A Sr-Nd-Hf isotope characterization of dust source areas in Victoria Land and the McMurdo Sound sector of Antarctica

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Abstract

Determining the geographical provenance of dust provides crucial insight into the global dust cycle. For the East Antarctic Ice Sheet (EAIS), the importance of Southern hemisphere potential dust sources has been thoroughly investigated using radiogenic isotopes, whereas proximal dust source areas located on the periphery of the ice sheet remain poorly documented from a geochemical standpoint. In this work, we expand the existing isotopic (Sr-Nd) catalogue of dust and sand-sized sediments from Victoria Land and the McMurdo Sound sector, and incorporate Hf isotopic data to place additional constraints on dust source identification. The isotopic field for materials considered in this study is characterized by $^{87}\text{Sr}/^{86}\text{Sr}$ ratios ranging from 0.703 to 0.783, ϵNd between -12.01 and 6.36, and ϵHf from -16.77 to 6.89. As reported in previous works, the data reveal close relationships between Antarctic sediments and distinct parent lithologies; in addition, our findings emphasize the background presence of very fine dusts originating from dominant global sources and regional volcanic activity as barriers to direct

source-to-sink comparison of isotopic signatures. Thus, geochemical characterizations of dust sources to the Antarctic ice sheet involving multiple size fractions, including coarser-grained particles more susceptible to short-range transport, can help us to rule out global sources of dust when examining local sediment cores and ice cores.

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1. Introduction

Eolian dust preserved in polar ice cores is an important indicator of past climatic conditions in the continents and the evolution of paleo-atmospheric circulation patterns (Basile et al., 1997; Biscaye et al., 1997; Bory et al., 2010; Bory et al., 2002; Bory et al., 2003; Delmonte et al., 2008; Delmonte et al., 2010; Delmonte et al., 2013; Delmonte et al., 2004a; Delmonte et al., 2004b; Delmonte et al., 2007; Gaiero, 2007; Grousset and Biscaye, 2005; Grousset et al., 1992; Marino et al., 2008); understanding the geographic provenance of ice core dust in particular has become important for informing global circulation models and deciphering past environmental conditions of the surrounding landmasses (e.g. Bory et al., 2003; Gabrielli et al., 2010; Mahowald et al., 1999; Mahowald et al., 2006; Svensson et al., 2000). Grounded on the rationale that sediments and soils derived from rocks of different lithologies and geologic ages tend to maintain the radiogenic isotope compositions (or “fingerprints”) of the rocks from which they were eroded, dust provenance can be determined by comparing the geochemical compositions of dust particles preserved in undisturbed sequences of snow and ice to the fingerprints of fine-grained sediments collected from target potential sources areas (PSA) (e.g. Grousset and Biscaye, 2005). In this way, it has been determined that southern South America (SSA) was likely the dominant supplier of dust to the entire East Antarctic Ice Sheet (EAIS) during the Last Glacial Maximum (LGM) (e.g. Delmonte et al., 2008; Delmonte et al., 2010; Delmonte et al., 2013; Delmonte et al., 2004a; Delmonte et al., 2004b; Delmonte et al., 2007; Grousset and Biscaye, 2005).

While the isotopic fingerprints of dust archived in central Antarctic ice cores (e.g. Vostok and EPICA-Dome C) have been matched with SSA source areas for glacial times (Basile et al., 1997; Delmonte et al., 2004a; Gaiero, 2007; Revel-Rolland et al., 2006; Sugden et al., 2009), little information exists on the origin of eolian dusts and sediments preserved in these ice cores for interglacials such as the Holocene (Delmonte et al., 2010; Delmonte et al., 2013). Numerous studies have suggested that ice-free areas located at the present-day margin of the EAIS could be

important generators of mineral material (dust, sediments), and yet limited data exists for these PSAs (e.g. Lanci et al., 2008; Vallelonga et al., 2010); in contrast to the cores drilled further inland, coastal ice cores TALDICE-Talos Dome and Taylor Dome may be especially influenced by local dust sources due to their proximity (see Figure 1).

In this work, we sought to deepen the current understanding of Holocene dust input to the periphery of the EAIS by expanding the existing geochemical catalogue of local Antarctic PSAs. Windborne dust derived from remote continental sources and deposited in Antarctic ice cores typically consists of particles smaller than $<5\ \mu\text{m}$ (diameter) (Gaiero, 2007; Lupker et al., 2010); however, contribution from local sources may include all size bins (*i.e.* larger particle diameters), which is why we chose to separately analyze a $<5\ \mu\text{m}$ (fine fraction) sample and a bulk sediment sample from each PSA. Considering both the grain size and isotopic composition of a PSA sample may help determine whether the sampling site represents *primary* or *secondary* source of dust (Delmonte et al., 2004a). In this same way, combining and comparing data from multiple isotope tracer systems greatly improves the chances of distinguishing PSA samples that have been weathered directly from a local deposit (e.g. moraine) from samples composed of a mixture of particles that have already been subject to eolian or fluvial transport. Here, we characterize an array of primary and secondary dust sources concentrated at the margin of the Antarctic ice sheet using the Sm-Nd, Rb-Sr, and Hf-Lu isotope systems in order to gain a more thorough understanding of regional dust provenance and transport pathways, and expand the pre-existing catalogue of southern hemisphere PSAs.

