

## Magnetization of Greenland ice and its relationship with dust content

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[1] We estimate the concentration of fine magnetic particles in ice samples from the North Greenland Ice Core Project core from the central Greenland ice sheet, using low-temperature (77K) isothermal remanent magnetization (IRM) analysis and compare it with the mass concentration of aerosol dust. Samples were taken from six climatic intervals, spanning the time from the Holocene (Preboreal) back to the Last Glacial Dansgaard/Oeschger cycle 5. The mean IRM intensity of the ice varies by a factor of 3 from glacial to interglacial stages, being lower during interglacials. The IRM acquisition curves of the ice do not quite saturate at the maximum available field of 0.8 T and show a relatively broad coercivity, which is compatible with a mixture of maghemite or magnetite and hematite. Comparison of the IRM intensity and total dust mass shows a remarkably good correlation but also reveals a large background magnetization, which may be essentially constant over the different climatic stages. IRM suggests that the dust properties are independent of the background signal and that the dust aerosol has a magnetization within about 30% of pristine loess from the Chinese Loess Plateau, which is considered to have the same source in the same east Asian deserts as dust in Greenland ice. Ice contamination and the flux of extraterrestrial dust particles were considered in order to explain the origin of the background magnetization. Nevertheless, we could not find a convincing explanation for this signal, which represents a considerable part of the IRM signal and is the dominant component during interglacial intervals, without invoking the presence of undetected dust mass. The alternative hypothesis of a varying magnetization of the ice dust at different climatic periods would suggest that different sources of aerosol are active during different climatic periods. This, however, has not proven to be the case so far for studies of the provenance of dust in Greenland ice. *INDEX TERMS:* 0305

Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0399 Atmospheric Composition and Structure: General or miscellaneous; 0394 Atmospheric Composition and Structure: Instruments and techniques; *KEYWORDS:* magnetization, IRM, Greenland, ice core, NorthGRIP, aerosol

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### 1. Introduction

[2] Dust present in ice cores consists of atmospheric continental aerosol that has been deposited on the surface of polar ice sheets. The concentration of polar ice dust varies with time and reflects changes in environmental conditions. Larger total mass and number of dust particles in Greenland ice are related to cold or glacial intervals [Hammer *et al.*, 1985; Zielinski and Mershon, 1997; Ruth *et al.*, 2003], and these variations are found to be synchro-

nous with the seasonal and long-term climate variations [Hamilton and Langway, 1967; Hammer *et al.*, 1978]. Dust concentration in polar ice can be measured directly with a variety of methods such as Coulter Counter or laser light scattering on meltwater or directly on the ice [e.g., Steffensen, 1997; Ram and Illing, 1994; Ram *et al.*, 1995; Ruth *et al.*, 2003]. From glacial to interglacial intervals, the concentration of ice dust has been found to vary by more than an order of magnitude in mass [Thompson, 1977; Hammer *et al.*, 1985; Hanson, 1994].

[3] Dust flux and particle size are probably controlled by a combination of factors such as source-area characteristics and extent, atmospheric transport dynamics and relative contribution of dry vs. wet deposition. Hamilton and Langway [1967] interpreted the dust variations in Greenland ice as mainly due to higher aridity in the dust source areas and/or a more vigorous atmospheric circulation in the Northern Hemisphere during cold intervals.

[4] On the basis of mineralogic and isotopic analysis of dust from Greenland ice cores, and comparison with

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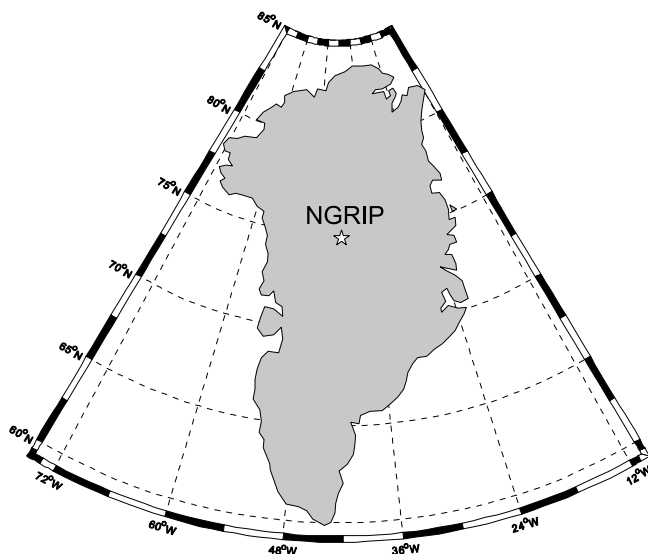


Figure 1. Location of the NorthGRIP ice core.

samples from possible Northern Hemisphere source areas, those arid source areas have been found to be the deserts of eastern Asia from the last glacial period through to the present [Biscaye *et al.*, 1997; Svensson *et al.*, 2000; Bory *et al.*, 2002, 2003]. Aeolian sediment transported from these source areas is considered to have been the source of the extensive loess deposits in the CLP [Liu, 1985], some of the fine-grained fraction of which has been transported as far as Greenland, especially during loess-depositing, glacial episodes [Biscaye *et al.*, 1997]. Similar to loess deposits, polar ice dust contains a significant fraction of highly magnetic iron oxides whose magnetization can be measured directly on ice samples [Lanci *et al.*, 2001]. The magnetic fraction can potentially be used as a tracer since it must reflect the availability of iron oxides in the dust source area as well as the total dust concentration. The concentration and mineralogy of the magnetic fraction in polar ice can be estimated by using some of the magnetic methods that have been successfully applied to environmental analysis of other wind-transported sediments such as Chinese loess [e.g., Heller and Evans, 1995, 2001; Verosub and Roberts, 1995]. Although reliable measurements on ice were found to be much more difficult to obtain than on loess [Sahota *et al.*, 1996; Lanci *et al.*, 2001], they allow us to investigate a novel property of ice and to estimate the concentration of iron oxides in the atmospheric aerosol. Moreover ice dust gives us the possibility to access the magnetic properties of a pure aerosol that remain unaffected by postdepositional weathering, compared to wind-blown material preserved in sedimentary deposits.

