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The origin of lithogenic sediment in the southwestern Ross Sea and implications for iron fertilisation

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Abstract

Austral summer iron (Fe) fertilisation in the Ross Sea has previously been observed in association with diatom productivity, lithogenic particles and excess Fe in the water column (Collier et al., 2000). This productivity event occurred during an early break out of sea ice via katabatic winds, suggesting that aeolian dust could be an important source of lithogenic Fe required for diatom growth in the Ross Sea. Here we investigate the provenance of size-selected dust deposited on sea ice in McMurdo Sound, southwestern (SW) Ross Sea. The isotopic signature of McMurdo Sound dust ($0.70533 < {}^{87}Sr/{}^{86}Sr < 0.70915$ and $-1.1 < \varepsilon_{Nd}(0) < 3.45$) confirms that dust is locally sourced from the McMurdo Sound debris bands and comprises a two-component mixture of McMurdo Volcanic Group and Southern Victoria Land lithologies. In addition, we investigate the provenance of lithogenic sediment trapped in the water column: the isotopic signature ($\varepsilon_{Nd}(0)=3.9$, ${}^{87}Sr/{}^{86}Sr=0.70434$) is differentiated from long-range transport dust originating from South America and Australia. Elevated lithogenic accumulation rates in deeper sediment traps in the Ross Sea, suggest that sinking particles in the water column cannot simply result from dust input at the surface. This discrepancy can best be explained by significant upwelling and remobilisation of lithogenic Fe from the sea floor.

Key words

Dust provenance, dust, iron, McMurdo Sound, Antarctica

1. Introduction

Atmospheric dust is potentially an important source of dissolved iron (DFe) which is the limiting nutrient required for primary production in vast regions of the remote Southern Ocean, including Antarctica's marginal seas (Boyd et al., 2010; Sedwick et al., 2000). Despite being seasonally iron (Fe) limited, the high-nutrient, high chlorophyll (HNHC) regime of the Ross Sea is the most biologically productive continental shelf region in Antarctica, and supports intense phytoplankton blooms in the austral summer (Arrigo et al., 2008). Although the flux of Fe into the Ross Sea plays a critical role in determining its productivity, the origin(s) of this Fe remains poorly constrained (Sedwick et al., 2011).

1.1. Dust deposition in Antarctica

Global 'background' dust is characterized by fine particles having a mass modal diameter <5 μm, long atmospheric residence time and modern mass deposition rates in the order of 0.001-0.02 g m^{-2} yr^{-1} in the Southern Ocean (Wagener et al., 2008; and references therein). The isolated, snow and ice-covered central East Antarctic Plateau (EAP) has proven to be an excellent location for investigating long-range transport of dust representative of the broader Southern Hemisphere, both at present and in the past (Delmonte et al., 2008; Delmonte et al., 2007). Moreover, the high East Antarctic Plateau (EAP) has much lower accumulation rates of around 0.0002-0.0006 g m^{-2} yr^{-1} during the Holocene (Albani et al., 2012). Recently, it has become apparent that peripheral areas of the Antarctic ice sheet, close to high-elevation ice-free mountain ranges, such as the Transantarctic Mountains (TAM), can receive significant additional dust inputs from exposed Antarctic sources, some of which have been ice-free for millions of years (Delmonte et al., 2013 and references therein).

The relative contribution of much smaller, patchy but proximal dust sources to the atmospheric dust load over Antarctica and the Southern Ocean is not well known. The largest expanse of contiguous ice-free ground in Antarctica is found in the McMurdo Dry Valleys - a series of west-to-east-oriented, glacially carved valleys located between the high EAP and the Ross Sea in Southern Victoria Land. However, the dustiest known place in Antarctica is located in the southwestern (SW) Ross Sea, associated with the so-called 'debris bands' area on the McMurdo Ice Shelf (Kellogg et al., 1990) (Fig. 1). In this region, dust deposition flux (~1 g m^{-2} yr^{-1}) is at least two orders of magnitude greater than fallout of long-range transport dust measured in ice cores from the EAP (Atkins & Dunbar, 2009; Chewings et al., 2014; Delmonte et al., 2013) and is, potentially, an important source of bioavailable Fe to the Ross Sea (Winton et al., 2014).

Dust provenance in Antarctica 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) (e.g. Delmonte et al., 2010). This geochemical method allows mantle-derived (basaltic rocks, tephra and soils derived from them, weathered and eroded mafic rocks) and crustal-derived sediments and soils to be identified. Both the geochemical fingerprint and particle size of dust deposited on the EAP suggests it originates from arid regions in southern South America during glacial periods (Delmonte et al., 2008; Gaiero et al., 2007). However, for dust deposited during interglacial periods, that is when dust input to inner Antarctica was extremely low, the source is less certain (Delmonte et al., 2007), and an Australian contribution is likely (Delmonte et al., 2008; Delmonte et al., 2007; Revel-Rolland et al., 2006). In addition

to atmospheric circulation, dust transport efficiency is dependent on particle size; for example, long-range dust deposited on the EAP has a mass-modal size of ~2-3 µm (Delmonte et al., 2002). When investigating the provenance of dust, the fractionation of Sr isotopes into different grain size fractions needs to be considered, as there is a correlation between grain size and $^{87}Rb/^{86}Sr$ ratios and thus $^{87}Sr/^{86}Sr$ ratios. In coarse (fine) grained suspended particulate matter Sr is enriched (depleted) in less radiogenic Sr isotopic ratios (e.g. Andersson et al., 1994). In contrast, Nd isotopic ratios are not influenced to the same extent by particle size (e.g. Andersson et al., 1994).

