The Holocene

HOLOCENE DUST IN EAST ANTARCTICA: PROVENANCE AND VARIABILITY IN TIME AND SPACE

Journal:	The Holocene
Manuscript ID	HOL-19-0005.R1
Manuscript Type:	Review
Date Submitted by the Author:	n/a
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Keywords:	Ice Cores, Dust, Holocene, East Antarctica, Provenance, Dust stratigraphy
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Revised for the Special Issue on Holocene Dust Dynamics

HOLOCENE DUST IN EAST ANTARCTICA:

PROVENANCE AND VARIABILITY IN TIME AND SPACE

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1 Department of Environmental and Earth Sciences, University of Milano-Bicocca, Milan, Italy 2 Physics and Astronomy, Curtin University, Perth, Western Australia, 6102, Australia 3 British Antarctic Survey , High Cross, Madingley Road, Cambridge CB3 0ET, United Kingdom 4 Aix Marseille Univ, CNRS, IRD, INRA, Coll France, CEREGE, Aix-en-Provence, France 5 Department of Physical Geography, Stockholm University, S-106 91 Stockholm, Sweden 6 Swedish Museum of Natural History, Frescativägen 40, 104 05 Stockholm, Sweden 7 Department of Earth Sciences, University of Pisa, Via S. Maria n. 53, 56126, Pisa, Italy 8 CNR-Institute of Geosciences and Earth Resources, Via G. Moruzzi n.1, 56124, Pisa, Italy 9 Department of Pure and Applied Science, University of Urbino "Carlo Bo", Via S. Chiara 27, 61029 Urbino (PU), Italy (*corresponding author: barbara.delmonte@unimib.it) Abstract In this paper we provide a comprehensive overview of the state-of-knowledge of dust flux and variability in time and space in different sectors of East Antarctica during the Holocene. By integrating literature data with new evidences, we discuss the dust flux and grain size variability during the current interglacial and its provenance in the innermost part of the East Antarctic plateau as well as in peripheral regions located close to the Transantarctic Mountains. The local

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INTRODUCTION

Mineral dust deflated from continental landmasses of the Southern Hemisphere reaches high elevation sites of central East Antarctica after transport in the mid-to-high troposphere over very long distances greater than 1000 km (Lambert et al., 2008; Petit et al., 1999). After deposition onto the polar plateau, dust buried in snow and ice layers can be studied through deep ice core stratigraphies and can be used to document past atmospheric circulation variability.

During the late Quaternary, dust influx to the polar East Antarctic plateau was a factor ca. 25 higher during glacial periods than during interglacials, when the dust concentration in snow and ice layers reached extremely low levels, especially in the interior of the ice sheet (Petit et al., 1999; Lambert et al., 2008). Among the causes responsible for the drastic glacial/interglacial change of dust concentration in polar ice, a primary role is played by changed environmental conditions within dust source areas modulating the so-called source "intensity" (Delmonte et al., 2017). Dust atmospheric lifetime and snow accumulation rate in Antarctica, which are factors related to some degree to climate and the hydrological cycle (Petit & Delmonte, 2009, Markle et al., 2018), additionally played a key role. Dust in central East Antarctica is remotely sourced from extra-Antarctic continental landmasses, including South America and possibly other areas in the Southern Hemisphere (Basile et al., 1997; Delmonte et al., 2007, 2008, 2010a; Revel-Rolland et al., 2006; Gaiero, 2007; Gaiero et al., 2007; Gili et al., 2016, 2017; Revel-Rolland et al., 2006; De Deckker et al., 2010). Given the remoteness of the sources and the very long transport distance, the Holocene dust depositional flux in central East Antarctica is the lowest on Earth, and the grain size of background mineral aerosol is very small (<5 μm diameter). Even so, slight (0.3-0.6 μm) variations in the modal value of dust grain size mass distribution have been observed and associated to the varying strength of air subsidence over Antarctica and air inflow from low latitudes.

On the periphery of the East Antarctic plateau, close to the Transantarctic Mountains or to ice-free surfaces outcropping from the ice sheet as mountain peaks (nunataks), an important contribution to the input of fine dust particles into the local Antarctic atmosphere is provided by high-altitude exposed regoliths and glacial drifts, providing non-cohesive sediments immediately available for wind mobilization and transport. In Northern Victoria Land, for example, indication of a long history of wind erosion is provided by some high-elevation relict surfaces, sometimes displaying aeolian deflation pavements, exposed for millions of years (Baroni et al., 2004; Oberholzer et al.,

2003, 2008; Di Nicola et al. 2012). During the Holocene, when the extremely low remote dust influx from extra-Antarctic sources is mixed in varying proportions with locally-sourced dust, the ice core dust record becomes sensitive to the regional climate and atmospheric circulation variability. This is the case for Talos Dome (Baccolo et al., 2018; Delmonte et al., 2010b, 2013; Albani et al., 2012) and Taylor Dome ice core records (Aarons et al., 2016), for example. The spatial dispersal of local sediments towards the interior of the ice sheet, however, is definitely negligible or null, as revealed by spatial studies on a number of firn cores from the pre-industrial period (Delmonte et al., 2013).

At low elevation, on the coastal fringes of the continent close to sea level, East Antarctic ice-free areas are more abundant although, on the whole, their extension represents only about 1% of Antarctica. The most extensive ice-free areas in East Antarctica are located in Northern and Southern Victoria Land and in the southern Transantarctic Mountains. Although discontinuously distributed, other ice-free areas much less extended than these exist in coastal Adélie Land, Wilkes Land, Prince Charles Mountains, Endreby Land and Dronning Maud Land (figure 1). In these regions, the local dust deposition at low elevation sites can be orders of magnitude greater than the pristine remote background measured in ice cores from the inner plateau (Atkins & Dunbar 2009, Chewings et al. 2014). In addition, in some settings Antarctic dust can become an important potential source of bio-available Fe to the ocean (Winton et al. 2014, 2016a, 2016b).

The intent of this paper is to provide a comprehensive overview of the state-of-knowledge of dust flux and of its variability in time and space in different sectors of East Antarctica during the Holocene. We divide this paper in three sections. First, we present an overview of the main issues coming from studies of dust influx and transport variability in central East Antarctica during the Holocene, preceded by a brief excursion of the most well-established methods for analyzing dust concentration and grain size in polar ice cores. In this first section, we also present some new, high-resolution results from a late Holocene section of the new SOLARICE ice core, central East Antarctica. Then, we discuss Holocene dust provenance both in the innermost part of the East Antarctic plateau, at different sites located along a transect following nearly similar latitude but different longitude, and in marginal sites of Victoria Land close to ice-free areas that are of unique importance in terms of regional paleo-climatic and paleo-environmental reconstructions. Finally, the third and last part of this paper is focused on the local importance of aeolian mineral dust aerosol deflated from dusty low-elevation areas of Antarctica such as McMurdo Sound, and new

data from Dronning Maud Land and the Bunger Hills Oasis, in Queen Mary Coast, Wilkes Land. All throughout this paper, literature data are integrated with new evidences from this work; we recommend the reader to refer to the supplementary material for details about samples and methods, and for additional data.

DUST CONCENTRATION, FLUX AND GRAIN-SIZE IN CENTRAL EAST ANTARCTICA THROUGHOUT THE HOLOCENE

TECHNIQUES FOR DUST CONCENTRATION AND GRAIN-SIZE MEASUREMENTS

Since the early 1980's microparticle counting and sizing on discrete ice core meltwater samples has been typically performed through the Coulter Counter technique (Petit et al., 1981; Fujii and Ohata, 1982), which measures the short-term changes in the electrical impedance across a very small aperture (typically 30 or 50 µm) through which particles suspended in a very diluted electrolyte solution are forced to flow. Since this change is proportional to the particle volume, a particle volume-size distribution spectrum - represented in terms of equivalent spherical diameter - can be obtained. Under the assumption of spherical particle shape and constant density (2.5 g/cm³), the number- and mass-size distributions can be also obtained. The high sensitivity, precision and accuracy of Coulter Counter measurements have made this technique a reference for dust counting and sizing in polar ice cores. Today it is possible to measure by Coulter Counter concentrations of a few ppb over a high-resolution size spectrum of 400 log-size channels (or more) on less than 5 mL of sample. Optical counters measuring particle scattering (e.g. Fujii et al., 2003; Kawamura et al., 2017), optical extinction cross section (e.g. Ruth et al., 2002, 2008) or both extinction coefficients and optical thickness (Potenza et al., 2006) have also found application in ice-core research. The systematic application of extinction (scattering plus absorption)-based optical devices (e.g. Klotz Abakus laser sensor) coupled to continuous flow systems for high-resolution analyses of ice cores, has produced the longest undisturbed record of mineral dust aerosol spanning the last 800.000 years (Lambert et al., 2008) from the EPICA Dome C (DC) ice core. However, while optical particle counters can be easily calibrated with Coulter Counter mass concentrations, the non-negligible size distribution differences between extinction-based optical sensors and the Coulter Counter measurements caused by the irregular shape of dust particles in ice core samples (Potenza et al., 2016), made it necessary to introduce a new calibration routine (Simonsen et al., 2018).