## 2. Methods and Materials

### 2.1. Magnetic Measurements

[5] We performed isothermal remanent magnetization (IRM) measurements on 72 samples from the North Greenland Ice Core Project (NorthGRIP) ice core that was drilled at 75.97°N, 42.32°W in north central Greenland (Figure 1). The ice samples were taken from six different climatic intervals including ice from the Preboreal to the Last Glacial

Maximum and across the Dansgaard/Oeschger cycle 5 transition. Figure 2 shows the depth position of the sampled intervals and a comparison with dust concentrations [Ruth *et al.*, 2003] and  $\delta^{18}\text{O}$  data (courtesy of S. Johnsen) as a proxy for climate.

[6] Although IRM is a simple, routine measurement in rock magnetism, obtaining reliable measurements on ice requires several critical precautions [Lanci *et al.*, 2001]. In particular, contamination arising from ice manipulation or the drilling process can be a major problem because of the very low magnetization of ice. The laboratory procedures and level of noise introduced by the measurement procedure were tested by measuring the IRM induced in 6 samples each made of about 50 g of high-purity distilled water prepared in a class 100 clean room. These samples were treated exactly as were the Greenland ice samples and have shown a very low magnetization level ( $0.3 \times 10^{-8} \text{ A m}^2 \text{ kg}^{-1}$ ) that can be regarded as a background level in our ice measurements. The results are shown in Table 1.

[7] Greenland ice samples were cut in pieces of about 5 cm lengths from 55 cm long core slabs (referred to by the Copenhagen group as “bags”) in the Lamont-Doherty freezer using a nonmagnetic phosphor-bronze saw. The topmost sample from each slab was not used because samples in this position are the most vulnerable to possible contamination due to penetration of drilling fluid. The prepared samples were stored in the cold room ( $-35^\circ\text{C}$ ) and transported in insulated containers as needed to the Paleomagnetic Lab located in a nearby building. Just prior to measurement 1–2 mm of ice was scraped from the surface of the sample using a phosphor-bronze knife; the

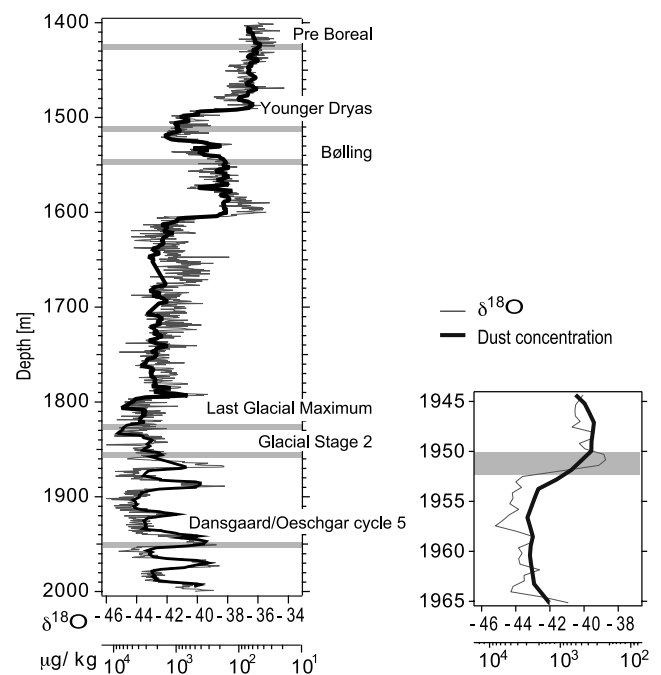


Figure 2. Measured intervals compared to an oxygen isotope ( $\delta^{18}\text{O}$ ) profile and dust concentration of the North Greenland Ice Core Project (NorthGRIP) ice core. The inset shows the detailed position of the samples taken from the Dansgaard/Oeschger cycle 5 transition.

**Table 1.** Average Values of Ice Magnetization IRM at 0.8 T (77K) and Dust Mass<sup>a</sup>

	Bag Depth, m	Section	IRM, A m <sup>2</sup> kg <sup>-1</sup>	IRM Standard Deviation, A m <sup>2</sup> kg <sup>-1</sup>	Dust, μm kg <sup>-1</sup>	Dust Standard Deviation, μm kg <sup>-1</sup>
Ultrapure ice	–	–	$0.39 \times 10^{-8}$	$0.23 \times 10^{-8}$	–	–
Preboreal	1425.8	2595	$3.12 \times 10^{-8}$	$1.93 \times 10^{-8}$	70.4	24.1
Younger Dryas	1512.3	2750	$3.65 \times 10^{-8}$	$1.66 \times 10^{-8}$	849.7	479.6
Bølling	1546.85	2813	$2.94 \times 10^{-8}$	$1.82 \times 10^{-8}$	273.1	198.0
LGM	1825.8	3320	$8.79 \times 10^{-8}$	$1.79 \times 10^{-8}$	6480.1	1649.0
Glacial Stage 2	1856.05 (1855.95)	3375 (3373)	$4.83 \times 10^{-8}$	$7.00 \times 10^{-9}$	2401.6	699.3
Dansgaard/Oeschger cycle 5	1950.2	3547	$2.15 \times 10^{-8}$	$1.09 \times 10^{-8}$	–	–
Dansgaard/Oeschger cycle 5	1951.2	3548	$2.50 \times 10^{-8}$	$2.01 \times 10^{-8}$	–	–
Dansgaard/Oeschger cycle 5	1952.2	3549	$2.52 \times 10^{-8}$	$1.01 \times 10^{-8}$	–	–
Dansgaard/Oeschger cycle 5 (mean of the 3 above)	1951.2	3547–3549	$2.68 \times 10^{-8}$	$2.13 \times 10^{-8}$	<b>701.7</b>	–

<sup>a</sup>Mass value from Dansgaard/Oeschger cycle 5 (bold) is averaged over three core sections and measured with a different technique (see text). Dust mass for the Glacial Stage 2 period has been measured in a different section at a slightly lower depth indicated in parentheses.

samples were then rinsed in a bath of pure (99.9%) propanol in order to melt, and therefore remove, even more potentially contaminated ice from the sample surface, and finally wiped with lint-free paper towels. From an initial sample mass of typically 50 g, the average sample mass after scraping and the propanol bath was around 30 g with the estimated removal of 3–4 mm of ice from the surface of each sample. The effectiveness of the cleaning by rinsing and final wiping was tested and confirmed by repeated measurements on a few pilot samples and corroborated by treatment of ice samples made of high-purity water.

