are two other known lamproite occurrences in Antarctica: Priestley Peak, located within the Napier Complex of Enderby Land, and Mt. Bayliss, in the southern Prince Charles Mountains of MacRobertson Land (Fig. 1A) (Bergman, 1987; Mitchell et al., 1991). It might be expected that other lamproite suites exist in these regions, but are covered by ice (Mitchell et al., 1991). Although limited in extent, the unusually low $^{87}\text{Sr}/^{86}\text{Sr}$ of Gaussberg lamproite (0.70692–0.70989), coupled with very low ϵNd (−15.0 to −12.2) and Sm/Nd ($^{147}\text{Sm}/^{144}\text{Nd} = 0.076\text{--}0.101$) (Murphy et al., 2002; Sushchevskaya et al., 2014), is too good a geochemical match to Siple Dome dust to discount (Figs. 3 and 4). Therefore, we propose that the low- ϵNd end-member contributing dust to Siple Dome may be material sourced from an Antarctic volcano like Gaussberg, in addition to sediment eroded from metamorphosed East Antarctic Archean-to-Proterozoic bedrock.

Transport of dust from coastal East Antarctica, including Enderby Land, MacRobertson Land, and Gaussberg to Siple Dome likely would occur via katabatic flow carrying dust from coastal sites into the circumpolar westerlies. Subsequent deposition would occur via cyclonic systems in the Ross Sea embayment. This hypothesized pathway is supported by air-mass modeling experiments in the modern atmosphere, which show that ~10–20% of air masses reaching the eastern Ross Sea arrive via long-range oceanic transport from the region offshore of Prydz Bay (Tuohy et al., 2015). Transport of dust from Patagonia also would occur via the westerlies. In this case, previous work has shown that ~5 μm diameter particles can travel from southern South America to central West Antarctica via the westerlies in a matter of days to a week (Koffman et al., 2017). While we are unable to identify the exact geographic sources supplying dust to Siple Dome, and thus cannot evaluate transport pathways more specifically, existing work suggests that dust transport from both Patagonia and coastal East Antarctica near Prydz Bay to the eastern Ross Sea is plausible. Additional work is needed to identify the specific sources and transport pathways supplying dust to Siple Dome.

In sum, Siple Dome Sr–Nd isotope compositions can be explained by a mixture of ‘young’ (WAIS/Ross Island/McMurdo Sound volcanics and/or Patagonia) and ‘old’ (Archean-Proterozoic bedrock and/or lamproitic compositions like Gaussberg) ϵNd sources (in the case of Gaussberg, the first derivation from the mantle would be ‘old’). A plot of $^{147}\text{Sm}/^{144}\text{Nd}$ vs ϵNd (Fig. 4) is useful to investigate these potential mixtures. For a mixture of Enderby Land and WAIS/Ross Island/McMurdo Sound volcanic compositions to explain Siple Dome dust, the data in Fig. 4 require that the Enderby Land samples with the lowest Sm/Nd and the lowest ϵNd contributed dust to Siple Dome (i.e., ϵNd of −53 to −45). If the suite of published analyses represents the proportions of outcropping rocks then we consider this as unlikely. If so, the Siple Dome samples would exhibit higher $^{147}\text{Sm}/^{144}\text{Nd}$, and also higher $^{87}\text{Sr}/^{86}\text{Sr}$ (Fig. 4). On the other hand, if the investigators intentionally analyzed Enderby Land samples to represent the range of compositions, and the lowest Sm/Nd – ϵNd are the ones that best represent the compositions of outcropping samples, then it is a possible dust source. It is noteworthy also that a Patagonia–Enderby Land mixture similarly does not work to explain the Siple Dome compositions, and would be subject to the same requirements. A mixing envelope for a Patagonia–Gaussberg mixture, in contrast, encompasses nearly all the Siple Dome data points and does not require the exclusion of any source samples (Fig. 4). Two Siple Dome points have ϵNd -values outside of the Patagonia–Gaussberg mixing array (indicated with white dots), lower than $\epsilon\text{Nd} = -15$, requiring a contribution from an Enderby Land-like source. Therefore, this scenario means that the Siple Dome dust samples mainly reflect a mixture of Patagonian dust and a Gaussberg-like