DUST FLUX VARIABILITY

Average Holocene and pre-industrial dust mass concentrations and fluxes are reported in table 1 for different ice and firn cores from the East Antarctic plateau, along with additional information such as the investigated time interval, the average accumulation rate (from AICC2012 timescale, Veres et al., 2013), the altitude of each site and references for dust data. Locations of ice core sites are shown in figure 1. For the same sites, in figure 2 we show average dust fluxes calculated respectively for the Early, Middle and Lower Holocene sections of the cores.

Holocene dust fluxes from high-altitude inner plateau ice cores are definitely very low compared to other parts of the polar ice sheet: at Dome B, one of the highest ice core drilling sites considered, Holocene dust fluxes display the lowest average Holocene levels (0.28 mg m⁻² per year; this work and Delmonte 2013). Similar average values are found at Vostok, Dome C (0.36 and 0.39 mg m⁻² per year respectively; Delmonte et al., 2005) and at Dome Fuji (0.54 mg m⁻² per year; Fujii et al., 2003; Kawamura et al., 2017). All these sites are located above 3000 m a.s.l. at similar latitude but different longitude, and receive dust from remote sources through high-altitude transport (Delmonte et al., 2004, 2005). The similarity is also confirmed by the general trend of dust flux variability during the Holocene at these sites. For all of them the temporal evolution of dust deposition reveals a general, slight, but continuous decrease across the Holocene. It was observed since the very first measurements on the EPICA DC ice core (Delmonte et al., 2002) and tentatively attributed to the gradual reduction of the dust reservoir available for wind deflation by progressive development of deflation pavement, pedogenesis, and increased vegetation cover on continents. Compared to the other high altitude inner sites, Dome Fuji shows an apparent flux increase during the Middle and Late Holocene, the reason for which is under investigation but could be an artifact of the lower resolution record (Kawamura et al., 2017).

When comparing inner plateau sites with peripheral sites such as Talos Dome and Taylor Dome, located close to the Transantarctic Mountains where important ice-free areas (nunataks, cliffs, ice free valleys, glacial moraines, etc) are located, some important differences arise. Compared to

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Dome C, the average Holocene flux of fine particles (<5 μ m) is ~4-5 times at Talos Dome and ~3 times at Taylor Dome. These differences are related to the local contribution from proximal ice-free areas within Victoria Land, which provide an additional regional dust input to the margin of the Antarctic plateau, as discussed below. Given these peculiarities, the stratigraphic records of mineral dust aerosol at Talos Dome and Taylor Dome (Albani et al., 2012; Baccolo et al., 2018; Aarons et al., 2016) is of exceptional importance to regional climate studies. Their analysis and interpretation made it possible to link the evolution of the local dust cycle with important climatic and environmental processes that are expressed in this Antarctic region. This is the case for the retreat of the Ross Sea ice shelf and the opening of the Ross Sea embayment occurred after the deglaciation and during the Early Holocene. In that period the drastic reduction of dust influx from remote glacial sources allowed local dust dynamics to emerge and become recognizable in ice core records.

13 In the South Atlantic sector of the Plateau, the dust flux at EPICA DML is also remarkably higher 14 than in central plateau sites, a factor ~3-4 compared to Dome C (figure 2). The higher flux is 15 related to the proximity of EPICA DML site to Southern South America, that is believed to be the 16 major dust source at the site (see below), and possibly to the further contribution from additional 17 sources (Wegner et al. 2015).

GRAIN-SIZE VARIABILITY CONTROLLED BY SECULAR-SCALE OSCILLATIONS

Holocene dust minerals deposited at Dome C (Gaudichet et al., 1986) typically consist of feldspars (k-feldspars and plagioclase), quartz and its polymorphs, phyllosilicates (e.g. micas such as muscovite, illite and other clay minerals), Fe, Al and Ti oxides in variable amounts. These particles display variable shape (Potenza et al., 2016) but systematically small grain size, as a consequence of the long and complex transport history, which occurred at high altitude prior to reaching the polar plateau (figure 3C). The maximum spherical equivalent of dust particles representing the typical background at Dome C is about 5 µm, while the volume-size distribution typically shows a lognormal distribution with a mode around 2 μ m and a geometric standard deviation (σ_g) of ~1.8 (Delmonte et al., 2002). An exception is given by the presence of occasional tephra particles. They are related to explosive volcanic eruptions and their size can lead to significant deviations from the

background values: in ice core samples it is not uncommon to observe volcanic particles with a
 diameter larger than 50 μm (Narcisi et al., 2005).

The background dust deposited in central East Antarctica displays only limited grain-size variations in time. In low-accumulation sites of central East Antarctica, where the effect of wet deposition (Unnerstad and Hansson, 2001) is negligible and deposition is essentially dry (Legrand and Mayewski, 1997), very small grain-size variations can be attributed to different atmospheric transport mechanisms. Relatively coarse dust (displaying abundance of coarse particles in the 3-5 µm diameter range) is related to dust-carrying air masses subjected to short and relatively low-altitude trajectories in the mid-troposphere. On the contrary, relatively fine dust (displaying abundance of fine particles in the 1-2 µm diameter range) is related to longer circumpolar atmospheric pathways, implying mass convergence in the middle troposphere and subsidence over the Antarctic Plateau (Krinner & Genthon, 2003). This possible explanation of dust grain size variability, proposed in some literature studies (Delmonte et al., 2005, 2017) can be applied both to Holocene and to glacial climate conditions. Actually, the dust transport time from the continental sources of the Southern Hemisphere to Antarctica, is definitely very long; according to a semi-empirical model (Petit & Delmonte, 2009), the mean dust transit time is about one month. This value is similar to the ones calculated using ²²²Rn air concentration: they span from 22 to 28 days (Maenhaut et al., 1979; Genthon & Armengaud, 1995; Jacob et al., 1997; Li et al., 2008).

The EPICA DC ice core reveals clear oscillations of the grain-size parameters (Coarse Particle Percentage, CPP; i.e., the percentage of dust mass represented by particles within 3-5 µm spherical diameter) throughout the Holocene (Figure 3A), similarly to Vostok (Delmonte et al., 2005). This feature suggests that transport patterns at a given site may vary in time. Spectral analyses of the low resolution (discontinuous, one sample every ~50 years) Holocene grain-size stratigraphic profile of the Dome C and Vostok ice core, revealed that the energy is mostly spread between secular-scale bands of periodicity from 330-380 years to about 120 years. Interestingly, both EPICA DC and Vostok share a common significant frequency band around 200 years (Delmonte et al., 2005).

To better understand these periodic patterns, we present here the grain-size record from a new ice core drilled in Dome C in the framework of the SOLARICE research project (figure 3B). For the first time, dust size is measured continuously along the core on discrete samples of about 4-5 cm each (see supplementary material), representing about 2 years of accumulation at 100-200 m depth. According to a very preliminary timescale established by transferring SOLARICE to

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AICC2012 chronology (Veres et al., 2013), the time period investigated here spans from about 3300 to 4500 years BP (in terms of depth from 125 to 160 m). The preliminary profile of CPP (figure 3B) confirms the presence of secular-scale oscillations (for example around ~360 years according to these preliminary data), similar to the ones already observed in low-resolution measurements. In addition, thanks to the higher temporal resolution, some high-frequency modes of variability arise. One of them is located around the ~80-90 years band, according to these preliminary data. These new results confirm the variable strength and localization of tropospheric air subsidence over Antarctica, which is related to local (< $\sim 10^4$ - 10^5 km²) and regional ($\sim 10^6$ - 10^7 km²) atmospheric dynamics, and highlight the importance of high-resolution ice core analyses for depicting high frequency modes of variability still hidden to discontinuous dust measurements. Studies on the SOLARICE ice core are going on in order to cover the entire Holocene; dust grain size variations will be compared, along with other climate and atmospheric proxies, to the cosmogenic-produced ¹⁰Be isotope, in order to investigate possible links between solar activity and climate.

DUST PROVENANCES

CENTRAL EAST ANTARCTICA

20 Dust-provenance studies have been extensively carried out over the last decades in central East 21 Antarctic ice cores. These studies were primarily focused on glacial periods because of the 22 relatively high abundance of aeolian particles in the samples. Important changes in dust 23 composition with respect to glacial periods arise during Holocene, although undoubtedly analytical 24 errors associated to geochemical measurements become higher with respect to glacial age, as a 25 consequence of the extremely low dust content, i.e. a few ng of dust per mL of sample.

Methods classically used include radiogenic isotope fingerprinting (Sr, Nd, Pb; Grousset et al., 1992; Basile et al, 1997; Delmonte et al., 2004, 2007; 2010a; Vallelonga et al., 2010), dust geochemical composition in terms of major, trace and Rare Earth Elements (Marino et al., 2008; Baccolo et al., 2018; Siggaard-Andersen et al., 2007; Gabrielli et al., 2010; Wegner et al., 2012), as well as mineralogical composition (Gaudichet et al., 1986, 1988) and dust ferromagnetic properties (e.g. Lanci et al., 2008).

