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Magnetic properties of aerosol dust in peripheral and inner Antarctic ice cores as a proxy for dust provenance

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ABSTRACT

We use laboratory-induced remanent magnetization of polar ice to measure the rock-magnetic properties of the aerosol dust directly in ice samples. Former studies on Vostok and EPICA-Dome C ice core, recovered on the inner East Antarctic ice sheet, revealed that glacial and interglacial periods of the latter are characterized by distinct magnetic mineralogies at Dome C, which might reflect different dust source areas. In this work we present the first results on glacial and Holocene samples from the TALDICE ice core, collected at the peripheral site of Talos Dome located at high-elevation on the ice sheet close to some ice-free areas of the Transantarctic mountains. Magnetic properties of interglacial samples from both Dome-C and Talos Dome ice cores turned out to have peculiar characteristics that suggest an enhanced concentration of Fe-rich minerals in the aerosol dust, compared to Vostok. The most likely explanation for the extremely high dust magnetization measured in interglacial samples is the presence of volcanic material, although occasional occurrence of meteoritic material (micrometeorites) cannot be ruled out. The volcanic nature of the Holocene aerosol dust and its variability between sites provides further constrains on dust geographic provenance that are complementary to geochemical and physical evidences. Moreover, the calculations of the flux of the highly magnetic dust provide information on wind transport toward the continent interior during the Holocene.

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1. Introduction

In central East Antarctica most of the variability of aerosol dust concentration in ice associated to glacial/interglacial cycles was related to the supply of mineral particles from the remote continental sources of the southern hemisphere, to the snow accumulation rate, and to the atmospheric transport efficiency (e.g., Petit and Delmonte, 2009). Aerosol dust suspended in the troposphere and transported over long distances is uniformly dispersed and can reach the most inner and high-elevation sites in East Antarctica.

According to the current view, the sources of the windborne dust reaching central East Antarctica during glacial times have uniform geochemical properties and consequently their major source area, South America, seem to be relatively well defined (e.g., Basile et al., 1997; Delmonte et al., 2004; Gabrielli et al., 2010). Conversely, the provenance of the aerosol dust reaching Antarctica during interglacial times is not well resolved. A major difficulty in the analysis of interglacial dust is related to the extremely low dust concentrations in firn and ice cores (Petit and Delmonte, 2009). This problem is overcome by magnetic measurements, which are sensitive to extremely small dust concentrations. Magnetic methods, which are particularly effective in recognizing Fe-rich material such as volcanic dust and highly oxidized soils, can thus be used as a tool to discriminate among aerosol dust sources. A considerable difference between glacial and interglacial dust properties was shown by magnetic measurements at Dome-C site (EPICA Dome-C ice core) by Lanci et al. (2008a, 2008b, 2012) which also suggest the volcanic nature of the interglacial dust based on its high values of the dust IRM. This finding is in accordance with the recent growing consensus to the hypothesis that local (Antarctic) source could contribute significantly to the dust supply during interglacial periods and that these sources are volcanic in nature (e.g., Gabrielli et al., 2010; Vallelonga et al., 2010; Delmonte et al., 2013). However, despite the general understanding of long-distance aeolian dust transport to Antartica, little information is available on the transport and deposition of mineral aerosol originating locally in marginal ice-free areas toward the Antarctic interior.

The aeolian transport of particles from Antarctic ice-free areas towards the interior of the ice cap can represent a significant additional aerosol source when the long-range transport is less efficient, such as during interglacials, and its study might provide information about the behavior of regional atmospheric circulation in the past. Understanding the spatial extent where local sources can influence the dust budget on the ice sheet and the climatic condition when this occurred is also helpful for better defining the atmospheric dust cycle in Antarctica and for the interpretation of the dust history at different sites. Here we show new results from TALDICE (TALos Dome Ice CorE drilling project) ice core and compare them with revised data from Vostok and Dome-C

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in order to gather information on the areal distribution during different climatic stages and, indirectly, on the aerosol dust provenance and transport.

