Multiple sources of soluble atmospheric iron to Antarctic waters

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Iron, dust, aerosol, black carbon, soluble iron, dissolved iron, Roosevelt Island, Ross Sea, Antarctica
Key points

1. Dust deposition at Roosevelt Island occurs primarily during spring-summer.
2. Local and remote sources contribute to Roosevelt Island dust deposition at present.
3. Dissolved Fe in Antarctic snow appears to be related to both dust and black carbon deposition.
4. Exceptionally high black carbon, dust and dissolved Fe concentrations in 2011/2012 austral summer.
5. Background fractional Fe solubility averaged 0.7% at Roosevelt Island between 2010 and 2012.
The Ross Sea, Antarctica, is a highly productive region of the Southern Ocean. Significant new sources of iron (Fe) are required to sustain phytoplankton blooms in the austral summer. Atmospheric deposition is one potential source. The fractional solubility of Fe is an important variable determining Fe availability for biological uptake. To constrain aerosol Fe inputs to the Ross Sea region, fractional solubility of Fe was analyzed in a snow pit from Roosevelt Island, eastern Ross Sea. In addition, aluminum, dust, and refractory black carbon (rBC) concentrations were analyzed, to determine the contribution of mineral dust and combustion sources to the supply of aerosol Fe. We estimate exceptionally high dissolved Fe (dFe) flux of $1.2 \times 10^{-6}$ g m$^{-2}$ y$^{-1}$ and total dissolvable Fe (TDFe) flux of $140 \times 10^{-6}$ g m$^{-2}$ y$^{-1}$ for 2011/2012. Deposition of dust, Fe, Al, and rBC occurs primarily during spring-summer. The observed background fractional Fe solubility of $\sim$0.7 % is consistent with a mineral dust source. Radiogenic isotopic ratios and particle size distribution of dust indicates that the site is influenced by local and remote sources. In 2011/2012 summer, relatively high dFe concentrations paralleled both mineral dust and rBC deposition. Around half of the annual aerosol Fe deposition occurred in the austral summer phytoplankton growth season; however the fractional Fe solubility was low. Our results suggest that the seasonality of dFe deposition can vary, and should be considered on longer glacial-interglacial timescales.
1. Introduction

The Ross Sea is the most biologically productive continental shelf region around Antarctica and an important region for atmospheric CO$_2$ sequestration [Arrigo and van Dijken, 2007; Arrigo et al., 1998; Arrigo et al., 2008; Smith and Gordon, 1997]. Each summer the Ross Sea blooms with phytoplankton [e.g. Smith and Gordon, 1997]. Two distinct blooms occur, with each characterized by differences in location, timing, water stratification and species [Arrigo and van Dijken, 2004]. The first bloom occurs in the central Ross Sea polynya north of the Ross Ice Shelf and is dominated by *Phaeocystis Antarctica*. This bloom develops in late October-November when sea ice is still present and terminates as early as mid-December. The second bloom occurs in the southwestern (SW) Ross Sea, is dominated by diatoms, and is much smaller in areal extent. In the Southern Ocean, environmental factors responsible for controlling the rates of phytoplankton production include: grazing [e.g. Banse, 1991], temperature [e.g. Bunt, 1963], light availability [e.g. Mitchell et al., 1991], water stratification [Tagliabue and Arrigo, 2006], sea ice extent [Smith and Nelson, 1986], trace metal availability [e.g. Fe; Martin et al., 1990; Sedwick and DiTullio, 1997; Sedwick et al., 2011], or a combination of these [e.g. Arrigo et al., 2000]. Due to the Ross Sea’s capacity to support intense early summer phytoplankton blooms, the seasonally Fe-limited high-nutrient high-chlorophyll (HNHC) regime of the Ross Sea in summer is distinct from the chronically Fe-limited high-nutrient low-chlorophyll (HNLC) offshore waters of the Southern Ocean [Tagliabue and Arrigo, 2005].

In-situ oceanic iron (Fe)-fertilization experiments have demonstrated a response of the ecosystem to relatively small additions of dissolved Fe (dFe) in the Ross Sea and
other sections of the Southern Ocean [Coale et al., 2003; Martin et al., 1990]. The seasonally Fe-limited HNHC regime in the Ross Sea requires continuous new inputs of dissolved Fe (dFe) to sustain these phytoplankton blooms [e.g. Fitzwater et al., 2000; Sedwick et al., 2011]. Inputs of new Fe to surface waters in the Ross Sea can occur through upwelling of deep waters, transport from continental margins by ocean currents, melt from sea-ice, icebergs and ice shelves, and atmospheric aerosol deposition [Atkins and Dunbar, 2009; de Jong et al., 2013; Gerringa et al., in press; Jacobs et al., 1970; Marsay et al., 2014; Sedwick and DiTullio, 1997; Sedwick et al., 2011; Winton et al., 2014].

In terms of an atmospheric source, the deposition of aerosol Fe to remote Southern Ocean surface waters is extremely low [e.g. Winton et al., 2015], and has been investigated in relation to the distribution and transport of mineral dust. Little aerosol Fe data exists for the Ross Sea region. Total dissolvable Fe (TDFe) aerosol fluxes to the Ross Sea from long-range transport dust have been estimated to range between 0.007 to 0.1 mg m\(^{-2}\) yr\(^{-1}\) [Edwards and Sedwick, 2001]. Measurements of local soluble Fe from local dust on sea ice in McMurdo Sound [Winton et al., 2014] show that this local Fe source is much larger (2-7 mg m\(^{-2}\) yr\(^{-1}\)) than that supplied by long-range transport sources [e.g. Wagener et al., 2008]. The supplied Fe, however, only contributes to phytoplankton blooms within the localized McMurdo Sound region. On longer timescales, enhanced glacial atmospheric Fe deposition has been linked to higher rates of Southern Ocean primary productivity as observed from recent Southern Ocean marine sediment studies [e.g. Martinez-García et al., 2014]. Furthermore, Antarctic ice core records associate higher dust deposition rates with Fe supply during glacial stages [Conway et al., 2015; Edwards et al., 2006; Gaspari et
Dust deposition to Antarctica can be sourced from both remote and local sources [e.g. Bory et al., 2010; Delmonte et al., 2013]. Dust provenance can be determined from the $^{87}$Sr/$^{86}$Sr and $^{143}$Nd/$^{144}$Nd radiogenic isotope composition of dust in snow and ice by comparison with potential source areas (PSAs) [Grousset and Biscaye, 2005]. This is because the Sr and Nd isotopic composition of dust are primarily related to lithology and the geologic history of parent materials [Faure, 1986].

