The 800 year long ion record from the Lomonosovfonna (Svalbard) ice core

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[1]   We present a high-resolution record of water-soluble ion chemistry from a 121 m ice core spanning about 800 years. The core is well dated to 2/3 depth using cycle counting and reference horizons and a simple but close fitting model for the lower 1/3 of the core. This core suffers from modest seasonal melt, and so we present concentration data in decadal running means to minimize percolation effects. Sea-salt ions (Na^+, Cl^-, Mg^{2+}, and K^+) account for more than 70% of all ions. In general, sea-salt ion concentrations are rather variable and have no clear association with climatic variations. Sulfate, with 74% being from non-sea-salt sources, has higher concentrations than seen on Vestfonna ice cap but lower than in Ny-Ålesund aerosols, suggesting central Spitsbergen receives more marine (westerly) air masses than Ny-Ålesund but more sulfate enriched (easterly) air masses than Nordaustlandet. Clear anthropogenic impacts are found for sulfate, nitrate, and ammonium (and probably excess chloride) after the mid twentieth century, with sulfate showing a significant rise by the end of the nineteenth century. Sulfate and methanesulfonate concentrations correlate well during the twentieth century, and it is clear that most of the preindustrial sulfate is of biogenic origin. Terrestrial component (Ca^{2+}) has the highest concentrations in the coldest part of the Little Ice Age, suggesting more windy conditions, transporting local terrestrial dust to the ice cap. All ion concentrations decrease at the end of the twentieth century, which reflects loss of ions by runoff, with non-sea-salt magnesium being particularly sensitive to melting.

1.   Introduction

[2]   Ice cores are well known as one of the best archives of past climatic and environmental changes [e.g., *Legrand and Mayewski*, 1997]. In Greenland and Antarctica these archives can span hundreds of thousands of years, but the smaller ice caps of the Arctic mostly contain only Holocene ice and typically records span a few hundred years [*Koerner*, 1997; *Delmas*, 1992; *Wolff*, 1990]. Water-soluble ions provide a lot of information on past atmospheric composition, with both short events (e.g., volcanic eruptions, storms, forest fires) and long-term changes (e.g., anthropogenic input, biogenic production, temperature variations) recorded. If no migration (postdepositional changes and melting) occurs, the ice core would contain a record of the original snowfall composition that becomes buried over time. However, in many locations, outside the cold polar interiors of the large ice sheets, some melting occurs in summer, and ions also diffuse as ice is compressed or deformed by flow. Nevertheless despite some melting, water-soluble ions provide a great deal of information on the past [*Kotlyakov et al.*, 2004].

[3]   The small ice caps of the Arctic are situated relatively close to large sources of anthropogenic pollution, and their low altitude provides more regional signal than found in the central Greenland ice cores. They are therefore valuable for assessing impacts and source histories in detail over the period since the industrial revolution [*Koerner et al.*, 1999]. Svalbard is in an interesting geographical position surrounded by Arctic Ocean, Barents Sea and North Atlantic and located at the southerly edge of the permanent Arctic sea ice. In addition it is near to the overturning point of the North Atlantic thermohaline circulation.

[4]   Several ice cores have been drilled on Svalbard ice fields producing records less than 1000 years long. Groups from the former Soviet Union drilled two ice cores on Lomonosovfonna in 1976 [*Gordiyenko et al.*, 1981; *Vaikmäe*, 1990] and in 1982 [*Zagorodnov et al.*, 1984] and five ice cores from other sites around Svalbard between 1975 and 1987 [*Punning et al.*, 1980; *Zagorodnov and*

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Figure 1. Map of Svalbard showing the Lomonosovfonna ice core site, Ny-Ålesund, and the Snøfjellafonna, Vestfonna, and Austfonna ice caps.