Investigations of Sr compositions in sediments of differing particle sizes began with Dasch's pioneering work (1969) on the behavior of Sr in weathering profiles, deep-sea sediments, and sedimentary rock, wherein it was observed that $^{87}\text{Sr}/^{86}\text{Sr}$ ratios tend to increase with decreasing grain size. In recent years, a number of studies have expanded upon grain size controls on $^{87}\text{Sr}/^{86}\text{Sr}$ in various sediments, including deep-sea NE Atlantic sediments (Meyer et al., 2011) and Asian dust (Kanayama et al., 2005). Similar to $^{87}\text{Sr}/^{86}\text{Sr}$, $^{143}\text{Nd}/^{144}\text{Nd}$ ratios of weathered dust are often studied and compared to compositions of corresponding source rock material (e.g. Basile et al., 1997; Bory et al., 2010; Delmonte et al., 2004a; Gaiero, 2007; Grousset et al., 1992). For instance, Feng et al. (2011) analyzed the Sr and Nd isotopic compositions of Chinese loess and dust-fall samples in seven different grain-size fractions, and found that ϵNd values for *homogenous* samples varied little between the size fractions. Similar

results have been reported in works by Grousset et al. (1992), Kanayama et al. (2005), Chen et al. (2007), and Meyer et al. (2011), to name a few.

Many works to date have also discussed the decoupling of Lu-Hf and Sm-Nd isotope systems in relation to continental weathering and sediment transport (e.g. Aarons et al., 2013; Bayon et al., 2009; Bayon et al., 2006; Carpentier et al., 2009; Garçon et al., 2013; Patchett and Tatsumoto, 1981; Piotrowski et al., 2000; van de Flierdt et al., 2007; Zhao et al., 2014). That is, while the two systems are closely coupled during magmatic processes and remain isotopically linked in whole rock and coarse sediment samples, forming a strong positive correlation known as the “terrestrial array” (Vervoort et al., 1999), the long-term alteration, erosion, and transport of crustal materials can lead to the gravitational separation of dense accessory mineral phases during transport, and therefore an eventual offset between the isotopic signatures of coarse-grained detrital sediments (*i.e.* sands) and finer-grained or clay-sized particles (*i.e.* dusts). If the mineral zircon (ZrSiO_4) is considered the main reservoir for Hf and Zr in terrestrial rocks, a significant variation in the quantity of zircons present in a sediment sample will modify the $\epsilon\text{Hf}/\epsilon\text{Nd}$ ratio, a phenomenon known as the “zircon effect” (e.g. Carpentier et al., 2009). Zircon grains are very high-density (not easily-transportable by wind), and contain large amounts of non-radiogenic Hf due to low initial Lu/Hf ratios and the slow radiogenic decay of ^{176}Lu to ^{176}Hf (Patchett and Tatsumoto, 1981). Thus, it can be generalized that the continental (non-marine) budget of Hf is effectively split between (1) a low ϵHf reservoir in zircon-bearing sediments, and (2) a more radiogenic, zircon-free reservoir (Bayon et al., 2009; Carpentier et al., 2009).

Although it may not necessarily be possible to pinpoint the exact nature of a PSA sample due to mixing of proximal and remote sources, the unique physical and isotopic properties of materials featured in this study have allowed us to narrow down and characterize a number of potential dust sources to snow and ice layers located at the periphery of the EAIS, including: the McMurdo Sound (MS) volcanics and glacial drifts; the Northern Victoria Land (NVL) regoliths, and; the regoliths, drifts, and sands of Southern Victoria Land’s (SVL) dry valleys, including the Taylor Valley moraines (Figure 3).

2. Regional setting

Virtually the entire Antarctic continent is covered by ice year round, with the exception of the ice-free valleys and nunataks of Victoria Land and the nearby McMurdo Sound/Ross Sea sector,

located along the north-central extent of the Transantarctic Mountains (TAM) (see Figure 1 inset). Here, freezing temperatures, hyper-arid conditions, and distinctive daylight cycles result in a diverse array of erosional and depositional landforms, most of which are characterized by underdeveloped soils or loose, unconsolidated material (Delmonte et al., 2013). Equally diverse is the local bedrock, ranging from Late Proterozoic-to-early Paleozoic granites and gneisses, Devonian-to-Triassic Beacon Supergroup sedimentary rocks, and sills of the Jurassic Ferrar dolerite (e.g. Stump and Fitzgerald, 1992). Below, we describe the three general regions from which the PSA samples discussed in this work were obtained, with additional geographic information summarized in Table 1 of the supplementary material.

2.1 Northern Victoria Land

Northern Victoria Land (NVL) is bounded by the Borchgrevink Coast (the western margin of the Ross Sea), the Rennick Glacier basin to the west, and the Pennel Coast (Pacific Ocean) to the north (Baroni et al., 2005b). Previous surface exposure dating studies conducted in Victoria Land have revealed that relict surfaces in NVL document a continuous and long lasting (millions of years) exposure to wind erosion (Armienti and Baroni, 1999; Di Nicola et al., 2012; Di Nicola et al., 2009; Oberholzer et al., 2008; Oberholzer et al., 2003; Schäfer et al., 1999; Strasky et al., 2009; Summerfield et al., 1999). Delmonte et al. (2010) suggested that mineral particles derived from the Mesa Range and other high-altitude ice-free areas of NVL may be favored for transport inland during times when maritime air masses sweep across the Antarctic plateau.

2.2 Southern Victoria Land

The McMurdo Dry Valleys (MDV) of Southern Victoria Land (SVL) are a series of east-west oriented, ice-free valleys carved by retreating glaciers, and make up the largest ice-free area in Antarctica (Fountain et al., 1999; Marchant and Head, 2007). Air flows seaward from the Polar Plateau and is forced over the TAM, cooling, condensing, and falling as snow at high elevations, a phenomenon that casts the intense rain-shadow to which the MDV owes its extreme aridity and winds (Fountain et al., 1999). The valley floors are covered with rocks and sediments from the Beacon Supergroup, as well as from older granites and gneisses of the Ross Orogeny. Although sandy deposits are found throughout the MDV, the only areas where major eolian sand accumulates are within the Victoria Valley, near the hypersaline Lake Vida and the Victoria

Lower Glacier (Selby et al., 1974). Here, windblown materials have accumulated into a series of transverse and whaleback dunes (Selby et al., 1974).