Aerosol Fe bioavailability information is required to constrain the biogeochemical impact of present and past atmospheric Fe variability. Reported values of fractional aerosol Fe solubility range from 0.01 to 90 % [Baker and Croot, 2010; Bowie et al., 2009; Edwards and Sedwick, 2001; Heimburger et al., 2013; Mahowald et al., 2005]. This large range may reflect differences in mineral dust concentrations, particle size, atmospheric weathering and aerosol leaching methods [Aguilar-Islas et al., 2010; Baker and Jickells, 2006; Bonnet and Guieu, 2004; Buck et al., 2006; Chen and Siefert, 2003; Spokes and Jickells, 1995; Trapp et al., 2010; Zhuang et al., 1990; Zhuang et al., 1992]. An alternative hypothesis for the observed solubility range is that it results from a mixture of aerosol Fe sources with different mineralogy and fractional Fe solubility [Sholkovitz et al., 2012]. Sholkovitz et al. [2012] showed that global scale fractional aerosol Fe solubility displays an inverse hyperbolic relationship with the total Fe concentration. This relationship is consistent with a low fractional Fe solubility for mineral dust (~1-2 %) and the presence of other soluble Fe sources, such as those originating from biomass burning and oil combustion with higher fractional Fe solubility [Chuang et al., 2005; Guieu et al., 2005; Ito, 2011; Kumar et al., 2010; Luo et al., 2008; Paris et al., 2010; Sedwick et al., 2007;
Estimates of fractional Fe solubility from fire combustion range from 1 to 60% and may vary in relationship to biomass and fire characteristics as well as that of the underlying terrain [Guieu et al., 2005; Ito, 2011; Kumar et al., 2010; Luo et al., 2008; Paris et al., 2010]. Over the time period investigated (2010-2012) in this study, Southern Hemisphere biomass burning emissions primarily occurred in the intertropical convergence zone (ITCZ) of Africa, Australia and South America [Giglio et al., 2013]. Of these regions, Australia is the closest to the Ross Sea. Biomass burning constitutes a large source of austral dry-season aerosol emissions over northern Australia, and episodic austral summer wildfires in southern and eastern Australia [e.g. Meyer et al., 2008]. Refractory black carbon aerosols (rBC) are emitted by biomass burning and fossil fuels [Reid et al., 2005]. In the Southern Hemisphere, rBC emissions are primarily from biomass burning [Giglio et al., 2013] and can be used as a proxy for the long-range transport of biomass burning aerosols to Antarctica. Bisiaux et al. [2012] investigated ice core records of rBC deposition to West Antarctica and found annual deposition consistent with austral dry season biomass burning. Thus, Fe associated with rBC may provide information with respect to biomass burning inputs of Fe to the Southern Ocean in this study.

An intermediate depth ice core (764 m) was recently drilled in the framework of the Roosevelt Island Climate Evolution (RICE) project Roosevelt Island (79.36086°S, 161.64600°W; Fig. 1), located on the opposite side of the Ross Sea to McMurdo Sound. Many aspects of atmospheric Fe deposition in marginal areas of Antarctica remain poorly known. In this respect, Roosevelt Island represents an ideal location to investigate the timing and source(s) of soluble Fe deposition to the eastern Ross Sea region. This study investigated the present-day seasonality of fractional Fe solubility,
and potential sources from mineral dust and refractory black carbon (rBC) at Roosevelt Island.

2. Methods

A detailed description of the snow sampling procedure, dust, rBC, trace metal solubility and Sr and Nd isotopic analysis can be found in Supplementary Information. Briefly, four parallel profiles of ultra-clean snow samples were collected at 3 cm resolution from a 1.5 m snow pit. The snow pit was located in a designated clean sector at Roosevelt Island during the 2012/13 RICE ice core drilling campaign. Parallel profile samples were measured for particle size and concentration (Coulter Counter [Delmonte et al., 2002]), rBC concentration (single particle intracavity laser-induced incandescence photometer (SP2 [Sterle et al., 2013]), $\delta^{18}$O isotopes (high-resolution laser absorption spectroscopy; Los Gatos Research Liquid-Water Isotope Analyzer), and dissolved and total dissolvable concentrations of sulfur (S), Fe and aluminum (Al). Dissolved Fe and Al fractions were determined by filtering a 10 mL aliquot of snow melt through a 0.2 µm filter [Lannuzel et al., 2008]. The remaining (unfiltered) sample was leached with 1 % HCl (ultra-pure) for three months to determine the total dissolvable trace metal fraction following Edwards et al. [2006] who showed that a three-month leaching period was required for TDFe concentrations to plateau. Total dissolvable solutions and dissolved leachates were analyzed by high resolution inductively coupled plasma mass spectrometry (HR-ICP-MS). HR-ICP-MS operating conditions and blank elemental levels are reported in Tables S1 and S2. Stringent trace metal practices were employed at all stages of sample processing and analysis. Sampling vials and filtration gear was rigorously acid cleaned prior to use.
following GEOTRACES protocols\textsuperscript{1}. Fractional Fe solubility was calculated using equation 1:

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\text{Fractional Fe solubility} = \frac{dFe}{TDFe} \times 100 \tag{equation 1}
\]

where \(dFe\) is the soluble or dissolved Fe fraction and \(TDFe\) is the total dissolvable fraction.

A reflected light optical microscope (BX51M) was used to confirm the presence of large particles (>10 µm) detected by Coulter Counter (Fig. S1). Adjacent to the snow pit, a large volume of surface snow was collected for Sr and Nd isotopic ratios and concentrations of dust for provenance attribution (Supplementary Information 1). Dust particles were separated from the snow, and two samples (RI1-2) were spiked, digested, separated from interfering elements and analyzed by TRITON Thermal Ionization Mass Spectrometer (TIMS) following Delmonte et al. [2008].

3. Results

3.1. Snow pit dating

The dating of the RICE snow pit was based on water stable isotope ratios \(\delta^{18}O\) and on concentrations of total dissolved non-sea-salt sulfur (nss-S) (Fig. 2) following Tuohy et al. [2015]. Non-sea-salt sulfur shows sharp, well defined summer peaks. The \(\delta^{18}O\) measurements in Antarctic snow and ice often reflect seasonal cycles in temperature \cite{Dansgaard, Johnsen et al.}. However, the upper snow pack at Roosevelt

\textsuperscript{1}http://www.geotraces.org/images/stories/documents/intercalibration/Cookbook.pdf
Island contains $\delta^{18}$O variability within seasons which reflects individual storm events [Tuohy et al., 2015]. Summers were determined as 1 January and were positioned where nss-S peaks aligned with peaks or shoulders of peaks in the $\delta^{18}$O record. Winters were determined as 1 July where a nss-S trough aligned with a $\delta^{18}$O trough. Annual cycle counting of nss-S layers shows the snow pit spans a two year period from summer 2012/13 to summer 2010/11 (Fig. 2), with an age uncertainty of ±0.5 year at the base of the snow pit. The snow accumulation rate of the snow pit is estimated to be 0.33 m yr$^{-1}$ water equivalent (w.e; estimated from nss-S concentration peaks between January 2011 and January 2012), assuming the average annual distribution of snowfall is uniform. These rates are consistent with an accumulation rate of ~0.27 m yr$^{-1}$ ice equivalent from snow stake measurements ~700 m from the snow pit site at Roosevelt Island between 2010 and 2012 [H. Conway, unpublished data, 2015].