1.2. Iron-fertilisation in the Ross Sea

The Ross Sea is one of the most productive regions in the Southern Ocean and an important oceanic sink for atmospheric carbon dioxide (CO_2) (e.g. Arrigo et al., 2008). The environmental factors responsible for controlling the rates of phytoplankton production and incomplete utilisation of inorganic macronutrients include: grazing (Banse, 1991), temperature (Bunt & Wood, 1963), light availability (e.g. Mitchell et al., 1991), micro-nutrient availability (e.g. Fe and Mn) (Sedwick & DiTullio, 1997; Sedwick et al., 2000), or a combination of these (e.g. Arrigo et al., 2000). Collier et al. (2000) show that a Ross Sea diatom productivity event, captured during the 1996-1997 deployment of moored Antarctic Environment and Southern Ocean Process Study (AESOPS) sediment traps, is correlated with elevated lithogenic particle accumulation rates, excess Fe (determined from high Fe/Al ratios), and with an early breakout of sea ice caused by katabatic winds. They go on to suggest that there may be a causal relationship between the retreat of sea ice, the supply of particulate Fe and diatom production and export. The source of this lithogenic Fe to the Ross Sea is unknown but could be derived from either dust released into the ocean

126 from melting sea ice from local and/or from distal sources, or new particulate Fe
127 derived from ice shelves, icebergs, upwelling of resuspended continental sediments
128 from the sea floor, circumpolar deep water or some combination thereof.
129
130 Multiple sources of new Fe to the Ross Sea region have been identified, which
131 include local dust sourced mainly from the McMurdo Ice Shelf (Atkins & Dunbar,
132 2009; Chewings et al., 2014; de Jong et al., 2013; Winton et al., 2014), sea ice melt
133 (de Jong et al., 2013; Sedwick & DiTullio, 1997), and lithogenic sediments
134 resuspended from the sea floor (de Jong et al., 2013; Gerringa et al., 2015; Marsay et
135 al., 2014; Sedwick et al., 2011). However, the relative importance of these sources for
136 stimulating primary production remains an open question. Winton et al. (2014)
137 estimate that the supply of soluble aeolian Fe in dust from the debris bands, southern
138 McMurdo Sound to the adjacent ocean could support up to ~15 % of primary
139 production in the area. The implication being that Fe supporting the remaining 85 %
140 of productivity was derived largely from other sources, such as lithogenic sediment
141 resuspended from the sea floor (de Jong et al., 2013; Gerringa et al., 2015; Kustka et
142 al., 2015; Marsay et al., 2014; McGillicuddy et al., 2015; Sedwick et al., 2011).
143
144 As Fe is critical for seasonal phytoplankton growth in the Ross Sea, this study aims to
145 further investigate the source(s) of lithogenic Fe as a driver of the vast austral summer
146 phytoplankton blooms in the SW Ross Sea. We do this by examining the provenance
147 of lithogenic material sinking in the upper 200 meters below sea level (mbsl) of the
148 water column and compare its origin to both known local and global sources. Here we
149 report the Sr-Nd isotopic composition of i) size-selected dust from snow samples on
150 sea ice from McMurdo Sound, and ii) sediment trap material from the Research on

Ocean - Atmosphere Variability and Ecosystem Response in the Ross Sea (ROAVERRS) moorings program (1996-98) that represents accumulation of sediment settling out of the water column. When investigating PSAs to Antarctica, previous studies have size-selected the PSA samples prior to Sr analysis to be comparable to that of the fine size range of dust deposited in Antarctica (Delmonte et al., 2008), and a similar approach is used here.

2. Methods

2.1. Samples used in this study

Previous studies have focused on dust flux and particle size distribution patterns in McMurdo Sound (Atkins & Dunbar, 2009; Chewings et al., 2014; Dunbar et al., 2009). This study is based on the following samples:

- Samples of dust-laden snow collected from sea ice along a south-north X-Y transect in McMurdo Sound, collected in November 2010 and described in Chewings et al. (2014) and in Winton et al. (2014) (Fig. 1b).
- A Ross Sea sediment trap sample from 200 mbsl, collected between 25 Dec. 1997 and 3 Jan. 1998 from the ROAVERRS program Chinstrap site (76° 20.5'S, 165° 1.78'E) in the SW Ross Sea. This site was anchored in 830 m water depth in the southern extension of the Drygalski Basin (Fig. 1a).