[8] Immediately after the cleaning, samples were immersed in liquid nitrogen (77K) and kept there except for the short time needed to magnetize and measure them. We found that maintenance of the sample at low temperature during measurement procedures is critical for obtaining reliable results [Lanci *et al.*, 2001]. No significant contamination due to the liquid nitrogen immersion was found in a test made by measuring a clean microscope slide before and after immersion and on high-purity water ice. Repeated measurements also ensured that no significant contamination arises from sample handling during the measurement procedure.

[9] The cleaning process that we applied is similar to that used on ice for geochemical analysis and was effectively tested with the high-purity ice. It is possible that residual contamination, originating (for instance) from iron-bearing particles in the drilling process, may have penetrated deeper into the ice core than the superficial part of the samples that we have removed with cleaning. We have tested this possibility by measuring ice from the center of the ice core, and comparing that to ice from the outer “rind” of the ice core on which most of our measurements were made. This “outer” rind ice shows no significant IRM differences compared to that of the inner-core ice.

[10] The IRM was induced using an AST pulse magnetizer with a custom-built 6.5 cm diameter coil in which even our larger samples could fit. The maximum field available with this setup is 0.8 T. Ice magnetization was measured using a 2G Model 760 3-axis DC-SQUID cryogenic magnetometer with a 6.8 cm room temperature access. Stepwise IRM acquisition was measured on about one sample from each core slab, but in the other samples, we measured only the IRM at the maximum field (0.8 T). The measurement procedure was repeated for each sample with the

applied field in opposite directions to check the reproducibility of the results. The IRM acquired at 0.8 T (IRM<sub>0.8T</sub>) was calculated from the averaged values. The samples were measured immediately after immersion in liquid nitrogen in order to obtain reproducible measurements [Lanci *et al.*, 2001], taking particular care to execute the IRM acquisition and the measurement quickly to avoid significant warming of the samples. Even with these precautions, the scatter in the repeated measurements is larger than that of a typical rock sample and is on the order of 5–10%. The most likely sources of noise in ice measurements are the slightly variable rewarming of the ice samples during the IRM acquisition and measurement process that could result in changes of magnetization due to relaxation of superparamagnetic components and variation of the spontaneous magnetization (Ms) due to its fundamental temperature dependence, some minor but variable contamination from handling and dust suspended in air during the measurement procedure.

[11] Given the between-sample variability in IRM<sub>0.8T</sub> intensity and dust contents, representative values were obtained by averaging sample data within each core section (bag), which corresponds to a climatic period, whereby each mean IRM<sub>0.8T</sub> consists of an average of nine samples measured independently. The results are listed in Table 1 where the between-sample variability within the same climatic period is expressed by the standard deviation.

[12] In addition to the ice samples, we measured IRM acquisition on two samples from the CLP (courtesy of F. Heller and R. Egli) that are representative of very pristine loess and paleosol. The IRM acquisition curve was generated by first giving the sample a backfield magnetization of 800 mT along the  $-z$  sample direction and then applying progressively increasing fields along the  $+z$  direction up to a maximum of 800 mT. IRM acquisition curves were generated at both room and liquid nitrogen (77K) temperatures to allow a direct comparison with the ice measurements; the results are listed in Table 2.

## 2.2. Dust Measurements

[13] We used two different estimates of the dust concentration in ice samples from NorthGRIP. One set of measurements was obtained using a Coulter Counter with 20 μm orifice and procedures described by Steffensen [1997]. The Coulter Counter sample values were averaged in a fashion

**Table 2.** Loess and Paleosol Isothermal Remanent Magnetizations at 0.8 T ( $IRM_{0.8T}$ )

	Room Temperature, $A\ m^2\ kg^{-1}$	77K, $A\ m^2\ kg^{-1}$
Sample BY55 (loess)	$4.53 \times 10^{-3}$	$6.84 \times 10^{-3}$
Sample SPS33-7 (soil)	$8.46 \times 10^{-3}$	$1.73 \times 10^{-2}$

similar to the magnetization measurements where each mean Coulter Counter dust mass estimate consists of an average of eight independently measured samples taken over a core section (bag) of 55 cm length. The mean value and the variability of the individual measurements, expressed as the standard deviation, are shown in Table 1. The only samples that were not measured with this procedure are the samples taken from the Dansgaard/Oeschger cycle 5 transition.

[14] The dust content in Dansgaard/Oeschger cycle 5 samples, as well as in the other intervals of the NorthGRIP core, was taken from *Ruth et al.* [2003], who measured the dust volume using a novel laser microparticle detector technique. To account for the presence of soluble minerals, calcite and gypsum, that are completely dissolved on melting of the ice and thus removed from the Coulter Counter measurements but remain mostly undissolved during the laser scatter measurements, we renormalized the Coulter Counter measurements to the laser scatter