To constrain the seasonality of nss-S with known sulfate emissions from phytoplankton [e.g. Rhodes et al., 2009], high-resolution chlorophyll-a satellite data were downloaded for the Ross Sea region (area defined by 161.175° to -151.125°, -70.078° to -80.0°) (Fig. 2k-l, SeaWiFS;  http://disc.sci.gsfc.nasa.gov/giovanni). The temporal development of Ross Sea polynya phytoplankton bloom (“bloom 2,” see Section 1.1) was used due to the dominant easterly transport within the Ross Sea [Sinclair et al., 2010]. Comparison of chlorophyll-a concentration data over the Ross Sea region shows that the bloom develops in November and declines in December in both 2010 and 2011 (Fig. 2l). In addition, a hoar layer (with a low snow density of 0.20 g m$^{-3}$) at ~55 cm depth in the snow pit was compared to nss-S summer maxima
3.2. Seasonal dust variability and particle size distribution

The Roosevelt Island snow pit records inter-annual variability in dust deposition over a two year period. The dust concentration in the snow pit ranges from 2 to 41 ng g\(^{-1}\) for the 1-5 µm fraction and 2 to 58 ng g\(^{-1}\) for the 1-30 µm particle size fraction. We note that two samples have low dust concentrations (<4 ng g\(^{-1}\)) close to exposure blank levels (~2 ng g\(^{-1}\)). Dust deposition in the snow pit is episodic. The dust record displays two maxima corresponding to summer-spring elevated levels of nss-S in January 2012 and January 2011 (Figs. 2g-h and 2k). Lower dust concentrations are observed in winter, however, an episodic dust event is captured during winter 2012. Seasonal dust deposition at Roosevelt Island, primarily during spring-summer, is coherent with an earlier snow pit study from the site that showed higher Al/Na ratios [Cohen, 2013] in correspondence with summer peaks of the \(\delta^{18}O\) record. At GV7, a peripheral site located on the South Pacific margin of the East Antarctic ice sheet in Northern Victoria Land (Fig. 1), dust deposition from a snow pit clearly shows spring-summer maxima and winter minima (B. Delmonte, unpublished data, 2015).

Dust deposition at other locations in Antarctica also exhibits seasonal variability with a maximum in summer (e.g. Berkner Island [Bory et al., 2010], Windless Bight, McMurdo Sound [Dunbar et al., 2009] and South Pole [Legrand and Kirchner, 1988]).

The particle size distribution of dust particles measured from 1 to 30 µm does not approximate a lognormal particle size distribution. Rather, the size distribution of dust
particles approaches the theoretical dust emission particle size distribution, parameterized by [Kok, 2011a], for particles between 1 and 5 µm in diameter in samples with elevated dust concentrations in both spring-summer periods. The particle size can be seen in some of these highly concentrated spring-summer samples in Fig. 3 and compared to the lognormal particle size distribution of long-range transport dust to the East Antarctic Plateau. For very low concentration samples, the size distribution of microparticles typically does not show a clear mode. For this reason, other size indicators (fine particle percentage (FPP) and the coarse particle percentage (CPP) parameters) were introduced to study long-term dust size variations in central Antarctica [Delmonte et al., 2002]. In this case, longer records of particle size distributions are required to investigate seasonality in particle size distribution at Roosevelt Island. Large particles with an equivalent spherical diameter between 5 and 10 µm were also observed (Fig. 3).

3.3. Isotopic composition of dust

The Sr and Nd isotopic composition of the two Roosevelt Island dust samples (RI1-2) are $0.7122 < \frac{^{87}\text{Sr}}{^{86}\text{Sr}} < 0.7156$ and $\varepsilon_{\text{Nd}}(0) = -9.6$. These data are reported in Table 1 and Figs. 4-5 with additional isotopic data from Antarctic PSAs. PSAs in Fig. 4 are grouped by geographic location.

3.4. Microscope observations

Mineral dust and volcanic glass (~10 µm in diameter) was present in Roosevelt Island samples, but only a few qualitative optical observations were performed on selected samples from Roosevelt Island (Fig. S1). Volcanic glass is a common component of background dust in Antarctica [Narcisi et al., 2005]. These microscopic observations
were useful for confirming the presence of particles having a diameter between 5 and 10 µm, thus a) eliminating the possibility of particulate contamination of sampling equipment and laboratory procedures, and b) validating Coulter Counter observations of relatively coarse particles in the samples.

3.5. Refractory black carbon

Deposition of rBC in the Roosevelt Island snow pit is highest during the 2011/2012 summer (Fig. 2d). Austral winter background values in 2011 average ~100 pg g\(^{-1}\), and the summer rBC concentrations reached four times above background levels in January 2011, and seven times the background levels in January 2012 (up to ~1.5 ng g\(^{-1}\)). The 2011/2012 summer peak in rBC was exceptionally high in comparison to rBC concentrations reported for the West Antarctic Ice Sheet (WAIS) Divide ice core (geometric mean ~80 pg g\(^{-1}\) [Bisiaux et al., 2012]). Additional smaller peaks of rBC concentrations were also found in late winter-spring in 2012 at Roosevelt Island, similar to the timing of rBC deposition at WAIS Divide.

3.6. Dissolved and total dissolvable iron and aluminum

With the exception of one layer (61.5 - 64.5 cm depth), TDFe and TDAl concentrations ranged from 0.01 ± 0.001 to 1.9 ± 0.1 ng g\(^{-1}\) and 0.01 ± 0.001 to 1.8 ± 0.1 ng g\(^{-1}\) respectively. The higher concentrations from the 61.5 - 64.5 cm layer were 5.4 ± 0.1 ng g\(^{-1}\) of TDFe and 6.7 ± 0.1 ng g\(^{-1}\) of TDAl. This depth also displayed high rBC concentrations of 1630 pg g\(^{-1}\). TDFe and TDAl co-vary in the snow pit (Fig. 2e-f). A strong relationship was found between the two trace metals (r\(^2\)=0.86; Fig. S3a), with maxima occurring in the summer at the same time as rBC and dust in 2011/2012 but not in 2010/2011.
Dissolved Fe concentrations were lower than TDFe, and ranged between 0.001 ± 0.0001 and 0.2 ± 0.001 ng g⁻¹ (Fig. 2b). Although the exposure blank dFe and dAl concentrations are exceptionally low (Table S2), the dFe and dAl blank background is similar to some of the snow pit samples. Due to the extremely low dissolved concentration of the snow pit samples, it is possible that the uncertainty on the blank concentrations could be responsible for some of the temporal variation. Nevertheless, dFe peaked four times above the recorded dFe background of 0.02 ng g⁻¹. Two of these maxima occurred in summer 2011/2012 at the same time as peaks in TDFe, TDAI, dust and rBC concentrations at Roosevelt Island, while the other two peaks occur in the winter when dust rBC, TDFe and TDAI are low (Fig. 2).