For the glacial periods of the late Quaternary, these methods (figure 4) converge towards a dominant provenance from Patagonia/Tierra del Fuego (Grousset et al., 1992; Basile et al., 1997; Delmonte et al., 2004) including the Argentinean continental shelf at least during the sea-level low stand of MIS2 (Delmonte et al., 2017), with additional inputs from lower-latitude areas in South America (Gaiero et al., 2007; Gili et al., 2016, 2017) such as the Pampean region in the southern part of central western Argentina.

When compared to the last glacial period, MIS 2, Holocene dust in central East Antarctic ice cores is enriched in Ti, Al and K (Marino et al., 2008), more variable in time in terms of Rare Earth Elements (REE) patterns (Gabrielli et al., 2010; Wegner et al., 2012), characterized by a higher Lithium solubility (Siggaard-Andersen et al., 2007), relatively low-radiogenic in terms of ¹⁴³Nd/¹⁴⁴Nd (figure 4; Delmonte et al., 2007), and more highly-magnetic (Lanci et al., 2008). In this respect, Pb isotope data are less indicative because the Pb input related to local (Antarctic) volcanoes veils the faint portion of Pb that is dust-related (Vallelonga et al., 2010).

All dust proxies cited above, agree on the variable dust composition during Holocene and likely during the last 15 ka, in opposition to the very uniform composition of dust during MIS2, and point towards a great mixture from multiple sources, an hypothesis that is also coherent with a possible contribution from different sub-sources inside the same continent.

The Holocene Sr-Nd radiogenic isotope composition of dust from Vostok and Dome C, compared with the vast literature data from potential source areas of the Southern Hemisphere available today, can be justified in different ways. A first possibility (Delmonte et al., 2007) implies a South American provenance only, from a weakened Patagonian source where periglacial dust production is drastically reduced, plus other low-latitude sources inside South America itself. These include loess from latitudes North of about 30°S, displaying isotopic characteristics very different from Patagonia/Southern Pampas aeolian sediments (Smith et al., 2003; Gaiero et al., 2004, 2007; Gili et al., 2017). Actually, while the similarity between loess samples from the southern Pampas and volcanic rocks of the southern Andes and Patagonia corroborates the possible Patagonian/southern Andean source for this loess (e.g. Smith et al., 2003), the isotopic fingerprint of loessic deposits further north becomes progressively distinct with respect to Patagonian/southern Andean basalts, and a possible role played by a source in the Puna Altiplano and/or the Bolivian Andes is also possible for these deposits (Iriondo et al., 1997), although debated (Sayago et al., 2001). An additional hypothesis to explain the isotopic signature of Holocene dust in central East Antarctica, is a direct transport from high-altitude Andean sources

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(such as the Altiplano-Puna plateau) to Antarctica via the jet stream (Gaiero et al., 2007; Delmonte
 et al., 2008; Siggaard-Andersen et al., 2007; Gili et al., 2016; 2017).

3 Other plausible scenarios for Holocene dust provenance in central East Antarctica imply a 4 dominant contribution from South America plus a secondary Eastern Australian source (Revel-5 Rolland et al., 2006; De Deckker et al., 2010; Du et al., 2018) possibly integrated with a New 6 Zealand source (Wegner et al., 2012; Neff and Bertler, 2015).

Almost certainly, the variable relative contribution from different sources in the course of the Holocene and the lack of one dominant dust source area account for the variable geochemical characteristics of Holocene dust in time. This marked temporal variability can be observed in different proxies, including dust magnetization (Lanci et al., 2008) which shows a very large variability with sometimes extremely high values that are unusual in crustal rocks. This suggests concentration of ferromagnetic minerals in association with Holocene and in general interglacial dust; one possible explanation relates high magnetization to an underestimated flux of micrometeorites (Lanci et al., 2012) in the size range of aeolian dust, which to date is still largely unconstrained (Duprat et al., 2007).

THE VICTORIA LAND MARGIN OF THE ICE SHEET

During glacial times the remote dust influx from South America influenced both the interior and the margin of the Antarctic ice sheet, overlooking any regional signal; after the massive deglacial dust decline and during Holocene, the margin of the East Antarctic plateau experienced a progressive regionalization of the dust cycle. This is clearly expressed by the Talos Dome (Baccolo et al., 2018; Delmonte et al., 2010, 2013; Albani et al., 2012) and Taylor Dome (Aarons et al., 2016) ice core dust records. These two ice cores offer a unique opportunity to study the regional history of marginal East Antarctica.

At Talos Dome in Northern Victoria Land, Holocene dust is not well-sorted, and is characterized by the occurrence of sporadic fine-silt grains of obvious local provenance. The isotopic (Figure 5, Delmonte et al., 2013) and elemental composition (Baccolo et al. 2018) of Holocene dust at Talos Dome is distinctly different from Dome C and Vostok (figure 5) and shows close similarity with local rock outcrops, mainly with regoliths developed on Ferrar dolerites. The importance of the doleritic outcrops as high altitude (2500–3000 m a.s.l.) dust sources in Northern Victoria Land was

recently explained in relation with the geomorphology of the outcrops. Doleritic reliefs usually culminate with tabular ice-free plateaus (Mesa), consisting of high-altitude relict flat structural surfaces that given their altitude, are not affected by glacial/interglacial changes of ice sheet height (Baroni et al., 2004). Such elevated surfaces are intensively weathered and produce material that is promptly injected into the middle troposphere by winds. As outlined in Baccolo et al. (2018), conversely the typical alpine morphology that characterizes granitic nunataks, with pronounced peaks and steep cliffs, is less suitable for deflation and promotes accumulation of debris and glacial drift at the bottom of the walls, where deflation is disadvantaged.

Under these conditions, strong atmospheric uplift is not necessary in order to transport mineral aerosol to the site, while only the direction of air masses is important. Modern back trajectories reanalysis shows that dust transport from high-elevation ice-free areas of Victoria Land towards the Talos Dome area mainly occurs in spring/summer months and is mainly driven by local atmospheric circulation in the western Ross Sea, involving mesoscale cyclogenesis and northward-flowing air masses (Delmonte et al., 2013). Such a regional atmospheric circulation pattern involves only the margin of the East Antarctic ice sheet, as demonstrated by the limited spatial extent of local dust influence.

Interestingly, a steady feature of background dust at Talos Dome is the presence of geochemically-heterogeneous volcanic material, incompatible with derivation from a primary volcanic eruption (Delmonte et al., 2013). This volcanic contribution represents an intrinsic characteristic of the background dust at Talos Dome, suggesting a role played by wind in the secondary remobilization of volcanic material accumulated from different volcanic sources of Victoria Land. This volcanic contribution also accounts for the more volcanogenic isotopic fingerprint that characterizes the Holocene dust at Talos Dome if compared to the central plateau (figure 5) and for the peculiar ferromagnetic features of TALDICE Holocene dust (Lanci & Delmonte, 2013).

Similarly to Talos Dome, the radiogenic isotope and REE composition of dust in the Taylor Dome ice core (Aarons et al., 2016, Figure 5) also point towards a local dust provenance after the deglaciation and throughout the Holocene. Dust geochemical composition is characterized by a marked variability in time, in opposition to the homogeneous South American-like signature of dust from the last glacial period. A detailed study on dust from the Taylor Glacier (Aarons et al., 2017), originating from its accumulation area at Taylor Dome and terminating in the McMurdo Dry Valleys, confirms these findings and provides evidence that changes in local dust inputs occurred approximately concurrently to the Ross Ice Shelf retreat.

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The documentation of the radiogenic isotope composition of dust and sandy sediments from Northern and Southern Victoria Land, as well as McMurdo Sound, was extended by Blakowski et al. (2016), that produced an interesting regional map showing the spatial variability of Sr and Nd isotopes in the area. The spatial patterns observed in the map are primarily related to parent lithologies (Beacon sandstone, Ferrar Dolerites, mixture of these with Kirkpatrick basalts and granites in some cases). Here we also integrate new Sr and Nd isotopic values of two moraine samples from Taylor Glacier (figure 5). As in the case of Talos Dome, however, the transport of volcanic material related to regional volcanic activity prevents any direct and definite source-to-sink comparison of isotopic signatures. From figure 5 it can be observed that some Holocene samples clearly fall within the Dry Valleys isotopic field, but the presence of volcanic material makes many Holocene data aligned towards a volcanic pole.

Reciperies

DUST CYCLE IN LOW ELEVATION ICE-FREE AREAS OF EAST ANTARCTICA

A number of ice free sites and other potential source of dust exist at low-elevation across East Antarctica; they consist of raised beaches, coastal oases and islands, cliffs and ice-free valleys, Late Pleistocene glacial drift and supraglacial debris on ice shelves (debris bands, Fig. 6A, 6B). Here we highlight their importance either as dominant source locally, or as potential contributors for atmospheric background dust at present day and in the past.