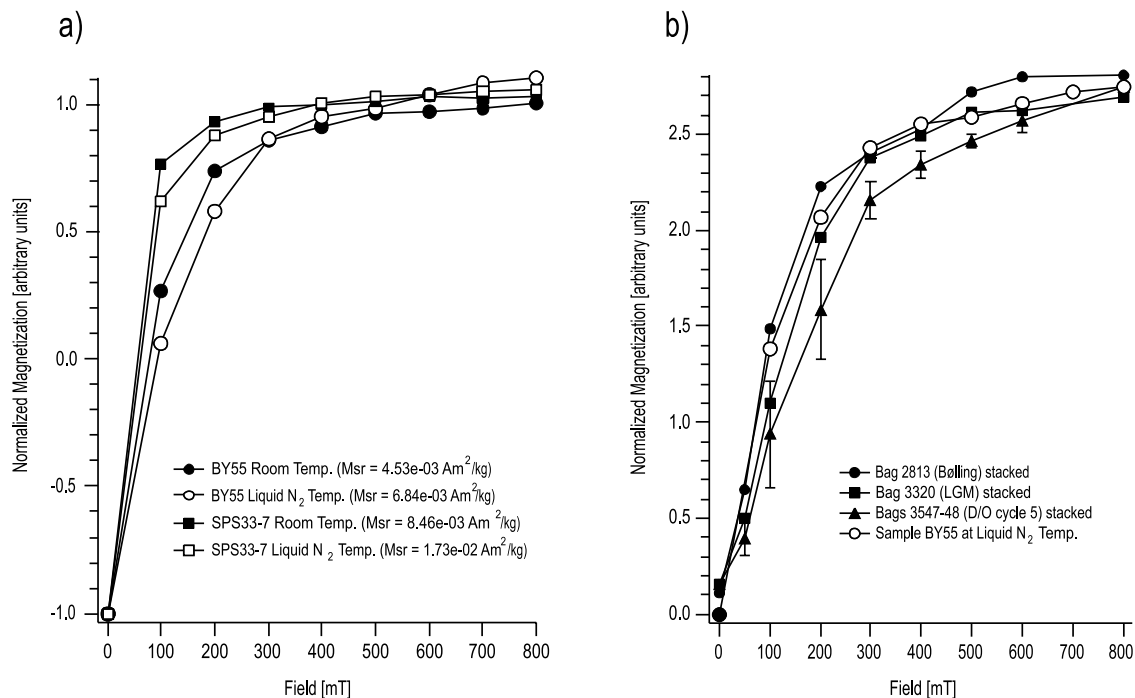
measurements by multiplying them by a factor of 2.5. This resulted in values similar in concentration and granulometry to that reported from GRIP [*Steffensen, 1997*] for equivalent climatic intervals. Each laser detector measurement represents an average of three sections (bags) of core and an estimate of the dispersion is not available. For these reasons we have chosen to keep separated the results measured with the two different techniques. The conversion from volume to mass for both techniques was done assuming a density of  $2600\ kg\ m^{-3}$ .

### 3. Results

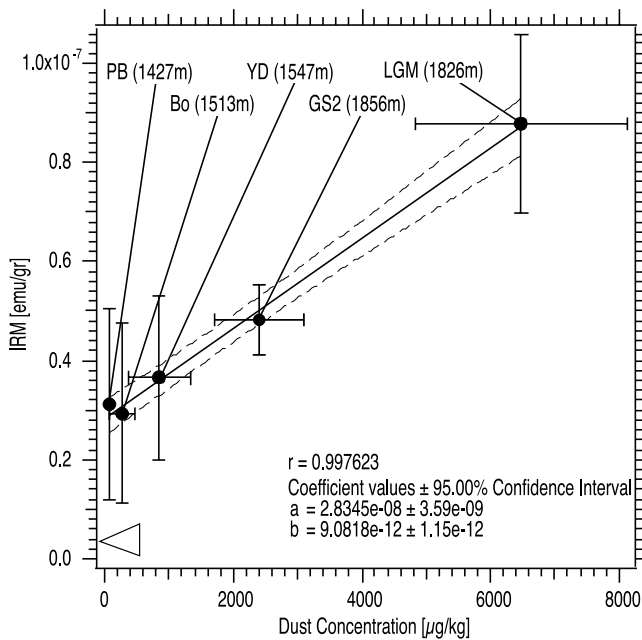
#### 3.1. IRM Acquisition

[15] The magnetic mineralogy of the ice dust was inferred from an analysis of IRM acquisition curves, which are shown in Figure 3b for the climatic periods of Bølling (warm), LGM (cold) and the Dansgaard/Oeschger cycle 5 transition. The data represent IRM acquisition curves of three or four samples from the same climatic period that has been stacked to reduce measurement noise and between-sample variability.

[16] The IRM acquisition curves do not completely saturate at the maximum available field of 0.8 T and exhibit a rather broad curvature that can be interpreted as due to a mixture of a lower-coercivity ferrimagnetic minerals, most likely magnetite/maghemite, as the main magnetic carrier in the ice dust, and a subordinate high-coercivity mineral, such



**Figure 3.** (a) Isothermal Remanent Magnetization (IRM) of loess (BY55) and paleosol (SPS33-7) samples at room and liquid nitrogen temperatures. In both samples the coercivities and the saturation magnetizations increase at low temperature. (b) IRM acquisition curves of ice samples from three different climatic stages (LGM, Bølling, and Dansgaard/Oeschger cycle 5 transition). The IRM curves were produced by averaging three or more curves from individual samples taken in the same climatic interval after normalizing by the total standard deviation. Standard deviation is shown as an error bar. For LGM and Bølling samples the error bars are smaller than the symbols used in the plot. IRM acquisition curve for sam loess (BY55 at 77K) is plotted with open symbols for comparison.



**Figure 4.** Linear correlation between Coulter measurements of total dust mass and  $IRM_{0.8T}$  for NorthGRIP ice core samples. Data sets are averaged within each climatic interval; error bars in the plot indicate the standard deviation of individual intervals, and dashed lines are the 95% confidence bands of the regression line. The open triangle indicates the magnetization measured in the ultrapure ice as a reference for noise level. The regression line has been computed by weighted least squares,  $r$  is the correlation coefficient, and  $a$  and  $b$  are the coefficients in the linear equation  $y = a + bx$ .

as antiferromagnetic goethite or, more probably, by comparison with Chinese loess, hematite. The extremely low concentration of dust practically precludes the possibility to perform rock-magnetic or thermomagnetic analyses on residue from melted ice because to obtain just a few mg of dust would require melting a very large amount of ice (about 1000 kg). Samples from the Dansgaard/Oeschger cycle 5 transition show the largest contents of the high-coercivity mineral and large between-sample variability in the coercivity spectra. However, the present data are insufficient to establish if there is a trend of decreasing contribution of high-coercivity minerals up core.

[17] IRM acquisition experiments on loess and paleosol samples confirm the typical results reported in the literature [e.g., Heller and Evans, 1985] (Figure 3a). The paleosol sample approaches saturation IRM by  $\sim 300$  mT, whereas the loess sample must contain a higher-coercivity fraction because the IRM acquisition curve has a broad curvature and does not completely saturate by 800 mT. The data also show enhanced IRM magnitude (about 50%–65%) and coercivity when acquired and measured at low temperature. In particular, the coercivity spectrum of the loess at 77K is very similar to that of ice dust (Figure 3b).