4. Discussion

4.1. Atmospheric dust deposition and provenance

Ice cores from the high-elevation East Antarctic Plateau represent a unique archive to investigate long-range dust transport [e.g. Delmonte et al., 2004]. In contrast, around the margin of Antarctica, the occurrence of sparse ice-free areas can represent a non-negligible dust source to the local atmosphere [Bertler et al., 2005; Delmonte et al., 2013]. At these sites, the interpretation of ice core dust records is related to mesoscale dust entrainment, advection and deposition driven by regional circulation. Roosevelt Island is a low-altitude location (550 m.a.s.l.), well outside the high-elevation polar plateau. Large expanses of ice-free areas occur around the margins of the Ross Sea (Fig. 1), and thus the dust cycle there is widely different from the East Antarctic Plateau in terms of abundance and origin.
Known dust sources to the western Ross Sea include the Transantarctic Mountains, Terra Nova Bay and McMurdo Sound [Atkins and Dunbar, 2009; Barrett et al., 1983; Dunbar et al., 2009]. Of these, southern McMurdo Sound is known to be the dustiest location in Antarctica [Chewings et al., 2014; Winton et al., 2014]. In terms of the western Ross Sea, Sr and Nd isotopic data is only available for McMurdo Sound and some areas in the Transantarctic Mountains [Cook et al., 2013; and references therein; Delmonte et al., 2004; Delmonte et al., 2013; Winton et al., 2014], although an expansion of the existing isotopic catalogue of Antarctic PSAs is in preparation [Blakowski et al., submitted] in an effort to deepen the current understanding of Holocene dust input to the periphery of the EAIS. The isotopic signature of these PSAs in the western Ross Sea is plotted in Fig. 4 and compared to the signature of Roosevelt Island dust measured in this study. The isotopic composition of one sample representing modern summer snow at Roosevelt Island is markedly different from the fingerprint of McMurdo Sound (Table 1, Fig. 4), i.e., Roosevelt Island dust (0.7122< $^{87}$Sr/$^{86}$Sr < 0.7156 and $\varepsilon_{Nd}(0) = -9.6$) has a more radiogenic $^{87}$Sr/$^{86}$Sr signature and less radiogenic $\varepsilon_{Nd}(0)$ signature compared to the volcanic sediments from McMurdo Sound [Winton et al., 2014]. Therefore, the isotopic data alone suggests that dust deposited on Roosevelt Island cannot be solely sourced from McMurdo Sound.

In addition, the dust concentrations at Roosevelt Island are lower and the particle size distribution is finer than the dusty McMurdo Sound [e.g. Chewings et al., 2014]. An annual dust flux at Roosevelt Island was estimated for the calendar year 2011 (the beginning of the calendar year was taken at each nss-S peak), and the water equivalent depth was calculated from the snow pit density measurements (Fig. S2).
The estimated dust flux of 6 mg m\(^{-2}\) yr\(^{-1}\) for the 1-5 µm particle fraction (8 mg m\(^{-2}\) yr\(^{-1}\) for the bulk 1-30 µm particle fraction) at Roosevelt Island in this study is seven times greater than the equivalent pre-industrial East Antarctic dust flux of 0.19-0.75 mg m\(^{-2}\) yr\(^{-1}\) [Delmonte et al., 2013], and also greater than modern modelled and empirical fluxes of dust to the Southern Ocean (0.3 to 2.5 mg m\(^{-2}\) d\(^{-1}\)) [Duce et al., 1991; Mahowald et al., 2005; Wagener et al., 2008]. The bulk 1-30 µm dust flux at Roosevelt Island is double that found in the WAIS Divide ice core: ~4 mg m\(^{-2}\) yr\(^{-1}\) over the past two millennia [Koffman et al., 2014]. By comparison, the dust fluxes for Roosevelt Island, East Antarctica and the Southern Ocean are orders of magnitude lower than the dust flux of the McMurdo Sound fine fraction reported by Winton et al. [2014], i.e. 460 mg m\(^{-2}\) yr\(^{-1}\) for the <10 µm fraction.

Larger dust particles (between 5-10 µm) represent a non-negligible contribution to the total dust input at Roosevelt Island. The particle size distribution of the dust at Roosevelt Island (Fig. 3) is coarser than the distally-sourced dust deposited on the East Antarctica Plateau. For example, at Dome C and other sites located in central Antarctica [Delmonte et al., 2002], dust particles are mostly within the size interval 1-5 µm in diameter. The only large particles deposited on the central East Antarctic Plateau have been previously associated with tephra layers [Basile et al., 2001; Narcisi et al., 2005]. Furthermore, the particle size distribution curve can provide additional information about the proximity to the dust source [Kok, 2011a; b]. Dust follows a lognormal distribution in ice cores from regions that primarily receive long-range transport dust to the East Antarctic Plateau or central Greenland [Delmonte et al., 2002; Ruth, 2002; Steffensen, 1997], for example, Dome C (Fig. 3). However, recent work by Kok [2011a; 2011b] has shown that dust near the emission source does
not necessarily follow a lognormal distribution. The spring-summer snow pit samples that approach the theoretical dust emission particle size distribution, parameterized by [Kok, 2011a], at Roosevelt Island and other sites near the margin of the ice sheet (e.g. WAIS Divide [Koffman et al., 2014]), suggest that dust is not travelling as far as long-range transported dust reaching the East Antarctic Plateau which has a lognormal distribution [Delmonte et al., 2002] (Fig. 3). These spring-summer samples that approach the theoretical dust emission distribution are indicative of local dust sources.

Compared to the East Antarctic Plateau, the higher dust flux, particle size distribution approaching the theoretical dust emission distribution and the presence of large particles (<10 µm) observed under the optical microscope and SEM suggests a local Antarctic dust input to Roosevelt Island. This local Antarctic dust input has a distinctly different isotopic fingerprint to modern McMurdo Sound dust emissions (Fig. 4).

The Sr and Nd isotopic signature of Roosevelt Island dust in Figs. 4 and 5 suggest that dust deposited at Roosevelt Island during summer 2012/2013 could be a mixture of at least two local sources. One end member is likely volcanic material with relatively high radiogenic Nd and less radiogenic Sr. The other end member is likely to be much older, with more radiogenic Sr and less radiogenic Nd. Given existing Sr and Nd isotopic data, the sources to Roosevelt Island include eastern Australia, parts of coastal Marie Byrd Land, and most of Victoria Land. Given the relatively high dust flux, the presence of large particles and particle size distribution approaching the theoretical dust emission distribution we first consider local dust sources upwind of Roosevelt Island. The fetch area of five-day air mass back trajectories for 2011 and 2012 arriving at Roosevelt Island are predominately from West Antarctica, the Pacific
Ocean sector of Antarctica and Victoria Land [Tuohy et al., 2015]. Air mass back trajectories and background climatological circulation shows prevailing winds cross exposed rock in Marie Byrd Land, West Antarctica before arriving at Roosevelt Island [Koffman et al., 2014; Tuohy et al., 2015]. Air masses can also arrive at Roosevelt Island via synoptic cyclonic circulation in the Ross Sea [Koffman et al., 2014; Neff and Bertler, 2015]. Therefore, two potential end member local dust sources for Roosevelt Island are West Antarctic volcanic material and Victoria Land Paleozoic rocks (i.e., the bedrock that comprises most of northern Victoria Land, effectively upwind of Roosevelt). Fig. 4 shows that the isotopic composition of Roosevelt Island dust falls between the isotopic fields of both these end members. Despite the isotopic similarity of Victoria Land Palaeozoic rocks and Roosevelt Island dust, we believe this West to East dust transport hypothesis very unlikely, because strong convective uplift would be necessary to uplift dust in the troposphere above the marine boundary layer, where dust is rapidly removed.

In terms of Victoria Land, previous studies have shown that although there is a high dust flux within the McMurdo Dry Valleys [Ayling and McGowan, 2006; Gillies et al., 2013; Lancaster et al., 2010; Selby et al., 1974], little dust exits the valleys [Winton et al., accepted]. The long exposure to katabatic winds has winnowed the surface sediment, resulting in a lack of dust and very fine sand-sized material over most of the valley floor [e.g. Delmonte et al., 2010; Selby et al., 1974]. These air masses do not travel high in the troposphere and when they encounter humidity from the ocean, the dust is scavenged. Although Bhattachan et al. [2015] suggest that dust from Taylor Valley in the McMurdo Dry Valleys could supply soluble iron to the wider Southern Ocean, other studies have discounted the McMurdo Dry Valleys as a
major dust (and dFe) source due to the limited transport in this region [Barrett et al., 1983; Bentley, 1979; Chewings et al., 2014; Winton et al., accepted]. Therefore, we suggest that dust sourced from Victoria Land is unlikely to be transported eastward to Roosevelt Island. While Victoria Land sources should not be completely discarded due to the large expanse of ice-free area and the isotopic signature Victoria Land Paleozoic rocks potentially acting as an end member dust source, we also consider local sources from Marie Byrd Land that are immediately upwind of Roosevelt Island.

