GEOPHYSICAL RESEARCH LETTERS, VOL. 33, L13308, doi:10.1029/2006GL026480, 2006

Meteoric smoke fallout revealed by superparamagnetism in Greenland ice

L. Lanci[1] and D. V. Kent[2,3]

Received 2 April 2006; revised 21 May 2006; accepted 31 May 2006; published 11 July 2006.

[1] Meteoric material reaching Earth contains an appreciable percentage of iron, much of which can be oxidized into nanometric-size particles produced by ablation and subsequent condensation in the atmosphere. New measurements of isothermal remanent magnetization (IRM) show that magnetic particles of extraterrestrial origin can be distinguished from terrigenous particles based on their smaller superparamagnetic (SP) size as inferred from magnetic relaxation and by the poor correlation of the SP fraction with dust contents. The magnetic relaxation data suggest that extraterrestrial magnetic particles are in the size range of about 7–17 nm, which is compatible with the expected size of condensed particles. The concentration of extraterrestrial material in Greenland ice was estimated from the magnetic relaxation data. Assuming an iron content typical of average chondritic composition, the results correspond to a particles concentration of 0.78 ± 0.22 ppb for Greenland, good agreement with results based on iridium concentrations in NGRIP ice samples. **Citation:** Lanci, L., and D. V. Kent (2006), Meteoric smoke fallout revealed by superparamagnetism in Greenland ice, *Geophys. Res. Lett.*, *33*, L13308, doi:10.1029/2006GL026480.

1. Introduction

[2] Atmospheric aerosol reaching remote regions like Greenland is mainly composed of soil dust originating in desert areas [e.g., *Biscaye et al.*, 1997; *Bory et al.*, 2003; *Delmonte et al.*, 2004]. However, there are other sources, like volcanic and extraterrestrial, whose contributions to atmospheric aerosol are generally poorly known. Dust from all these sources will include a variable percentage of iron [*Jickells et al.*, 2005]. *Lanci et al.* [2001] have shown that IRM measurements on ice samples provide an indirect estimate of the amount of iron oxides in atmospheric dust. The IRM data and dust concentrations in Greenland ice accumulated during the last glacial and the Holocene showed a good linear correlation, giving an average dust magnetization compatible with that of Chinese loess [*Lanci et al.*, 2004], which originates from the same source material [*Biscaye et al.*, 1997]. However, the regression line did not go to the origin, indicating that Holocene ice has an inordinately large magnetization compared to its independently measured low dust concentration. *Lanci et al.* [2004] proposed two possible explanations for this "background magnetization": either there is a significant amount of systematically undetected dust mass or there is a nearly constant flux of more highly magnetic extraterrestrial particles (volcanic contributions, which may also be highly magnetic, can be excluded due to their very episodic occurrence). Much of extraterrestrial material may be in the form of interplanetary dust particles of micrometric size, or meteoric smoke consisting of meteor ablation products that occur as nanometric-size particles [*Hunten et al.*, 1980]. Although direct measurements of meteoric smoke providing information on composition, hence proving their extraterrestrial origin, are not available, recent papers yielded indirect evidences for the existence of these particles [*Lynch et al.*, 2005; *Rapp et al.*, 2005]. Such small particles are rare in terrestrial aerosols, which are usually characterized by much larger grain-sizes, for example, the size of common soil and desert dust particles that are transported over long distances in the atmosphere ranges from 0.1 to 20 μm, with a median diameter of 1.5–3 μm [*Duce*, 1995; *Jickells et al.*, 2005; *Pye*, 1987]. Such aerosols can stay suspended for hours to weeks, allowing long-range transport on scales of thousand of kilometers. In contrast, the nanometric-size particles produced as meteoric smoke can be transported at high altitude in the atmosphere and have very long residence times, and possibly be removed efficiently only by wet deposition [*Tegen and Fung*, 1994].