2.2. Nd and Sr isotopic ratios and concentrations

2.2.1. Sample processing

McMurdo Sound surface snow on sea ice samples

The samples analysed in this work were size-selected in order to be comparable to provenance measurements made on dust from ice core PSAs and with similar studies on dust in Antarctica (e.g. Delmonte et al., 2008; Delmonte et al., 2010; Delmonte et al., 2004). We analysed both the bulk (all particle sizes) and fine (<10 µm) fraction of McMurdo Sound dust to check for particle size induced bias in the isotopic fractionation of samples. The coarse fraction was removed from bulk samples by using a pre-washed 10 µm SEFAR Nitex® open mesh while the fraction 0.4µm<∅<10µm was collected on 0.4 µm Isopore™ polycarbonate membranes. After filtration, the membranes were put into pre-cleaned Corning® tubes filled with ~10 ml of ultra-pure water, and micro-particles were removed from the filter by sonication. Samples were transported to the Department of Geosciences, Swedish Museum of Natural History, Sweden where the liquid was evaporated in acid-cleaned 15 ml Savillex® beakers. Dry dust samples, ranging between 0.1 and 1.2 mg, were weighed a minimum of five times to obtain a mean weight, which was used for subsequent calculations.

Sediment trap samples

Isotopic analysis of bulk sediment revealed that the biogenic fraction of the sediment (~up to 70 % total mass estimated from AESOPS sediment trap data reported in Collier et al. (2000)) incorporated marine Sr and thus the isotopic signature could not be distinguished from that of seawater (Table 1). To remove the biogenic silica and calcium carbonate fraction of the sediment we leached the sediment with 6 M HCl in Savillex® beakers and centrifuged following the method of Freydier et al. (2001). The lithogenic residue was then rinsed three times with ultra-pure water and dried.

201

202 *2.2.2. Sample digestion*

203 The chemical treatment of the dust samples and leached sediment, including digestion
204 and elemental separation (Rb-Sr and Sm-Nd) using ion exchange chromatography,
205 was performed at the Swedish Museum of Natural History following the established
206 method Delmonte et al. (2008). The samples were spiked with a mixed $^{147}Sm/^{150}Nd$
207 spike and ^{84}Sr-enriched spike for the isotope dilution determination of the
208 concentrations. Samples were digested in an acid mixture of 1.5 ml, of HNO_3, HF and
209 $HClO_4$ heated to 90 ºC in closed Savillex® beakers for 24 h. The solution was
210 evaporated to complete dryness on a hot plate, and the residue re-dissolved in 4 ml 6
211 M HCl.

212

213 *2.2.3. Ion exchange*

214 To achieve separation of potential interfering elements (Fe, Ba, Rb, Sm, Ce, and Pr),
215 and obtain high column yield and low blanks, the residue was subjected to chemical
216 procedures described in Delmonte et al. (2008). The total blank, including dissolution,
217 chemical separation and mass spectrometry, was frequently monitored in each ion
218 exchange batch and blank concentrations were <5 pg for Nd and <130 pg for Sr.

219

220 *2.2.4. Mass spectrometry*

221 Isotopic analysis of Nd and Sr was performed with a Thermo Scientific TRITON
222 Thermal Ionisation Mass Spectrometer (TIMS). Neodymium was loaded mixed with
223 colloidal graphite, Alfa Aesar, on double rhenium filaments and analysed as metal
224 ions in static mode using rotating gain compensation. Concentrations and ratios were
225 calculated assuming exponential fractionation. The calculated ratios were normalised

226 to $^{146}Nd/^{144}Nd = 0.7219$. Epsilon units are calculated as follows:

227

228 $\varepsilon_{Nd}(0) = [(^{143}Nd/^{144}Nd)_{sample}/(^{143}Nd/^{144}Nd)_{CHUR} -1] \times 10^4$;

229 CHUR, chondritic uniform reservoir with $(^{143}Nd/^{144}Nd)_{CHUR} = 0.512638$

230

231 The external precision for $^{143}Nd/^{144}Nd$ is estimated from analysis of the nNdß
232 standard (Wasserburg et al., 1981) by analysing a range, 4-12 ng loads, of nNdß
233 standard. The external precision becomes larger for smaller loads, with an estimated
234 precision of about 40 ppm for small loads, for intermediate size about 30 ppm and
235 about 20 ppm for larger loads. These values have been used to estimate the errors for
236 the samples in Table 1. The mean $^{143}Nd/^{144}Nd$ ratio for the nNdß was 0.511895±22
237 (n=20). Literature values for repeated analysis of standard nNdβ (Andreasen &
238 Sharma, 2006) yielded $^{143}Nd/^{144}Nd = 0.511892±3$ (2σ, n = 23) and thus no accuracy
239 correction was applied.