Zotikov, 1981; *Vaikmäe et al.*, 1984; *Kotlyakov*, 1985; *Punning et al.*, 1985; *Punning and Tyugu*, 1991; *Tarussov*, 1992]. Most ice core results presented have been stratigraphic studies. The only ion analyses done were of chloride and sulfate [*Punning et al.*, 1987; *Vaikmäe*, 1990]. However, the low concentrations in the ice were not measured very precisely because of a lack of sophisticated instruments. The Japanese have also drilled several ice cores at other sites in Svalbard since 1987, [e.g., *Goto-Azuma et al.*, 1995; *Watanabe et al.*, 2001; *Matoba et al.*, 2002]. Only a few ion records have been published because most of the ice cores have not yet been analyzed. Published records cover only short time periods for certain ions. The most significant ion results from the ice cores show increases in sulfate and nitrate in the mid twentieth century in Snøfjellafonna [*Goto-Azuma et al.*, 1995], Austfonna [*Watanabe et al.*, 2001] and Vestfonna [*Matoba et al.*, 2002] (Figure 1).

[5] In 1997 we drilled a 121 m long ice core (spanning about the 800 years) on Lomonosovfonna (Figure 1), the highest ice field in Svalbard (1255 m above sea level (asl)), where the current annual temperature range is from $0°C$ to about $-40°C$. Total ice depth from radar sounding was 123 m, and the site is close to the highest point of the ice cap with roughly radial ice flow. Published data from a previous, lower-elevation ice core on Lomonosovfonna drilled in 1976 indicated better preserved stratigraphy than the other sites on Svalbard [*Gordiyenko et al.*, 1981]. Therefore we selected the summit of Lomonosovfonna as our coring site with the aim of making higher-resolution sampling and better stratigraphy than accomplished in previous cores from this area.

[6] Studies so far indicate that our Lomonosovfonna ice core contains a reliable record of isotope and chemical concentrations that can be successfully used in climate and environmental studies [e.g., *Isaksson et al.*, 2001]. Interpretation of the ion records from the upper part of the core covering the period 1920–1997 is presented by *Isaksson et al.* [2001]. *Isaksson et al.* [2001] and *Kekonen et al.* [2002] reported increases in sulfate and nitrate in the mid twentieth century. Although local sources of pollution have to be taken into account [*Simões and Zagorodnov*, 2001], there are also clear anthropogenic signals from long-range transport to the Lomonosovfonna ice cap [*Kekonen et al.*, 2002], probably from both Eurasia and also from North America. *Vehviläinen et al.* [2002] found PAH compounds in the core and they concluded that these come from long-range sources. *Isaksson et al.* [2003] suggested that the twentieth century was the warmest during the past 600 years based on $\delta^{18}O$ record from both the Lomonosovfonna and also the Austfonna ice cores (Figure 1). *O'Dwyer et al.* [2000] and *Isaksson et al.* [2005] discussed methanesulfonic acid variations in terms of Barents Sea conditions. The annual accumulation is calculated back to 1715A.D. by *Pohjola et al.* [2002b]. The borehole temperature profile is modeled by *Van de Wal et al.* [2002] and shows that the nineteenth century was about $2°-3°C$ cooler than the twentieth century. Chemical analytical methods are described by *Kekonen et al.* [2002, 2004] and *Jauhiainen et al.* [1999]. Generally, ions have a certain order of elution and runoff [e.g., *Davies et al.*, 1982]. Melting in the Lomonosovfonna ice core was studied in detail by *Pohjola et al.* [2002a] and showed that nitrate and sulfate are most easily percolated, while ammonium and oxygen isotopes are virtually unaffected. Calcium is one of the most conservative species.

[7] In this paper we focus to the chemistry of water-soluble ions in the Lomonosovfonna ice core from Svalbard. We present ion concentration profiles over an 800 year period that includes the whole of the Little Ice Age. In earlier studies, only the upper 36 m, post-1920 period has been discussed in detail.