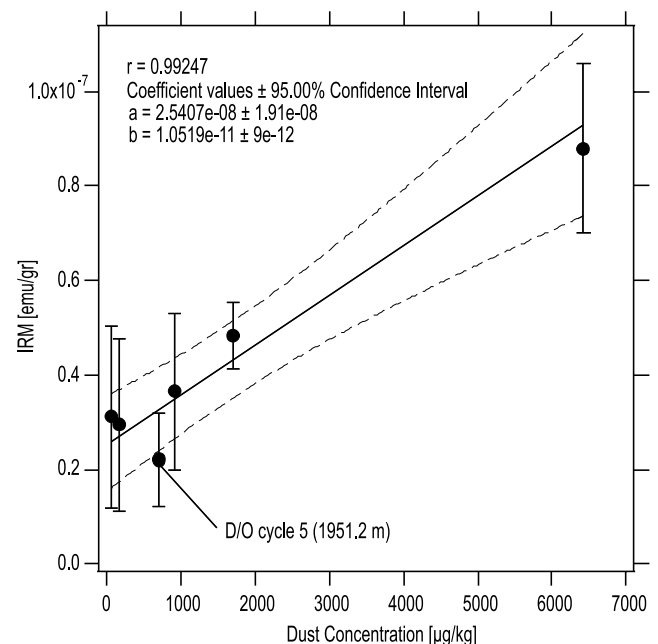
### 3.2. Comparison With Dust Measurements

[18] Even though the maximum field available was unable to completely saturate the samples, we believe that the

IRM magnetization at 0.8 T ( $IRM_{0.8T}$ ) can be reasonably interpreted, within the uncertainties discussed above, as a parameter proportional to the concentration of magnetic minerals. The mean  $IRM_{0.8T}$  intensity varies approximately by a factor of 3 between glacial and interglacial stages (Figures 4 and 5).

[19] Since the magnetic particles in Greenland ice are a fraction of its dust content, the comparison with dust mass is of particular interest. However, making quantitative comparisons of the results is not straightforward because the magnetic and dust measurements involve rather different ranges of grain size. The range of grain sizes measured with the Coulter Counter using the 20  $\mu\text{m}$  orifice tube is limited approximately to between 800 nm and 10,000 nm. In magnetic measurements most of the IRM will be carried by single-domain and pseudosingle domain ferrimagnetic (magnetite and maghemite) particles that have a narrow size range between about 50 and 500 nm [Buttler and Bannerjee, 1975]. Only weakly magnetic antiferromagnetic minerals, like hematite, can be efficiently magnetized at much larger grain sizes, but their contribution to the IRM is usually small because of their much lower value of spontaneous magnetization. Therefore, in general, the IRM will tend to be most sensitive to the concentration of the smallest magnetic grains, which might be grossly underrepresented or even undetected in the total dust mass measurements.

[20] Nevertheless, the  $IRM_{0.8T}$  magnetization and the total dust concentration, averaged within the same climatic periods, show a very good correlation. A linear regression between magnetization and Coulter dust mass is obtained by weighted least squares (also referred to as  $\chi^2$  minimization), taking into account the standard deviation of the  $IRM_{0.8T}$



**Figure 5.** Linear correlation between laser detector dust mass (data from Ruth *et al.* [2003]) and  $IRM_{0.8T}$  (see caption to Figure 4 for explanation). Note that samples from the Dansgaard/Oeschger cycle 5 transition have a lower magnetization and fall below the regression line.

**Table 3.** Concentration of Magnetic Minerals<sup>a</sup>

	Section	Maghemite Concentration in Dust (Mass)	Hematite Concentration in Dust (Mass)	Fe-Oxides Concentration in Dust (Mass)	Fe-Oxides Concentration in Ice (Mass)
Preboreal	2595	$1.42 \times 10^{-2}$	$2.40 \times 10^{-1}$	$2.54 \times 10^{-1}$	$1.79 \times 10^{-8} \pm 1.11 \times 10^{-8}$
Younger Dryas	2750	$1.38 \times 10^{-3}$	$2.33 \times 10^{-2}$	$2.47 \times 10^{-2}$	$2.09 \times 10^{-8} \pm 9.55 \times 10^{-9}$
Bølling	2813	$3.46 \times 10^{-3}$	$5.83 \times 10^{-2}$	$6.18 \times 10^{-2}$	$1.68 \times 10^{-8} \pm 1.05 \times 10^{-8}$
LGM	3320	$4.37 \times 10^{-4}$	$7.35 \times 10^{-3}$	$7.78 \times 10^{-3}$	$5.04 \times 10^{-8} \pm 1.03 \times 10^{-8}$
Glacial Stage 2	3375	$6.47 \times 10^{-4}$	$1.09 \times 10^{-2}$	$1.15 \times 10^{-2}$	$2.77 \times 10^{-8} \pm 4.02 \times 10^{-9}$
Dansgaard/Oeschger cycle 5	3547–3549	$8.91 \times 10^{-4}$	$3.83 \times 10^{-2}$	$2.92 \times 10^{-2}$	$2.05 \times 10^{-8} \pm 9.05 \times 10^{-9}$

<sup>a</sup>We assume that IRM is carried by 85% maghemite and 15% hematite except in the Dansgaard/Oeschger cycle 5 interval where we used 75% and 25%, respectively. The value of the slope and offset are  $5.13 \times 10^{-3} \pm 1.85 \times 10^{-3}$  and  $1.60 \times 10^{-8} \pm 1.35 \times 10^{-8}$ , respectively, and are calculated using dust data from Coulter Counter except for Dansgaard/Oeschger cycle 5, where laser detector was used (data from *Ruth et al.* [2003]).

measurements (Figure 4). The linear correlation is significant at the 99% confidence level according to a Student's *t* test. No significant difference in the correlation between magnetization and dust is found when using dust mass measured with the laser detector (Figure 5). Even if these data include the datum from the Dansgaard/Oeschger cycle 5 transition, which has lower magnetization intensity, the linear regression has practically identical coefficients.