240

241 Purified Sr samples were mixed with tantalum activator and loaded on a single
242 rhenium filament. Two hundred 8 s integrations were recorded in multi-collector
243 static mode, applying a rotating gain compensation. Measured ^{87}Sr intensities were
244 corrected for Rb interference assuming $^{87}Rb/^{85}Rb=0.38600$ and ratios were calculated
245 using the exponential fractionation law and $^{88}Sr/^{86}Sr=8.375209$. External precision for
246 $^{87}Sr/^{86}Sr$, estimated from analysing NBS SRM987 standard, was calculated as
247 ±0.000016 (n=12) while repeated measurements of prepared CIT #39 sea water gave
248 a reproducibility of ±0.0000082 or 12 ppm (n=21) which was taken to be the best
249 estimate of the external precision. Accuracy correction was applied to the $^{87}Sr/^{86}Sr$
250 ratios corresponding to a $^{87}Sr/^{86}Sr$ ratio of 0.710245 for NBS SRM 987 standard

(NBS 987: literature value 0.710245, Department of Geosciences value 0.710217 ±16 (n=12); difference: 0.000028).

The accuracy of the Nd and Sr isotopic composition of small dust samples was determined using the Basalt Columbia River rock standard (BCR-2), a certified reference material. Preparation and analysis of 150 to 600 µg aliquots of BCR-2 in each batch of ion exchange resulted in a recovery of >79 % (n=6) for concentration and >99 % (n=6) for isotopic composition. Due to the small dust samples and the difficulty of weighing such small masses we estimate a Sr and Nd concentration error of ±10 % estimated by repeated weighing of BCR-2 standards (~0.3 mg).

3. Results

The Sr and Nd isotopic composition of dust are primarily related to lithology and geologic age of parent materials, although the Sr isotopic composition, for particles between 2-50 µm can also be influenced by their size. The Sr and Nd isotopic composition of the fine (<10 µm) and bulk (all sizes included) dust samples collected in this study are well characterised and reported in Table 1 and Fig. 2 along with additional isotopic data from McMurdo Sound measured in an earlier study (Winton et al., 2014). Samples in Fig. 2 are grouped by geographic location. The samples collected from the snow on sea ice in McMurdo Sound display a narrow isotopic composition ($0.70533 < {}^{87}Sr/{}^{86}Sr < 0.70915$ and $-1.1 < \varepsilon_{Nd}(0) < 3.45$). Our fine dust (<10 µm) samples have relatively higher ${}^{87}Sr/{}^{86}Sr$ ratios compared to the bulk samples (Fig. 2), consistent with previous studies of size-dependent fractionation (Andersson et al., 1994). The Nd isotopes do not fractionate with the particle size (with the exception of MPN10-5 which could be related to the different size fractions

originating from different sources; Fig. 2), also consistent with previous studies. The $\Delta^{87}Sr/^{86}Sr$ is about 0.00115, that is slightly smaller than the $^{87}Sr/^{86}Sr$ increase of ~0.0028 units observed between 63 µm and 2 µm dust particles by Gaiero et al. (2007). The isotopic ratios of leached (lithogenic sediment fraction) and unleached (lithogenic and biogenic sediment fraction) Ross Sea sediment obtained from the upper Chinstrap sediment trap (200 mbsl) are reported in Table 1. Leaching had a significant effect on the Sr isotopic ratio of this sample and removed a Sr seawater overprint from the sediment. After leaching, the remaining lithogenic sediment has an isotopic signature between the McMurdo Volcanic Group and Southern Victoria Land PSAs (Fig. 2).

4. Discussion

4.1. Dust provenance

4.1.1. McMurdo Sound

The overwhelming majority of dust deposited in snow on sea ice in McMurdo Sound is locally sourced. It is not possible to detect any contribution from South American or Australian sources with our approach. Sedimentological, meteorological and geochemical evidence consistently points to the debris bands on the McMurdo Ice Shelf (Kellogg et al., 1990) as the dominant local dust source in the McMurdo Sound region (Fig. 1) (Atkins & Dunbar, 2009; Chewings et al., 2014; Winton et al., 2014). Studies of spatial variability of dust and particle size for the greater McMurdo Sound region show a distinct decrease in particle size and dust flux along transect X-Y (Fig. 1b) as part of a dust plume extending northwards from the debris bands (Atkins &

Dunbar, 2009; Chewings et al., 2014). As the plume extends northwards away from the debris bands the particle size and dust accumulation rate decrease, although secondary elevated patches of both occur near coastal headlands. Overall dust accumulation declines exponentially from 55 g m^{-2} yr^{-1} near the debris bands to ~0.2 g m^{-2} yr^{-1} 120 km north of the debris bands (Atkins & Dunbar, 2009; Chewings et al., 2014). This northward dust dispersal is consistent with the local meteorology whereby the highest wind speeds, i.e. those most competent with respect to entraining silt and fine sand, are predominately from the south (Fig. 1c), dispersing dust from the debris bands north along the Southern Victoria Land coastline.