2. Sample Preparation and Analysis

[8] Ice core sections were transported in a frozen state (in plastic sleeves) from Svalbard and stored in a cold room ($-22°C$) at Norwegian Polar Institute in Tromsø and then transported to the Rovaniemi Research Station of the Finnish Forest Research Institute. The ice core was cut into 5 cm sections and the outer parts of the samples were removed in a cold room with strict contamination control. Artificial cores were also made by freezing ultra pure water in similar size of tube as the drilled ice core. Artificial cores were handled in the same manner as the real ice core, and no significant contamination was found. All samples (over 1000 in total) were melted just before chemical analyses. Ion analyses (Na^+, K^+, NH_4^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^-, SO_4^{2-}, $CH_3SO_3^-$) were made using a Dionex ion chromatograph with conductivity detector and suppressor housed in a clean laboratory. All sampling, melting and ion analyses were done under a laminar flow hood. The middle part of the ice core (36–60 m) was analyzed in the glaciology clean room at the British Antarctic Survey in 1999, and the rest of the core was analyzed in Rovaniemi in 1999 and 2000 in a similar manner. *Kekonen et al.* [2004, 2002] and *Jauhiainen*

et al. [1999] describe details of core handling and analytical methods. Sea-salt and non-sea-salt (nss) ratio calculations are based on ion ratios to sodium, assuming that all sodium comes from seawater [*Wilson*, 1975].

[9] In addition to ion analysis, oxygen isotopes ($\delta^{18}O$) and stratigraphic melt index (SMI) were also measured at similar resolutions. *Isaksson et al.* [2003] present the complete $\delta^{18}O$ profile back to 1400 and show how it is related to regional climate and local sea ice changes. SMI has been widely used in Arctic ice cores as a proxy for temperature [e.g., *Koerner*, 1997]. *Pohjola et al.* [2002a] discuss SMI for the upper 40 m of the core, and here we present the record to 81 m depth, which is the limit of the method used.

3. Dating

[10] The ice core was drilled at the summit of the Lomosovfonna dome and GPS surveys of the area suggest ice flow is essentially radial. As this position is the center of the ice cap a simple ice flow model [*Nye*, 1963] can be used to establish an age/depth relationship within the ice cap from the surface all the way to bedrock. This approach requires only knowledge of ice thickness and accumulation rate and therefore has the fewest number of free parameters possible with which to date the ice core. The radar travel time to from surface to bedrock was measured with 50 MHz radar along five transects within 20 m of the borehole. The ice cap density profile can then be used to convert this travel time to ice thickness. Detailed density measurements on the upper 18 m of the core combined with density from visual stratigraphy of the whole core allows computation of the actual ice cap thickness of 123 ± 1 m at the core site. Thus the core reached to within a $2-3$ m of bedrock (drilling was halted because of logistic constraints). We slightly modified the Nye dating model, which only requires a constant accumulation rate, to utilize two accumulation rates for different parts of the core found from two known reference horizons. These horizons are both in the upper half of the core: the 1963 radioactive layer and the 1783 Laki volcanic sulfate layer and volcanic ash particle. The mean accumulation rate for the $1997-1963$ period is 0.41 m water equivalent (w eq) per year (*Pinglot et al.* [1999] presented a value of 0.36 m w eq yr^{-1} but this neglected layer thinning with depth) and a somewhat lower value of 0.31 m w eq yr^{-1} for the period $1963-1783$. The model age profile can be independently checked by comparison with automated seasonal cycle counting in stable isotopes and ions down to 81 m [*Pohjola et al.*, 2002b]. The model age at 81 m depth is 1705, while the cycle counting method gives a date of 1715. There is a thus a discrepancy of 10 years in about 75 years between the Laki horizon and the limit of cycle counting. However, the cycle counting method will always tend to miss a fraction of low accumulation rate years because of the resolution of the data and isotope diffusion effects, so a good model dating should be more reliable (on average) than cycle counting. Even so, if we assume that 1715 is the correct date at 81 m depth, mean accumulation rates from 1783 to 1715 are virtually the same as between 1963 and 1783. Thus the mean accumulation rate appears surprisingly constant prior to 1960, despite both anthropogenic impacts and the end of the Little Ice

Age. An increase in accumulation around 1960 is also observed over much larger spatial scales by radar internal reflection horizons [*Pälli et al.*, 2002].