2. Material and methods

We measured new ice samples from TALDICE ice core, drilled at Talos Dome (72°49′S, 159°11′E; 2315 m a.s.l.) in the Ross Sea sector of East Antarctica, Northern Victoria Land (Fig. 1). A total of 26 TALDICE ice samples weighting from ~20 to ~40 g were taken at core depths ranging from 490 m to 1292 m, corresponding to the current interglacial period and glacial Marine Isotopic Stage (MIS) 3 and 4 (Stenni et al., 2011). Dating for TALDICE (TALDICE-1 chronology) was set up using stratigraphic tie points, implementing a new probabilistic inverse approach for ice core dating based on Bayesian inference aiming at finding the best compromise between an ice flow model scenario setup a priori and chronological information from tie points, and evaluating the quality of the chronology a posteriori (Buiron et al., 2011).

Standard sample preparation consisted of core cutting and decontamination with ultra-pure water in clean room (e.g., Lanci et al., 2012).

The Isothermal Remanent Magnetization (IRM) acquisition and measurement procedure for ice samples from Talos Dome ice core are equivalent to that described in previous papers (e.g., Lanci and Kent, 2006; Lanci et al., 2012). IRMs were induced in whole-ice samples at low temperature. In order to avoid the immersion of ice samples in liquid nitrogen with consequent fracturing, samples were cooled in liquid nitrogen vapours until reaching a stable temperature of about 170 K. Repeatable results indicate that this temperature is sufficient to prevent the physical rotation of magnetic particles in the ice matrix (Lanci et al., 2001, 2012). Ice samples were magnetized using a pulse magnetizer and the remanent magnetization was measured using a 2G superconducting magnetometer with DC-SQUID sensors at the ALP laboratory. The measurement procedures were performed as quickly as possible to avoid significant re-warming of the samples.

An IRM in a maximum field of 1 T was first induced in each sample; subsequent IRMs were induced in the opposite direction with stepwise increasing fields to allow the calculation of the coercivity of remanence (H_{cr}) ; the exact field value of H_{cr} is interpolated between zero-crossing IRM values. The maximum IRM was also remeasured after allowing the sample to re-equilibrate to the freezer temperature (~255 K) for about



Fig. 1. Location map of the three Antarctic cores considered in this study. The dashed polygon represent the possible source area of volcanic dust according to Delmonte et al. (2013)).

4 h; the increase in temperature from 170 K to 255 K causes thermal relaxation of the remanent magnetization carried by very small magnetic particles, thereby decreasing the remanent magnetization. The fraction of magnetic particles whose remanent magnetization relaxes at freezer temperature is referred to as superparamagnetic and the IRM carried by this fraction can be calculated as the difference between magnetic measurements taken before and after thermal relaxation. We refer to the fraction of IRM that remains after warming to ca. 255 K as IRM_{255 K} to distinguish it from the IRM acquired at 170 K (IRM_{170 K}), which includes the superparamagnetic fraction. Previous studies (Lanci and Kent, 2006; Lanci et al., 2007, 2012) have argued that most of the superparamagnetic fraction of the polar ice is carried by particles of extraterrestrial origin and for this reason it is disregarded in this study. Ice samples were returned after magnetic measurements for dust concentration measurements.

Insoluble Dust Concentration (IDC) and size distribution were analyzed on the same sample measured for magnetic properties, after additional decontamination of ice in clean room and melting at room temperature. Insoluble microparticles were measured with a Multisizer[™] 3 Coulter Counter[®] which can detect insoluble material with equivalent spherical diameters of 1 to about 30 µm. The dust mass was calculated assuming that mineral grains have an average density of 2.5 g/cm^3 . The dust concentrations obtained in our samples are perfectly compatible with the extensive measurements published by Albani et al. (2012). The dust volume-size distribution from TALDICE shows the typical lognormal distribution with a variable modal value around 2 µm, as observed in Dome C, but also peculiar characteristics related to the presence of dust particles larger than 5 µm in diameter, representing only a very low number of counts but contributing significantly to the total dust mass and flux (Albani et al., 2012; Delmonte et al., 2010, 2013).