2. Measurements Procedure and Results

[3] To help resolve the origin of the background magnetization, we performed new experiments on 32 ice samples from the N-GRIP ice core (Greenland) taken at various levels spanning the Holocene (interglacial) and last glacial periods. Measurements presented here (Table 1) were made on a significant subset of surviving samples for which IRM measurements and dust contents were reported previously [*Lanci et al.*, 2004]. In those measurements, IRMs were induced with a pulse magnetizer with the ice samples at liquid nitrogen temperature (77 K) and measured immediately. The ice magnetizations and dust concentration were then averaged over about 60 cm of core length to address the likelihood of inhomogeneous distribution of dust and magnetic mineral concentrations on the centimeter scale. For the new measurements, the cleaning procedure was significantly improved by adding multiple baths in pure propanol and by handling the samples under a class 100 laminar flow hood. Moreover, a quicker measurement routine reduced the warming of the samples in the magnetometer. The new measurements of ice magnetization were made after the cleaned and reweighed samples were

---

[1] Faculty of Science and Technology, University of Urbino, Italy.

[2] Department of Geological Sciences, Rutgers, The State University of New Jersey, Piscataway, New Jersey, USA.

[3] Also at Lamont-Doherty Earth Observatory of Columbia University, Palisades, New York, USA.

0094-8276/06/2006GL026480$05.00

Table 1. Mean and Standard Deviation of Dust Contents and Magnetization (±1σ Standard Deviation) for Different Climatic Periods at NGRIP

Climatic Period	Depth, m	Dust, μg/kg [*Lanci et al.*, 2004]	Total IRM, Am^2/kg [*Lanci et al.*, 2004]	Total IRM, Am^2/kg	Stable IRM, Am^2/kg	SP IRM, Am^2/kg
Pre-Boreal	1425.8	70.4 ± 24.1	$3.12 \pm 1.93 \times 10^{-8}$	$1.65 \pm 0.73 \times 10^{-8}$	$4.14 \pm 2.04 \times 10^{-9}$	$1.23 \pm 0.67 \times 10^{-8}$
Younger Dryas	1512.3	849.7 ± 479.6	$3.65 \pm 1.66 \times 10^{-8}$	$2.52 \pm 0.75 \times 10^{-8}$	$8.56 \pm 3.94 \times 10^{-9}$	$1.66 \pm 0.57 \times 10^{-8}$
Bølling	1546.85	273.1 ± 198	$2.94 \pm 1.82 \times 10^{-8}$	$1.74 \pm 0.58 \times 10^{-8}$	$3.64 \pm 2.71 \times 10^{-9}$	$1.38 \pm 0.49 \times 10^{-8}$
LGM	1825.8	6480.1 ± 1649	$8.79 \pm 1.79 \times 10^{-8}$	$9.68 \pm 2.09 \times 10^{-8}$	$5.34 \pm 1.12 \times 10^{-8}$	$4.34 \pm 1.16 \times 10^{-8}$
Glacial Stage 2	1856.05	2401.6 ± 699	$4.83 \pm 0.70 \times 10^{-8}$	$4.29 \pm 0.63 \times 10^{-8}$	$2.10 \pm 0.44 \times 10^{-8}$	$2.17 \pm 0.34 \times 10^{-8}$

given another maximum IRM (0.8T), measured, and then allowed to re-equilibrate to the freezer temperature (−20 C, ca. 256 K) for a minimum of a few hours to overnight. For practical reasons [see *Lanci et al.*, 2001] the samples were cooled again to 77 K inside a magnetically shielded room just before remeasurement. The remeasured values always show a decrease in remanent magnetization. We attribute the decrease to thermal relaxation of the finest magnetic particles, referred as the super-paramagnetic (SP) fraction, resulting from storage of the sample at the higher ambient temperature. The contribution of the SP fraction in each sample was computed from the difference between measurements taken before and after thermal relaxation by warming at 256 K; the latter is referred to as the "stable magnetization" (SM). The total magnetization is thus the sum of the SP and SM components.