[21] A positive intercept of the regression line with the *y* axis indicates a relatively large background IRM signal that is uncorrelated with the dust concentration. In the Coulter Coulter measurements, this background signal is  $2.7 \pm 0.13 \times 10^{-8} \text{ A m}^2 \text{ kg}^{-1}$  and the slope of the regression line has a value of about  $9.0 \pm 0.48 \times 10^{-3} \text{ A m}^2 \text{ kg}^{-1}$  with a correlation coefficient  $r > 0.99$ . Similar values are obtained for the laser detector measurements (Figure 5).

[22] The between-sample variability of  $\text{IRM}_{0.8\text{T}}$ , as indicated by the standard deviations (Figure 4 and Table 1), is comparable to that of dust measurements during glacial intervals but it is much higher during the interglacials, especially during the Bølling and Preboreal. No comparison with the dust variability is possible during the Dansgaard/Oeschger cycle 5 transition because of the different technique of dust measurement.

## 4. Discussion

### 4.1. Estimate of Fe-Oxides

[23] Fe-oxide (i.e., ferri- and antiferromagnetic) concentrations can be coarsely estimated from the ice IRM measurements. According to the IRM acquisition curves (Figure 3b), the fraction of  $\text{IRM}_{0.8\text{T}}$  carried by the magnetic mineral with coercivity below 300 mT accounts for about 85% of the entire IRM in all climatic periods except Dansgaard/Oeschger cycle 5. Assuming that this fraction is carried by maghemite ( $\gamma\text{-Fe}_2\text{O}_3$  with saturation magnetization,  $M_s = 82.5 \text{ A m}^2 \text{ kg}^{-1}$  and the ratio of saturation remanence to saturation magnetization,  $\text{Mr}/M_s = 1/3$ ) and the remaining 15% of  $\text{IRM}_{0.8\text{T}}$ , with coercivity above 300 mT, is carried by hematite ( $\alpha\text{-Fe}_2\text{O}_3$  with  $M_s = 0.47 \text{ A m}^2 \text{ kg}^{-1}$  and  $\text{Mr}/M_s = 1/2$ ), it is possible to estimate the concentration of these magnetic minerals. Similarly, Fe-oxide concentration in the Dansgaard/Oeschger cycle 5 is calculated assuming that 75% of IRM is carried by maghemite and 25% by hematite, as suggested by the IRM acquisition curves. Results are shown in Table 3 and the calculated Fe-oxide concentration is plotted versus dust concentration in Figure 6. The absolute values of these

calculations must be taken with caution due to the incomplete saturation of IRM and uncertainties in the  $\text{Mr}/M_s$  ratios. However, the linear regression in Figure 6 is tightly constrained; even the sample from Dansgaard/Oeschger cycle 5 is well aligned on the regression line, signifying that the lower magnetization of this interval as observed in Figure 5 is a consequence of a different magnetic mineralogy.

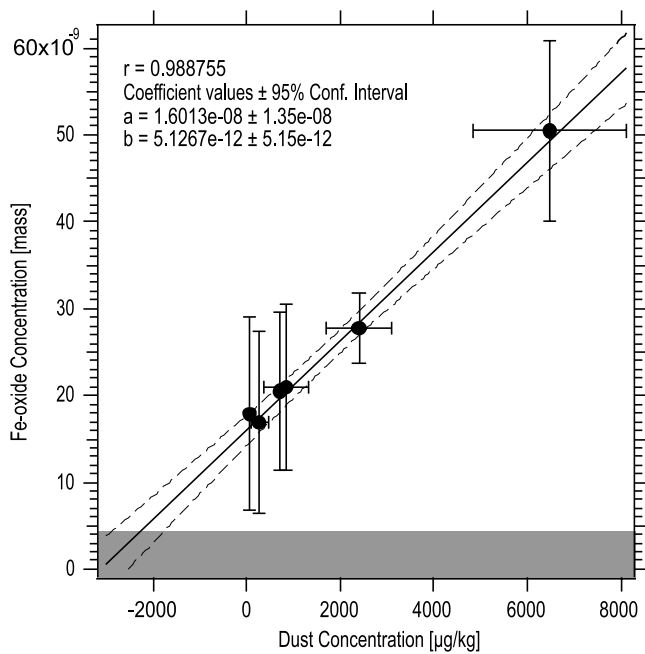
[24] The total concentration of Fe-oxides in the measured dust calculated from the regression slope of Figure 6 is about 5‰ (Table 3). Compared with the typical total iron concentration of 5% of crustal material this indicates only about 10% of the iron occurs as an oxide phase. The concentration of Fe-oxides in ice due to the background signal is rather large and estimated at about  $1.60 \pm 1.35 \times 10^{-8} \mu\text{g kg}^{-1}$  provided that the magnetic mineralogy is the same as in all climatic periods except Dansgaard/Oeschger cycle 5.

### 4.2. Rock Magnetic Record in Ice

[25] Except for Dansgaard/Oeschger cycle 5, we did not find evidence for systematic changes of the magnetic mineralogy or grain size from glacial to interglacial periods. IRM acquisition curves for Bølling and LGM periods are representative of the magnetic mineralogy of interglacial and glacial stages and do not show any appreciable difference (Figure 3b). Although some mineralogical changes are possible, they could not be reliably distinguished from the IRM measurements available for this study due to the large between-sample variability of the IRM and the few IRM acquisition curves made on interglacial samples. Samples from the Dansgaard/Oeschger cycle 5 transition show the largest between-sample variability in IRM acquisition curves and, on average, the largest contents of a high-coercivity mineral. The slightly different magnetic mineralogy might explain the lower average magnetization of the Dansgaard/Oeschger cycle 5 transition samples compared to the mean value for the ice dust (see also Figure 5).

[26] The good linear correlation suggests that the IRM of ice is, in general, proportional to the measured dust contents. The linear regression, however, does not go to the origin but shows an offset of about  $2.7 \pm 0.13 \times 10^{-8} \text{ A m}^2 \text{ kg}^{-1}$  (as calculated on Coulter Counter data). Linear regression on the laser data gives practically identical results (offset of  $2.5 \pm 0.13 \times 10^{-8} \text{ A m}^2 \text{ kg}^{-1}$ ) and the same high statistical significance. This background component of IRM represents a significant fraction of the total ice magnetization that becomes predominant in interglacial ice.