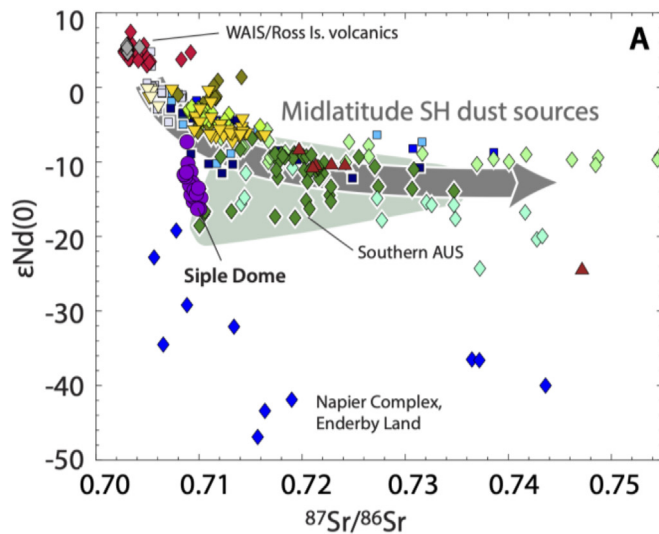
source, with some additional dust sourced from East Antarctic Archean bedrock such as that exposed in Enderby Land. A mixture of WAIS/Ross Island/McMurdo Sound volcanics with a Gaussberg-like source would have similar constraints as a Patagonia–Gaussberg mixture. While we cannot rule out this mixture, we favor the former scenario based on the REE data (explained below).

We also assessed potential mixtures of these four end-members using rare earth element (REE) patterns, plus Sr (Fig. 5), as any inferred mixture that explains Siple Dome dust compositions should account for both isotopes and trace elements. The Siple Dome data are characterized by high Sr and LREE and low HREE, with essentially no Eu anomaly. Of these potential end-members, Patagonia sediment has lower Sr than Siple Dome and a REE profile with a positive PAAS-normalized trend from La to Lu (HREE are higher than LREE) and a small positive Eu anomaly (Gili et al., 2017). East Antarctic high-grade metamorphic rocks have lower Sr than Patagonia, and a fairly flat PAAS-normalized REE profile with LREE and HREE both higher than Patagonia, and a negative Eu anomaly (Liu et al., 2014). Of the likely Siple Dome end-members, only the Gaussberg lamproite has Sr and LREE higher than the Siple Dome dust. The Gaussberg data show a steep downward trend from La to Lu, punctuated by a strong positive Eu anomaly (Murphy et al., 2002; Sushchevskaya et al., 2014). Siple Dome dust values therefore are intermediate between Gaussberg (high LREE, low HREE, positive Eu anomaly), Patagonia (low LREE, high HREE, positive Eu anomaly), and East Antarctic metamorphic (intermediate LREE, high HREE, negative Eu anomaly) compositions. Importantly, East Antarctic metamorphic rocks are the only one of these potential end-members to display a negative Eu anomaly. The contribution of dust from this source therefore helps to explain the lack of Eu anomaly seen in the Siple Dome data. In contrast, WAIS volcanic REE compositions are a relatively poor fit as an end-member, with a large positive Eu anomaly and relatively low PAAS-normalized LREE and HREE (Fig. S4). Considered together, the trace elements thus favor the Patagonia–lamproite–Precambrian source mixing scenario.

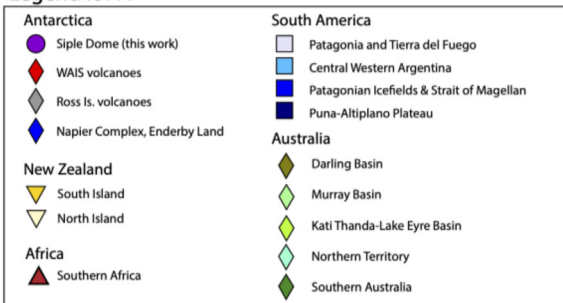
We used median end-member REE values to calculate theoretical mixtures (Table S5). Siple Dome dust is broadly consistent with a 15% contribution from East Antarctic metamorphic sources and ~25–60% contributions from Patagonia and Gaussberg-like compositions. These mixtures produce REE patterns that represent the Siple Dome values fairly well, though the ice core Sr concentrations are higher than would be expected, looking more like Gaussberg (Fig. 5). The theoretical three-end-member mixtures produce reasonable isotope ratios as well. For example, a mixture that is 15% from East Antarctica, 50% from Patagonia, and 35% from Gaussberg yields $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.7093 and ϵNd of −13.4, very close to the median Siple Dome values (0.70936 and −13.4, respectively).