[4] Particle volume rather than mineralogy is the main factor controlling thermal stability of magnetic particles [*Neel*, 1955], hence it was possible to estimate the size of the SP particles from the thermal relaxation. Chemistry of Fe in the mesosphere can be rather complicated [*Helmer et al.*, 1998] but IRM acquisition curves in Greenland ice [*Lanci et al.*, 2004] and analysis of atmospheric dust particles [*Rietmeijer*, 2001] suggest that the particles are mostly made of maghemite (γFe_2O_3) or magnetite (Fe_3O_4). Assuming this composition with a coercivity, H_c ranging from 100 mT to 200 mT [*Tauxe*, 2002] and using a frequency factor, $f_0 = 1 \times 10^9$ [*Moskowitz et al.*, 1997], calculations suggest that the SP particles range from about 7 nm to 17 nm in equivalent diameter for spherical shapes. Similar calculations for high-coercivity minerals such as hematite (αFe_2O_3) or goethite ($\alpha FeOOH$) that could represent minor magnetic minerals in ice would result in diameters ranging from 5 nm to 30 nm; the wider size range is due to their more variable magnetic properties. However, because of their low magnetization and high coercivity these minerals are not expected to give a large contribution to the magnetization measured in ice.

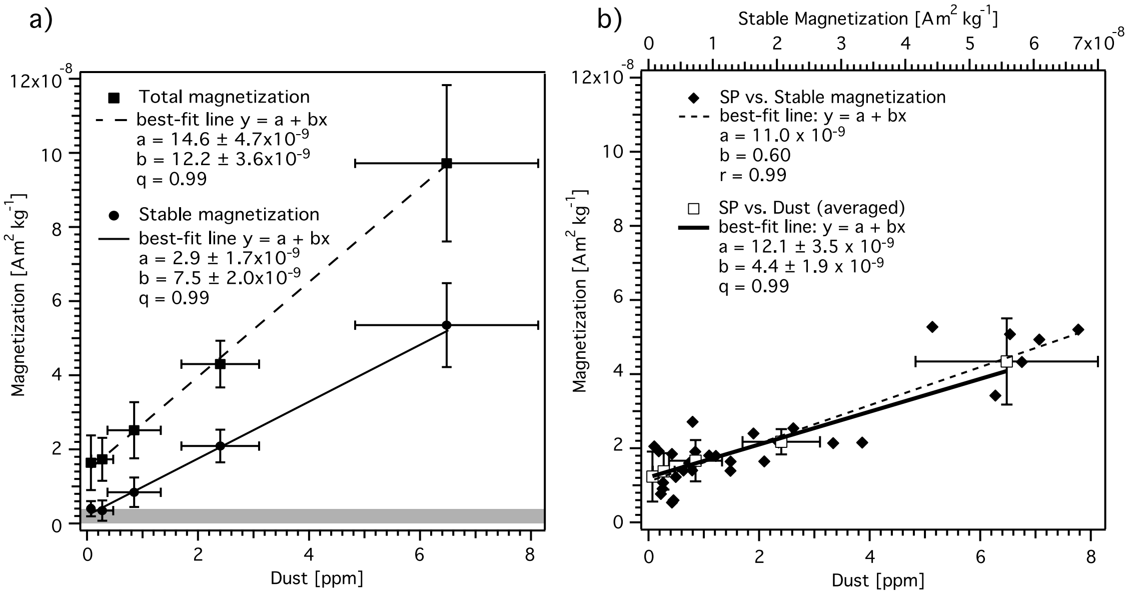

Figure 1. (a) Ice magnetization versus dust concentration in N-GRIP ice core, based on remeasurements and new experiments on samples reported by [*Lanci et al.*, 2004]. The y-axis intercept of the best-fit line for total magnetization is significant but for the stable magnetization (SM, after magnetic relaxation) is indistinguishable from experimental noise (gray band), which is of about 4×10^{-9} A m^2/kg [*Lanci et al.*, 2004]. Best-fit lines were calculated taking in account errors in both coordinates [*Press et al.*, 2002]. (b) SP magnetization plotted with respect to dust concentration and stable magnetization (SM). Data in the SP-dust correlation represent average values within 60 cm-thick intervals [*Lanci et al.*, 2004]. The ±1σ (standard deviation) values are plotted as error bars and used to estimate the errors in the best-fit line, which was calculated taking in account errors in both coordinates. Single data points are plotted in the SP−SM correlation.