Although Roosevelt Island is completely ice-covered, there are patchy ice-free areas (Rockefeller Mountains and Alexandra Mountains, and extensive volcanics e.g. the Executive Committee Range) along the north-westernmost part of Marie Byrd Land, on the King Edward VII Peninsula (Fig. 1b), that is adjacent to the eastern Ross Sea. According to Adams et al. [1995], the King Edward VII Peninsula is characterized by presence of three main rock units: i) a low-grade metasedimentary suite of late Ordovician age, correlated with the Swanson Formation of the Ford Ranges; ii) a granitoid suite correlated to the Byrd Coast granite and particularly developed in the Rockefeller Mountains, which includes monzogranites and syenogranites, and iii) the Alexandra Metamorphic Complex formed by migmatitic paragneiss. The Sr isotopic signature of these ice-free areas on the eastern margin of the Ross Sea is compared to Roosevelt Island dust in Fig. 5. Whole-rock Rb-Sr geochemistry of the Rockefeller and Alexandra Mountains of Edward VII Peninsula show $^{87}\text{Sr}^{86}\text{Sr}$ higher than about 0.721 (Swanson Formation) and higher than 0.712 (Byrd Coast Granite) (Fig. 5) while Nd isotopic data are not available [Adams et al., 1995]. On the contrary, volcanic rocks from Marie Byrd Land and other volcanic provinces show very unradiogenic Sr
McMurdo Sound and Marie Byrd Land volcanic rocks ($0.7026 < ^{87}\text{Sr}/^{86}\text{Sr} < 0.7032$ and $2.0 < \varepsilon_{\text{Nd}(0)} < 6.9$); Fig. 5) [Futa and Le Masurier, 1983; Hole and Le Masurier, 1994]. On the basis of Sr isotopic data and particle size distribution data indicating that dust at Roosevelt Island may be locally-sourced at present-day, we suggest that the summer 2012/2013 Roosevelt Island snow dust sample represents a mixture of local volcanic dust and crustal material likely deriving from the neighboring granites and metasedimentary rocks of western Marie Byrd Land and possibly Paleozoic rocks from Victoria Land (Figs. 4 and 5).

We also consider a remote Southern Hemispheric contribution of long-range transported dust to Roosevelt Island. Distally-derived larger particles could reach Roosevelt Island but are not transported further inland to the high elevation Antarctic interior. The Roosevelt Island isotopic signature is distinctly different to dust deposited on East Antarctica during the pre-industrial (1400 A.D. - 1800 A.D.; $^{87}\text{Sr}/^{86}\text{Sr}$ ranging between 0.707468 - 0.708468) and Holocene periods ($^{87}\text{Sr}/^{86}\text{Sr}$ ranging between 0.70769 - 0.711200 [Delmonte et al., 2013]), which suggests the eastern Ross Sea has a different dust source to the East Antarctica Plateau. Roosevelt Island falls within the Australian isotopic field, which is characterized by $^{87}\text{Sr}/^{86}\text{Sr}$ ratios ranging from 0.709 to 0.732 and $\varepsilon_{\text{Nd}(0)}$ between $-3$ and $-15$ [Delmonte et al., 2004; Revel-Rolland et al., 2006]. Revel-Rolland et al. [2006] and De Deckker et al. [2010] suggested that Australia could contribute to the dust input in central East Antarctica. Modelling studies of dust transport to Antarctica from Southern Hemispheric continents [Krinner et al., 2010] show that the annual mean concentration of dust in the eastern Ross Sea region of Antarctica is mostly represented by dust derived from Australia, in agreement with former modelling
studies [Albani et al., 2012; Li et al., 2008; McGowan and Clark, 2008]. We note, in addition, that the seasonality of Australian dust export to high southern latitudes occurs during spring and summer [Boyd et al., 2004; Mahowald et al., 2005], hence it is synchronous with the dust peaks we observed in the Roosevelt Island snow pit. Krinner et al. [2010] clearly showed that the concentration of Australian continental dust tracers is at a maximum at about 150 °W, close to Roosevelt Island. Although, dust appears to be mostly concentrated at higher tropospheric levels (about 6000 m) that are well above the altitude of the Roosevelt Island [Krinner et al., 2010]. Neff and Bertler [2015] and Tuohy et al. [2015] highlight the likelihood of air parcels transported to Roosevelt Island: both air mass back trajectories for the 2011/2012 summer season and the average seasonal air mass back forward trajectories from Southern Hemisphere dust sources for the previous thirty-five years indicate that air can be derived from the South Pacific which includes transport from Australia and New Zealand. Based on the Sr and Nd isotope data of Australian and New Zealand rocks [Delmonte et al., 2004; Revel-Rolland et al., 2006], an Australian contribution is more likely.

In summary, geochemical evidence excludes McMurdo Sound dust inputs as the dominant dust source to Roosevelt Island at present. The isotopic data and the presence of coarse particles at Roosevelt Island, compared to East Antarctic Plateau dust, suggest a local contribution from coastal regions of Antarctica. Potential dust sources include Marie Byrd Land rock outcrops, West Antarctic outcrops which are currently not well characterized in terms of their Nd and Sr isotopic signature, or other ice-free areas that have an old crust-like signature. These ice-free areas could include Victoria Land, given that the synoptic cyclonic circulation pattern in the Ross
Sea region could allow for dust transport from the western Ross Sea to the eastern Ross Sea. We cannot exclude the possibility of an Australian dust contribution, as the isotopic field overlaps with that of Marie Byrd Land and Victoria Land Paleozoic rocks. Both Bory et al. [2010] and Delmonte et al. [2013] show that local dust sources on the periphery of the ice sheet can significantly influence the dust composition at coastal, low elevation Antarctic ice core sites. Thus, Roosevelt Island could similarly be sourced from a mixture of local and distal dust sources. Further geochemical measurements from the RICE ice core are required to better constrain the dust provenance in the Ross Sea.