Dust magnetization in ice samples was computed by dividing the ice $IRM_{255 \text{ K}}$ by the IDC measured in the very same samples, in the case of Talos Dome, or in adjacent ice samples, as in the case of EPICA-Dome-C (Lanci et al., 2008a, 2008b). In the Vostok ice core, the averaged values of dust concentration for each group of samples were obtained from the published data of Petit et al. (1999) as explained in Lanci et al. (2012). Unfortunately only an incomplete set of H_{cr} measurements was available for the Vostok samples.

Both magnetic and dust concentration measurements have analytical errors that, in samples with the lowest concentrations, might lead to notable uncertainties in the values of dust IRM_{255 K}. Possible sources of errors are: i) errors in determination of the magnetic moment, and ii) errors in the determination of the dust mass. Between these two the second one is generally the largest; the magnetic moment can be measured very precisely with cryogenic magnetometers and the routinely repeated measurements, which would account for possible temperature bias and laboratory contamination, demonstrate that differences do not exceed 10%. Errors in dust mass measurements made with the Coulter counter are related to the extremely small dust concentrations in Holocene ice; for example, in the case of EPICA-Dome C the internal variability among three replicate and consecutive dust mass measurements was around 20% for the Holocene (Delmonte et al., 2002). Furthermore, possible errors arise from the unknown dust density, and from the insensitivity to very fine (<1 µm) grains and the consequent underestimation of the dust mass in the smallest size bins. We approximate that, in the worst-case scenario and considering the 2 sources of errors as independent, the dust IRM_{255 K} could be overestimated by a maximum of 25%.

3. Results and discussion

A strong linear relationship between Ice-IRM_{255 K} and IDC indicates uniform magnetic properties and the slope of the regression line can be taken as the average measurement of the Dust-IRM_{255 K}. We use this criterium on the Vostok ice core, where the correlation is highly significant (Fig. 2a), to argue that no major magnetic mineralogy changes



occurred during the last glacial to Holocene transition at this site. In other words, the magnetic properties of the dust in the Vostok ice are very similar during the last glacial and interglacial stages. This situation, which is completely equivalent to what was observed in the Greenland ice sheet (Lanci and Kent, 2006), justifies the use of averaged values for Vostok H_{cr} and dust-IRM_{255 K} as plotted in Fig. 3. More peripheral sites have a different behavior. Samples from Dome-C plotted in the same ice-IRM_{255 K} vs. IDC graph (Fig. 2b) show two different trends for glacial and interglacial samples. The linear relationship between ice-IRM and IDC is partially preserved with samples from the same climatic stage, although with lower correlation coefficients (Fig. 2b). The different slopes of the two regression lines in Fig. 2b indicate a higher magnetization of interglacial dust, indicating that higher magnetic dust fluxes reach Dome-C during interglacial periods, as also pointed out by Lanci et al. (2008a, 2008b, 2012). Samples from Talos Dome (Fig. 2c) are characterized by a more complicated situation with largely variable dust magnetic properties even within the same climatic stage. This variability prevents any significant correlation between ice-IRM and IDC, and reflect an increased source mixing compared to Dome-C.

Source mixing effects are best illustrated by a dust-IRM vs. H_{cr} plot (Fig. 3) that summarizes dust magnetic properties in ice samples and where both parameters are independent from dust concentration and snow accumulation rates. For comparison purpose, the statistical distributions of H_{cr} and saturation IRM of antarctic micrometeorites computed from Miller Butte, Walcott Neve and Frontier Mountain data sets (Suavet et al., 2009) are also shown. Micrometeorite saturation IRM from Miller Butte, Walcott Neve and part of Frontier Mountain sets are taken directly from published data, while the missing saturation IRM data from the Frontier Mountain set were inferred using the linear relationship between magnetic susceptibility (χ) and saturation magnetization (M_s) ($\chi = M_s 1.53 \times 10^5$) and the published Mrs/Ms ratio (Suavet et al., 2009).

