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**Microbes, organic
aerosols, dust, and
msa in polar ice**

P. B. Price et al.

Fluxes of microbes, organic aerosols, dust, and methanesulfonate onto Greenland and Antarctic ice

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Abstract

Using a spectrofluorimeter with 224-nm laser excitation to measure fluorescence intensity at 300- μ m depth intervals, we report results of the first comparative study of concentrations of microbial cells (using the spectrum of protein-bound tryptophan (Trp) as a proxy) and of aerosols with an autofluorescence spectrum different from Trp as a function of depth in ice cores from west Antarctica (WAIS Divide and Siple Dome) and Greenland (GISP 2). The ratio of fluxes of microbial cells onto Antarctic Greenland ice is 0.23 ± 0.11 and of non-Trp aerosols is 0.17 ± 0.08 , both of which are comparable to the ratio of fluxes of mineral dust at Antarctic and Greenland sites (0.09 ± 0.06). In contrast, the ratio of fluxes of methanesulfonate (MSA) onto Antarctic relative to Greenland sites is 1.86 ± 0.4 , a factor 20 higher. The lower fluxes of microbes, non-Trp aerosols, and dust onto Antarctic ice may be due to the smaller areas of their source regions, together with less favorable wind patterns for Antarctic ice than Greenland ice. We attribute the higher fluxes of MSA in Antarctic ice to the concentration of haptophytes, a phylum of marine algae, in the far more extensive sea ice margin around Antarctica than around Greenland. The similarity of flux ratios of microbes and non-Trp aerosols to dust flux ratios suggests that their source regions overlap with dust sources rather than with MSA sources. A new version of the spectrofluorimeter with additional channels for mapping chlorophyll and volcanic tephra will be used to map WAIS Divide ice at 1 mm intervals to bedrock.

1 Introduction

In recent years biologists have determined concentrations and taxa of microorganisms at a few depths in polar ice (e.g. Miteva et al., 2004; Priscu and Christner, 2004; Price, 2007) and permafrost (Gilichinsky, 2002). Due to the labor involved in extracting microbes from ice and analyzing them under sterile conditions, no systematic study has until now been made of the concentrations of microbes and their rates of deposition

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onto the two polar caps as a function of depth. By contrast, because of their value as monitors of Earth's climate, atmospheric gases such as CH₄, N₂O, δ¹⁸O, CO₂ (e.g. Chappellaz et al., 1997; Sowers et al., 2003; Grootes et al., 1993; Bender et al., 1994; Petit et al., 1999), CH₃SO₃H (e.g. Legrand et al., 1991; Saltzman et al., 1997), and mineral dust (e.g. De Angelis et al., 1997; Delmonte et al., 2004) have been thoroughly studied as a function of depth in both Antarctic and Greenland ice.

Our group is now using scanning spectrofluorimetry to map concentrations of microbes and non-Trp aerosols at sub-mm depth intervals in ice cores. Elsewhere we have discussed our fluorimetric method of determining those concentrations based on the spectral shapes in six channels of emission wavelength resulting from fluorescence excited by our 224-nm laser (Rohde and Price, 2007; Rohde et al., 2008). We calibrated the instrument by relating the intensity of protein-bound tryptophan (Trp) autofluorescence in microbial cells to direct counts of stained cells in ice cores from Summit, Greenland (GISP2) (Tung et al., 2005).

Among the new results obtained with fluorimetry, our group (Rohde and Price, 2007; Rohde et al., 2008) presented evidence that some cells are located in liquid veins at triple junctions in the polycrystalline ice, that metabolizing cells are often located in the ice lattice rather than in veins, and that at some depths the microbial concentration is so high that products of their metabolism give rise to localized excesses in N₂O that interfere with their role as a climate proxy. We also showed that both microbes and non-Trp aerosols are deposited in discrete bursts with peak values that fluctuate on seasonal to decadal scales. One sees from Figs. 1 and 2 that large fluctuations are common, even when averaged over 1-m depth intervals, which led us to suggest that scanning fluorimetry may provide meteorological information on abrupt changes of wind speed or direction over the last 10⁵ years for which ice core records exist.

Herein we compare concentrations and fluxes of microbes and non-Trp aerosols measured with our fluorimeter on Greenland and Antarctic ice cores, together with dust and MSA measured by others, and draw conclusions about likely sources.