4.2. Atmospheric iron in the Ross Sea region

4.2.1. Atmospheric iron fluxes

Annual dissolved and total dissolvable Fe and Al fluxes for 2011 were calculated using the same method for dust (see section 4.1). Summer and winter dissolved Fe and Al fluxes for the period 2011 to 2012 were also estimated and are reported in Table 2. We estimate an annual dFe flux of $1.2 \times 10^{-6}$ g m$^{-2}$ y$^{-1}$ and annual TDFe flux of $140 \times 10^{-6}$ g m$^{-2}$ y$^{-1}$. The TDFe flux estimate for Roosevelt Island is higher than published Holocene values in Antarctic ice cores, for example, a TDFe flux of 45 x $10^{-6}$ g m$^{-2}$ y$^{-1}$ was reported for coastal Law Dome [Edwards et al., 2006]. The TDFe flux at Roosevelt Island is also higher than acid leachable fluxes of $7 \times 10^{-6}$ g m$^{-2}$ y$^{-1}$ at Dome C, East Antarctic Plateau [Gaspari et al., 2006] and 90 x $10^{-6}$ g m$^{-2}$ y$^{-1}$ at Talos Dome [Vallelonga et al., 2013]. Talos Dome is known to be influenced by local dust sources [Delmonte et al., 2010], and the higher TDFe flux at Roosevelt Island is likely due to the contribution of local dust sources from Marie Byrd Land and/or
Victoria Land. Local dust sources located on the coast of East Antarctica are also known to influence the concentration of aerosol Fe samples collected over marginal waters [Gao et al., 2013]. The TDFe flux at Roosevelt Island is orders of magnitude lower than locally derived aeolian Fe from McMurdo Sound (HF and HNO₃ digestible total Fe: 37 - 121 mg m⁻² yr⁻¹; water-soluble Fe: 2 - 7 mg m⁻² yr⁻¹ [Winton et al., 2014]). We acknowledge that these studies are not directly comparable as different methods were employed to determine the total and soluble Fe fractions. It has been demonstrated that both the acid leachable Fe method [Gaspari et al., 2006; Vallelonga et al., 2013] and the TDFe method [Edwards et al., 2006; this study] underestimate the total Fe fraction in snow and ice [Conway et al., 2015].

4.2.2. Atmospheric fractional iron solubility

Fractional Fe solubility was calculated using equation (1). Fractional Fe solubility ranged from 0.1 - 30 %. Fractional Fe solubility was fairly constant at ~0.7 % throughout the record, however dramatically rose above this background to 10 and 30 % during winter in 2012 and 2011 respectively. The background fractional Fe solubility (Fig. 2a) parallels δ¹⁸O (Fig. 2i) suggesting this variability in Fe deposition could be driven by synoptic weather conditions. We note that it is possible that the higher uncertainty associated with extremely low dFe and dAl concentrations in the snow pit could be responsible for some of the variability in the fractional Fe solubility. Additionally, precipitation of Fe(III) during sample melting or removal of oxyhydroxide complexes during filtration through a 0.2 µm filter could lead to underestimation of dFe concentrations and fractional Fe solubility. However, these processes would have had to occur rapidly as snow melting took <2 hours and
samples were filtered, acidified and analysed immediately after melting to minimise such processes.

The data in this study displayed an inverse hyperbolic relationship between the TDFe concentration in snow and fractional Fe solubility (Fig. 6). This relationship has been attributed to the mixing of low Fe solubility mineral dust and other soluble Fe aerosols from sources such as biomass burning and oil combustion [e.g. Sedwick et al., 2007]. Sholkovitz et al. [2012] reported global fractional Fe solubility data sets and concluded that the characteristic inverse hyperbolic relationship is common over large regions of the global ocean. This relationship was also found for baseline air at the Cape Grim Baseline Air Pollution Station (CGBAPS), Tasmania, Australia representative of the Southern Ocean [Winton et al., 2015]. Results here are similar to or greater than CGBAPS with respect to the extremely low TDFe in the exceptionally clean air. The inverse hyperbolic relationship at Roosevelt Island also suggests a mixture of mineral dust and combustion sources of Fe.

Reported values for global fractional Fe solubility of mineral dust are around 1 - 2 % [e.g. Baker and Croot, 2010]. Aerosol Fe deposition to remote Southern Ocean surface waters has previously been investigated in relation to the distribution and transport of mineral dust [e.g. Edwards and Sedwick, 2001; Martinez-Garcia et al., 2014; Wagener et al., 2008; Winton et al., 2014]. High-elevation East Antarctic ice core records also link higher rates of Fe and dust deposition during glacial periods [Conway et al., 2015; Vallelonga et al., 2013]. The background fractional Fe solubility of ~0.7 % at relatively high TDFe mass concentrations at Roosevelt Island is consistent with a mineral dust source.
Alternatively, the variability in fractional Fe solubility could be driven by changes in
dust mineralogy and grain size without necessarily being related to biomass burning
sources of soluble Fe. Changes in dust source could supply dust with a higher dFe
fraction, or where the TDFe dissolves less easily in acid. Furthermore, TDFe can be a
highly variable portion of total Fe [e.g. Conway et al., 2015; Tian et al., 2008].

Seasonal changes in dust provenance at Roosevelt Island are an important topic that
deserves further investigation. Some studies have shown that cloud chemistry and
atmospheric processing by oxalate and sulfate can enhance the solubility of mineral
dust [Desboeufs et al., 1999; Kumar et al., 2010; Meskhidze et al., 2003; Spokes et al.,
1994]. However, in the remote Atlantic and Pacific Ocean and the Southern Ocean
south of Australia, no relationship was observed between acid species and fractional
Fe solubility [Baker et al., 2006; Hand et al., 2004; Winton et al., 2015]. Even if the
dust source to Roosevelt Island in 2012 was continental Antarctica, e.g. Marie Byrd
Land, it is unlikely that atmospheric processing by combustion aerosols (observed in
polluted air masses where concentrations of organic acids are high [Chuang et al.,
2005; Ito, 2015; Ito and Shi, 2015; Kumar et al., 2010]) would enhance the solubility
of the iron contained in the mineral dust. The air masses over Antarctica and the
Southern Ocean contain trace concentrations of sulfate and oxalate [Keywood, 2007].
However, little is known about the enhancement of fractional Fe solubility in these
pristine air masses [Chance et al., 2015]. In addition, any enhancement of fractional
iron solubility by oxalate may not be sufficient to account for the high fractional Fe
solubility observed in the snow pit. For example, Paris et al. [2011] showed that
although oxalate complexation increased fractional Fe solubility from 0.0025 to 0.26
% in African dust minerals, the fractional Fe solubility remained low. No relationship
was found between the samples with a lognormal size distribution and high fractional Fe solubility. Longer records of particle size distribution and fractional Fe solubility at Roosevelt Island are required to investigate whether particle size can explain the variability in fractional Fe solubility.

### 4.3. Multiple sources of atmospheric dissolved iron to the Ross Sea

The temporal variability of mineral dust and the biomass burning tracer, rBC, parallels Roosevelt Island atmospheric dFe deposition (Fig. 2). Similar to TDFe concentrations, dust and rBC concentrations displayed an inverse hyperbolic relationship with fractional Fe solubility (Fig. S3b-c). Dissolved Fe deposition to Roosevelt Island is semi-annual with elevated deposition in summer and winter during the study period (Fig. 2b). The rest of this discussion focusses on the two largest deposition events of dFe highlighted in blue in Fig. 2. There were two intervals with high dFe concentrations. There were exceptionally high rBC concentrations in the first interval in summer 2011/2012. While during the second interval in winter 2011, concentrations of rBC were near background levels. In both cases, dust deposition was high indicated by high concentrations of dust, TDFe and TDAI. During the study period, it appears that dust is the primary source of dFe with additional rBC contributions. In both intervals, the excursion of δ¹⁸O to more negative values suggests that the process by which dFe is deposited at the site is storm related [Tuohy et al., 2015]. Multiple sources of dust and rBC may contribute to the atmospheric Fe supply to the Ross Sea, and the absence of an rBC source in winter highlights that the relative importance of the different sources varies seasonally.