[27] Given the linear relationship observed between measured dust mass and magnetization, the background mag-



**Figure 6.** Linear correlation between dust mass and an estimate of iron-oxide concentration in ice based on IRM measurements (see text). Error bars indicate the standard deviation of individual measurements, and dashed lines are the 95% confidence bands of the regression line. Dust mass was measured with Coulter Counter except for Dansgaard/Oeschger cycle 5 samples, which were measured with laser detector (data from *Ruth et al.* [2003]). The value of  $r$  represents the correlation coefficient, and  $a$  and  $b$  are the coefficients in the linear equation  $y = a + bx$ . Regression lines have been computed by weighted least squares. The plot shows that iron oxide concentration in the Dansgaard/Oeschger cycle 5 interval is aligned with that of other intervals, suggesting that the lower magnetization is due to a different magnetic mineralogy. The correlation line has been extrapolated to negative value to illustrate the amount of apparently missing dust, which is represented by the intersection of the best fit line with the  $x$  axis. The shaded region at the bottom of the plot is an estimate of the magnetization that can be accounted to external sources such as interplanetary dust particles.

netization, which is well above the pure-ice noise level, may be due to a fraction of very fine grained dust that is not detected by the measurements. If the effect of the incomplete measurements of the smaller fraction of the dust (<800 nm diameter) indeed resulted in an underestimate of the dust mass of about 10–30%, this could account for the measured difference only if the missing fraction has a strong magnetization such as from single-domain magnetite or maghemite grains. This would, however, not be consistent with the apparently uniform magnetic mineralogy of the Bølling and LGM, where the background fraction represents about 90% and 30% of the total magnetization, respectively.

[28] If the background fraction of the ice magnetization is taken as constant among the different climatic intervals and is subtracted; the slope of the regression line ( $9.0 \pm 0.48 \times 10^{-3} \text{ A m}^2 \text{ kg}^{-1}$ ) can be used as the best estimate of the

average magnetization. On the other hand, if the background magnetization is not removed, the dust from interglacial intervals shows a much higher concentration of magnetic minerals compared to the glacial periods.

[29] *Lanci et al.* [2001] reported two interglacial samples with a magnetization larger than LGM samples. This result cannot be confirmed here where interglacial samples have a mean magnetization smaller than the mean glacial samples. The earlier result can be explained by the heterogeneity of the samples, by their small number together with the large between-sample variability that is observed in ice magnetization and perhaps by a higher level of laboratory contamination in those initial measurements.

### 4.3. Comparison With Chinese Loess

[30] Since Chinese loess is considered to have had its source in the same East Asian deserts as the dust in Greenland [*Biscaye et al.*, 1997; *Svensson et al.*, 2000; *Bory et al.*, 2002, 2003], it seems appropriate to compare its magnetization to that of Greenland ice. The IRM acquisition of a pristine loess sample is compared directly to the ice IRM acquisition in Figure 3b and suggests a similar magnetic mineralogy. The IRM intensity of ice dust calculated from the regression slope (i.e., neglecting the background component) is also comparable to that of loess. Taking the mass magnetization of  $6.84 \times 10^{-3} \text{ A m}^2 \text{ kg}^{-1}$  for  $\text{IRM}_{0.8T}$  measured at 77K in our loess sample as representative for typical Chinese loess, the magnetization of ice dust, calculated after removal of the background signal, is less than that of the loess by a factor of approximately 1.3. Coercivity distributions of ice and loess samples derived from the IRM acquisition experiments are not distinguishable within the measurement uncertainties, suggesting a very similar magnetic mineralogy. We suggest that the small differences between ice dust and loess magnetization can be attributed to enhancement, perhaps by selective winnowing, of the magnetization of the aerosol that traveled 10,000 km to Greenland compared with that of the loess, which traveled perhaps only 1000 km from the source area to the CLP.

[31] Alternatively, if one chooses to not subtract the background signal, the large magnetization of the interglacial ice dust would be difficult to explain. In this case we calculate a magnetization of LGM and Preboreal ice dust of about  $1.35 \times 10^{-2} \text{ A m}^2 \text{ kg}^{-1}$  and  $4.45 \times 10^{-1} \text{ A m}^2 \text{ kg}^{-1}$ , respectively. These values are about 2 and 65 times larger, respectively, than the magnetization of our loess sample ( $6.84 \times 10^{-3} \text{ A m}^2 \text{ kg}^{-1}$ , also measured at 77K). Comparison with paleosol samples from the CLP does not seem appropriate because no pedogenesis is expected to occur in Greenland ice; however, even the comparison of the Preboreal ice with paleosol (Table 2) would lead to very large discrepancy (factor of about 25). Any selective winnowing during the transport process is unlikely to produce such a large difference during warm (Preboreal) times, even if magnetic grains are not proportionately distributed in the total sediment grain size distribution. For example, measurements on grain size fractions of Hungarian loess [*Sartori et al.*, 1999] show that the magnetic susceptibility is higher in the grain size fraction below  $1 \mu\text{m}$  and peaks at about  $0.1 \mu\text{m}$  [e.g., *Steffensen*, 1997] with a value that is less than 3 times the bulk susceptibility of the whole sediment. This suggests that atmospheric aerosol, which has a grain

size distribution peaking at or below about 1  $\mu\text{m}$ , could concentrate the more magnetic fraction of the loess and, although susceptibility may not be directly proportional to the IRM, it is conceivable to expect an enhancement in IRM of similar magnitude.

#### 4.4. Dust-Independent Background

[32] Given the nearly uniform magnetic mineralogy suggested by the IRM acquisition curves from Bølling and LGM (Figure 3b), we do not consider the variations in magnetic mineralogy of the dust from glacial to interglacial intervals to be a major cause of changes in dust magnetization. Moreover, the IRM acquisition suggests that the magnetic mineralogy of the background component is very similar to that of the overall magnetic component of the dust.

[33] With a quasi-constant magnetic mineralogy, the large difference in magnetization of glacial and interglacial ice dust could be simply justified with a larger concentration of magnetic particles in interglacial dust. A larger number of small magnetic particles would probably be difficult to detect since such ultrafine magnetic particles are too small for Coulter Counter measurements or, as is often common in magnetite or maghemite grains of terrestrial origin, they could be electrostatically attached to larger clay mineral particles. This hypothesis is in agreement with the observed uniform magnetic mineralogy, but it is not supported by the result obtained comparing ice and loess magnetization. Moreover, it seems unlikely to produce the linear correlation and apparently constant background that is observed in the correlation of magnetization versus dust mass.