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2 Results and discussion

We measured fluorescence spectra at intervals of $\sim 300 \mu\text{m}$ in 76 GISP2 ice cores at selected depths from 286 to 3042 m (Rohde et al., 2008). During that run we obtained data for cores from two sites in West Antarctica consisting of portions of 20 cores from depths 70 to 295 m at the WAIS Divide and portions of 16 cores from depths 58 to 1003 m at Siple Dome. We measured cores from all three sites during the same run so as to ensure consistent performance of the calibrated fluorimeter. The WAIS and Siple data have not yet been published.

For the $\sim 3 \times 10^5$ depths where we measured fluorescence, we divided the spectra into two categories, denoted by red points and blue points, as shown in Fig. 1. We attributed the red category, with emission peaked at 320 to 340 nm, to protein-bound Trp in microbial cells (Rohde and Price, 2007). See Rohde et al. (2008) for a discussion of how we used ground truth measurements of stained cells to relate cell concentration to fluorescence intensity. We attributed the second category, with emission spectra peaked at other wavelengths (denoted “non-Trp”), to non-microbial aerosols, which included the few mineral grains with fluorescent intensity above background. Rohde et al. (2008) showed that, of the non-Trp particles, the spectra with strongest fluorescence monotonically decreased with emission wavelength, which matched that of dissolved organic matter (DOM) that had been transported from near-surface ocean water (Mopper and Schultz, 1993) into the atmosphere and thence onto ice. The lower panel of Fig. 1 shows examples. Miteva and Brenchley (2005) and Tung et al. (2005, 2006) found from direct observations that bacteria in GISP2 ice had sizes typically 0.2 to $0.5 \mu\text{m}$, and Priscu (2007, private communication) found from flow cytometry that both biotic particles (those containing DNA that stains with Syto 60) and abiotic particles in the WAIS Divide ice core had sizes peaked at $0.3 \mu\text{m}$. The bacteria and abiotic particles in both GISP2 and WAIS Divide ice were similar in size to the smallest oceanic microbes. By contrast, fluorescence spectra of individual aerosol particles were sensitive only to particles with supermicron sizes (e.g. Pan et al., 2007) and thus may not

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have been representative of the particles trapped in glacial ice.

Some of our Trp spectra may have been due to free Trp or to proteins not associated with live cells. Free Trp has a rather sharp emission peak at 353 nm, whereas the emission peak for protein-bound Trp is ~ 330 nm, the downshift being the result of binding into proteins. About 10% of the spectra we characterized as Trp-like had a peak more consistent with that of free than of bound Trp. Based on ground-truth calibrations in which we compared counts of particles with Trp-like spectra with direct counts of stained cells at various depths in the GISP2 ice (Tung et al., 2005), we conclude that the great majority of the points with Trp-like spectra were due to proteins in microbial cells. Using a live/dead stain for their direct counts of cells, Miteva et al. (2006) found that the fraction alive in GISP2 ice (i.e., with uncompromised membranes) at various depths ranged from $\sim 2.5\%$ to $\sim 80\%$. Even those that failed the live/dead test were cell-like in shape, from which we surmise that they would have showed Trp-like spectra.

Figures 2 and 3 display our data, averaged over core lengths of up to ~ 1 m to avoid clutter. The lines are power law fits to cell counts (from spectra of protein-bound Trp) and non-Trp fluorescence counts for WAIS, Siple, and GISP2 ice cores. The abscissae show ages converted from sample depths using established age vs depth relations. Systematic errors, taken as $\pm 30\%$ for each point, dominate over statistical errors. They are mainly due to uncertainty in the distribution of cell sizes with depth and to the uncertain fraction of the Trp-like spectra that might be due to proteins not in cells. The large scatter of fluorescence intensities with depth exemplifies the stochastic nature of the deposition of microbes and non-Trp aerosols onto the ice. The smaller the volume of illuminated ice from which fluorescence was able to enter the fluorimeter, the larger the fluctuations. For the present work the volume sampled at a single location was a cylinder $\sim 200 \mu\text{m}$ in diameter and ~ 0.5 cm in depth. In an ongoing unpublished study with a new fluorimeter designed by R. A. Rohde, the volume sampled was more than 10^2 times greater (1 to 2 mm beam diameter, depth 1 to 2 cm), as a consequence of which the fluctuations in microbial and non-Trp concentrations were much smaller.