There is little information on transport distance of dust from proximal ice-free sources around the margin of Antarctica, and future work is required to understand the dust cycle on the margin of the ice sheet throughout the Holocene.

McMURDO SOUND

The McMurdo Sound is the dustiest known location in Antarctica. The dust accumulation there is spatially and temporally variable: the present-day dust flux is in the order of $\sim 1 \text{ g m}^{-2} \text{ yr}^{-1}$, which is orders of magnitude greater than long-range transport of remote dust in the central plateau (Chewings et al., 2014; Winton et al., 2014). Additionally, a modal particle size in the fine sand range and geochemical affinity with local rocks, confirm a local dust provenance (Winton et al., 2016a; Winton et al., 2014). Geochemistry and sedimentological studies of dust deposited on snow on McMurdo Sound sea ice (figure 6C) demonstrate that dust is locally-sourced from the debris bands on the McMurdo Ice Shelf. The debris bands (figure 6A) comprise an area of unconsolidated sediment, whereby sediment is lifted from the sea floor by anchor ice and frozen into the base of the ice shelf and eventually exposed by surface ablation (Kellogg et al., 1990). As such, the debris bands are considered to be an effectively unlimited source of aeolian dust (Atkins and Dunbar, 2009).

A progressive decrease in dust flux (55 to ~ 0.2 g m⁻² yr⁻¹) and modal particle size (primary mode: 130 to 25 μm; fine silt mode relatively constant between 4-10 μm) has been observed along a dust 55 29 plume extending >120 km northwards of the debris bands onto the sea ice (Atkins and Dunbar, 57 30 2009; Chewings et al., 2014; Winton et al., 2014; Macpherson, 1987). Dust dispersal northwards onto the sea ice is consistent with the local meteorology, since the fastest and most common winds that can entrain fine sand and silt, are predominantly from the South (katabatic flow). A

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local source for the supraglacial debris found on McMurdo ice shelf, is confirmed by Al/Fe elemental ratios (Atkins and Dunbar, 2009; De Jong et al., 2013) and Sr-Nd radiogenic isotopes (Winton et al., 2016a). The Sr and Nd isotopic composition of dust deposited along the sea ice dust plume (Winton et al., 2016a; Winton et al., 2014) forms a linear array resulting from a two component mixture of the isotopically-distinct McMurdo Volcanic Group (0.703<⁸⁷Sr/⁸⁶Sr<0.710; - $0.17 < \epsilon_{Nd}(0) < 5.7$) and southern Victoria Land (0.708 $<^{87}$ Sr/ 86 Sr<0.740; -12.5 $< \epsilon_{Nd}(0) < -2.0$) (Delmonte et al., 2004, 2010b, 2013; Blakowski et al., 2016) end members. Despite the significant spatial variability in the region, dust on the McMurdo Sound sea ice represents the isotopic signature of McMurdo Sound (Winton et al., 2016a) that can be uplifted and transported to other Antarctic sites. The isotopic composition of McMurdo Sound dust source (figure 5) confirms, not surprisingly, that sediments are dominated by volcanogenic material.

Secondary elevated patches of dust occur within embayments and near coastal headlands along the south Victoria Land coastline. Sourced from the McMurdo Dry Valleys, the dust is not widely dispersed and thus the McMurdo Dry Valleys represent only a minor source of dust to the sea ice (Bentley, 1979; Barrett et al., 1983; Chewings et al., 2014; Winton et al., 2016a; Winton et al., 2014; Ayling and McGowan, 2006).

Ice-core records of dust accumulation in the McMurdo Sound region only span the past 35 years, where higher dust fluxes in winter are related to more frequent local storms (Ayling and McGowan, 2006; Dunbar et al., 2009). This region represented a local Antarctic low elevation dust source over the Pleistocene and the entire Holocene, since it has been ice-free for millions of years (Sugden et al., 1995). Dust accumulation measurements up to 120 km from the debris bands highlight that McMurdo Sound represents an important source of dust for the immediate southwestern Ross Sea area today (Winton et al., 2016a). However, while the large fraction dust flux is comprised of coarse-grained particles with limited transport distance, the still-unknown transport distance of the finer-grained particles potentially extends far beyond.

DRONNING MAUD LAND

Dronning Maud Land (DML) is located in the Weddell Sea sector of East Antarctica, directly downwind of the South American continent. Large drainage systems divide DML into an eastern, a central and a western part. Although the majority of DML is covered by ice, some outcrop massifs occur in this region: Heimefrontfjella and Vestfjella, Kirwanveggen and Mühlig-Hofmann

Mountains, Schirmacheroase, Steingarden and SørRondane Mountains, and Yamato-Belgica
 mountains. Their morphology resembles typical alpine landscapes, with U-shaped and hanging
 valleys that indicate a glacial origin, related to ancient wet-based glaciations (Holmlund & Näslund,
 1994). These mountain ranges and nunataks represent important ice-free lands in DML.

5 Heimefrontfjella (10-13°W, 74-75°S) and Vestfjella (13-16°W, 73-74°S) are two important ranges 6 of nunataks about 150 km apart, trending nearly parallel to the coastline. Heimefrontfjella, 7 composed of various gneisses and schists of Precambrian age, represents an effective barrier to 8 the ice flow, which is channeled in several outlet glaciers. Vestfjella, conversely, is a range of 9 basaltic nunataks situated about 120 km from the coast. Basen is the northernmost nunatak, while 10 nearby ones are Plogen and Fossilryggen. This latter is the only nunatak where Permian 11 sedimentary rocks outcrop through the ice.

Here we present new Sr and Nd isotopic data (figure 7A) of: (1) regolith from the Vestfjella mountain range in DML and (2) ice core dust from a 100 m deep ice core that was drilled 140 km from the coast in the Vestfjella at Camp Maudheimvidda (CM, 73°06'19"S, 13°09'54"W; 360 m a.s.l) during 1997-1998 as part of the EPICA pre-site survey. The site is within tens of kilometres of the Basen, Plogen and Fossilriyggen nunataks, and ~200 km north of Svea Station. Dating of the CM core shows that it covers a period from AD 1997-1700 (Jonsell et al., 2005). Two dust samples were extracted from the ice core sections: sample DML#1 represents the upper 55 m of the core, that at this depth consists of firn, its age spans 30-100 years before 1997; DML#2 was prepared extracting the dust from the lower part of the core, from 55 to 105 m depth, it roughly covers the time period between AD 1890 and 1790. The dust samples from both the CM ice core and the Basen, Ploggen and Fossilryggen nunataks, display a similar isotopic composition, restricted in a relatively narrow interval (0.710378 $<^{87}$ Sr/ 86 Sr <0.715801 and -7.3< ϵ_{Nd} (0)<-8.5). This interval is more restricted than bulk rock values from the area (Vestfjella basalts, andesites and tholeiites, Coats Land dolerites), and less radiogenic in Sr with respect to samples from the Svea station, where lithologies are different (granites, gneiss).

Besides indicating that dust at CM is locally-sourced, these data allow us to fingerprint the isotopic composition of this local, low-elevation area within DML. Isotopic values from CM ice core are distinctly different from values registered in central East Antarctica during pre-industrial times (figure 4, Delmonte et al., 2013), which represents the pristine remote input to the polar plateau, and can be useful to assess the regional importance of local dust transport even in more inland areas. So far, no radiogenic Sr and Nd isotope fingerprint exist for Holocene ice core dust from the

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EPICA DML ice core drilled at Kohnen station (0° 04'E, 75°00'S) at about 2882 m a.s.l. However, Wegner et al. (2012), by using REE patterns, identified a clear contribution from multiple sources during the Holocene and generally after 15 ka BP. Surely, the high dust flux (figure 2) and atmospheric nssCa²⁺ concentration at the site, used as a proxy for mineral dust (Fischer et al., 2007), can be partly explained with the greater proximity of the South American dust source to the EPICA DML site. However, the possibility of a contribution from other sources must be also taken into account, as suggested by Wegner et al. (2015) on the basis of the phasing between dust emission and transport. The local importance of the contribution from Antarctic sources in the DML area can be also appreciated when considering the very high dust flux of 30-120 mg m⁻² yr⁻¹ registered for the last 50 years in a low elevation coastal ice core (IND-25/B5; 1300 m a.s.l., 71° 20' S, 11° 35' E), that is orders of magnitude higher than on the plateau (Laluraj et al., 2014).