In addition to sedimentological considerations, geochemical evidence also points to dust being locally sourced. The Sr isotopic of modern seawater is homogenous ($^{87}Sr/^{86}Sr$=0.70924; Elderfield (1986)), and has a similar Sr isotopic composition to the geology in McMurdo Sound (Fig. 2). However, the Nd isotopic composition of the local geology and Ross Sea seawater (-10<$\varepsilon_{Nd}(0)$<-6; Basak et al. (2015)) is distinguished in Fig. 2, and combined with other provenance indicators (coarse particle size, high dust flux and Fe/Al elemental ratios (Atkins & Dunbar, 2009; de Jong et al., 2013); see below, allows tracing of dust to local PSAs. Winton et al. (2014) report two Sr and Nd isotopic ratios of the bulk sediment from snow on sea ice in southern McMurdo Sound and one from Granite Harbour (Fig. 1). The values are consistent with dust originating from McMurdo Volcanic Group (MVG), although within the Granite Harbour embayment there is also evidence for dust-derived from TAM lithologies. We rule out the possibility that the volcanic signature of McMurdo Sound dust on sea ice is derived from volcanic rocks in Marie Byrd Land (0.7026< $^{87}Sr/^{86}Sr$ <0.7032 and 1.99< $\varepsilon_{Nd}(0)$ <6.87) (Futa & Le Masurier, 1983; Hole &

LeMasurier, 1994) due to the northerly direction of the prevailing winds (Chewings et al., 2014).

Consequently, only local potential source areas (PSAs) are considered for comparison to the new isotopic dataset. Overall, the Sr isotopic ratios for McMurdo Sound samples analysed in this study and in Winton et al. (2014) are tightly grouped and range between $0.705 < {}^{87}Sr/{}^{86}Sr < 0.709$ while $\varepsilon_{Nd}(0)$ ranges between $3.45 < \varepsilon_{Nd}(0) < -1.1$. These new isotopic data form a linear array in Fig. 2. McMurdo Sound dust can be considered the result of a two-component mixture derived from isotopically distinct end-members: i) the MVG volcanic rocks and ii) Southern Victoria Land lithologies found in the TAM such as Ferrar Dolerites and Beacon sandstone (Fig. 2). The narrow range of isotopic ratios of McMurdo Sound dust along the south-north transect X-Y represents northwards dust dispersal downwind from the debris bands, that is a mixture of TAM and MVG sources, with minor localised additions of TAM dust input from coastal outcrops from New Harbour and Marble Point that contribute to the dominant south to north dust plume (Fig. 1). This is consistent with field observations showing sediment on the McMurdo Ice Shelf debris bands is itself a mixture of MVG and TAM lithologies (Kellogg et al., 1990).

Previous studies have shown dust deposited within embayments or adjacent to headlands along the Southern Victoria Land coastline is not widely dispersed (Barrett et al., 1983; Chewings et al., 2014; de Jong et al., 2013). Within the narrow range of isotopic ratios of McMurdo Sound dust, GH9 is isotopically distinct and displays a dominant TAM signature (Fig. 2). This sample is not situated under the main northward-directed dust plume and hence represents localised dust accumulation

within the Granite Harbour embayment (Fig. 1.). In contrast, the isotopic composition of GH2 lies within the tight cluster of McMurdo Sound dust and thus highlights that the mass of dust on the sea ice immediately seawards of Granite Harbour originates from the south. A single-source from the debris bands is also consistent with Fe concentrations within dust samples that were uniform along the transport pathway (Winton et al., 2014).

4.1.2. Southwestern Ross Sea

The isotopic signature of the lithogenic fraction of sediment from the upper 200 mbsl Chinstrap sediment trap, located ~170 km north of the debris bands ($^{87}Sr/^{86}Sr$=0.704, $\varepsilon_{Nd}(0)$=3.9), falls outside of the isotopic range of dust originating in Australia ($0.709<^{87}Sr/^{86}Sr<0.763$, $-2.9<\varepsilon_{Nd}(0)<-15.4$; Delmonte et al. (2004); Revel-Rolland et al. (2006)) and South America ($0.704<^{87}Sr/^{86}Sr<0.713$, $-8.9<\varepsilon_{Nd}(0)<-8.3$; Delmonte et al. (2004)). These two potential Southern Hemisphere sources supply dust to the high elevation EAP at very low deposition rates (e.g. Delmonte et al., 2008). The signature of the lithogenic fraction of sediment from SW Ross Sea (Chinstrap) matches that of the local geology and dust on sea ice in McMurdo Sound. Thus, the lithogenic particles, and their associated Fe, collected here is 'locally' sourced from the Ross Sea region (Fig. 2).

4.1.3. Dust transport and deposition in the southwestern Ross Sea

Deposition of local dust into the SW Ross Sea can occur by direct atmospheric fallout into ice-free surface waters, and released into surface waters by sea ice melt associated with subsequent northwards advection (Atkins & Dunbar, 2009; Chewings et al., 2014; de Jong et al., 2013). The geographical area over which local dust is

transported into the Ross Sea and hence contributes to Fe-fertilisation is potentially large. Although dust accumulation measurements only exist up to 120 km from the debris bands and decrease exponentially from the source, local dust deposition likely extends far beyond this point. Extrapolating the dust flux trend observed by Chewings et al. (2014) suggests that the Chinstrap site may represent a northern extension of the dust and DFe dispersal pattern reported by Winton et al. (2014). We estimate an annual accumulation rate of ~0.01 g m^{-2} yr^{-1} at the Chinstrap site from aeolian dust, although we do not have accumulation rate data from the Chinstrap sediment trap with which to compare this estimate. Whether or not locally sourced aeolian sediment this is the main source of the lithogenic sediment in the Chinstrap site remains somewhat of an open question. However, when the relationship between DFe and phytoplankton productivity in McMurdo Sound is considered we suggest this is unlikely to be the case (Fig. 3; Section 4.2.).