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Table 1 compares our measurements of relative concentrations of microbial cells and non-Trp aerosols in various ice cores with relative concentrations of mineral dust and MSA reported in the references. Concentration ratios (column 2) are shown for the locations in column 1. The ratios of fluxes into the ice (column 4) were obtained by multiplying the concentration ratios by the ratios of ice accumulation rates (column 3). Accumulation rates vary in a complex way with glacial stage and with abrupt climate change, and we have used the most authoritative values in the literature. See Cuffey and Clow (1997) for a discussion of the complexities of how accumulation rates are determined from data on annual layer thicknesses. In part 1 of Table 1, our data on microbes in ice at the west Antarctic sites (WAIS and Siple Dome) relative to those at GISP2 are based on values of the averages in Fig. 2 taken at ~ 1.5 ka (1500 years) before present where data from the three sites overlap. The data for non-Trp aerosols at ~ 1.5 ka are obtained from Fig. 3. Also shown in part 1 are ratios of mass concentrations of microbes to dust concentrations in Greenland ice and in WAIS and Siple Dome ice, using the dust data in Fig. 4. In the absence of direct measurements of dust at GISP2, we used dust data at NGRIP (some 200 km north of the GISP2 site), and in the absence of dust data for WAIS or Siple Dome, we used an average of dust data at Dome C, Vostok, and EDML (Dronning Maud Land) (Delmonte et al., 2004; Ruth et al., 2008). The ratios of mass concentrations of microbes to dust were $\sim 10^{-2}$ for both Greenland and Antarctica.

Part 2 of Table 1 gives ratios we calculated for mineral dust in ice cores from various Antarctic and Greenland sites for part of the Holocene period (3 to 5 ka) and for the LGM (Last Glacial Maximum, 20 to 25 ka), using the data in Fig. 4. Part 3 of Table 1 gives ratios we calculated for MSA in ice cores from Antarctic and Greenland sites, using the data in Fig. 5.

Table 1, along with Figs. 2 and 3, provides an overview of results of our use of scanning spectrofluorimetry to infer microbial concentrations and non-Trp aerosols in glacial ice. As the table shows, fluxes of microbes, non-Trp aerosols, and mineral dust onto Antarctic ice are only $\sim 10\%$ to 20% as great as onto Greenland ice. To account

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for the similarities in the ratios, we note that all of them have roughly similar sizes (sub-micron to micron). As a result, it is likely that microbes, aerosols, and dust particles are swept up from desert and ocean surfaces with similar efficiency and are transported along similar flight paths. The small values of the ratios (0.23, 0.17, and 0.09) suggest that Antarctic sources have a smaller source area and/or less direct flight paths than Greenland sources. Li et al. (2008) concluded from modeling that Patagonia (South America) and Australia are the main sources of the dust deposited in Antarctica but that they differ zonally, with each one dominating half of a hemisphere along 120° E–60° W: the half comprising the Atlantic and Indian Oceans in the case of Patagonian dust and the Pacific half in the case of the Australian dust.

Another subtlety is that there may have been a shift in the origin of dust between interglacial and glacial periods (Revel-Rolland et al., 2006). Unfortunately, detailed dust data are not yet available for west sites, and detailed microbial data are not yet available for east Antarctic sites (Vostok and Dome C), so we are not yet able to compare fluxes of dust and microbes at the same sites.

For Greenland ice, the Taklimakan and Gobi deserts in China are the dominant sources of the dust for both glacial and interglacial periods (Bory et al., 2003).

By contrast, the average flux ratio, 1.86 ± 0.4 , for MSA (Part 3, Table 1) is a factor 20 higher than the ratios for microbes, aerosols, and dust. MSA is one of the atmospheric oxidation products of dimethyl sulfide, a metabolic byproduct of dimethylsulfoniopropionate (DMSP), produced primarily by haptophytes, a phylum of sea algae. The DMSP is emitted at the sea ice margin by the haptophytes, which bloom following sea ice decay. To minimize the role of climate in MSA production, which is still not settled (Abram et al., 2007; Saltzman et al., 1997), we separately compare the flux ratios for the Holocene and for the Last Glacial Maximum. The MSA signal in ice cores seems to be determined by atmospheric transport strength and to a lesser extent by sea ice conditions (Abram et al., 2007). Antarctica is surrounded by sea ice, on the lower surfaces of which haptophytes live. Greenland is surrounded by far less sea ice than Antarctica. We infer from the low Antarctic/Greenland flux ratio of the microbes whose

fluorescence we measured that they are most likely bacteria and archaea that originate primarily not on the lower surfaces of sea ice but in sources of dust and aerosols, most likely in terrestrial desert and soil and in surfaces of the open ocean far from sea ice margins.