BUNGER HILLS

Bunger Hills (Queen Mary Land; 66°S; 100°E; 165 m a.s.l.), also known as the Bunger Hills Oasis, is a low elevation ice free area of about 950 km² composed of a southern landmass and several large islands and marine inlets to the north. The Bunger Hills emerged from the ice sheet during the latest Pleistocene and early Holocene, and according to Augustinus et al. (1997) the Bunger Hills and the adjacent Islands to the west might have been ice-free throughout the Last Glacial Maximum. This seems corroborated by sand luminescence and radiocarbon dating described in Gore et al. (2001) and Augustinus and Duller (2002) (Table 1). The area consists of granulite-facies metamorphic rocks with a variety of compositions intruded by voluminous plutonic rocks (from gabbro to granite), and a variety of mafic dykes (Stüwe and Powell, 1989; Stüwe and Wilson, 1990; Sheraton et al., 1992; Ravich et al., 1968). The geochemistry of Bunger Hills rocks is described by Sheraton et al. (1993). In this work we document for the first time the Sr and Nd isotopic composition of some Bunger Hills fossil beach ridges of Holocene age (except one sample from the last glacial maximum), representing a low elevation potential source for dust since the last glacial period. Details about the samples, including their age, are reported in the supplementary material. Results are plotted in figure 7B. The isotopic range spanned by these samples $(0.720 < {}^{87}Sr / {}^{86}Sr < 0.761$ and $-24.3 < \epsilon_{Nd}(0) < -19.7$) is very similar to one moraine sample from Bunger Hills reported by Basile et al. (1997) and distinctly different from other Antarctic sources. Also, these values show no similarity with coeval dust archived in central East Antarctic ice cores,

confirming that the Bunger Hills ice-free area has only a local relevance with respect to the atmospheric dust emission and deposition. These data will be useful for future studies aimed at assessing the regional dust cycle in the area during the late Quaternary.

CONCLUSIONS

The Holocene dust cycle in East Antarctica shows regional differences between high-altitude sites of the central East Antarctic plateau, marginal Victoria Land locations close to the Transantarctic Mountains, and low elevation coastal sites. The former display the lowest dust deposition fluxes on Earth, given the long atmospheric transport from remote, extra-Antarctic sources. High accuracy measurements have provided evidence that the grain size of mineral dust displays some variations on secular and decadal timescale, although it is always very small. This can be associated to variable atmospheric circulation patterns including the varying strength of air subsidence over Antarctica. Aeolian dust reaching these remote sites represents the well-mixed atmospheric background of the Southern Hemisphere. Given the extremely reduced contribution from remote sources, the Holocene dust history at the margin of the polar Plateau close to the Transantarctic Mountains, becomes sensitive to regional climate changes. While field evidences suggest that the areal extent of ice-free areas at high elevation (mountain tops, nunataks) did not change drastically since the last glacial maximum, the atmospheric circulation changes related also to the opening of the Ross Sea, surely played a role on local dust dispersal to the margin of the plateau during the Holocene. The contribution from patchy, low elevation sites of marginal East Antarctica is much less studied, except for the southwestern Ross Sea, the dustiest known place in Antarctica, where the so-called "debris bands" on the McMurdo ice shelf are found. Detailed studies in other sectors of the continent (DML, Bunger Hills, etc.) are presented, but need further investigation in order to assess the contribution of these sites to the atmospheric dust load over the Continent and the Southern Ocean.

1		Revised for the Special Issue on Holocene Dust Dynamics 2019								
2 3	1									
4 5	1 2	Acknowledgments								
6 7 8 9 10 11 12 13 14 15 16	2	This paper is a contribution to the Franco-Italian project SOLARICE (IPEV/11/15, PNRA16, 00008)								
	1	The field work at Concordia benefited from logistical support from the French and Italian Polar								
	т 5	Agoncios IDEV and DNPA and the C2EN for drilling activities								
	5	Agencies, in EV and FINRA, and the CZFN for drining activities.								
	0	Museum" Grant This article is an outcome of Project "MULD Disartimenti di Escollenza 20								
	/	iviuseum Grant. This article is an outcome of Project "MIUR-Dipartimenti di Eccellenza 201								
10 17	8	2022". We all acknowledge P. Augustinus for providing raised beach deposit samples from Bung								
18 19	9	Hills and P. Biscaye for providing samples from Dronning Maud Land.								
20 21	10									
22	11	Tables and figure captions								
23 24	12									
25 26 27 28	13	Table 1: Ice core drilling sites with coordinates and age of the dust record considered, average dust								
	14	concentration (ppb) for the time period indicated with standard deviation, average accumulation ra								
29	15	expressed in cm of water equivalent per year deduced from the AICC2012 timescale, average dust								
30 31	16	calculated from dust concentration and accumulation at each site, expressed in mg of dust per m ² per ye								
32 33	17	altitude of each site and references for dust data.								
34 25	18									
36	19	Figure 1: Map of Antarctica with zoom on the McMurdo Sound area, Bunger Hills and Dronning Maud								
37 38	20	Land. The location of the most important ice core drilling sites cited in the text is indicated with a blue								
39 40	21	star. Toponyms are abbreviated as follows: EDC: EPICA Dome C; VK: Vostok; DB: Dome B; DF: Dome Fuji;								
41	22	TD: Talos Dome; TY:Taylor Dome; TG: Taylor Glacier; CM: Camp Maudheimvidda; EDML: EPICA Dronning								
42 43	23	Maud Land.								
44 45	24									
46 47	25 26	Figure 2: Early Holocene (11.7-8.2 kyr BP), Middle Holocene (8.2-4.2 kyr BP) and Late Holocene (4.2-2 kyr								
48	26	BP) dust flux (particles Ø<5 μm) measured in seven different ice cores located on the East Antarctic								
49 50	27	plateau. The blue circle refers to data from the new SOLARICE-Dome C ice core (see text). Black boxes								
51 52	28 20	refer to preindustrial levels (between about AD 1800 and 1400). See Table 1 for data references.								
53	29									
54 55	30	Figure 3: Holocene dust grain-size variability at Dome C. (A) discontinuous, low resolution record of coarse								
56 57	31	particle percent (CPP) from the EPICA DC ice core (Delmonte et al., 2005); (B) running average of CPP dust								
58 59 60	32	grain size index measured at high resolution (4-5 cm long samples) on the new SOLARICE ice core. The red								

curve represents a low frequency smoothing of data. (C) SEM images reporting examples of Holocene particles in central East Antarctic ice. Scale bar=5 µm. Figure 4: 87 Sr/ 86 Sr versus $\varepsilon_{Nd}(0)$ isotopic composition of East Antarctic ice core dust from the Holocene and from MIS2 and of fine sediments (S) and aeolian dust (AD) from potential source areas. Holocene ice core data are from Dome C and Vostok; MIS2 ice core data are from Dome C, Vostok, Dome B. One additional point for the previous interglacial (MIS 5.5) is also reported for comparison. Source data are referred to the fine fraction of aeolian dust (AD) and sediments (S) of different typology (topsoil, loess, alluvial fans, ephemeral lakes, lacustrine clay/silt, fluvial suspended load, salar edges, etc.) from South America, Australia and New Zealand. Data from the Illimani ice core (Bolivian Altiplano, 16°37'S, 67°46'W) are also reported for comparison. Data sources: Basile et al., 1997; Delmonte et al., 2004; 2007; 2010; 2013; 2017; Smith et al., 2003; Gaiero, 2007; Gaiero et al., 2004, 2007, 2013; Gili et al., 2017; Sugden et al., 2009; Revel-Rolland et al., 2006; Gingele and De Deckker 2005. Figure 5: ⁸⁷Sr/⁸⁶Sr versus $\varepsilon_{Nd}(0)$ isotopic composition of Holocene and MIS2 ice core dust from marginal East Antarctic plateau sites in Northern and Southern Victoria land (Talos Dome, Delmonte et al., 2010, 2013; Taylor Dome, Aarons et al., 2016) and Taylor Glacier (Aarons et al., 2017). Ice-core data are compared with fine sediments from local Antarctic dust sources (Blakowski et al., 2016; Delmonte et al., 2010, 2013, Winton et al., 2014, 2016a, 2016b, and this work), bulk rocks (Fleming et al., 1992, 1995) and the volcanic end-member (Rocholl et al., 1995). The black ellipse highlights the isotopic field for McMurdo Sound dust identified by Winton et al., 2016a. The two black circles refer to new data from the Dry Valleys analyzed in this work (see text and supplementary material). Figure 6: (6A) Late Peistocene glacial drift (in foreground) with granite erratic (white) on Bratina Is. and supraglacial debris band on McMurdo Ice Shelf (in background). (6B) Section in the Late Pleistocene glacial drift (younger drift) on the Dailey Island West. Note the aeolian deflation pavement on top. (6C) Southern McMurdo Sound, November 2009: snow on sea ice sampling for dust provenance studies. Photo credit: James Pinchin. Fig. 7: Sr-Nd isotopic composition of Holocene ice core dust from low elevations sites. (A) Camp Maudheimvidda (CM) ice core in coastal DML, local nunataks (Basen, Ploggen, Fossilryggen) and Svea station. Isotopic fields are drawn from literature data on bulk rocks (basalts, tholeiites, andesites) from the DML area of Vestfjella, and Coats Land dolerites (Luttinen & Furnes, 2000; Harris et al., 1990; Riley et al., 2005; Brewer et al., 1992). (B) Bunger Hills. Sr-Nd isotopic data from bulk (all size included) and fine (<5

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2 3 1 4 2 5 2	μ m) dust samples from the Bunger Hills raised beaches in Wilkes Land (see text) as well as one data point from a local moraine (Basile et al. 1997).
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Figure 1: Map of Antarctica with zoom on the McMurdo Sound area, Bunger Hills and Dronning Maud Land. The location of the most important ice core drilling sites cited in the text is indicated with a blue star. Toponyms are abbreviated as follows: EDC: EPICA Dome C; VK: Vostok; DB: Dome B; DF: Dome Fuji; TD: Talos Dome; TY:Taylor Dome; TG: Taylor Glacier; CM: Camp Maudheimvidda; EDML: EPICA Dronning Maud Land.