### 4.3.1. Atmospheric iron sourced from mineral dust
A mineral dust source in summer is evident through i) co-variation of dFe maxima and high dust in 2011/2012 summer-spring (Figs. 2e and 2g), ii) strong correlation between the crustal elements TDFe and TDAl ($r^2 = 0.86$; Fig. S3a), iii) co-variation of TDAl and dust in 2011 (Fig. S3d-e), and iv) low fractional Fe solubility during summer (~0.7 %). For the majority of the record, TDFe and TDAl tracks dust concentration. However, not every dust deposition event leads to higher dFe, for example in spring 2011. This could be related to differences in the mineralogy of the dust, which varies depending on the dust provenance. Whether the dust provenance switches between seasons is unknown, but it is an important topic that deserves more attention in future. The coarser particle size distribution in summer-spring and Sr and Nd isotopic ratios matching that of nearby Marie Byrd Land, Victoria Land and Australia suggests that a mixture of different types of local and possibly remote dust sources influence dFe deposition at present.

4.3.2. Atmospheric iron sourced from biomass burning

Biomass burning may also contribute to the 2011/2012 summer deposition of dFe at Roosevelt Island. Refractory black carbon deposition to Antarctica is linked to Southern Hemispheric long-range transport of biomass burning [Bisiaux et al., 2012]. The high fractional Fe solubility during winter in 2011 and 2012 (up to 30 %; Figs. 2a and 6) indicates that additional atmospheric sources, other than mineral dust or biomass burning, are responsible for high fractional Fe solubility in the winter periods observed in this record. Thus, biomass burning can only account for the high fractional Fe solubility observed in background Southern Hemispheric air during the 2011/2012 summer period covered by the snow pit. Ice core records report rBC as a late winter-spring phenomena in West Antarctica [Bisiaux et al., 2012]. Consistent
with WAIS Divide, a small late winter-spring-time rBC peak is observed in 2012 in this study. However, the exceptionally large 2011/2012 summer rBC peak does not overlap with the time period covered by WAIS Divide rBC record. Longer records of rBC and fractional Fe solubility are needed to test the hypothesis that biomass burning sources of dFe can account for high fractional Fe solubility of aerosols over the Ross Sea.

4.3.3. Timing and supply of iron deposition

Sea ice is a source of dFe to the ocean in the SW Ross Sea, where high dFe concentrations have been observed in the surface waters after considerable sea ice melt [de Jong et al., 2013; McGillicuddy et al., 2015; Sedwick and DiTullio, 1997]. This snow pit record from Roosevelt Island demonstrates that spring-summer dFe deposition occurs during ice-free open water conditions in the Ross Sea, when phytoplankton are blooming (Fig. 2l). Around half of the annual TDFe and dFe for 2011 was deposited in the summer, and, therefore, the seasonality of dFe deposition needs to be considered on longer glacial-interglacial time scales. Despite high dFe and TDFe deposition in spring-summer, fractional Fe solubility was only ~0.7 % at this time. Higher fractional Fe solubility occurs in winter when TDFe is low, and the Ross Sea is seasonally ice covered. Snow on sea ice is a repository for aerosol Fe [Lannuzel et al., 2010; Winton et al., 2014]. Therefore, deposition of aerosol Fe, with relatively high dFe content, is stored in the surface snow of sea ice during winter. In spring-summer, aerosol Fe with a higher fractional Fe solubility is released and supplied to the ocean as sea ice melts. Regardless of the timing and mechanism in which aerosol dFe is supplied to the Ross Sea, i.e., deposition directly into open water in summer and deposition onto surface snow on sea ice in winter with subsequent release into the
ocean during spring-summer sea ice melt, both mechanisms act as a new source of dFe to phytoplankton blooms in spring-summer.

5. Conclusions

A dust flux of 6 mg m\(^{-2}\) yr\(^{-1}\) for the 1-5 µm fraction and 8 mg m\(^{-2}\) yr\(^{-1}\) for the bulk 1-30 µm has been estimated for modern dust deposition at Roosevelt Island. Snow pit data from Roosevelt Island reveal dust deposition occurs primarily in the spring-summer season. The higher dust flux at Roosevelt Island compared to the East Antarctic Plateau, the presence of coarse dust particles (>10 µm), the particle size distribution that approaches the theoretical dust emission distribution and the Sr and Nd isotopic ratio of dust deposited on Roosevelt Island suggest a mixture of local and possibly remote dust sources for the present day. The Sr isotopic composition (0.7122 < \(^{87}\)Sr/\(^{86}\)Sr < 0.7156) of summer dust in the snow at Roosevelt Island suggests a possible mixing of volcanic and crustal rocks of local origin (i.e., older material, e.g. Paleozoic rock from Victoria Land and parts of Marie Byrd Land, and Mesozoic to Cenozoic volcanics). Additional input from remote dust sources cannot be discounted. In this respect, Australia best matches the Roosevelt Island isotopic composition of Sr and Nd. Advection of Australian dust is consistent with modelling studies of modern Australian aeolian transport for the present day. These dust data from Roosevelt Island provide useful context for the interpretation of the dust record in the 764 m long RICE ice core.

An annual dFe flux of 1.2 \(\times 10^{-6}\) g m\(^{-2}\) y\(^{-1}\) and an annual TDFe flux of 140 \(\times 10^{-6}\) g m\(^{-2}\) y\(^{-1}\) have been estimated for Roosevelt Island for 2011. Deposition of dFe is semi-annual occurring in the summer and winter, with half of dFe and TDFe deposition
occurring in the summer. The inverse hyperbolic relationship between TDFe concentrations and fractional Fe solubility shows that additional atmospheric sources, other than mineral dust, are responsible for high Fe solubility at different times in the year. There were two intervals with high dFe concentrations in the snow pit: one with exceptionally high rBC concentrations in summer 2011/2012, and the other with rBC concentrations near background levels in winter 2011. In both cases, dust was high. Therefore, mineral dust, from both local and remote sources, is the primary source of dFe to Roosevelt Island with additional input from long-range transport of biomass burning aerosols. Biomass burning may account for the high fractional Fe solubility observed in background Southern Hemispheric air during the summer period. The semi-annual nature of Fe deposition to Antarctic waters should be considered when interpreting longer glacial-interglacial time scales of aerosol Fe deposition to the Southern Ocean.
Acknowledgements

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Table 1: Nd and Sr concentrations [in parentheses] and isotopic composition of Roosevelt Island surface snow samples analyzed in this study. n.d.: no data.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Size (µm)</th>
<th>$^{143}$Nd/$^{144}$Nd</th>
<th>$^{143}$Nd/144Nd</th>
<th>$^{143}$Nd/144Nd</th>
<th>[Nd] (ppm)</th>
<th>$^{143}$Nd/144Nd</th>
<th>$^{143}$Nd/144Nd</th>
<th>[Sr] (ppm)</th>
<th>$^{143}$Sr/$^{86}$Sr</th>
<th>$^{143}$Sr/$^{86}$Sr</th>
</tr>
</thead>
<tbody>
<tr>
<td>RI-1</td>
<td>Bulk</td>
<td>n.d.</td>
<td>n.d.</td>
<td>n.d.</td>
<td>3</td>
<td>0.3</td>
<td>0.712173</td>
<td>17</td>
<td>0.712201</td>
<td>17</td>
</tr>
<tr>
<td>RI-2</td>
<td>0.512146</td>
<td>50</td>
<td>-9.6</td>
<td>0.3</td>
<td>12</td>
<td>1.2</td>
<td>0.715560</td>
<td>48</td>
<td>0.715588</td>
<td>48</td>
</tr>
</tbody>
</table>

a) Internal precision, 2 standard errors of the mean.