4.2. Implications for iron-fertilisation

4.2.1. Contribution of local dust to lithogenic iron

By extrapolating the dust flux trend observed by Chewings et al. (2014) and its associated contribution to DFe (Winton et al. (2014) into the SW Ross Sea, we can estimate the upper bound of the DFe at the Chinstrap site from the debris bands (Fig. 3). Assuming a lithogenic dust flux of ~0.01 g m^{-2} yr^{-1} and an associated total Fe content of 4 % and 11 % of this Fe is soluble (Winton et al., 2014), we estimate a maximum DFe flux of ~2 nmol m^{-2} d^{-1} to the Chinstrap site (Fig. 3). However, when this is considered relative to the spatial distribution of primary production in the SW Ross Sea, using averaged annual Sea-Viewing Wide Field-of-View Sensor

(SeaWiFS) satellite chlorophyll-*a* data, we find that the gradient in increasing chlorophyll-*a* concentration with distance from the debris bands within the McMurdo Sound polynya does not match the pattern of decreasing dust accumulation (Fig. 3). This pattern suggests that DFe from dust not regulate growth in the SW Ross Sea. A seasonal phytoplankton bloom occurs in the McMurdo Sound polynya, SW Ross Sea each summer and is dominated by diatoms. The rate of primary production is greatest in the centre of the McMurdo Sound Polynya. As the dust flux decreases and primary production increases with distance from McMurdo Sound, it is difficult to reconcile these patterns at the Chinstrap site assuming only a local dust source (Fig. 3).

4.2.2. Supporting evidence from the wider Ross Sea

Some further insight into the origin of sediment in the water column in the SW Ross see may be inferred from the data published by Collier et al. (2000). They show a significantly elevated lithogenic accumulation rate in deep AESOPS sediment traps compared to accumulation rates measured in the upper AESOPS sediment traps at other sites in the Ross Sea (e.g. MS-7; 76°30'S, 178°1'W). In addition, lithogenic Fe fluxes between 1-90 µg m^{-2} d^{-1} have been measured for the upper 200 mbsl AESOPS trap (MIS-7b) and 40-850 µg m^{-2} d^{-1} for the deep trap (MS-7a). The greater mass of sediment and lithogenic Fe flux in the deep traps in the Ross Sea highlight that concentrations of suspended sediment in the water column at these sites cannot simply result from sediment input at the surface (whereby the accumulation in each trap would be the same regardless of depth). Instead, the increase in accumulation with depth likely reflects resuspension and horizontal near-bottom transport processes. Whilst we do not have the data to constraint these processes at Chinstrap we infer, based on the provenance of the lithogenic sediment and distance from

known local sources, that the lithogenic sediment accumulating there is most likely dominated by resuspended bottom sediments, potentially from sills either side of the Drygalski Basin but also locally sourced material falling through the water column sourced from atmospheric deposition or ice rafting. Although, no Fe flux data for the Chinstrap site are available, future work could examine the relationship between Fe fluxes in the Chinstrap sediment trap and those DFe fluxes reported for locally derived dust at McMurdo Sound.

Despite McMurdo Sound representing the upper bound of locally derived dust and associated DFe to Antarctic waters, previous studies have ruled out local dust as the major source of DFe supply for phytoplankton blooms in the Ross Sea. Dissolved Fe in McMurdo Sound dust can only support up to 15 % of primary production in the region (Winton et al., 2014). Furthermore, based on regional scale estimates of dust deposition to the Southern Ocean, primary production triggered by long-range transport dust is likely to be less significant than local dust (e.g. Edwards & Sedwick, 2001). Evidence from the extrapolation of the mass accumulation rate to the upper 200 mbsl Chinstrap trap, the sedimentological study of Chewings et al. (2014), sedimentation in the water column, and the low contribution of local aeolian DFe to phytoplankton blooms, suggest that it is unlikely that aeolian dust deposition is the dominant process by which lithogenic Fe is supplied to the water column in the SW Ross Sea. Considered together, these lines of evidence point to a combination of resuspended bottom sediments with smaller additions of local dust sourced from atmospheric deposition or ice rafting as the sources of Fe-bearing sediment to the water column in the SW Ross Sea.