Table 2. Magnetization, Iron and Meteoric Smoke Concentrations, (±1σ, Standard Deviation) Calculated Using Magnetic Relaxation Method[a]

Description	Value
Average SP (intercept), 10^{-9} A m^2/kg	12.1 ± 3.5
Fe mass concentration, ppb	0.19 ± 0.06
Meteoric smoke concentration assuming ordinary chondrites with Fe 24%, ppb	0.78 ± 0.25
Meteoric smoke concentration assuming CI chondrites with Fe 19%, ppb	1.0 ± 0.29

[a] Iron concentration was computed assuming it is carried by magnetite, meteoric smoke concentration was computed from Fe concentration assuming chondritic composition.

[5] With the removal by thermal relaxation of the nanometric SP contribution, the resulting average SM component shows a good correlation with average dust concentration [from *Lanci et al.*, 2004] and importantly, the intercept of the best-fit line becomes statistically indistinguishable from measurement noise with no significant background magnetization (Figure 1a). This is expected for magnetic particles that have the same provenance as the dust. We therefore conclude that there is no undetected dust mass for particles larger than about 17–30 nm (depending upon composition). Moreover, the distribution curve of aerosols in Greenland ice, with its much larger average grain size [*Steffensen*, 1997], also cannot accommodate a large fraction of such small particles. Compared to the previous data [*Lanci et al.*, 2004], the new measurements of total magnetization from N-GRIP have a smaller scatter as indicated by the standard deviations, especially for the Holocene samples which also show a ~30% lower average magnetization as a result of improvements in cleaning and measuring techniques. Nevertheless, the dust magnetization of about 1.2×10^{-2} A m^2/kg calculated for the SM is close to that previously computed from the total magnetization (0.90×10^{-2} A m^2/kg [*Lanci et al.*, 2004]) and compatible with that of Chinese loess.

3. Discussion and Conclusion

[6] The background magnetization is carried by the SP fraction with magnetic grain sizes estimated to range from about 7 nm to 17 nm, which are compatible with particles of extraterrestrial origin in meteoric smoke. There is still a weak dependence of the SP on dust (Figure 1b), suggesting that part of the SP component, probably from particles at the higher end of the estimated grain-size range, is of terrigenous origin. Nevertheless, the y-axis intercept can be taken as the best estimate of the dust-independent background magnetization, which we interpret as due to meteoric smoke. Alternatively, the SM fraction could be assumed to be directly proportional to and thus a proxy for the terrigenous contribution. Both approaches (Figure 1b) give practically equivalent results suggesting that the average SP magnetization that is predominantly due to meteoric smoke is about 12×10^{-9} A m^2/kg.

[7] At liquid nitrogen temperature (77K), the SP magnetic particles behave as stable single domain grains. Iron concentrations corresponding to the SP remanent magnetization values were calculated assuming that this fraction is carried by magnetite with a remanent to saturation magnetization ratio $M_r/M_s = 0.5$, which is appropriate for a random dispersion of single domain particles. We obtained Fe concentrations of 0.19 ± 0.06 ppb from the SP_S component (Table 2). *Gabrielli et al.* [2004] estimated the concentration of meteoric smoke in Greenland ice core from measurements of platinum-group elements. From Ir concentrations ranging from 0.3 to 0.6 fg/g they measured in Holocene ice and using a CI chondritic composition for Ir (481 ppb) and Fe (19.04%) [*Anders and Grevesse*, 1989], the expected Fe concentration in ice from meteoric smoke would be from about 0.12 ppb to 0.24 ppb. These values are compatible with our estimates based on the magnetic relaxation method, which is thus considered effective in estimating the amount of extraterrestrial material.