[11] Below 81 m (i.e., about 2/3 total ice thickness) we have no direct dating. However, there are a number of observations that give confidence to simple flow modeling: The basal ice layers show no disturbances in stratigraphy, no basal debris bands are observed, and the crystals are large and interlocked. Time series analysis of ions reveals several multidecadal cycles that exhibit statistically significant power throughout the whole length of the core; these would not be present if the timescale were in gross error. Furthermore, the Little Ice Age glacier advance began in the fourteenth and fifteenth centuries on Svalbard, culminating in the nineteenth century [*Svendsen and Mangerud*, 1997], while sea ice in the western Barents Sea retreated dramatically around 1920. These climatic indicators are in good agreement with the Lomonosofonna $\delta^{18}O$ profile which has values before 1400 (T. Martma, unpublished data, 2000) that are comparable to those of the twentieth century. Therefore we base our ice core stratigraphy on a modified Nye flow model using an accumulation rate of 0.41 m w eq yr^{-1} for $1963-1997$ and 0.31 m w eq yr^{-1} for earlier periods. Figure 2 shows the timescale including the marker horizons and the independent timescale from layer counting. The close fit of our model using well-constrained and few free parameters added to the independent cycle counting increases confidence in the extension of the timescale to the bottom of the core.

4. Results

[12] Recent calculations suggest that, though for most the Lomonosovfonna core ions are preserved within an annual or biannual layer [*Pohjola et al.*, 2002a], in the warmest years (such as post-1990), percolation lengths appear to be $2-8$ annual layers. Therefore to remove most of the percolation effects we present the ion data as 10 year running means. There is evidence of loss of ions under present conditions, most likely as runoff during summers, but we argue that this is much less important in the past based on the ion concentrations and $\delta^{18}O$ profiles.

4.1. Fluxes and Concentrations

[13] *Pohjola et al.* [2002b] show that annual cycles can be observed in the stable isotopes profile to at least 81 m depth, and could be used to calculate accumulation rates for the past 300 years. We expect that wet deposition is the dominant mechanism of chemical species removal from the atmosphere to the snow in a marine, relatively high precipitation, location such as Lomonosovfonna. If dry deposition is important on Lomonosovfonna we would expect to see a clear relationship between ion concentrations and accumulation rate [*Delmas*, 1992]. This is not observed for any ion; for example, Figures 3a and 3b show the accumulation rate dependences of nitrate and calcium concentrations. With these two ions, the highest concentrations do tend to decrease with increasing accumulation rate (though there is no statistically significant trend), hinting at some dry deposition perhaps as large discreet particles (as may be expected from locally derived terrestrial sources). We also tested the relationship separately over the post-1950
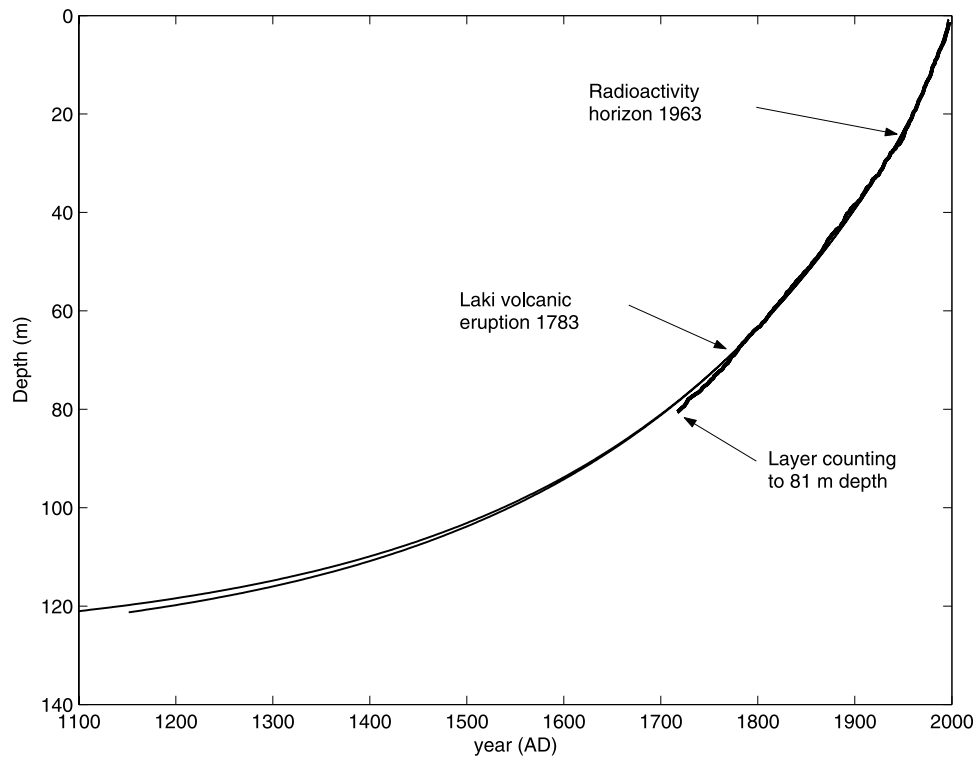
Figure 2. Timescale of the core. Reference horizons are marked: 1963 radioactive layer and 1783 Laki volcanic sulfate layer. The model curves extending to bedrock show two lines representing the dating uncertainty arising simply from the radar errors in ice thickness. The irregular curve extending to 81 m depth is the independent timescale obtained by automated layer counting.