4. Clark glacier dust provenance

In contrast to the low Sr–Nd isotopic signatures of Siple Dome dust, our two Sr–Nd data points from the Clark Glacier ice core taken from near the eastern Ross Sea (Fig. 1) have both higher $^{87}\text{Sr}/^{86}\text{Sr}$ (0.7142–0.7145) and ϵNd (−9.3 and −8.2) values (Fig. 2). These values are consistent with the local bedrock in the McMurdo Dry Valleys and in fact closely reflect the ice core’s location near the contact of the Permian to Cretaceous Ferrar Large Igneous Province (FLIP) with the Lower Paleozoic terranes of coastal Victoria Land (Figs. 1B and 3B) (Cook et al., 2013). Additional Sr–Nd isotope data from Victoria Land fine-grained sediments also bracket our Clark Glacier data (Delmonte et al., 2004; Blakowski et al., 2016) (Fig. 3B). Considering that the FLIP and Lower Paleozoic terranes are the predominant bedrock units in southern Victoria Land, and that there is no evidence of additional dust sources supplying Clark



Legend for A



*Please see Supplementary Information for a full list of data citations

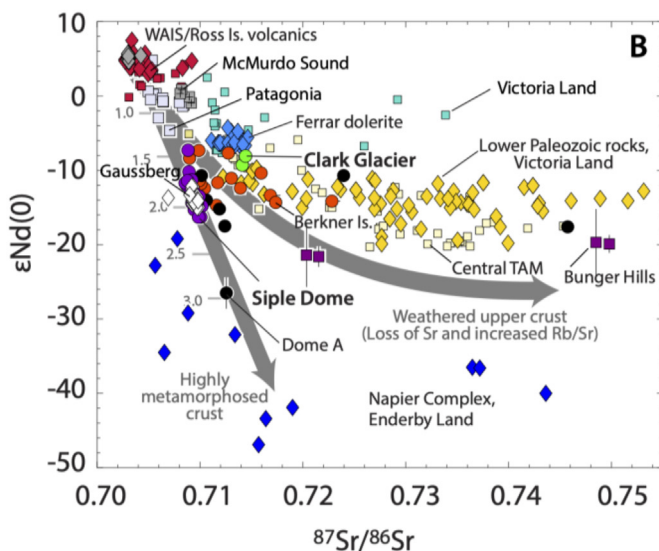


Fig. 3. Siple Dome Sr–Nd compositions compared to potential source area sediments (<5 μm diameter) and selected bedrock samples. (A) Mid-latitude dust sources cannot on their own account for the low Sr–Nd composition of Siple Dome dust, which requires a more ancient end-member, though Patagonia may be the younger end-member. The green highlighted area shows the compositional range of sediments from southern Australia (not including the Kati Thanda-Lake Eyre or Murray-Darling Basins). The field overlaps the Siple Dome samples, but tends toward $^{87}\text{Sr}/^{86}\text{Sr} > 0.715$, and thus cannot be a primary source. (B) Siple Dome dust can be explained by a mixture of young (WAIS and/or Ross Island/McMurdo Sound volcanic material or Patagonia dust) mixed with old (derived from Enderby Land and/or Gaussberg lamproite) sources. Other sources, such as sediments from the central Transantarctic Mountains (TAM) and Bunker Hills, have $^{87}\text{Sr}/^{86}\text{Sr}$ too high to be a viable source to Siple Dome. Clark Glacier dust composition reflects local input of dust derived from Ferrar dolerite and lower Paleozoic terranes, both in Victoria Land. The