Dust in Antarctic ice displays a moderate variability of H_{cr} and a very large variability of dust-IRM, whose values span over two orders of magnitude. Measurement errors and laboratory contamination, which are the possible source errors in dust-IRM measurements can be disregarded because, as discussed before, in the worst-case scenario their contribution to dust-IRM variability does not exceed 25%. The dust-IRM variability is thus to be regarded as a characteristic of the ice core dust record and can be related to the spacial and temporal variability of atmospheric dust transport and deposition patterns at the site. We note that saturation IRM intensities >0.5 A m²/kg are unusually large for geologic materials and rare in sedimentary materials, including unconsolidated sediment, but typical of highly magnetic volcanic rocks. Values > 2 A m²/kg are even rarer and typical in micrometeorites (Suavet et al., 2009). Large dust-IRM values of interglacial samples could therefore be explained by large volcanic or meteoritic contributions to aerosol dust during this climatic stage.

The expected contribution from micrometeorites can be estimated from the empirical cumulative size frequency distribution (i.e., the number of micrometeorites with a mean diameter larger or equal to diameter D falling per year per square meter) based on Antarctic micrometeorite collections (Suavet et al., 2008). The distribution of the finest grain sizes of 40 μ m \leq D \leq 200 μ m is described by the equation F(\geq D) = $6 \times 10^{-5} D^{-0.87}$ (Suavet et al., 2008) from which we extrapolate the mean average volume of micrometeorites in the diameter range from 20 nm to 1 μ m found in a 50 g ice sample. The spherical equivalent volume of micrometeorites is computed using a conservative snow

Fig. 2. Ice-IRM_{255 K} versus dust concentration at the three different sites. The good degree of correlation (r = 0.94) at Vostok (a) indicates constant magnetic properties of aerosol dust during the investigated period spanning though the lasts glacial and interglacial. The low the degree of correlation in Dome-C (b) and the clear lack of correlation at Talos Dome (c) are interpreted as a consequence of a mixed sources of magnetic minerals. The slope coefficient b in panels (a) and (b) can be considered as average values of dust-IRM. Also note that glacial (MIS 3 and 4) samples at Talos Dome do not reach the full-glacial dust concentrations.



Fig. 3. Representation of the magnetic properties of Antarctic aerosol dust measured in ice samples by the H_{cr} versus $IRM_{255 \text{ K}}$ diagram. Vostok data correspond to the average of 47 samples out of the 70 ice samples measured in Lanci et al. (2007) for which H_{cr} measurements are available. Error bars represent $\pm 1\sigma$. Dome-C and Talos Dome date are divided in interglacial, glacial (MIS 3 and 4) and fully glacial (MIS 2) samples in order to differentiate the magnetic properties according to climatic stages. Histograms at the top and right represent the distribution of saturation IRM and H_{cr} , respectively, of Antarctic micrometeorites from Suavet et al. (2009).

accumulation rate of 2 cm/yr is \sim 5.5 \times 10⁻²¹ m³ and their maximum IRM contribution, generously estimated using the largest value of micrometeorite IRM distribution of 10 A m²/kg (Fig. 3) and a density of 7000 kg/m³, is $\sim 3.8 \times 10^{-16}$ A m²/kg. Alternatively one can use the typical magnetite parameters ($M_s = 480,000 \text{ A/m}, M_{rs}/M_s = 0.3$) and obtain the comparable value of $\sim 7.9 \times 10^{-16}$ A/m (or A m²/kg since ice density is ~1). These estimates are 7 orders of magnitude smaller than the smallest magnetization measured in ice. Considering the average flux in the range between 1 µm and 10 µm diameter and applying the above considerations we obtain an average magnetization of ~5.1 \times 10⁻¹⁴ A m²/kg, or ~1.0 \times 10⁻¹³ A/m using the magnetite hypothesis, that are still negligible compared to the ice IRM ranging from 4×10^{-9} to 1×10^{-7} A m²/kg. The chance to find one occasional particle with $D \ge 5 \ \mu m$ in an ice sample, calculated from the size distribution above and sample volume, is ~0.006, which can be considered a rare event.

The above considerations suggest that, despite compatible magnetic properties (Fig. 3), micrometeorites cannot explain the magnetic properties of Talos Dome interglacial samples because, according to the current flux estimates, even the weak interglacial dust-IRM (averaging ~40 × 10⁻⁹ A m²/kg) is too large to be the consequence of primary micrometeorites deposition. Furthermore, micrometeorite fallout could not account for differences between ice-IRM intensities of Vostok and other sites.