2.3 McMurdo Sound

Numerous PSA samples were collected from the low-elevation, coastal area inland of the Ross Sea, bounded by the MDV and the Polar Plateau, the McMurdo Ice Shelf extending to McMurdo Sound, and Ross Island; we hereafter refer to this region as the McMurdo Sound sector (MS). This area—specifically Ross Island—is home to Mt. Erebus, an active composite volcano from which about 4520 km³ of volcanic material has erupted over the past ~4 Ma (Esser et al., 2004). Because the MS geology is dominated by Late Cenozoic, alkaline volcanics belonging to the McMurdo Volcanic Group (Kyle et al., 1990; Sims et al., 2008), a relatively straightforward geochemical distinction was anticipated between MS PSAs closer to Mt. Erebus and the Victoria Land PSAs.

3. Materials and methods

3.1 Sample collection

Closely following the sampling strategies described in Delmonte et al. (2013) (as well as previous works by these authors), sediments were collected just below the surficial deflation eolic pavement (1-3 cm), with the collection campaign primarily centered around finding high-elevation sites (including old glacial deposits and relict rock surfaces with well-developed regolith) that could represent sources for loose sediments available for wind transport. Locating many of these potential sites was made possible by previous geological and geomorphological studies carried out in the Victoria Land area over the last decades (Armienti and Baroni, 1999; Baroni et al., 2005a; Baroni et al., 2008; Baroni et al., 2004; Baroni et al., 2005b; Hall and Denton, 2000a, b; Meneghel et al., 1999; Orombelli, 1990; Sugden and Denton, 2004; Sugden et al., 1999; Summerfield et al., 1999). The authors collected additional samples from glacial deposits of various ages, and at elevations ranging from the sea level to more than 2000 m.a.s.l. In SVL, some PSA samples were collected with the assistance of B. L. Hall (glacial deposits) and P. Augustinus (dune sand). Below, we describe the PSA samples analyzed in this work, with further geographic information provided in Table 1 of the supplementary material.

Two regolith samples were obtained from Frontier Mountain, a large nunatak situated at the inland catchment of the upper Rennick Glacier, near the edge of the Polar Plateau (Perchiazzi et al., 1999). This area is characterized by strong katabatic winds and abundant scattered fragments of local granitoids, pegmatites, and aplites associated with the Granite Harbor Intrusive Complex (Ordovician) (Gunn and Warren, 1962; Perchiazzi et al., 1999). Samples were collected from elevations 2200 and 2700 m.a.s.l., and appear to be directly weathered from the local granites. An additional granitic regolith was collected from the Lichen Hills, about 60 km southeast of Frontier Mountain.

To the east, two regoliths were sampled from Mount Masley of Pain Mesa, where the local bedrock is dominated by flat layers of Kirkpatrick basalt and intrusions of the Jurassic Ferrar dolerite (Baroni et al., 2004; Baroni et al., 2005b; Mensing and Faure, 1996). The Mesa Range is series of flat-topped, scree-covered mesas located at the head of the Rennick Glacier (Mensing and Faure, 1996). These samples were collected from 2520 and 2533 m.a.s.l., the first of which consists of topsoil on top of regolith, the second of weathered dolerite. A set of four samples was also collected from the Vantage Hills area, approximately 50 km south of Pain Mesa, nearer the edge of the Polar Plateau. Three of these samples consist of regolith weathered from continental feldspathic sandstone of the Beacon Supergroup, while the southernmost regolith is composed of a metamorphosed version of this sandstone (Capponi et al., 1997; Carmignani, 1989). The fourth sample from the Vantage Hills contains a mixture of glacial drift materials. Finally, two regolith samples were collected from the nearby Illusion Hills and Intermediate Hills, both weathered from the Ferrar dolerite.

Courtesy of P. Augustinus, two sediment samples were selected from the dune fields adjacent to Lake Vida, both of which are weathered from the continental sandstone (Bristow et al., 2010a; Bristow et al., 2010b). With the assistance of B. L. Hall, three additional samples were selected from an area slightly south of the dune fields: the first was obtained from an elevation of 1350 m.a.s.l., and consists of older glacial drift material varying from clast-supported to matrix-supported diamicton with a sandy-silty matrix (Baroni et al., 2008; Delmonte et al., 2010); the next sample is represented by a sandy-silt matrix from LGM glacial drift (correlated to the “Ross Sea I” glaciation, also referred to as the Terra Nova Drift by Baroni and Orombelli (1991)), along with older glacial drift material perhaps from the early- to mid-Pleistocene (Hall and Denton, 2000b; Kelly et al., 2002), and was collected much nearer sea-

level at 283 m.a.s.l.. A final sample from this set consists of regolith and thin topsoil, a mixture of dolerite fragments with possible traces of Beacon sandstone (1000 m a.s.l.).

From Taylor Valley, the southernmost of the ice-free valleys, five additional sediment glacial drift samples were collected. The first sample was obtained from one of the Marr Glacier moraines at 754 m.a.s.l. East of the Marr, two samples were gathered from a large moraine composite near the Goldman Glacier and Goldman Ponds, both from around 370 m.a.s.l. The two final samples were collected from a large, very old Taylor Glacier moraine, the second of which appears to be basalt-rich and possibly influenced by old Ross Sea drift; these were sampled at 420 and 438 m.a.s.l., respectively (Denton et al., 1993; Hall et al., 1993; Sugden and Denton, 2004).