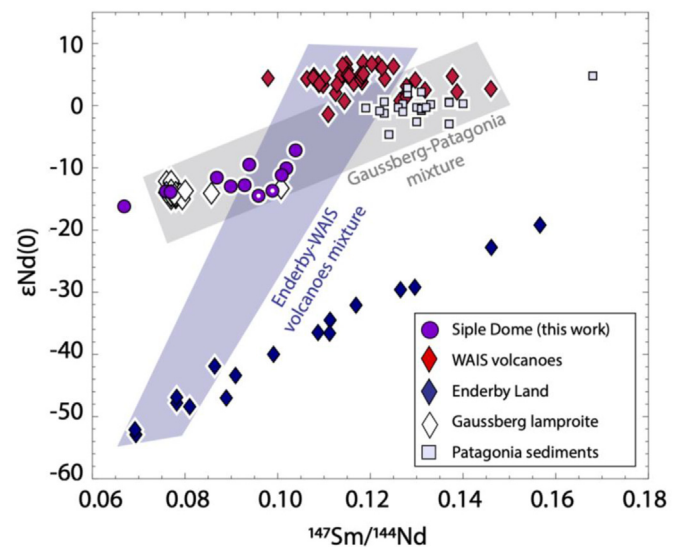


Fig. 4. ϵNd vs $^{147}\text{Sm}/^{144}\text{Nd}$ showing two potential end-member mixtures that could explain Siple Dome compositions. For a mix of Enderby Land and WAIS volcanic materials to work, it would require sediment to be derived from Enderby Land representing the lowest Sm/Nd and ϵNd values of the suite. A mix of Gaussberg lamproite with Patagonia dust, on the other hand envelopes nearly all the Siple Dome dust compositions. Two Siple samples with $\epsilon\text{Nd} < -15$ fall slightly below the range of Patagonia-Gaussberg mixtures (white dots) and their composition indicates some contribution of material derived from East Antarctic Archean or Proterozoic bedrock. Data sources are listed in the Supplement.

Glacier, we consider the Clark Glacier data to represent an integrated signal of dust emitted from the surrounding Dry Valleys. This interpretation is supported by the coarse grain sizes we observed.

5. Implications for regional dust transport

The dust that settled in the western and eastern Ross Sea (Clark Glacier and Siple Dome, respectively) show significantly different compositions. This demonstrates that the Dry Valleys, presumed to be the only dust supplier to Clark Glacier, cannot have been an important source of dust to Siple Dome and hence West Antarctica during the past millennium. This finding is consistent with the limited Dry Valleys dust signal found on McMurdo Sound sea ice, as well as data from Roosevelt Island in the Ross Sea (Chewings et al., 2014; Winton et al., 2016a,b), but contradicts estimates based on forward-trajectory modeling (Bhattachan et al., 2015). Similarly, the new data from Siple Dome show that neither Australia nor New Zealand is a major dust supplier to the western WAIS during the late Holocene (~1030–1800), in contrast to modeling predictions (Li et al., 2008; Neff and Bertler, 2015). This finding highlights the value of geochemical data for ground-truthing modeling studies. Finally, differences in Sr–Nd isotope compositions between late Holocene Siple Dome samples and modern Roosevelt Island samples (Fig. 2) (Winton et al., 2016b) point to the need for additional data through space and time to better constrain the regional dust cycle.

gray curves and text in (B) indicate time evolution of Archean granulite and weathered upper crust (see text). All source area data in (A) are fine-grained sediments (<5 μm diameter) except for WAIS and Ross Island volcanoes and Enderby Land rocks. Source area data in (B) are distinguished as diamonds (bedrock) and squares (fine-grained sediments), and all regions are labeled directly in the figure. Source area references are listed in the Supplement. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