[8] We estimated smoke concentration from the magnetic method by assuming that all Fe available in CI chondrites is oxidized. Although this may not be completely realistic, the large majority of Fe in the atmosphere is available for oxidation [*Plane*, 2003] and in any case, the error introduced with this assumption is small compared to the uncertainly caused by the type of deposition. The estimated smoke concentration from the magnetic method is 1.0 ± 0.3 ppb; the slightly higher contents of iron available for oxidation (FeO and metallic iron) of about 24% in ordinary chondrites [*Wilkison and Robinson*, 2000], which are the most common type of meteorites and thus the principal source of meteoric smoke, would suggest a somewhat lower expected smoke concentration of 0.78 ± 0.25 ppb. Following [*Gabrielli et al.*, 2004], we can calculate the accretion rate assuming a snow deposition rate of 160 kg/m^2/yr for the N-GRIP ice core [*North Greenland Ice Core Project members*, 2004] and obtain an accretion rate of 64 ± 20 kt/yr. Our result is in substantial agreement with the estimate of 78 ± 30 kt/yr made by [*Gabrielli et al.*, 2004], which are somewhat higher yet not significantly different than accretion rate estimates of 40 ± 20 kt/yr from direct measurements on the long-duration exposure facility satellite [*Love and Brownlee*, 1991]. An important source of uncertainty in our estimate of accretion rate is the type of deposition (wet or dry) for the meteoritic smoke. We have assumed dry deposition to allow direct comparison with the estimate made by [*Gabrielli et al.*, 2004] even though wet deposition may be more common [*Tegen and Fung*, 1994] but which requires further assumptions that are presently difficult to evaluate.

[9] **Acknowledgments.** We thank Pierre Biscaye for help with sample handling, the journal reviewer for constructive comments, and the U.S. National Science Foundation-Office of Polar Programs (grant OPP-0424940) for support of this research. Lamont-Doherty Earth Observatory contribution 6914.

References

Anders, E., and N. Grevesse (1989), Abundances of the elements: Meteoritic and solar, *Geochim. Cosmochim. Acta*, *53*, 197–214.

Biscaye, P. E., F. E. Grousset, M. Revel, S. Van der Gaast, G. A. Zielinski, A. Vaars, and G. Kukla (1997), Asian provenance of glacial dust (stage 2) in the Greenland Ice Sheet Project 2 Ice Core, Summit, Greenland, *J. Geophys. Res.*, *102*(C12), 26,765–26,781.

Bory, A. J.-M., P. E. Biscaye, and F. E. Grousset (2003), Two distinct seasonal Asian source regions for mineral dust deposited in Greenland (NorthGRIP), *Geophys. Res. Lett.*, *30*(4), 1167, doi:10.1029/2002GL016446.

Delmonte, B., I. Basile-Doelsch, J.-R. Petit, V. Maggi, M. Revel-Rolland, A. Michard, E. Jagoutz, and F. Grousset (2004), Comparing the Epica and Vostok dust records during the last 220,000 years: Stratigraphical correlation and provenance in glacial periods, *Earth Sci. Rev.*, *66*(1–2), 63–87.

Duce, R. A. (1995), Sources, distributions, and fluxes of mineral aerosols and their relationship to climate, in *Aerosol Forcing of Climate*, edited by R. J. Charlson and J. Heintzenberg, pp. 43–72, John Wiley, Hoboken, N. J.

Gabrielli, P., et al. (2004), Meteoric smoke fallout over the Holocene epoch revealed by iridium and platinum in Greenland ice, *Nature*, *432*, 1011–1014.

Helmer, M., J. M. C. Plane, J. Qian, and C. S. Gardner (1998), A model of meteoric iron in the upper atmosphere, *J. Geophys. Res.*, *103*(D9), 10,913–10,926.