We have reported here our first step toward a comprehensive record of microbial and non-microbial aerosol fluxes over $>10^5$ years in glacial ice. A new version of our fluorimeter has the ability to detect chlorophyll and volcanic ash in addition to protein-bound Trp. In the next several years we will use the new fluorimeter to enumerate phototrophs, microbial cells, non-microbial aerosols, and mineral grains and rock fragments suggestive of volcanic tephra throughout the entire depth of the ~ 3600 m WAIS Divide ice core.

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Table 1. Relative Concentrations and Fluxes of Microbial Cells, non-Trp Aerosols, Mineral Dust, and MSA in Antarctic/Greenland Ice.

Part 1. Fluorimetry: microbes and non-Trp aerosols at 1.5 ka	Concentration ratio	Accumulation ratio	Flux ratio
(microbes in WAIS + Siple ice*) ÷ (microbes in GISP2 ice*)	0.33±0.17	16/23	0.23±0.11
(non-Trp aerosols in WAIS+Siple ice*) ÷ (non-Trp aerosols in GISP2 ice*)	0.25±0.12	16/23	0.17±0.08
(microbes in GISP2 ice*) ÷ (dust in NGRIP ice ^a)	0.01±0.06 g/g	n/a	n/a
microbes ÷ dust in Antarctic ice**	0.01±0.06 g/g	n/a	n/a
Part 2. Mineral dust			
Dome C ^b /NGRIP ^a (Holocene)	0.26±0.04	3.05/24	0.033
EDML ^c /NGRIP ^a (Holocene)	0.54±0.1	6.4/24	0.14
Vostok ^b /NGRIP ^a (Holocene)	0.43±0.04	2.2/24	0.039
Dome C ^b /NGRIP ^a (LGM)	0.23±0.08	1.5/6.5	0.053
EDML ^c /NGRIP ^a (LGM)	0.33±0.1	4.1/7	0.19
Vostok ^b /NGRIP ^a (LGM)	0.33±0.1	1.2/7	0.057
Average Antarctic dust/Greenland dust	0.35±0.11	–	0.09±0.06
Part 3. Methanesulfonate (MSA)			
Siple ^d /GISP2 ^e (Holocene)	4±1	0.48	1.9±0.5
Siple ^d /GISP2 ^e (LGM)	4.6±0.7	4/7	2.6±0.45
Vostok ^f /GRIP ^g (LGM)	9±1.5	1.2/7	1.5±0.25
Vostok ^f /NGRIP ^h (LGM)	8±1.6	1.2/7	1.4±0.3
Average Antarctic MSA/Greenland MSA	6.4±2.8	–	1.86±0.4

* This work.

** Microbes were measured in WAIS and Siple Dome ice; Antarctic dust was taken as average of Dome C, EDML, and Vostok dust.

^a Ruth et al. (2003); ^b Delmonte et al. (2004); ^c Ruth et al. (2008); ^d Saltzman et al. (2006); ^e Saltzman et al. (1997);

^f M. Legrand et al. (1991); ^g Legrand et al. (1997); ^h Jonsell et al. (2007).

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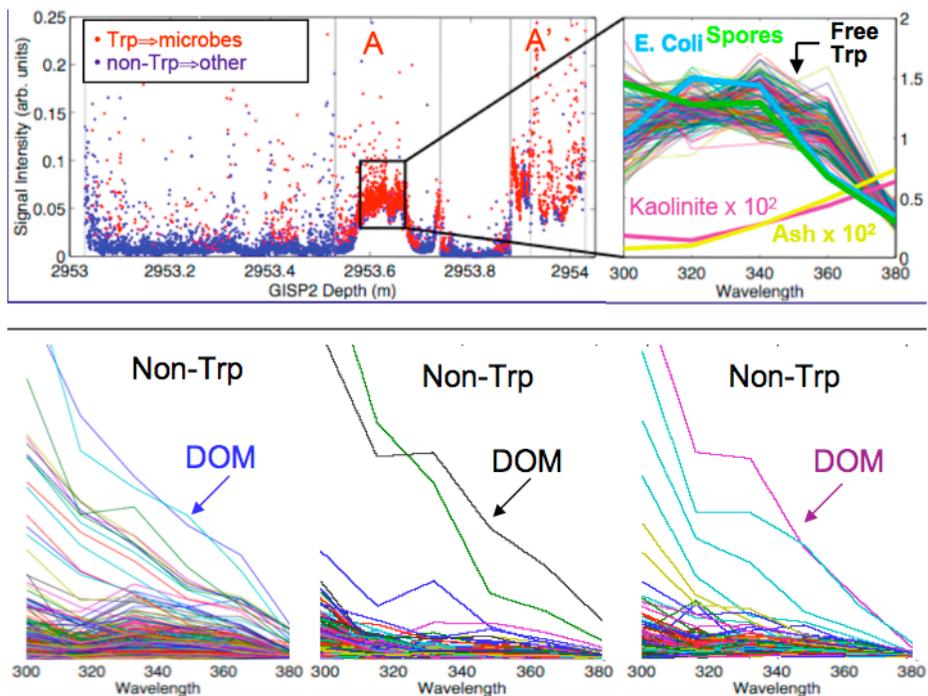