Two regolith samples were collected from Tent Island (Erebus Bay) and Knob Point: a porphyric phonolite and an olivine basalt with xenolithic inclusions. Next, a total of seven glacial drift samples identified as LGM Ross Sea Drift (Hall and Denton, 2000a; Stuiver et al., 1981) were obtained from a variety of sampling sites, all within fairly close proximity to one another. These sites are as follows: Bratina Island; Dailey Island; Walcott Bay; Gneiss Point; Cape Crozier (Ross Island), and Cape Crozier's Bomb Peak (Ross Island). Six samples were from Cape Bird, including a volcanic regolith, additional Ross Sea Drift materials from the LGM (these potentially containing volcanic scoria or glasses), and two samples from a Holocene ice-cored moraine. The final MS site, Cape Royds, provided two regolith/drift samples composed of porphyric phonolite and volcanic scoria, as well as one ash sample.

3.2 Sample preparation and dissolution

Samples were prepared for isotopic analysis in a 10,000 level clean room at the University of Michigan, inside class 10 laminar flow hoods. Fine fraction PSA samples (particle size <5 µm) were selected according to separation procedures outlined in Delmonte et al. (2004a) at the University of Milano-Biocca, and checked using a Laser Particle Counter. Once separated, the dusts (approximately 10 mg of sample for either grain size fraction) were weighed into 3 mL Teflon beakers in preparation for Parr bomb digestion.

It has been demonstrated that standard hot plate digestion procedures using concentrated acids do not guarantee the complete dissolution of whole-rock samples containing refractory minerals such as zircon and garnet (Blichert-Toft et al., 2004; Mahlen et al., 2008; Patchett and

Tatsumoto, 1981). Because these same minerals tend to be highly concentrated in hafnium (Hf), an incomplete digestion of a sample could skew Hf concentrations and $^{176}\text{Hf}/^{177}\text{Hf}$ ratios, thereby excluding the application of the Lu-Hf isotope system from the work. In this study, following the advice of Patchett and Tatsumoto (1981) and many others (e.g. Blichert-Toft et al., 2004; Lapen et al., 2004; Mahlen et al., 2008), all samples were dissolved in highly pressurized, steel-jacketed Parr bombs using a two-phase acid attack. Once dissolved, the column chemistry procedures established by Aciego et al. (2009) were used to isolate the elements Sr, Nd, and Hf for isotopic analyses.

2.3 Neodymium isotope analysis

The initial-stage column, filled with 50-100 μm mesh TruSpec resin, split each sample into three cuts, of which the second cut containing the high field strength elements (HFSE) and the rare earth elements (REE) was set aside for subsequent sets of Nd chemistry. First, columns loaded with 100-200 μm LnSpec resin were used to separate the REEs from the HFSE, including Hf. Then, in the second separation stage, REE cuts were loaded into columns containing 50-100 μm LnSpec resin and exposed to a sequence of hydrochloric acids of varying normality in order to isolate the Nd, described in greater detail by Aciego et al. (2009).

Once dried, 1 μL each of 3M HCl and 3M HNO_3 was added to the sample, and the solutions were loaded onto outgassed rhenium double filaments. A current of 0.8 A was run through the filament until the sample dried. The current was then increased to 1.8 A and held for 60 seconds, after which it was briefly flashed at 2.2 A before returning to 0 A. Isotopic ratios were measured at the Glaciochemistry and Isotope Geochemistry Laboratory at the University of Michigan using a Thermo Scientific Triton PLUS Thermal Ionization Mass Spectrometer (TIMS), following methods detailed by Aarons et al. (2013). To correct for mass fractionation, Nd was normalized to $^{146}\text{Nd}/^{144}\text{Nd} = 0.7129$, and mass 149 was monitored for Sm interference. The Nd isotopic standard JNdi-1 (10 ng) was measured to be $^{143}\text{Nd}/^{144}\text{Nd} = 0.5121008$ ($2\sigma = \pm 2.37601 \times 10^{-5}$). During the course of this study, the BCR-2 USGS basalt rock standard yielded a $^{143}\text{Nd}/^{144}\text{Nd}$ ratio of 0.512625 ($2\sigma = \pm 1.41174 \times 10^{-5}$), and measured blanks were considered to be negligible. Below, Nd compositions are reported in terms of their deviations from the Chondritic Uniform Reservoir (CHUR) evolution line, wherein $\epsilon\text{Nd}(0) =$

$((^{143}\text{Nd}/^{144}\text{Nd})_{\text{meas}}/(^{143}\text{Nd}/^{144}\text{Nd})_{\text{CHUR}}) - 1 \times 10^4$, using the present-day CHUR value of 0.512638 (Jacobsen and Wasserburg, 1980).

2.4 Strontium isotope analysis

The first set of the three TruSpec cuts, containing the Sr, Pb, and Ra fractions was dried down and brought up in 7.5M HNO₃. Samples in solution were subsequently loaded into columns filled with 50-100 m mesh Eichrom Strontium Spec resin, and processed according to the recipe dictated by Aciego et al. (2009).

The dried strontium samples were re-dissolved in 1 μL of 7.5M HNO₃, and loaded mixed with 0.8 μL of TaF₅ activator onto outgassed rhenium single filaments to enhance ionization efficiency (Charlier et al., 2006). Drying procedures resembled those previously described for Nd, with samples heated and left to dry down at a current of 0.8 A, followed by a brief flash to 2.2 A. Radiogenic Sr isotopic ratios were then measured on the University of Michigan TIMS, following methods described by Stevenson et al. (2014). All measurements were corrected for mass fractionation using $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$. The isotopic standard NBS-SRM-987 yielded a value of $^{87}\text{Sr}/^{86}\text{Sr} = 0.710291$ ($2\sigma = \pm 8 \times 10^{-6}$), and the USGS reference material BCR-2 measured $^{87}\text{Sr}/^{86}\text{Sr} = 0.705033$ ($2\sigma = \pm 7.86 \times 10^{-6}$), in good agreement with reported literature values (e.g. Aarons et al., 2013; Aciego et al., 2009).