New data from Talos Dome share two clusters of H_{cr} and dust-IRM values for glacial and interglacial samples that are similar to those of Dome C. Interglacial samples at Dome-C and Talos Dome are characterized by lower H_{cr} and especially by high $IRM_{255 K}$ intensities. The cluster formed by full glacial, MIS 3 and MIS 4 samples is centered on the Vostok average value indicating that dust properties during cold climatic periods are virtually identical at all sites, an evidence that is coherent with the very similar isotopic composition of dust at the three sites during glacial periods (Delmonte et al., 2010). In line with geochemical and physical evidences (e.g., Basile et al., 1997; Delmonte et al., 2010) we consider this flux of magnetic dust particles, which are characterized by low dust-IRM_{255 K} and relatively high H_{cr}, prevalently controlled by long-distance transport. By contrast the absence of high IRM_{255 K} and low H_{cr} dust in Vostok interglacial ice suggests that the highly-magnetic interglacial dust in not transported over long distance and thus advocate for shorter distance transport from local (i.e., Antarctic) volcanic sources. This hypothesis is consistent with the evidence of higher superchondritic fluxes of siderophilic elements to Dome-C than Vostok during the Holocene (Gabrielli et al., 2006).

For these reasons we favor a volcanic origin of the highly-magnetic dust accumulated at Talos Dome and Dome C during the Holocene. The extensive presence of geochemically-heterogeneous volcanic particles probably remobilized from different volcanic sources was recently found in Holocene snow from Talos Dome (Delmonte et al., 2013) by microscopic observations. The volcanic origin of the highly magnetic dust trapped in interglacial ice from Talos Dome indirectly gives constrains on its possible geographic provenance, which is conceivably located on the Transantarctic mountains (Fig. 1), as proposed by Delmonte et al. (2013) on the basis of geochemical and grain size data. We suggest that the extremely large dust-IRM observed in a few samples could be explained either by the presence of a few large volcanic particles undetected by coulter counter or by the presence of a significant amount of micrometeorites that, given the low probability of direct deposition, have originated from aeolian reworking of micrometeorites deposits or from concentration due to ice ablation.

The variability of dust magnetic properties (H_{cr} and dust-IRM) can be explained with the mixing of two or more sources with distinct end-member properties. The simplest case of two end-members would include a long-distance source that is dominant during glacial times, and a second source contributing to interglacial dust. Mixing ratios cannot be determined because samples containing only one end-member are not available. However, careful comparison of Talos Dome and Dome-C sites show a few differences that might offer a clue for a more complete interpretation. Although no samples from MIS 2 have been measured yet on the TALDICE core, the MIS 3-4 samples are more dispersed compared to Dome-C (Fig. 3) and sometimes have rather high dust-IRM_{255 K} intensities. The plot of dust-IRM_{255 K} versus age for the last glacial and interglacial stages (Fig. 4) gives a different view of the variability of dust-IRM_{255 K} and shows that during MIS 3–4 periods the dust-IRM_{255 K} of Talos Dome dust is often higher than Dome-C. Although the observation are limited to a few samples, we argue that ingression of highly-magnetic dust could have started earlier at Talos Dome compared to Dome-C.

The flux of highly-magnetic dust can be estimated from the ice-IRM_{255 K}. Because of the large natural variability affecting the dust transport and deposition process we have considered the averaged value over the last 10,000 years, a long period with relatively uniform climatic conditions, and use the standard deviation as a rough measure of this natural variability. Since highly-magnetic particles do not reach Vostok it seems reasonable to assume that the average magnetization flux during the last 10,000 years in Vostok, which is calculated as the average ice-IRM_{255 K} multiplied by ice accumulation rate, represents a good estimate of the long-transported Holocene background. The same calculations made for the Dome-C and Talos Dome sites show that this background is small compared to the fluxes at these sites, which are more than 6 times and 30 times larger, respectively (Table 1). Furthermore they show that the Holocene high-magnetization flux and its variability are strongly site-dependent, thus corroborating the hypothesis

of a local source. Our calculations indicate that the flux of highlymagnetic dust reaching Talos Dome during the Holocene is, on average, ca. 5.8 fold higher than that reaching the more internal – and higher altitude – site of Dome-C, providing new valuable information on the dust transport mechanism from the outer to the inner continental area.