Fig. 1. Upper panels: calibration of spectrofluorimeter to microbes. (Left side) intensity of fluorescence along a 1-m section of GISP2 ice. Points in red indicate protein fluorescence; points in blue have spectra inconsistent with proteins. (Right side) Spectra for measurements in the boxed region compared with spectra for lab specimens for bacteria and minerals. Arrow indicates wavelength for which free Trp would show a maximum. Curves in lower three panels show examples of non-Trp spectra.

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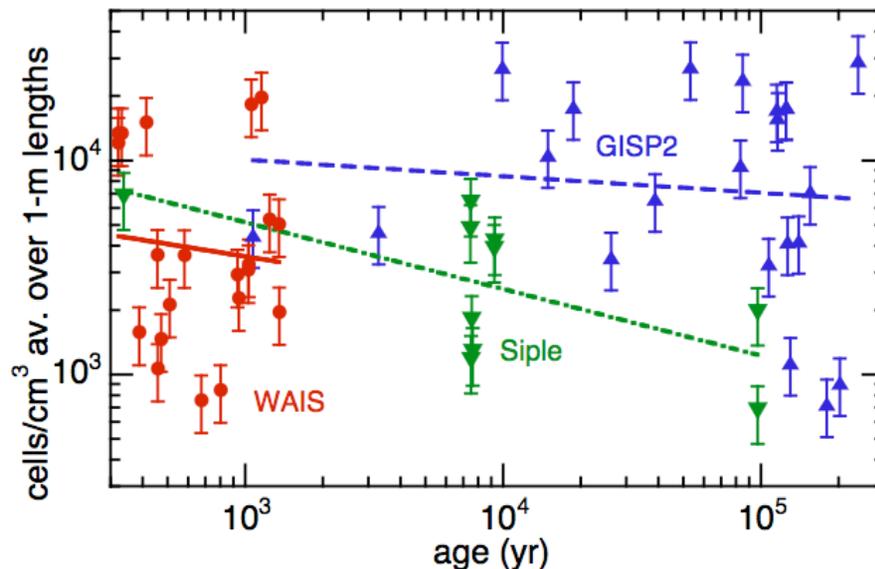


Fig. 2. Microbial cell concentrations inferred from Trp fluorescence in ice cores from WAIS, Siple, and GISP2. Symbols are average values in individual core sections up to 1 m in length. Microbial cells from some depths in GISP2 ice with ages $>1.1 \times 10^5$ yr may have originated in basal ice and been transported upward by turbulent flow. Lines are power law fits with points for a given core given equal weight. Because GISP2 ice with ages $>1.1 \times 10^5$ yr are uncertain, those data are excluded from the fits.

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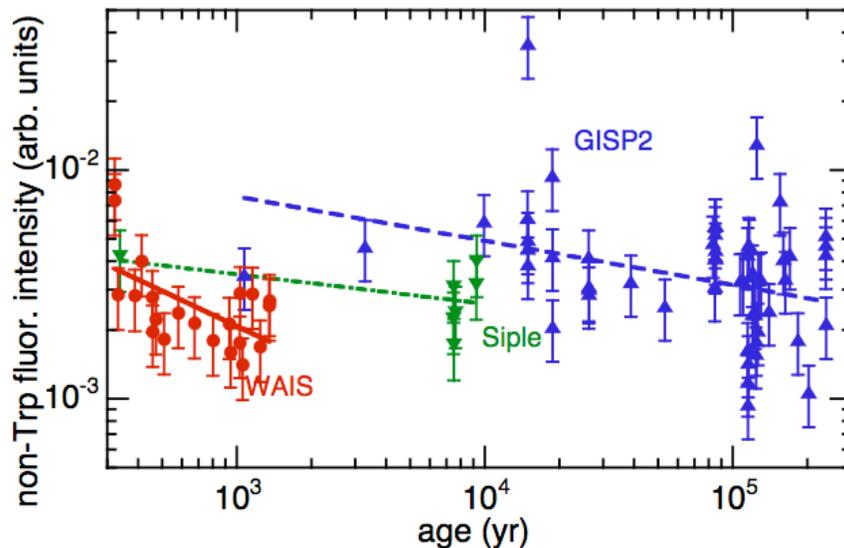