2.5 Hafnium radiogenic isotope analyses

Hf, along with the other HFSEs, was separated from the REEs during Nd column chemistry; Hf cuts were passed through an additional LnSpec column to isolate from the elements Lu, Yb, and W to prevent isobaric interferences. After this stage, samples were dried down and transferred to the University of Wyoming's High-Precision Isotope Laboratory, where they were brought up in 1M HNO₃ and measured on a Thermo Scientific Neptune Plus Multiple Collector Inductively Coupled Plasma Mass Spectrometer (MC-ICP-MS) as detailed in Arendt et al. (2014). During analysis, instrumental mass fractionation was corrected using $^{179}\text{Hf}/^{177}\text{Hf} = 0.7325$ and the exponential fractionation law and masses 182, 175, and 172 were monitored for W, Lu and Yb interferences. Baselines were measured prior to each sample, and amplifier gains were measured at the start of every day. Interference corrections to $^{176}\text{Hf}/^{177}\text{Hf}$ were less than 100 ppm. All sample measurements were further normalized by bracketing each sample with the JMC-475 Hf

reference material (accepted value of $^{176}\text{Hf}/^{177}\text{Hf} = 0.28216$) (Vervoort and Blichert-Toft, 1999). Hf isotopic compositions obtained for the BCR-2 rock standard measured $^{176}\text{Hf}/^{177}\text{Hf} = 0.28288$ ($2\sigma = \pm 4 \times 10^{-5}$), similar to reported literature values (e.g. Jweda et al., 2015). As with Nd, Hf isotope ratios will throughout the text be presented in epsilon notation, with $\epsilon\text{Hf}(0) = ((^{176}\text{Hf}/^{177}\text{Hf})_{\text{meas}} / (^{176}\text{Hf}/^{177}\text{Hf})_{\text{CHUR}} - 1) \times 10^4$, wherein CHUR = 0.282785 (Nowell et al., 1998).

3. Results

The isotopic compositions of fine ($<5 \mu\text{m}$) and bulk fraction PSA samples analyzed in this work are reported in Table 1 of the supplementary material. New Sr-Nd fingerprints plot along the same “hyperbolic envelope” as previously measured Victoria Land PSA data (Delmonte et al., 2010; Delmonte et al., 2013), which have been shaded in gray and plotted along with the new data in Figure 2 (samples measured by Delmonte and coworkers were measured exclusively in the finer ($<5 \mu\text{m}$) grain size fraction). Also included in Figure 2 are the Sr-Nd isotopic compositions of lava and tephra samples from Erebus volcano (Sims et al., 2008), grouped in color along with our “McMurdo Volcanics/glacial drift” samples based on close isotopic similarities.

It can be observed that, despite having been collected from a small area, the Antarctic PSAs overall are quite heterogeneous. For instance, while the materials collected near the McMurdo Sound and Mt. Erebus are relatively constrained on the Sr-Nd plot (blue circles in Figure 2), the isotopic compositions of the Victoria Land regoliths and drifts are scattered throughout the plot. Measured Sr isotopic ratios range from $0.703041 < ^{87}\text{Sr}/^{86}\text{Sr} < 0.782734$. As for variations in ϵNd , the range is also quite broad, with $-12.01 < \epsilon\text{Nd} < 6.36$. These same Sr-Nd data have been incorporated into the regional isotope contour map of fine-grained ($<5 \mu\text{m}$) source materials (Figure 3).

The new PSA data from this study are plotted in the three panels of Figure 4, separated by geographic location (MS, SVL, NVL, TV). In this case, MS volcanics remain indicated by blue circles, with orange diamonds representing NVL PSA samples and dark squares for SVL samples, with the exception of TV moraine materials, which are pictured as tan triangles. The first panel (Figure 4a) features the Sr-Nd plot for new bulk fraction samples, the second panel (Figure 4b) Sr and Nd ratios of complimentary fine fraction ($<5\mu\text{m}$) material. In the third panel, we have calculated and plotted the isotopic difference between fine and coarse-grained samples.

Here, it is shown clearly that NVL PSAs are most variable in their Sr and Nd isotopic compositions. Meanwhile, TV and other SVL samples display little variation in their $^{87}\text{Sr}/^{86}\text{Sr}$ signatures. As for sites outside Victoria Land, Figure 5a takes a closer look at $^{87}\text{Sr}/^{86}\text{Sr}$ differences for the MS samples with respect to distance from the Erebus volcano. Particularly for the bulk fraction, it appears that the sites closest to Mt. Erebus (Cape Royds) exhibit the least radiogenic Sr signatures (Figure 5a). Conversely, the data shifts away from more radiogenic Nd signatures when plotted against increasing distance from the volcano, with the exception of the fine-fraction Knob Point outlier ($\epsilon\text{Nd} = -0.17$), located near the bottom left corner of Figure 5b.

The combined Hf and Nd isotopic signatures of fine and bulk samples are plotted in Figure 6. The first panel (Figure 6a) shows that the bulk material plots near the igneous and “zircon-bearing” sediment arrays (as defined by Bayon et al. (2009)). For the bulk fraction, the MS samples plot slightly below the “zircon-bearing” line, while the remaining Victoria Land samples tend towards lower ϵHf and ϵNd values. A sample of lacustrine sand from the Dry Valleys and the two granitic Frontier Mountains samples sit at the bottom left corner of Figure 6a with the least radiogenic isotopic compositions. In contrast, Figure 6b illustrates more radiogenic Hf signatures for the fine-fraction Victoria Land samples, with the majority plotting between the “seawater” and “zircon-free” sediment arrays (Bayon et al., 2009). The MS samples occupy a similar region in Hf-Nd space for both size fractions.