4.2.3. Implications for iron-fertilisation

More broadly, Sedwick et al. (2011) noted that the phytoplankton-Fe limitation must be overcome by continuous replenishment from new sources to sustain the significant biomass observed over summer. They considered the following as potential sources of new DFe: vertical mixing, lateral advection, aerosol input, and dissolution of particulate Fe from any or all of these sources. Consistent with upwelling of DFe as a major source of DFe in the Ross Sea, Marsay et al. (2014) reported the highest DFe concentrations are found within 50 m of the seafloor in the austral summer 2012. Most recently, Gerringa et al. (2015) measured seawater DFe concentrations in the 2013-2014 austral summer, and concluded that DFe from the seafloor and land mass sediments are the main sources of DFe which support phytoplankton in the upper mixed layer of the Ross Sea Polynya in the early summer. Similarly, phytoplankton blooms in the Pennell Bank region of the Ross Sea are supported by upwelling of DFe (Kustka et al., 2015). However, Kustka et al. (2015) also highlight the spatial variability of processes supplying DFe in the Ross Sea. For example, circulation patterns around bathymetric features can alter the input of DFe from increased upwelling rates and higher concentrations of DFe. Numerical modelling of DFe supply by McGillicuddy et al. (2015) suggests that the largest sources to the euphotic zone are wintertime mixing and melting sea ice (e.g. de Jong et al., 2013; Sedwick & DiTullio, 1997) with smaller inputs from Circumpolar Deep Water and from melting glacial ice.

Conclusions

Dust extracted from surface snow on McMurdo Sound sea ice enables us to document the present day provenance of dust reaching the SW Ross Sea. Based on our

476 measurements of Sr and Nd isotopic ratios of dust deposited in surface snow on sea
477 ice at McMurdo Sound and in the Chinstrap sediment trap in the SW Ross Sea we
478 conclude the following:
479
480 1. The Sr and Nd isotopic signature of lithogenic sediment from the upper Chinstrap
481 sediment trap in the SW Ross Sea ($\varepsilon_{Nd}(0)$=3.9, $^{87}Sr/^{86}Sr$=0.70434) matches local dust
482 sources.
483
484 2. McMurdo Sound has been well characterised in terms of the Sr and Nd isotopic
485 composition of locally derived dust deposited on sea ice. Dust found there displays a
486 narrow isotopic field between 0.70533< $^{87}Sr/^{86}Sr$ <0.70819 and -1.1< $\varepsilon_{Nd}(0)$ <3.45 for
487 the bulk fraction and 0.70807< $^{87}Sr/^{86}Sr$ <0.70915 and -0.94< $\varepsilon_{Nd}(0)$ <0.86 for the fine
488 fraction. Due to Sr isotopic fractionation with particle size, the signature of the fine
489 fraction reference adds to the PSA database for comparison to Antarctic ice core dust
490 provenance studies.
491
492 3. Locally derived dust from McMurdo Sound is unlikely to be the major source of
493 DFe for seasonal phytoplankton blooms in the SW Ross Sea. Although, Sr and Nd
494 isotopic ratios of local dust on sea ice show similarities to lithogenic marine sediment,
495 we acknowledge the limited transport distance of coarse-sized dust in this region. As
496 dust transport varies from year to year, we cannot completely exclude the possibility
497 that local dust can contribute to DFe to the greater Ross Sea region although this is
498 not the dominant source of lithogenic Fe.
499

4. We surmise that there is significant remobilisation and upwelling of Fe from the sea floor that contributes to Fe-fertilisation of phytoplankton during the austral summer in the SW Ross Sea.

5. Source information of dust inputs to regions, such as the Ross Sea, improves the ability to predict how such supply will change as the climate changes. As local sources are important to the SW Ross Sea, this data could be included in models that predict changes in snow and ice cover in the region.

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Author Contributions

549 V.H.L.W, G.B.D, C.B.A and N.A.N.B designed the research; G.B.D and C.B.A
550 collected the samples; V.H.L.W, B.D and P.S.A prepared the samples and analysed
551 the data; V.H.L.W, G.B.D, C.B.A, B.D and P.S.A evaluated the data; all authors
552 contributed to the interpretation of the data and the writing of the manuscript.

553 **Table 1: Nd and Sr concentrations and isotopic composition of McMurdo Sound and Chinstrap sediment trap samples analysed in this study.**

Sample	Size (µm)	Location	Date sampled	$^{143}Nd/^{144}Nd$	[a] $\pm 2\sigma_{mean}$ *10^6	[b] $\varepsilon_{Nd}(0)$	[c] $\pm 2\sigma$	C_{Nd} (ppm)	$^{87}Sr/^{86}Sr$	[d] $\pm 2\sigma_{mean}$ *10^6	[e] $^{87}Sr/^{86}Sr$ corrected	[f] $\pm 2\sigma$ *10^6	C_{Sr} (ppm)	Reference
McMurdo Sound														
MPR11-5	Bulk	77° 35.44 S 164° 31.36 E	Nov 2011	0.512580	12	-1.1	0.2	110	0.708158	6.0	0.708186	8	140	This study
MPR11-5	<10	77° 35.44 S 164° 31.36 E	Nov 2011	0.512590	7	-0.94	0.2	220	0.709119	5.3	0.709147	8	320	This study
CRG7-5	Bulk	77° 05.44 S 163° 41.86 E	Nov 2011	0.512633	11	-0.10	0.3	38	0.707607	4.2	0.707635	8	510	This study
CRG7-5	<10	77° 05.44 S 163° 41.86 E	Nov 2011	0.512632	8	-0.12	0.3	68	0.708943	8.0	0.708971	8	570	This study
MPN10-5	Bulk	77° 24.52 S 164° 18.60 E	Nov 2011	0.512735	6	1.9	0.4	40	0.706705	13	0.706733	13	6600	This study
MPN10-5	<10	77° 24.52 S 164° 18.60 E	Nov 2011	0.512682	8	0.86	0.4	20	0.708188	6.4	0.708216	8	910	This study
GH2	Bulk	76° 55.33 S	Nov 2009	0.512650	11	0.23	0.3	76	0.707299	12	0.707327	12	890	This study