[34] Instead of assuming a compositionally variable ice dust with different concentrations of magnetic minerals, we favor the interpretation in which the ice dust magnetization has magnetic properties similar to that of the pristine Chinese loess and its magnetization is well approximated by the slope of the best-fit line. This interpretation leaves open the origin of the background component, which seems to be carried by particles undetected by the dust measurements, and that have a magnetic mineralogy similar to that of dust, as suggested by the IRM acquisition.

[35] Independent sources of magnetic minerals can be invoked to explain the background component assuming that its particles are small enough to pass undetected by the Coulter Counter. One possible source is the fallout of interplanetary dust particles (IDPs), which consist of cosmic dust encountering the Earth and mostly originating from the asteroid belt [Kortenkamp and Dermott, 1998]. IDPs generally have a chondritic composition, and thus contain a large amount of iron, with particle diameters ranging from a few nanometers to several microns (see Rietmeijer [1998] for a review). The common occurrence of IDPs and micrometeorites on the surface of ice sheets is well known [e.g., Mauvette *et al.*, 1986; Taylor *et al.*, 1998; Karner *et al.*, 2003]; therefore, a certain contribution of extraterrestrial materials to ice magnetization should be expected. Estimates of IDP flux on Earth vary by orders of magnitude depending on the proxy and the assumptions used in the calculations; however, it is usually believed that extraterrestrial dust is accreting to Earth at levels between 1 and  $12 \times 10^7 \text{ kg yr}^{-1}$  [Muller, ]. Generally accepted results

based on the study of micrometeorite impact craters on the long-duration exposure facility satellite [Love and Brownlee, 1991; Grün *et al.*, 1985] estimate that  $4 \pm 2 \times 10^7 \text{ kg}$  of IDPs from the zodiacal cloud are accreted to Earth every year. Moreover, measurements on deep-sea sediments of extraterrestrial  $^3\text{He}$ , a proxy for IDPs, indicate that the flux has been relatively constant over at least the past 200 ka [Farley, 1995; Marcantonio *et al.*, 1999]. The expected concentration of extraterrestrial materials calculated using the Love and Brownlee [1991] estimate ( $4 \times 10^7 \text{ kg}$  divided by Earth's surface area) gives a flux of  $7.83 \times 10^{-8} \text{ kg m}^{-2} \text{ yr}^{-1}$ . In the Holocene, with an ice accumulation rate in the NorthGRIP core of about  $1.8 \times 10^2 \text{ kg m}^{-2} \text{ yr}^{-1}$  (J. P. Steffensen, personal communication, 2003), this gives a mass concentration of IDPs of  $4.4 \times 10^{10}$  ( $0.44 \mu\text{g kg}^{-1}$ ). This is a small fraction (<1%) of the dust accumulation measured in Greenland ice and therefore, based on this estimate, IDPs could account for a significant part of the background magnetization only if they consisted mainly of a highly magnetic mineral such as maghemite or magnetite. However, IRM acquisition indicates that this is unlikely, and hence the contribution of IDPs to the total magnetization is probably smaller than 10%.

[36] The possibility of a penetrative contamination of magnetic particles into the ice core has been rejected after comparing the magnetization of ice from the inner and the outer part of the core. Since no external source seems capable to explain the background magnetization we have to conclude this is carried by a fraction of the dust that is not detected by the current measurements probably because of its extremely small grain size. This “invisible” dust is carrying a relevant portion of the ice magnetization and, assuming that its magnetization is similar to that of the measured dust fraction, as suggested by IRM acquisition, it may constitute a major contribution to the concentration of interglacial dust. If so, a major implication would be that the contrast between the dust concentration between glacial and interglacial periods is much smaller than has been heretofore measured.

## 5. Conclusion

[37] The magnetization of ice from the NorthGRIP core correlates linearly with the dust concentration in the ice. The regression line, however, does not go through the origin, implying that there is a background magnetization that is not proportional to the measured dust concentrations.

[38] We speculate that part of the dust background fraction of magnetic minerals could be of extraterrestrial origin and due to IDP flux to the ice, but this could not fully account for the background component of the magnetization. We have ruled out an alternate source of contamination, possibly due to penetration of drilling fluids bearing iron particles into the ice cores.

[39] Given the observed constant magnetic mineralogy and the highly significant linear correlation, we have two possible interpretations to explain the large background signal: (1) The concentration of magnetic minerals in the terrestrial aerosol is extremely different from glacial to interglacial stages, or (2) there is a fraction of dust in ice that is carrying a large part of the ice magnetization and that is mostly undetected by current dust concentration



measurements. Both interpretations are plausible, but each leaves open questions.

[40] Interpretation 1 requires variable concentration of magnetic minerals and must face the difficulty of explaining the linear correlation between dust mass and magnetization and the extremely high magnetization of interglacial dust, which is not easily reconcilable with loess or paleosol data. We favor interpretation 2 in which the climatically controlled aerosol in the ice has a nearly constant concentration of the magnetic fraction over the investigated time interval and suggests that there is a dust fraction with similar magnetic properties that is not detected in our measurements. Considering the different transport conditions and natural variability of the samples, we believe that the magnetization of the ice aerosol computed according to this hypothesis (i.e., after the removal of the background fraction) is compatible with that of the dust source areas indicated by other studies, the Chinese and Mongolian deserts of eastern Asia [Biscaye *et al.*, 1997; Svensson *et al.*, 2000; Bory *et al.*, 2002, 2003]. This interpretation is in agreement with the current idea that the enhancement of paleosol magnetization in the CLP is due to in situ pedogenic processes. Nevertheless, it requires that a considerable amount of dust mass is missed in the Coulter Counter and laser measurements. If verified, the presence of this dust would drastically change the ratio between glacial and interglacial dust masses and could have important consequence in climate reconstructions. Independent of the interpretation, the linear relationship found with the aerosol mass suggests that IRM of the ice, and hence the concentration of magnetic minerals, can potentially be used as a climatic proxy in ice.

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