Hunten, D. M., R. P. Turco, and O. B. Toon (1980), Smoke and dust particles of meteoric origin in the mesosphere and stratosphere, *J. Atmos. Sci.*, *37*, 1342–1357.

Jickells, T. D., et al. (2005), Global iron connections between desert dust, ocean biogeochemistry, and climate, *Science*, *308*, 67–71.

Lanci, L., D. V. Kent, P. E. Biscaye, and A. Bory (2001), Isothermal remanent magnetization of Greenland ice: Preliminary results, *Geophys. Res. Lett.*, *28*(8), 1639–1642.

Lanci, L., D. V. Kent, P. E. Biscaye, and J. P. Steffensen (2004), Magnetization of Greenland ice and its relationship with dust content, *J. Geophys. Res.*, *109*, D09104, doi:10.1029/2003JD004433.

Love, S. G., and D. E. Brownlee (1991), A direct measurement of the terrestrial mass accretion rate of cosmic dust, *Science*, *262*, 550–553.

Lynch, K. A., L. J. Gelinas, M. C. Kelley, R. L. Collins, M. Widholm, D. Rau, E. MacDonald, Y. Liu, J. Ulwick, and P. Mace (2005), Multiple sounding rocket observations of charged dust in the polar winter mesosphere, *J. Geophys. Res.*, *110*, A03302, doi:10.1029/2004JA010502.

Moskowitz, B. M., R. B. Frankel, S. A. Walton, D. P. E. Dickson, K. K. W. Wong, T. Douglas, and S. Mann (1997), Determination of the preexponential frequency factor for superparamagnetic maghemite particles in magnetoferritin, *J. Geophys. Res.*, *102*(B10), 22,671–22,680.

Neel, L. (1955), Some theoretical aspects of rock magnetism, *Philos. Mag.*, suppl. 4, 191.

North Greenland Ice Core Project members (2004), High-resolution record of Northern Hemisphere climate extending into the last interglacial period, *Nature*, *431*, 147–151, doi:10.1038/nature02805.

Plane, J. M. C. (2003), Atmospheric chemistry of meteoric metals, *Chem. Rev.*, *103*, 4963–4984.

Press, H. W., A. S. Teukolsky, T. W. Vetterling, and P. B. Flannery (2002), *Numerical Recipes in C: The Art of Scientific Computing*, 2nd ed., Cambridge Univ. Press, New York.

Pye, K. (1987), *Aeolian Dust and Dust Deposits*, 334 pp., Academic, New York.

Rapp, M., J. Hedin, I. Strelnikova, M. Friedrich, J. Gumbel, and F.-J. Lübken (2005), Observations of positively charged nanoparticles in the nighttime polar mesosphere, *Geophys. Res. Lett.*, *32*, L23821, doi:10.1029/2005GL024676.

Rietmeijer, F. J. M. (2001), Identication of Fe-rich meteoric dust, *Planet. Space Sci.*, *49*, 71–77.

Steffensen, J. P. (1997), The size distribution of microparticles from selected segments of the Greenland Ice Core Project ice core representing different climatic periods, *J. Geophys. Res.*, *102*(C12), 26,755–26,764.

Tauxe, L. (2002), *Paleomagnetic Principles and Practice*, 300 pp., Springer, New York.

Tegen, I., and I. Fung (1994), Modeling of mineral dust in the atmosphere: Sources, transport, and optical thickness, *J. Geophys. Res.*, *99*(D11), 22,897–22,914.

Wilkison, L. S., and S. M. Robinson (2000), Bulk density of ordinary chondrite meteorites and implications for asteroidal internal structure, *Meteorit. Planet. Sci.*, *35*(6), 1203–1213.

D.V. Kent, Department of Geological Sciences, Rutgers, The State University of New Jersey, Wright-Rieman Labs, 610 Taylor Road, Piscataway, NJ 08854, USA.

L. Lanci, Faculty of Science and Technology, University of Urbino, Campus Scientifico, I-61029 Urbino, Italy. (llanci@uniurb.it)