266x355mm (96 x 96 DPI)



Figure 2: Early Holocene (11.7-8.2 kyr BP), Middle Holocene (8.2-4.2 kyr BP) and Late Holocene (4.2-2 kyr BP) dust flux (particles ø<5 µm) measured in seven different ice cores located on the East Antarctic plateau. The blue circle refers to data from the new SOLARICE-Dome C ice core (see text). Black boxes refer to preindustrial levels (between about AD 1800 and 1400). See Table 1 for data references.

256x181mm (300 x 300 DPI)



4.4

Figure 3: Holocene dust grain size variability at Dome C. (A) discontinuous, low resolution record of coarse particle percent (CPP) from the EPICA DC ice core (Delmonte et al., 2005); (B) running average of CPP dust grain size index measured at high resolution (4-5 cm long samples) on the new SOLARICE ice core. The red curve represents a low frequency smoothing of data. (C) SEM images reporting examples of Holocene particles in central East Antarctic ice. Scale bar=5 µm.

190x300mm (96 x 96 DPI)





Figure 4: 87Sr/86Sr versus □Nd(0) isotopic composition of East Antarctic ice core dust from the Holocene and from MIS2 and of fine sediments (S) and aeolian dust (AD) from potential source areas.

Holocene ice core data are from Dome C and Vostok; MIS2 ice core data are from Dome C, Vostok, Dome B. One additional point for the previous interglacial (MIS 5.5) is also reported for comparison. Source data are referred to the fine fraction of aeolian dust (AD) and sediments (S) of different typology (topsoil, loess, alluvial fans, ephemeral lakes, lacustrine clay/silt, fluvial suspended load, salar edges, etc.) from South America, Australia and New Zealand. Data from the Illimani ice core (Bolivian Altiplano, 16°37'S, 67°46'W) are also reported for comparison.

Data sources: Basile et al., 1997; Delmonte et al., 2004; 2007; 2010; 2013; 2017; Smith et al., 2003; Gaiero, 2007; Gaiero et al., 2004, 2007, 2013; Gili et al., 2017; Sugden et al., 2009; Revel-Rolland et al., 2006; Gingele and De Deckker 2005.

270x269mm (300 x 300 DPI)

http://mc.manuscriptcentral.com/holocene



Figure 5: 87Sr/86Sr versus □Nd(0) isotopic composition of Holocene and MIS2 ice core dust from marginal East Antarctic plateau sites in Northern and Southern Victoria land (Talos Dome, Delmonte et al., 2010, 2013; Taylor Dome, Aarons et al., 2016) and Taylor Glacier (Aarons et al., 2017).

Ice-core data are compared with fine sediments from local Antarctic dust sources (Blakowski et al., 2016; Delmonte et al., 2010, 2013, Winton et al., 2014, 2016a, 2016b, and this work), bulk rocks (Fleming et al., 1992, 1995) and the volcanic end-member (Rocholl et al., 1995). The black ellipse highlights the isotopic field for McMurdo Sound dust identified by Winton et al., 2016a. The two black circles refer to new data from the Dry Valleys analyzed in this work (see text and supplementary material).

300x270mm (300 x 300 DPI)



Figure 6: (6A) Late Peistocene glacial drift (in foreground) with granite erratic (white) on Bratina Is. and supraglacial debris band on McMurdo Ice Shelf (in background). (6B) Section in the Late Pleistocene glacial drift (younger drift) on the Dailey Island West. Note the eolic deflation pavement on top. (6C) Southern McMurdo Sound, November 2009: snow on sea ice sampling for dust provenance studies. Photo credit: James Pinchin.

150x350mm (96 x 96 DPI)

http://mc.manuscriptcentral.com/holocene



Fig. 7: Sr-Nd isotopic composition of Holocene ice core dust from low elevations sites. (A) Camp Maudheimvidda (CM) ice core in coastal DML, local nunataks (Basen, Ploggen, Fossilryggen) and Svea station. Isotopic fields are drawn from literature data on bulk rocks (basalts, tholeiites, andesites) from the DML area of Vestfjella, and Coats Land dolerites (Luttinen & Furnes, 2000; Harris et al., 1990; Riley et al., 2005; Brewer et al., 1992). (B) Bunger Hills. Sr-Nd isotopic data from bulk (all size included) and fine (<5 μm) dust samples from the Bunger Hills raised beaches in Wilkes Land (see text) as well as one data point from a local moraine (Basile et al. 1997).

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	Reference		Delmonte, 2003; This work	Delmonte et al., 2005	Delmonte et al., 2002	This work	Fujii et al., 2003 Kawamura et al., 2017	Wegner et al., 2015	Albani et al., 2012 Baccolo et al., 2018	Aarons et al., 2016				Delitionte et al., 2013						
	Altitude (m a.s.l.)		3650	3480	3233	3233	3810	2882	2315	2365		3230	2795	2455	2315					
	Dust flux (mg m ⁻² yr ⁻¹)		0.28	0.36	0.39	0.31	0.54	1.50	1.76	1.18		0.20	0.19	0.50	0.75					
	Acc.rate (cm w.eq yr ⁻¹)	ections	2	2	2.7	2.6	2.7	9	104	9	sections	2.5	2.0	3.6	8.7					
	Dust conc. (ppb) ±St.Dev.	lce s	14.2±8	17.8±8	14.3±8	11.7±5	20±11	25±14	25.1±10	21±14	Firn	8±4	9±4	14±3	8±4					
	Age		2.0-11.7 Kyr BP	3.6-9.8 Kyr BP	2.0-11.7 Kyr BP	3.3-4.5 Kyr BP	3.2-11.7 Kyr BP	2.0-11.7 Kyr BP	2.0-11.7 Kyr BP	2.0-11.7 Kyr BP		1570-1800 AD	1420-1700 AD	1620-1800 AD	1420-1810 AD					
	Coordinates						77°05′S, 94°55′E	78°28'S, 106°48'E	75°06′S, 123°21′E	123°21′ 48′′E 75°06′31′′S	77°19'01"S, 39°42'12"E	75°00'S, 00°04'E	72° 49'S, 159°11'E	77°47′47″S, 158°43′26″E		75°06'S, 123°21'E	75°35'S, 135°49'E	75°32'S, 145°510E	72° 49'S, 159°11'E	
	lce core		DOME B	VOSTOK-BH7	EPICA-DC	SOLARICE-DC	DOME FUJI	EPICA-DML	TALDICE	TAYLOR DOME		DC ITASE	D4 ITASE	MdP-A ITASE	TALDICE					
Holocene Holocene									0 -	2 3 Pre-industrial	5 5	0 1 8								

Supplementary information SAMPLES ANALYZED IN THIS STUDY

1- SOLARICE ice core samples

The SOLARICE project is a multi-year Franco-Italian scientific initiative aimed at retrieving and studying Holocene climate and solar variability from a new ice core drilled at Dome C, Concordia Station (East Antarctic Plateau). One of the main goals of SOLARICE is the reconstruction of solar activity across the entire Holocene from high-resolution ¹⁰Be analyses and the investigation of the relationship between ¹⁰Be and climate/atmospheric circulation changes that are reconstructed through a multiproxy approach including mineral dust aerosol grain size.

The project lead to the recovery of a 204 m deep ice core during the Antarctic summer 2015/16 at about 1.5 km from Concordia station (123° 21,793'E, 75° 06,511'S). In this study, we present preliminary dust grain size data (figure **3B**) obtained from ~5 cm long samples from about 125 to 160 m depth.

2- Camp Maudheimvidda ice core samples

Two ice cores were drilled at Camp Maudheimvidda (CM, 73°06'S, 13°09'W; 360 m a.s.l) in Dronning Maud Land during the Antarctic summer 1997-1998, as part of the EPICA pre-site survey. The cores were drilled 5 m apart and are 100 m deep. Camp Maudheimvidda is situated 140 km from the coast in the Vestfjella mountain range. The site is within tens of kilometers range of several nunataks (Basen, Plogen, Fossilriyggen) and approximately 200 km north of Svea Station. The total time span of the CM core is 300 years (Jonsell et al., 2005).

Two dust samples were extracted from the ice core. Sample DML#1 is the upper 55 m of firn, spanning 30-100 years BP. Sample DML#2 is the lower section, from 55 to 105 m depth, spanning 100-200 years BP. The snow accumulation rate at CM is 220 mm w.eq year⁻¹. (Holmlund et al., 2000). Dust was extracted by filtration and analyzed for Sr and Nd isotopic composition. Samples were collected by Margareta Hansson and Pierre Biscaye.