For the purposes of determining provenance based on isotopic composition in a dust “sink,” such as an ice core (e.g. Grousset et al., 1992) or sediment core (e.g. Di Vincenzo et al., 2010), we list below a summary of our isotopic findings: (1) of all the PSAs considered in this study, materials representing the MS and Ross Island region are by far the most isotopically constrained. While the majority of Victoria Land samples exhibit relatively low, non-radiogenic Nd compositions, ϵNd for MS samples are comparatively quite radiogenic (*i.e.* greater than zero). Both Hf and Nd isotopic compositions are highly radiogenic, with approximate ranges of $0 < \epsilon\text{Nd} < 6$, and $2 < \epsilon\text{Hf} < 7$. Conversely, Sr values obtained for these materials trend towards non-radiogenic Sr, ranging approximately from $0.703 < ^{87}\text{Sr}/^{86}\text{Sr} < 0.709$; (2) for each of the Sr, Nd, and Hf isotope systems, the doleritic NVL PSAs occupy a moderate range, with $^{87}\text{Sr}/^{86}\text{Sr}$ falling just outside the MS range (most frequently around $^{87}\text{Sr}/^{86}\text{Sr} = 0.711$). Based on our limited selection of samples, we would expect radiogenic Nd and Hf signatures to range from approximately $-5 < \epsilon\text{Nd} < -1$, and $-2 < \epsilon\text{Hf} < 2$, respectively; (3) although some NVL PSA

samples plot closely to SVL values in $^{87}\text{Sr}/^{86}\text{Sr}$ space, regoliths weathered directly from the older granites and gneisses of the Ross Orogeny are particularly radiogenic with respect to Sr (*i.e.* $^{87}\text{Sr}/^{86}\text{Sr} > 0.726$). Our data indicates that granitic and other non-dolerite PSAs occupy a similar Nd isotopic range to the NVL dolerites; (4) the dataset implies that SVL sands have a fairly defined Sr fingerprint, with $0.712 < ^{87}\text{Sr}/^{86}\text{Sr} < 0.714$. Nd isotopic compositions range from around $-10 < \epsilon\text{Nd} < -5$ for bulk PSA samples, and slightly more radiogenic (around $-8 < \epsilon\text{Nd} < -3$) for the finer size fraction. As for Hf isotopes, all clay-sized particles fall around $\epsilon\text{Hf} = 0$, while the bulk material falls along, above, and below the zircon-bearing sediment array (Bayon et al., 2009), ranging from $-17 < \epsilon\text{Hf} < -6$, and; (5) such as with the other SVL PSAs, moraine material collected from the Taylor Valley occupies a narrow range of Sr isotopes, with $0.710 < ^{87}\text{Sr}/^{86}\text{Sr} < 0.712$ (slightly less radiogenic than the SVL sands). Hf isotopic ratios for bulk TV samples fall in the range of $-12 < \epsilon\text{Hf} < -5$, while Hf data for fine-fraction materials is limited in this region.

4. Discussion

4.1 Sr and Nd isotopes

Due to the variety of lithologies and ages of exposed rocks in Victoria Land and the McMurdo Sound areas, $^{87}\text{Sr}/^{86}\text{Sr}$ and ϵNd vary broadly for the Antarctic PSAs. For the NVL regolith samples, isotopic compositions resemble existing whole rock data for NVL granites, which suggest that $^{87}\text{Sr}/^{86}\text{Sr}$ may vary from 0.705 to 0.730, with ϵNd between -12.7 and 1.2 (Rocchi et al., 1998, and sources therein). Likewise, the doleritic regolith samples from NVL and SVL fall within the same isotopic ranges as Ferrar dolerite whole rock data presented in Faure et al. (1974) and Hergt et al. (1989). Southern Victoria Land (SVL, Dry Valleys) samples analyzed in this work tend to closely mimic the $^{87}\text{Sr}/^{86}\text{Sr}$ values obtained for PSA samples derived from Beacon sandstone reported in Delmonte et al. (2013). However, our range of ϵNd for SVL materials extends to more radiogenic values than obtained by B. Delmonte and coworkers; that is, while lacustrine and dune sands from this work fall in a similar, confined range ($-8.1 < \epsilon\text{Nd} < -7.6$) likely due to intense eolian reworking and sorting over time, the new glacial drift materials from SVL and TV are far more diverse, and may be as radiogenic as $\epsilon\text{Nd} = 2.5$ for the basalt-rich TAYLOR-5 ($< 5 \mu\text{m}$), and $\epsilon\text{Nd} = 0.51$ for sample TAYLOR-1 (also $< 5 \mu\text{m}$) obtained from an alpine moraine of the Marr Glacier.

Due to their close proximity to Mt. Erebus and the other volcanic centers of Ross Island, it was expected that MS glacial drifts would be composed at least partially of volcanic materials, yielding the lowest $^{87}\text{Sr}/^{86}\text{Sr}$ signatures and the highest ϵNd . This prominent volcanic signature is clearly illustrated by green $^{87}\text{Sr}/^{86}\text{Sr}$ contours on the isotope map (Figure 3) near Ross Island and the McMurdo Sound. It is well understood that radiogenic Sr and Nd isotopic compositions are distinct for mantle-derived or crust-derived rocks (Faure, 1986); given the distinctive isotopic composition of this PSA, regional dust or ash transport from the McMurdo Sound/Erebus region should be readily identifiable if it is a significant component of the dust flux in local ice cores (Delmonte et al., 2010; Delmonte et al., 2013).

As illustrated in Figure 4c, the majority of the MS samples, as well as some of the Victoria Land dusts, are more radiogenic with respect to $^{87}\text{Sr}/^{86}\text{Sr}$ in the finer versus the bulk size fraction, suggesting that the size-dependent fractionation for Sr isotope composition may be important and should continue to be taken into consideration in future PSA and other provenance studies (Kanayama et al., 2005; Meyer et al., 2011). Still, in cases where $\Delta ^{87}\text{Sr}/^{86}\text{Sr}$ (Fine – Bulk) is much less than 0 (see again Figure 4c), and/or $\Delta \epsilon\text{Nd}$ (Fine – Bulk) veers appreciably from 0 (*i.e.* the Frontier Mountains granites), we recognize that variations between the bulk and fine samples are completely unrelated to grain-size fractionation. Rather, we suggest that the widespread differences in ϵNd between these fine- and bulk-fraction samples can be attributed to variations in/mixing of source rock materials (*i.e.* a sampling site may serve as both a source and a sink of mineral dust).