Fig. 3. Concentrations of aerosols (dominantly organic) from non-Trp fluorescence spectra (same symbols and lines as in Fig. 2). Symbols are average values in 1-meter depth intervals. Non-Trp particles from some depths in GISP2 ice with ages $>1.1 \times 10^5$ yr may have originated in basal ice and been transported upward by turbulent flow. Lines are power law fits with points for a given core given equal weight; because GISP2 ice with ages $>1.1 \times 10^5$ yr are uncertain, those data are excluded from the fits.

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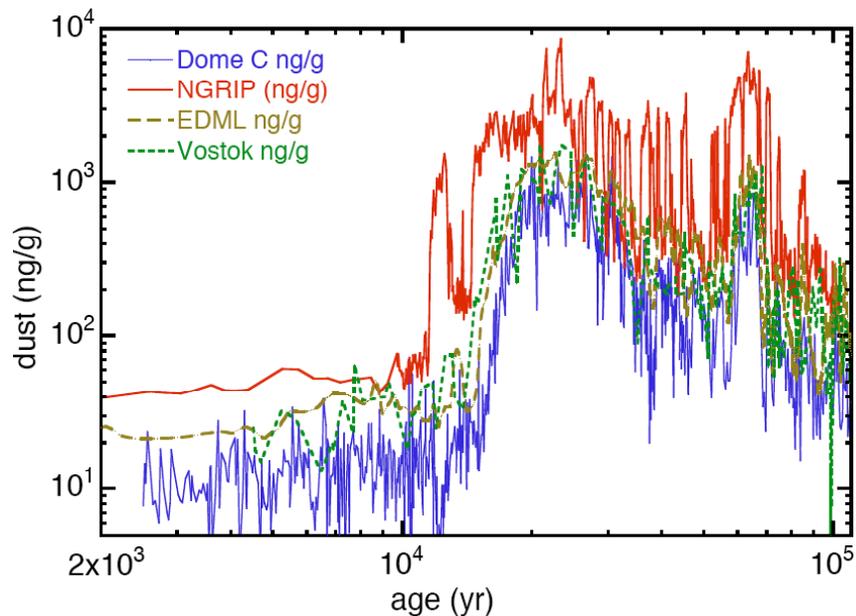


Fig. 4. Concentration of mineral dust at Greenland and Antarctic sites. See Table 1 for references.

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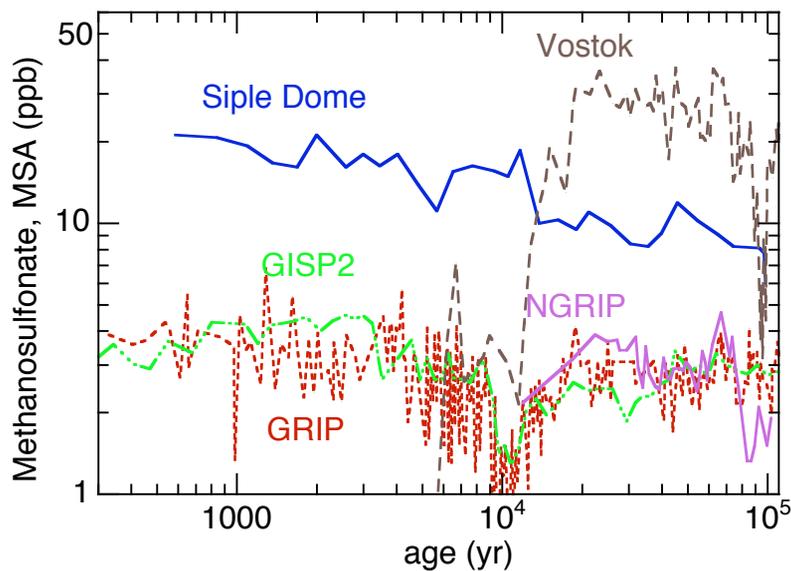


Fig. 5. Concentration of MSA at Greenland and Antarctic sites. See Table 1 for references.

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