		163° 6.17 E											
GH2	<10	76° 55. 33 S 163° 6.17 E	Nov 2009	0.512654	6	0.31	0.3	72	0.708045	5.8	0.708073	8	620	This study
GH9	Bulk	76° 58.36 S 162° 52.80 E	Nov 2009	0.512321	5	-6.18	0.3	37	0.712260	5.0	0.712288	8	290	(Winton et al., 2014)
MIS4	Bulk	77° 40.03 S 166° 35.97 E	Nov 2009	0.512815	4	3.45	0.3	56	0.705303	5.0	0.705331	8	800	(Winton et al., 2014)
MIS23	Bulk	77° 40.03 S 164° 35.79 E	Nov 2009	0.51276	6	2.38	0.3	51	0.705608	5.0	0.705636	8	520	(Winton et al., 2014)
Ross Sea Sediment														
CT1	Bulk	76° 20.5 S 165° 1.78 E	1997	0.512715	7	1.5	0.3	0.4	0.709008	5.1	0.709036	8	110	This study
CT1-leach	Bulk	76° 20.5 S 165° 1.78 E	1997	0.512836	8	3.9	0.3	0.5	0.704314	4.4	0.704342	8	20	This study

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

555 [b)]Nd isotopic ratios expressed as epsilon units $\varepsilon_{Nd}(0) = [(^{143}Nd/^{144}Nd)_{sample}/(^{143}Nd/^{144}Nd)_{CHUR}-1]x10^4$; CHUR, chondritic uniform reservoir.

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

557 [d)]Internal precision, 2 standard errors of the mean.

558 [e)]Corrected to a NBS987 $^{87}Sr/^{86}Sr$ ratio of 0.710245.

559 [f)] Uncertainty estimates based upon external precision for standard runs. Internal precision is used if it exceeds the external.

Figure captions

Fig. 1: a) Map of the SW Ross Sea showing the location of SW Ross Sea Chinstrap sediment trap (CT1). b) Insert of McMurdo Sound within the SW Ross Sea showing location of McMurdo Sound snow on sea ice samples (solid: this study, cross: Winton et al. (2014) and shaded: exposed areas of unconsolidated sediment. Samples are named based on their location i.e. MP: Marble Point; CR: Cape Robert; GH: Granite Harbour; MIS: McMurdo Sea Ice. EAIS: East Antarctic Ice Sheet, MDV: McMurdo Dry Valleys, TAM: Transantarctic Mountains. Transect X-Y shown in red. c) Wind roses illustrating the direction of storm events at Pegasus North and Marble Point automatic weather stations (AWS). Locations of AWS shown in Fig. 1c). Modified from Winton et al. (2014).

Fig. 2: Nd and Sr isotope signature of fine (black triangles) and bulk (white triangles) McMurdo Sound dust, including bulk McMurdo Sound data (GH9, MIS4 and MIS23; Winton et al. (2014)) and leached and unleached Chinstrap sediment trap material. Also plotted are data from Victoria Land potential dust sources that include different parent lithologies located in Fig. 1. (Delmonte et al., 2013; Delmonte et al., 2010; Delmonte et al., 2004) and the isotopic composition of Ross Sea seawater (Basak et al., 2015; Elderfield, 1986). Insert top right: McMurdo Sound dust highlighting the fractionation between fine and coarse particle sizes and a hypothetical mixing line between the two end members MVG and Southern Victoria Land, TAM.

Fig. 3: a) Extrapolation of the annual DFe flux from McMurdo Sound into the SW Ross Sea. DFe data sourced from: Winton et al. (2014). The predicted dust flux at the

585 Chinstrap site is estimated at ~0.01 g m^{-2} yr^{-1} with a corresponding DFe flux of ~2
586 nmol m^{-2} d^{-1}. b) Also shown is the rate of primary production with distance from
587 McMurdo Sound. As the dust flux exponentially decreases, the rate of primary
588 production increases. Primary production inferred from the annual mean chlorophyll-
589 *a* concentration (2000-2009) in the McMurdo Sound polynya (72.0 °S - 78.083 °S,
590 160.916 °E - 179.040 °W) from SeaWiFS satellite data
591 (http://giovanni.gsfc.nasa.gov).

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