To further explain, isotopic differences plotted in Figure 4c show that several Victoria Land samples are less radiogenic with respect to Sr for the fine versus bulk size fraction; in cases where this particular change in $^{87}\text{Sr}/^{86}\text{Sr}$ is combined with an also variable ϵNd signature, especially for fine fraction samples, we propose that the continental dusts and sediments have been mixed with very particles from different lithological terrains (e.g. Delmonte et al., 2013). For example, although coarser grained particles from TV isotopically resemble the other SVL materials, and some of the fine grained TV materials overlap well with SVL fine grained data, there appears to be a volcanic component resulting in higher ϵNd values for two of five of the TV PSAs (the previously mentioned TV-1 and TV-5), wherein isotopic signatures begin to resemble the MS volcanics; Delmonte et al. (2013) cautioned that this background volcanic component complicates the source to sink comparison of fine fraction PSAs to ice core dusts,

and thus, techniques other than Sr-Nd isotopic analysis should be used for a clearer discrimination.

Moreover, the isotopic similarities between other southern hemisphere PSAs and local dust sources (see Figure 7) prevent us from determining whether or not southern South America (or other remote sources) remain significant contributors of dust to the EAIS during the Holocene (Delmonte et al., 2010) (Figure 7), which is why we chose to further characterize the PSA samples using the Hf isotope system. Aarons et al. (2013) found that the quantity of non-radiogenic Hf present in an atmospheric dust sample can be correlated to the distance traveled from source area, and thus may be used to differentiate near and far sources of eolian material.

4.2 Hf isotopes

On the basis of Vervoort et al.'s "terrestrial array" (1999), we would anticipate negative ϵ_{Hf} and ϵ_{Nd} values to correspond with primary rocks or bulk sediments derived from old continental crust, such as the Antarctic granites and sandstones. Figure 6 illustrates that indeed all bulk fraction Victoria Land samples have $\epsilon_{\text{Hf}} < -5$, discounting the five NVL samples composed of or containing dolerite fragments (see gray boxes in Figure 6). Meanwhile, only two PSA samples (FM-1 and VH-3, both from NVL) display $\epsilon_{\text{Hf}} < -5$ in their fine fraction; from Figure 6a to Figure 6b, NVL, SVL, and TV samples have shifted away from the zircon-poor area of the plot (lower left), and towards the radiogenic MS samples in the clay-rich upper-right quadrant. The observed differences between the two size fractions considered in this study may indicate that dense zircon particles have fallen out of the air column during wind transport (Aarons et al., 2013). Indeed, differences between the fine and bulk fractions for PSA materials collected from the same sampling locale in terms of their radiogenic Hf isotope compositions may be diagnostic of inconsistent source rocks; that is, while the coarse fraction may have been weathered from the underlying bedrock, the finer fraction may have originated from a more distant source, (*i.e.* sample was already subjected to eolian transport and deposited at the PSA sample site).

In Figure 6, we note that the NVL dolerites do not show a significant shift in Hf isotope composition between the two size fractions (especially in comparison to the granitic materials, also labeled on the plot, although we are limited by a small sample size); based on homogeneous signatures for the dolerite samples, we recommend that the fine and coarse grain size fractions of dolerite materials are truly representative of the same PSA composition, having been derived

from the same parent rock source. Yet, we note that zircon (and therefore Hf) is likely present in smaller quantities for these dolerites than with the other rock types (*i.e.* sandstone, granite), in which case ϵ_{Hf} may not be the most ideal isotopic tracer for this particular lithology. For the MS samples on the other hand, we did not observe major differences in Hf isotopic compositions between the size fractions, in this case due to the materials being more highly weathered and well-sorted, and of course, volcanic in nature. Finally, although we have noted that the Sr-Nd isotopic ranges of SVL and TV materials tend to resemble other southern hemisphere dust sources, should particles archived in a natural “sink” such as a local, coastal ice core fall within Hf isotopic ranges reported in this study (most importantly, with non-radiogenic Hf), we believe that the material is most likely sourced locally from Victoria Land and not from a remote source.

In summary, Nd-Hf isotopes may be used to study the proximity of dust or sediments to the original source (see Aarons et al., 2013 for a more extensive discussion), and can potentially be a useful proxy to track changes in wind velocity and/or direction in relation to proximal and distal depositional sites (Garçon et al., 2013). Coarse (particle diameter > 5 μm) PSA materials generated from zircon-rich sandstones and granites are likely to be highly non-radiogenic in Hf, whereas fine-grained particles more susceptible to long-range versus short-range transport will have a more radiogenic signature with respect to Hf due to absence of the mineral zircon. Measurements of Hf isotopes from new or pre-existing PSA sites should provide further constraints on the application of this proxy; currently, our ability to use Hf isotopic ratios to identify dust provenances is limited by the lack of geochemical data in dust collected from potential source regions. The new Antarctic PSA data presented here greatly enriches the limited global catalogue of Nd-Hf isotopic signatures.