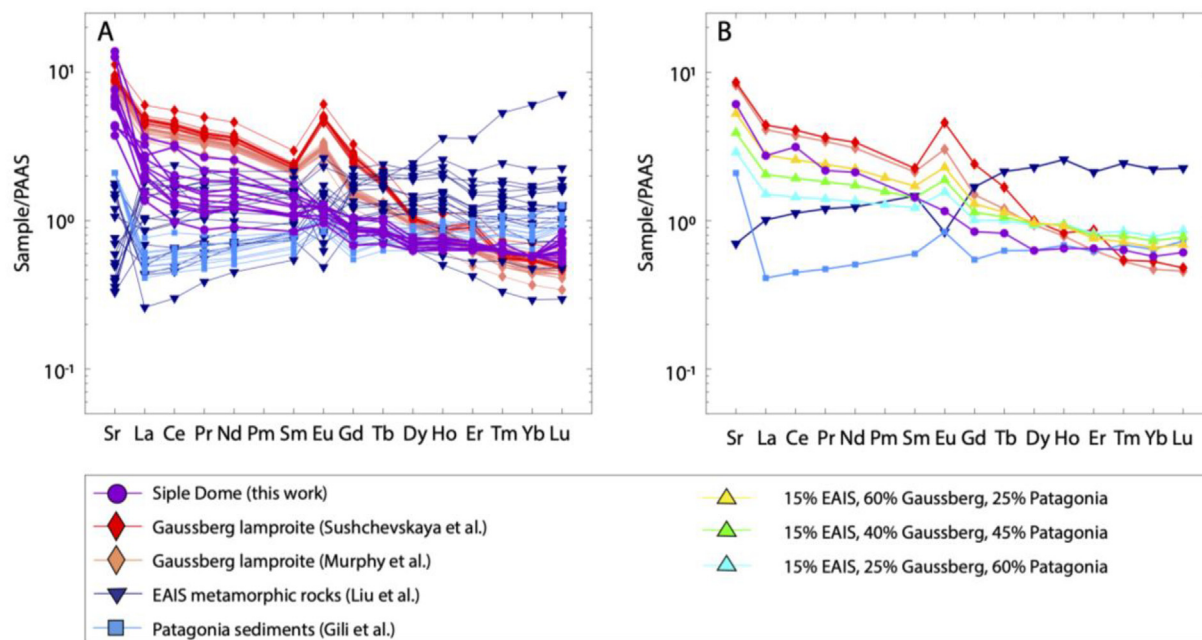


Fig. 5. Strontium and rare earth element (REE) data from Siple Dome compared to data from (A) inferred end-members including Patagonia, East Antarctic metamorphic rocks, and Gaussberg lamproite. (B) Shows median end-member values and theoretical three-end-member mixtures. Values are normalized to Post-Archean Australian Shale (PAAS) (Nance and Taylor, 1976; Taylor and McLennan, 1985; Pourmand et al., 2012). Siple Dome Sm concentrations have been corrected (see Supplement). Data sources are listed in the Supplement.

Interestingly, the data indicate that Siple Dome shares dust sources both with Berkner Island, in the Weddell Sea embayment, and with a transect of sites extending from coastal East Antarctica near Prydz Bay to Dome A, the highest-elevation site in East Antarctica (Figs. 1A and 2). At both locations, arrays of snow pit Sr–Nd data follow a parallel trend to the Siple Dome dataset, indicating a largely two-component mixture of young and old sources. At Berkner Island, Sr–Nd isotope analyses suggest that Patagonia may be one end-member, but the dust cannot be sourced exclusively from mid-latitude dust source regions (Bory et al., 2010). Low ϵNd combined with a range of $^{87}\text{Sr}/^{86}\text{Sr}$ values indicate that at least some dust (those samples with the lowest $^{87}\text{Sr}/^{86}\text{Sr}$) likely derives from material with an Enderby Land-like signature, such as we observe at Siple Dome. This interpretation is supported by air-mass modeling trajectories, which show transport from coastal Enderby Land and Dronning Maud Land (Bory et al., 2010). The majority of the Dome A transect data similarly fall along a mixing line between Patagonia (and/or other younger volcanic rocks) and Enderby Land bedrock (Fig. 2 and Du et al., 2018). Intriguingly, one sample from an inland site (800 km from the coast) has $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.7126 and ϵNd of -26.5 (Fig. 2), values that could point to the composition of the older end-member supplying dust to Siple Dome. These values are consistent with many of the existing Enderby Land data points (McCulloch and Black, 1984; DePaolo et al., 1982; Asami et al., 2002; Suzuki et al., 2006), and hint at a source of fine-grained material with this Sr–Nd isotopic composition. In this context, it would be valuable for future work to locate and map glacial deposits in ice-free areas around the Prydz Bay region that may be supplying dust to the high-latitude atmosphere, as the sediment from this region has a unique Sr–Nd isotopic composition and at the same time may serve as an important dust source.