3- Taylor Glacier moraines

In this work we analyzed the Sr and Nd isotopic composition of the fine fraction (<5 micron) of two
 sediment samples from Taylor Glacier, Transantarctic Mountains, provided by Dr. Luca Lanci (TG1, TG9).
 They consist of mixed material from a lateral moraine of the Taylor Glacier where the dominant lithology is
 Ferrar Dolerite possibly mixed with other lithologies. Results from these two samples (Supplementary Table

S1) have been integrated in Figure 5 inside the "Dry Valleys" group. TG1 coordinates: 161° 45.445'E77°46.152'S; TG9 coordinates: 161° 47.782' E, 77° 43.714'S.

4- Dronning Maud Land nunataks

Nine samples from Basen (73°02'S, 13°24'W), Plogen (73°13'S, 13°50'W) and Fossilriyggen(73°23'S, 13°02'W) nunataks in the Vestfjella mountain range, western DML, were sampled for this study and analyzed for their Sr and Nd isotopic composition. Bedrock in the area is basalt (Sohlenius et al., 2004). Also, two regolith samples were collected close to Svea Station (74°34'S, 11°13'W), located in the Heimefrontsfiella mountain range. There, bedrock consists of metamorphosed red granite and augengneiss (Sohlenius et al., 2004). Samples were collected by Margareta Hansson. The samples were sieved using a 150µm nylon mesh and the fine fraction was used for Sr and Nd analyses.

5- Bunger Hills fossil beach ridges

Seven raised beach deposits from Bunger Hills (Gore et al., 2001), East Antarctica (100°20'E to 101°28'E; 65°58'S to 66°20'S) were analyzed for Sr and Nd isotopic composition (Supplementary Table S1). Both the bulk (all size fractions) and the fine (<5 micron) size fractions were measured. At EUROCOLD (University Milano-Bicocca, UNIMIB), the coarse fraction was removed from bulk samples by using a pre-washed 5 µm SEFAR Nitex[®] open mesh, while the fraction 0.4µm<∅<5µm was collected on 0.4 µm Isopore[™] Polycarbonate membranes. After filtration, the membranes were put into one pre-cleaned Corning tube filled with ~10 mL MQ water, and microparticles were removed from the filter by sonication.

These samples were kindly provided by Dr. P.Augustinus. Organic matter was removed with hydrogen peroxide and carbonates removed with hydrochloric acid. The age of these sediments (Optically Stimulated Luminescence (OSL), Gore et al., 2001) spans from about 21900±3400 years BP to 4800±700 years BP.

METHODS

1- DUST CONCENTRATION AND GRAIN SIZE

About 600 samples were analyzed for concentration and grain size by using in parallel a Beckman Coulter Multisizer 4 and a Beckman Coulter Multisizer 4e, strictly intercalibrated, available in clean rooms ISO6 at EUROCOLD Laboratory, UNIMIB. Analytical protocols are described in Delmonte et al., 2005. Analyses were performed on about 4-5 mL per sample and from the dust volume (mass) size distribution the CPP (Coarse Particle Percentage) parameter was calculated. CPP represents the relative (%) volume or mass fraction included in the size interval between 3-5 μm.

1- STRONTIUM AND NEODYMIUM ISOTOPIC RATIOS

At the Department of Geosciences, Swedish Museum of Natural History (NRM, Sweden) the Taylor Glacier and Bunger Hills samples were evaporated in acid-cleaned 7 mL Savillex beakers. The chemical treatment of the samples including mineral dust digestion and an elemental separation (Rb-Sr and Sm-Nd) using ion exchange chromatography was performed. Here, a line dedicated to the treatment of small dust samples (1-10 ng of Nd and 5-100 ng of Sr in dust for Antarctic ice core samples) was developed (Delmonte et al., 2008). Briefly, the samples were spiked with mixed ¹⁴⁷Sm/¹⁵⁰Nd spike and ⁸⁴Sr enriched spike for isotope dilution determination of the concentrations. Samples were digested in an acid mixture of 1.5 mL of HNO₃, HF and HClO₄ and heated to 90 °C in closed Savillex beakers for three days. The solution was evaporated to complete dryness on a hot plate and the residue re-dissolved in 4 mL 6M HCl. Bunger Hills bulk samples were ground to a powder using a steel mortar and spiked as above. Bulk samples were digested in a mixture of 2 mL HF and 20 drops of HNO₃ in steel bombs in an oven at 205 °C for three days. The solution was evaporated to complete dryness on a hot plate. The residue repeatedly re-dissolved in concentrated HNO₃ (Seastar) and evaporated to remove fluorides. The residue was re-suspended in 5 mL of 6 M HCl and heated in steel bombs at 205 °C for 24 hours. The samples were evaporated and re-suspended in 4 mL 6 M HCl.

To achieve separation of potential interfering elements (Fe, Ba, Rb, Sm, Ce, and Pr), and obtain high column yield and low blanks, the residue was subjected to chemical procedures described in Delmonte et al. (2008). The total blank, including dissolution, chemical separation and mass spectrometry, was frequently monitored in each ion exchange batch and blank concentrations were <4pg for Nd and <80pg for Sr.

³⁰ 34 Isotopic analysis of Nd and Sr ratios was performed with a Thermo Scientific TRITON Thermal Ionisation
 ⁶⁰ 35 Mass Spectrometer (TIMS). Neodymium was loaded mixed with Alfa Aesar colloidal graphite, on double

rhenium filaments and analyzed as metal ions in static mode using rotating gain compensation. Concentrations and ratios were calculated assuming exponential fractionation. The calculated ratios were normalized to 146 Nd/ 144 Nd = 0.7219. Epsilon units are calculated as follows: $\varepsilon_{Nd}(0) = [({}^{143}Nd/{}^{144}Nd)_{sample}/({}^{143}Nd/{}^{144}Nd)_{CHUR} - 1] \times 10^4;$ CHUR, chondritic uniform reservoir with $(^{143}Nd)^{144}Nd)_{CHUR} = 0.512638$ The external precision for ¹⁴³Nd/¹⁴⁴Nd is estimated from analysis of the nNdß (small samples) and the La Jolla standard. The resulting precision is \pm 0.4 ϵ –for small samples (<15ng) and \pm 0.2 ϵ for larger samples (>15 ng), as reported in Supplementary Table 2. Accuracy correction was not applied since the mean standard 143 Nd/ 144 Nd ratios were within error of accepted literature values (nNdß 143 Nd/ 144 Nd ratio 0.511895 ± 22 (n=20) and LaJolla ¹⁴³Nd/¹⁴⁴Nd ratio 0.512863 ± 08 (n=12)). Total procedural blank levels (blank associated with the digestion in Teflon beakers, ion exchange and instrument) were <25 pg of Nd and are considered negligible. Strontium samples were spiked with a ⁸⁴Sr enriched spike and analyzed on a Thermo Scientific TRITON TIMS 30 17 using a load of purified sample mixed with tantalum activator on a single rhenium filament. Two hundred 8s integrations were recorded in multi-collector static mode, applying rotating gain compensation. Measured ⁸⁷Sr intensities were corrected for Rb interference assuming 87 Rb/ 85 Rb = 0.38600 and ratios were calculated using the exponential fractionation law and 88 Sr/ 86 Sr = 8.375209. The external precision for ⁸⁷Sr/⁸⁶Sr estimated from analyzing NBS SRM 987 standard was ±16 ppm for large samples (0.710221 ± 11 ; 200 ng loads; n=16) and \pm 30 ppm for small samples (0.710222 \pm 21; 10-50 ng loads n=12). An accuracy correction was applied to the ⁸⁷Sr/⁸⁶Sr ratios corresponding to a ⁸⁷Sr/⁸⁶Sr ratio of 0.710245 for NBS SRM 987 40 23 42 24 standard. Total procedural blank levels were <80 pg of Sr and are considered negligible. Each preparation batch of Nd and Sr from small dust samples was checked using the basaltic rock, BCR-2,

certified reference material. Preparation and analysis of 150 to 600 µg aliquots of BCR-2 gave a concentration precision of ±10% and with literature values highly comparable isotopic results (⁸⁷Sr/⁸⁶Sr -50 29 0.705011 ±22).