5. Conclusions

For geochemical studies of dust provenance, interpreting the information provided by radiogenic isotopes requires first a keen understanding of isotopic fractionation due to physical and chemical weathering, and internal sorting processes during transport. While the Sr-Nd isotopic compositions of sediments from Victoria Land and the McMurdo Sound sector described in this work—as well as those presented in previously published studies—generally correspond as expected to source rock lithologies, the potential background presence of aerosols transported from long-range southern hemisphere sources or from regional volcanic activity in some cases

complicates the direct source-to-sink comparison of isotopic signatures. In particular, the Sr-Nd isotopic fingerprints for fine-grained SVL and TV materials alone may be indistinguishable from those of several other (remote) southern hemisphere sources, such as South America and New Zealand. For these sites, incorporating Hf isotope analysis may lead to more accurate source identification, although additional sampling campaigns in both Antarctica and other southern hemisphere continents will be required in order to achieve a more precise Nd-Hf characterization of potentially important dust sources. Furthermore, because many PSA samples analyzed in this work are more radiogenic with respect to $^{87}\text{Sr}/^{86}\text{Sr}$ in the finer versus the bulk size fraction, we suggest that the grain size effect for radiogenic Sr isotopes should continue to be taken into consideration in future dust and sediment provenance studies. On the other hand, some samples from our study (especially the NVL granites) are quite different in terms of $^{87}\text{Sr}/^{86}\text{Sr}$, ϵNd , or both in their different size fractions in a way that does not suggest isotopic fractionation was at play; rather, it is highly possible that some selected PSA sites are both sources and sinks of mineral material, and may not be ideal sampling locales for future studies hoping to identify primary sources of mineral material. Moving forward, we advocate that the expansion of an isotopic catalogue for Antarctic PSAs should include multiple size fractions, as obtaining signatures for coarser-grained particles more susceptible to short-range transport may better allow us to rule out global sources of dust when examining local sinks such as sediment cores and ice cores.

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Figures and Figure Captions

Figure 1. Location map indicating PSA sampling regions and individual sites, with the East Antarctic Ice Sheet (EAIS) shaded in white, the Ross Sea in blue, and exposed rock surfaces in brown. Note: orientation of north arrow. Inset provides context for Antarctic ice core drilling sites Dome C, Vostok, Taylor Dome, and TALDICE.

Figure 2. Sr-Nd plot of PSA dust from Antarctica's Victoria Land and McMurdo Sound regions. In color, PSA data measured in this study, including volcanic Knob Point end-member (MS), and granitic Frontier Mountains end-member (NVL); gray symbols (as labeled), previously measured PSAs from Holocene raised beaches, eolian silt and sand traps, glacial drifts, and regoliths from Delmonte et al. (2010); also in color, volcanic samples from Sims et al. (2008).

Figure 3. Isotope contour map of fine-fraction (<5µm diameter) PSA samples in Antarctica's McMurdo Sound and Victoria Land regions, paired with previously published PSAs from Delmonte et al. (2010). Nd and Sr contours were determined by inverse distance weighted interpolation using ArcGis, with variations in Sr isotopic ratios defined by color, and Nd ratios separated by black contour lines. The MS and Ross Ice Shelf areas (southern samples; note orientation of north arrow) appear to be most radiogenic in terms of Nd, and least radiogenic for Sr, whereas the high-elevation nunataks of NVL exhibit the most radiogenic Sr compositions.

557 **Figure 4.** (a) Sr-Nd isotopic compositions of bulk fraction material measured in this study.
 558 Light-blue circles, glacial drift strongly influenced by McMurdo Volcanics; orange diamonds,
 559 Northern Victoria Land; dark-blue squares, Dry Valleys sand; and tan triangles, Taylor Valley
 560 moraine material; (b) Sr and Nd isotopic ratios of complimentary fine fraction ($<5\mu\text{m}$, more
 561 highly weathered) material; (c) difference between fine- and course-grained samples (i.e. bulk
 562 ratio subtracted from fine ratio), with $^{87}\text{Sr}/^{86}\text{Sr}$ differences multiplied by 1000 to better detect
 563 trends visually.

564
 565 **Figure 5.** (a) Strontium isotopic compositions of volcanic/glacial drift material with respect to
 566 distance from the Mt. Erebus volcano. Red circles, fine fraction ($<5\mu\text{m}$) dust; and black
 567 hexagons, bulk fraction samples; (b) ϵ_{Nd} isotopic compositions of the same PSA materials, also
 568 with respect to distance from Mt. Erebus.

Figure 6. (a) Nd-Hf isotopic signatures of bulk PSA material from this study, plotted along the steep, zircon-bearing sediment ($\epsilon_{\text{Hf}} = 1.80 \epsilon_{\text{Nd}} + 2.35$) and igneous arrays ($\epsilon_{\text{Hf}} = 1.37\epsilon_{\text{Nd}} + 2.89$) as defined by Bayon et al. (2009). The dashed line represents our calculated linear trend for the bulk fraction Victoria Land samples (NVL, SVL, and TV) ($\epsilon_{\text{Hf}} = 0.99 \epsilon_{\text{Nd}} - 1.07$; $r^2 = 0.54827$). Doleritic samples are surrounded by gray boxes, with samples BULK-MR1 and BULK-SM1 labeled for comparison to the corresponding fine fraction data. For this same purpose, granitic samples BULK-FM1, BULK-MW1, and BULK-DV2 are also labeled; (b) fine fraction dust plotted in Nd-Hf space, accompanied by the seawater ($\epsilon_{\text{Hf}} = 0.39\epsilon_{\text{Nd}} + 6.2$), zircon-free ($\epsilon_{\text{Hf}} = 0.91\epsilon_{\text{Nd}} + 3.1$), igneous, and zircon-bearing sediment arrays (2009). Again, dolerites are surrounded by gray boxes, although a smaller sample size and/or concentration for these materials resulted in markedly less reportable data than for the more abundant bulk materials, and therefore only two data points (MR1 and SM1) are reported. Fine fraction granitic samples FM1, MW1, and DV2 are also labeled.

Figure 7. Sr-Nd isotopic signatures of Antarctic PSA dust from this study and previous works, plotted alongside dust from remote southern hemisphere sources.

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