b) Nd isotopic ratios expressed as epsilon units $\varepsilon_{Nd}(0) = \left(\frac{^{143}Nd/^{144}Nd}_{sample}/^{143}Nd/^{144}Nd_{CHUR-1}\right) \times 10^4$; CHUR, chondritic uniform reservoir with $^{143}Nd/^{144}Nd=0.512638$.

c) Uncertainty estimates based upon external precision for standard runs. Internal precision is used if it exceeds the external.

d) Error due to difficulty of measuring small sample masses, estimated by repeat measuring of weighting BCR-2 standards (~0.3 mg).

e) Internal precision, 2 standard errors of the mean.

f) Corrected to a NBS 987 $^{87}Sr/^{86}Sr$ ratio of 0.710245.

g) Uncertainty estimates based upon external precision for standard runs. Internal precision is used if it exceeds the external.

Table 2: Summer and winter trace metal dissolved, total dissolvable and dust fluxes.

<table>
<thead>
<tr>
<th></th>
<th>dFe flux ($10^{-6}$ g m$^{-2}$ y$^{-1}$)</th>
<th>TDFe flux ($10^{-6}$ g m$^{-2}$ y$^{-1}$)</th>
<th>dAl flux ($10^{-6}$ g m$^{-2}$ y$^{-1}$)</th>
<th>TDAl flux ($10^{-6}$ g m$^{-2}$ y$^{-1}$)</th>
<th>Dust flux 1-5 µm (mg m$^{-2}$ y$^{-1}$)</th>
<th>Dust flux 1-30 µm (mg m$^{-2}$ y$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Summer</td>
<td>0.4</td>
<td>39</td>
<td>0.5</td>
<td>50</td>
<td>2.7</td>
<td>4.1</td>
</tr>
<tr>
<td>Winter</td>
<td>0.2</td>
<td>50</td>
<td>0.4</td>
<td>63</td>
<td>2.4</td>
<td>3.2</td>
</tr>
<tr>
<td>Annual</td>
<td>1.2</td>
<td>140</td>
<td>2.9</td>
<td>195</td>
<td>5.9</td>
<td>8.8</td>
</tr>
</tbody>
</table>
**Figure captions**

Fig. 1: a) Map of Antarctica showing the location of the Ross Sea. b) Insert of Marie Byrd Land showing West Antarctica potential source areas and Roosevelt Island dust sample locations (RI1-2). EAIS: East Antarctic Ice Sheet, MDV: McMurdo Dry Valleys, TAM: Transantarctic Mountains.

Fig. 2: Roosevelt Island snow pit profile showing a) fractional Fe solubility, b) dFe concentration, c) dAl concentration, d) rBC concentration, e) TDFe concentration, f) TDAI concentration, g) dust concentration for the 1-5 µm fraction, h) total dust concentration for the 1-30 µm fraction, i) δ¹⁸O, j) snow density, k) nss-S concentration, l) chlorophyll-a concentration. Grey bars indicate summer periods. Blue bars highlight the largest periods of dFe deposition. Black lines indicate smoothed data using a 0.15 loess model \cite{Cleveland and Devlin, 1988}.

Fig. 3: Examples of particle size distributions in the snow pit from Roosevelt Island and comparison to the lognormal particles size distribution of Holocene dust from the Dome C ice core on the East Antarctic Plateau. Blue: a) Normalized size distribution of the number of particles (dN/dlogD), b) normalized volume size distribution (dV/dlogD). Samples shown are RI_14, Black: RI_6, Red: RI_10. Dome C Holocene particle size distribution from Delmonte et al. [2002].

Fig. 4: Nd and Sr isotope signature of Roosevelt Island surface snow dust sample. Note only one sample is plotted as there is no Nd data for the second sample. Also plotted are data from Victoria Land sediments from potential dust sources (regolith, ...
glacial deposits, aeolian sediments) [Delmonte et al., 2004; Delmonte et al., 2013; Delmonte et al., 2010] that include different parent lithologies (Victoria Land Ferrar Igneous Provence [Antonini et al., 1999; Delmonte et al., 2004; Elliot et al., 1999; Fleming et al., 1995] and Victoria Land Paleozoic rocks [Cox et al., 2000; Schüssler et al., 1999; Talarico et al., 1995]) and volcanic rocks from the McMurdo Volcanic Group and West Antarctica [Delmonte et al., 2004; Futa and Le Masurier, 1983; Hole and LeMasurier, 1994].

Fig. 5: $^{87}$Sr/$^{86}$Sr isotopic composition of Roosevelt Island dust compared to McMurdo Sound dust [Winton et al., 2014; Winton et al., in review], Antarctic volcanic rocks [Delmonte et al., 2004; Futa and Le Masurier, 1983; Hole and LeMasurier, 1994] and King Edward VII Peninsula, Marie Byrd Land, West Antarctic Granites and metasedimentary basement rocks [Adams et al., 1995].

Fig. 6: Scatterplot of Roosevelt Island total dissolved Fe concentration versus fractional Fe solubility. Top right insert: low concentration data expanded.

Fig. S1: Scanning electron microscope image of coarse particles in a snow pit from Roosevelt Island (RI24).

Fig. S2: Roosevelt Island snow pit temperature and snow density profile.

Fig. S3: Relationships between variables in the snow pit. a) Correlation between TDAI concentrations and TDFe concentrations, b) inverse hyperbolic relationship between fractional Fe solubility and rBC concentrations, c) inverse hyperbolic
relationship between fractional Fe solubility and dust concentrations, d) relationship between dust concentrations and TDAl concentrations, e) relationship between dust concentrations and TDFe concentrations.
Particle diameter (μm)

Normalised number concentration (dN/d\log D μm⁻³ ml⁻¹)

R1_6
R1_10
R1_14
Dome C Holocene

Normalised volume concentration (dV/d\log D μm⁻³ ml⁻¹)

a)

b)
McMurdo Volcanic Group

West Antarctic volcanics

Beacon Sandstone

Ferrar Dolerite

Kirkpatrick basalts

Victoria Land Paleozoic rocks

McMurdo Sound dust

Roosevelt Island

Victoria Land Granite

Priestly Schist

Victoria Land Mixture of Beacon sandstone, Ferrar Dolerite, Kirkpatrick basalts

Mixture of Beacon sandstone, Ferrar Dolerite, Kirkpatrick basalts

McMurdo Volcanic Group
Coastal Marie Byrd Land Granite

Swanson Formation

Mt. Nilsen (630 m), La Gorce Peak (1030 m) and Drummond Peak (780 m)

Mt. Paterson, Mt. Butler, Mt. Tennant, Gould Peak

Mt. Frazier, Mt. Jackling, Mt. Fitzsimmons, Mt. Shideler

McMurdo Sound dust

RI1

RI2

MBL

MVG

King Edward VII Peninsula (Marie Byrd Land, West Antarctica)

Antarctic Volcanic rocks

Antarctic

87Sr/86Sr