4. Conclusions

With the exception of the innermost Vostok site, magnetic properties of ice show a clear difference between glacial and interglacial stages, which are more pronounced at the external site of Talos dome. The interglacial periods are characterized by low H_{cr} and high dust-IRM_{255 K}, but the exact timing and abundance of the highly magnetic dust flux appear to be site dependent. Rare events of highly magnetic dust inputs occur at the Talos dome starting during MIS 3. On the other hand, similar events are found only during the interglacial Holocene period at Dome-C. The highly magnetic dust never reaches the innermost high elevation Vostok site even during interglacials.

The magnetics analysis points toward a volcanic nature of Holocene dust source and shows that it gave an important contribution to the dust supply during interglacial ages. Even though volcanic dust seems to be the most likely source of the highly magnetic aerosol dust, an occasional contribution of meteoritic dust (micrometeorites) cannot be ruled out and it might be a major contributor in few occasional samples. Parallel results from Delmonte et al. (2013) who found the pervasive presence of volcanic particles in Holocene ice from Talos Dome corroborated our finding. An important consequence of the recognition of the volcanic nature of aerosol dust is the precise constrains on its source area which is possibly located on the Transantarctic Mountains.

The picture revealed by magnetic analysis of aerosol dust suggests thus the coexistence of a main long-distance dust flux and a sitedependent flux from local sources, in agreement with independent observations (Delmonte et al., 2010; Albani et al., 2012). The former being the most active during the glacial times possibly because of the enhanced transport and availability of dust sources, the latter that became predominant only in interglacial time due to a combined effect of increased local flux and decreased long-distance flux.

We used the averaged magnetization flux as a proxy of the intensity and frequency of volcanic dust transport at the different sites in order to gather constrains on the main wind transport toward the continent interior in the past. According to back trajectory calculation (Delmonte et al., 2013), the frequency of events suitable for peripheral dust transport to Talos Dome appears relatively high, while transport to Dome C is rarer for present day conditions and seems to be related to random events. In contrast our results suggest that during



Fig. 4. Intensity of IRM_{255 K} at Talos Dome and Dome-C versus time and climatic stages. Although the interpretation is based on a limited number of samples, the diagram suggests larger magnetization peaks at Talos Dome, and highly magnetized samples starting during MIS 3. Talos Dome ages are from Albani et al. (2012) for the Holocene and Buiron et al. (2011) for the interstadial samples; Dome-C ages are from Lemieux-Dudon et al. (2010).

Table 1

lce magnetization and magnetization fluxes during the last 10,000 years. The 1σ does represent the natural variability within the investigated time period and not the uncertainty of the measurements.

	Ice Equivalent. Acc. Rate (kg $m^{-2} yr^{-1}$)	Average ice-IRM _{255 K} \pm 1 σ (10 ⁻⁹ A m ² kg ⁻¹)	Magnetization flux $\pm 1\sigma$ (10 ⁻⁹ A yr ⁻¹)	Magnetization flux with Vostok background removed $(10^{-9} \text{ A yr}^{-1})$
Vostok	25 (Petit et al., 1999)	2.47 ± 1.28	61.7 ± 32.0	_
Dome-C	29 (Lemieux-Dudon et al., 2010	13.9 ± 6.8	403.1 ± 197.2	341
Talos Dome	72 (Buiron et al., 2011)	28.1 ± 22.7	2023.2 ± 1634.4	1961

the Holocene dust transport toward the continent interior was not uncommon even for more internal sites such as Dome-C but, at least in the Holocene, they never reached the innermost Vostok site. The magnetic flux calculation shows that Holocene volcanic dust transportation to Talos Dome is 1–6 times larger than that to Dome-C, which is comparable to the ratio of total dust flux (e.g., Albani et al., 2012) as expected when dust magnetic properties are similar. This ratio is significantly different from the ca. 1 to 40 frequency ratio of dust transport event according to back trajectory calculation in present day conditions (Delmonte et al., 2013) and provide new experimental data to model dust transport under average Holocene conditions.

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