A key finding of this study is that both Siple Dome and Clark Glacier have dust compositions that are distinct from the majority of East Antarctic high elevation plateau ice core sites, where most prior provenance data have been measured. Interglacial dust

samples from Vostok and Dome C show higher ϵNd (~ -10 to -4), but a similar range of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (Fig. 2). Located at elevations >3500 m, the Vostok and Dome C ice cores sample far-traveled dust transported at altitudes of ≥ 4000 m, and are less influenced by local sources (Delmonte et al., 2004, 2013; Krinner et al., 2010). Surprisingly, the Dome A (~ 4000 m) snow pit data have lower ϵNd (mostly -18 to -10 , but as low as -27), more similar to Siple Dome, likely indicating the input of Antarctic dust (Du et al., 2018). Lower-elevation sites such as Talos Dome, at 2300 m in northern Victoria Land, and Taylor Glacier, with a snow deposition elevation of ~ 1500 m in southern Victoria Land (Fig. 1), both likely receive dust from both local and distal sources (Delmonte et al., 2010; Aarons et al., 2017). The influence of ice-free areas near the Antarctic ice sheet margin is thus strongest at low-elevation and coastal ice core sites (Bertler et al., 2005; Delmonte et al., 2013, 2020; Krinner et al., 2010). Importantly, Siple Dome contrasts with Clark Glacier in that it does not appear to receive dust from surrounding areas (e.g., coastal WAIS volcanoes; Fig. 1A). If more of West Antarctica were ice-free, we would expect this local volcanic-sourced material to have a greater influence on Siple Dome dust compositions, comparable to the influence of Dry Valleys-sourced dust on Clark Glacier dust compositions.

6. Conclusions

In summary, our results provide the first characterization of late Holocene dust provenance in the Siple Dome ice core, located at the boundary of the Ross Sea embayment and continental West Antarctica. The data exclude Australia and New Zealand as dust sources. Instead, we find that Patagonia is the only potential mid-latitude dust source supplying Siple Dome, plus strong evidence that Antarctic ice-free areas supply about half of the dust reaching Siple Dome and West Antarctica; however, this does not include the Dry Valleys and the Transantarctic Mountains as a major source. Instead, the composition of Siple Dome dust is consistent with derivation from a mixture of dust from West Antarctic/Ross Island/

McMurdo Sound volcanics and East Antarctic Archean-to-early Proterozoic granulite-grade metamorphic rocks (an Enderby Land-like source), or dust from Patagonia and a Gausberg (lamp-rite) volcano-like source, plus some material also derived from East Antarctic metamorphic rocks. For reasons given in the text, we lean toward the latter scenario. Although the areal extent of Gausberg is very limited, and its last known eruption was 56 ± 5 ka, its Sr–Nd isotope and REE compositions are compatible with the unusual Siple Dome data. In particular, the unusually low $^{87}\text{Sr}/^{86}\text{Sr}$ (0.7069–0.7099) and very low ϵNd (–16.3 to –12.2) and $^{147}\text{Sm}/^{144}\text{Nd}$ (0.076–0.101) of Gausberg make its composition a good match as an end-member for Siple Dome. Similar material also could be sourced from other known lamproite volcanoes in East Antarctica, or from lamproite suites suspected to be buried under the ice sheet (Mitchell et al., 1991). Further work, therefore, is needed to characterize potential dust sources in coastal East Antarctica.

Given isotopic similarities of dust from Siple Dome and Berkner Island – located in opposing sectors of the Antarctic continent – as well as data from Dome A, representing the highest elevations in East Antarctica, coastal East Antarctic sources of Archean to early Proterozoic age appear to be more influential than previously recognized. We infer that fine-grained sediment may be produced from this bedrock source by glacial erosion, transported by the ice sheet, and subsequently exposed to the winds, delivering dust to >4000 m elevations in East Antarctica, as well as low elevations in both the Atlantic and Pacific sectors of the continent. This dust may serve as a distinct geochemical tracer of atmospheric circulation patterns around Antarctica.

Data availability

The data presented in this paper are available in the supplementary tables. In addition, they have been uploaded to the EarthChem Library (<https://www.earthchem.org/>), and are available as: <https://doi.org/10.26022/IEDA/112176>.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.quascirev.2021.107271>.

Author contributions

B.G.K., P.B., A.B., S.L.G., and M.R.K. designed the project; P.B., A.B., and K.J.K. collected the samples; B.G.K. and A.B. performed the labwork and analyses with help from L.B. B.G.K. and S.L.G. developed the interpretations with input from A.B., M.R.K., and G.W. B.G.K. wrote the drafts working closely with S.L.G. The final paper includes contributions from all authors.

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