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COMMENT: RESULTS ON FINE AND BULK FRACTIONS OF BUNGER HILLS SAMPLES

Bunger Hills potential source area (PSA) samples were size-segregated to account for Sr fractionation with particle size. With On the whole, the fine Bunger Hills PSA samples have relatively high⁸⁷Sr/⁸⁶Sr ratios (0.720372<⁸⁷Sr/⁸⁶Sr<0.761802 and -19.7<ε_Nd(0)<-21.6) compared to the bulk samples $(0.722543 < {}^{87}Sr / {}^{86}Sr < 0.730972$ and $-23 < \epsilon_{Nd}(0) < -24.3$; fig. 2). The ${}^{87}Sr / {}^{86}Sr$ variations with particle size in the Bunger Hills samples are consistent with previous studies of size-dependent fractionation, finer fractions having a higher Rb/Sr and thus a higher ⁸⁷Sr/⁸⁶Sr (Andersson et al., 1994; Winton et al., 2016a). The Δ87Sr/86Sr is ~0.028310 units, which is larger than the ⁸⁷Sr/⁸⁶Sr increase of ~0.0028 units observed between 63 µm and 2 µm of dust particles reported by Gaiero (2007)and 0.00115 units between 63 µm and 10 µm observed by Winton et al. (2016a). Two samples do not fractionate with particle size (BHO95 and BHO98). This could be related to different size fractions originating from different sources. There is also a size effect on the ¹⁴³Nd/¹⁴⁴Nd isotopes with an increase of around 3 $\varepsilon_{Nd}(0)$ units for the finer particle size, although this fractionation is less pronounced than that of the ⁸⁷Sr/⁸⁶Sr fractionation. Most studies found that 143 Nd/ 144 Nd in dust is independent of particle size. Fine (<5 μ m) and bulk Sr and Nd isotope ratios reported in Delmonte et al. (2004); Grousset et al. (1992) also show that ⁸⁷Sr/⁸⁶Sr are greater in fine particle size while $\varepsilon_{Nd}(0)$ variation with particle size has no clear trend. However, similar to this study, Yokoo et al. (2004) reports Nd isotope ratios in loess were higher in finer grained samples.

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Supplementary Table S1: Bunger Hills samples

SAMPLE CODE	LOCALITY	AGE						
BHO86	Kashalot Island	21900±3400	yr BP					
BHO96	Ostronaya Bay	4800±700	yr BP					
BHO78	Thomas island	7370±90	¹⁴ C yr BP					
BHO98	Krylatyy inlet	7110±130	¹⁴ C yr BP					
вно95	Ostronaya bay	6370	¹⁴ C yr BP					
BHO65+66	L.Dolgoe	11000±1200	yr BP					
BHO82/83	Thomas island	5780±80	¹⁴ C yr BP					

5780±80

			· · · ·			-	-	r –	-	-	1	-	1	,		-		r —	r	-	-		
[Sr] ppm		140	110		46		'				,	,	,	1	,	ı		160	940		860		120
⁸⁾ 2σ*10 ⁶		27*	54*		21	11	11	12*	15*	27*	11	11	11	11	11	11		21	21		11		21
		+1	+1		+1	+1	+1	+1	+1	+1	+1	+1	+1	+1	+1	+1		+1	+1		+1		+1
^{f) 87} Sr/ ⁸⁶ Sr corrected		0.712386	0.710378		0.715777	0.712609	0.711081	0.713126	0.716941	0.717059	0.716970	0.715899	0.708436	0.708446	0.739869	0.770459		0.718544	0.712162		0.730972		0.748524
^{e)} 2σ _{mean} *10 ⁶		27	54		19	07	60	12	15	27	10	60	60	10	11	60		08	12		05		20
		+1	+1		+1	+1	+1	+1	+1	+1	+	+1	+1	+1	+1	+1		+1	+1		+1		+1
⁸⁷ Sr/ ⁸⁶ Sr		0.712386	0.710378		0.715777	0.712609	0.711081	0.713126	0.716941	0.717059	0.716970	0.715899	0.708413	0.708423	0.739869	0.770459		0.718521	0.712139		0.730949		0.748501
^{d)} 2σ		0.4	1.3*		0.4	0.2	0.2	0.2	0.2	0.2		0.2	0.2	0.2	0.2	0.2		0.4	5.7*		0.2		0.5*
		+1	+1		+1	+1	+1	+1	+1	+1	0	+1	+1	+1	+1	+1		+1	+1		+1		+1
^{c)} ε _{Nd} (0)		-7.3	-8.1		-8.5	-9.6	-9.5	-9.2	-9.2	-9.4		-9.2	-14.4	-14.4	-6.5	-2.2		-9.8	-6.0		-24.3		-19.7
^{b)} 2σ _{mean} *10 ⁶		15	65		15	01	07	05	07	07		60	07	06	08	04		15	290		07		23
		+1	+1		+1	+1	+1	+1	+1	+1		+1	+1	+1	+1	+1		+1	+1		+1		+1
¹⁴³ Nd/ ¹⁴⁴ Nd		0.512264	0.512222		0.512204	0.512149	0.512151	0.512168	0.512165	0.512154		0.512164	0.511899	0.511900	0.512304	0.512524		0.512134	0.512350		0.511390		0.511628
(mqq) ±(°		2	1		1	4	<1	<1	<1	<1 		-1	-1	<1	<1	<1		2	-		4		æ
[PN]		18	11		7	37	26	31	47	47		43	15	18	140	57		20	10		21		30
fraction		Bulk	Bulk		Bulk	<150	<150	<150	<150	<150		<150	<150	<150	<150	<150		<5	5		Bulk		<5
type		ICD	ICD		nunatak	regolith	regolith	regolith	regolith	regolith		regolith	regolith	regolith	station	station		moraine	moraine		Fossil	beach ridge	Fossil beach
Sample	Camp Maudheimvidda	DML#1	DML#2	DML area	Innen Eoreilavaon	Basen 1 NO	Basen 2 NW	Basen 6 N	Basen Aboa 1	Basen Aboa 2	Basen Aboa 2 duplicate	Basen Aboa 3	Plogen toppen 1	Plogen toppen 2	Svea 1	Svea 2	Taylor Glacier	TG 1	TG 9	Bunger Hills	BHO 83		BHO 83

ridge	6 Fossil Bulk 20 <1 0.511453 beach ridge	6 Fossil <5 60 6 0.511560 beach ridge	8 Fossil Bulk 62 <1 0.511393 beach ridge	8 Fossil <5 50 5 0.511626 beach ridge	8 Fosšil Bulk 37 <1 0.511461 beach ridge	8 Fossil <5 50 5 0.511530 beach ridge	5 Fossil Bulk 22 <1 0.511424 beach ridge	5 Fossil <5 60 6 0.511542 beach ridge	5+66 Fossil Bulk 29 <1 0.511450 beach ridge	5+66 Fossil <5 80 8 0.511617 beach ridge	6 Fosšil Bulk 38 <1 0.511455 beach ridge	6 Fossil <5 100 10 0.511616
	+ 07	+ 10	± 02	+ 19	± 04	± 07	+	± 22	+ 04	60 +	+ 04	+ 06
	-23.1	- 21.0	-24.3	-19.7	-23.0	-21.6	-23.7	-21.4	-23.2	-19.9	-23.1	-19.9
	0.2	£ 0.4	F 0.2	0.4	0.2	t 0.2	0.2	0.4	E 0.2	F 0.2	t 0.2	0.2
	0.722871	0.760460	0.729460	0.758841	0.722520	0.721545	0.725008	0.720349	0.725945	0.761779	0.728599	0.749795
	+ 05	± 23	+ 04	60 +	- 00 +	90	+ 05	+ 05	+ 04	+ 13	+ 05	+
	0.722894	0.760483	0.729483	0.758864	0.722543	0.721568	0.725031	0.720372	0.725968	0.761802	0.728622	0.749818
	+ 11	± 23*	+ 11	± 21	+ 11	± 21	+ 11	± 21	+ 11	± 21	+ 11	± 21
	360	170	280	130	340	380	320	520	310	160	280	240

Supplementary Table S2: Nd and Sr concentrations[in parentheses] and isotopic composition of ice core/snow samples and potential source area samples analyzed in this study.

 ICD= Ice Core Dust sample ^{b)}Error due to difficulty of measuring small sample masses, estimated by repeat weighting BCR-2 standards (~0.3 mg). ^{b)}Internal precision, 2 standard errors of the mean. ^{b)}Internal precision, 2 standard errors of the mean. ^{c)}Nd isotopic ratios expressed as epsilon units ε_{Nd}(0) = [(¹⁴³Nd/)¹⁴⁴Nd)sample/(¹⁴³Nd/)¹⁴⁴Nd)CHUR-1]x10⁴; ^{c)}Uncertainty estimates based upon external precision for standard runs. Internal precision is used if it exceeds the external. ^{f)}An accuracy correction was applied to Taylor Glacier and Bunger Hills samples, ^{f)}An accuracy correction was applied to Taylor Glacier and Bunger Hills samples, ^{f)}An accuracy to the NBS 987 standard was 0.710222 ±22 (n=12). Corrected to a NSB 987 ⁸⁷Sr/⁸⁶Sr ratio of 0.710245. ^{g)}Uncertainty estimates based upon external precision for standard runs. * Internal precision is used if it exceeds the external. 		http://mc.manuscriptcentral.com/holocene
ICD= Ice ^{a)} Error d ^{b)} Interná ^{c)} Nd isot CHUR, c ^{c)} Nd isot ^{d)} Uncert ^{e)} Interna ^{e)} Interna ^{g)} Uncert		
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