period to remove effects from changes in species sources, and again found no significant dependence of concentration on accumulation rate. Similar results are also found for ice cores from Antarctica [*Reusch et al.*, 1999; *Sommer et al.*, 2000]; we are not aware of any corresponding analyses for other Svalbard or Arctic ice cores. Because concentrations appear to be independent of accumulation in our records, we will use the concentrations instead of fluxes in the rest of this paper.

4.2. Sodium, Chloride, Potassium, and Magnesium

[14] The ice core is dominated by the large contribution from sodium and chloride ions, on average they account for 63% of the whole ice core soluble ion concentrations (in μeq L^{-1}), and they vary between 52% and 75% in different time periods (Table 1). The mean sodium chloride ratio of 0.93 (in μeq L^{-1}) is near the bulk seawater ratio (0.86). Given the maritime location of the ice core and the measured sodium chloride ratio, we presume that the ocean is the major source for sodium ions. Svalbard aerosols also appear to be marine origin with only minor terrestrial sources of sodium [*Teinilä et al.*, 2003]. Chloride mainly comes from the ocean but is not as good a sea-salt marker as sodium, because chloride is more sensitive to postdepositional processes [e.g., *Legrand and Delmas*, 1988; *Davies et al.*, 1982] and may as well be affected by anthropogenic input of HCl [*Legrand et al.*, 2002].

[15] The complete sodium, chloride, potassium and magnesium profiles along the entire core illustrate rather similar trends (Figure 4). Potassium and magnesium do not show any great changes in average concentrations until in the twentieth century (Table 1). Sodium and chloride concentrations are high between fifteenth and mid sixteenth centuries. The period corresponds to the warmest part of the Lomonosovfonna δ^{18}O profile (Figure 4). This suggests that smaller sea ice extent because of warmer temperatures, allowed more efficient transport of sodium and chloride aerosols from the ocean. In several Greenland ice cores, sodium has higher concentrations during cold periods of the Little Ice Age [*Fischer et al.*, 1998]. However, the Lomonosovfonna δ^{18}O profile shows that the coldest part of the Little Ice Age (LIA) occurs from the mid eighteenth century to the beginning of the twentieth century in this region, and no sea-salt ions (Na$^+$, Cl$^-$, K$^+$, Mg^{2+}) show significantly higher concentrations in our ice core records at that time (Table 1). The Mount Logan ice core from Canadian Arctic shows a slight decreasing trend in magnesium concentrations from eighteenth to twentieth century [*Kang et al.*, 2003], and a south Greenland ice core (20D) shows an opposite trend during these three centuries [*Kang et al.*, 2003]. In this Lomosovfonna ice core magnesium concentrations show no systematic change in concentration. This may be explained by sea-salt magnesium being a much larger fraction of total magnesium than in more inland or high-elevation sites, and local dust rather than long-distance transport being the source of the non-sea-salt magnesium.

[16] Sodium concentrations may be linked to the natural variability of sea-salt input, caused by varying efficiency of transport by wind transport (storms) [*Hara*, 2000]. The Vestfonna ice core (Figure 1) shows high sea-salt
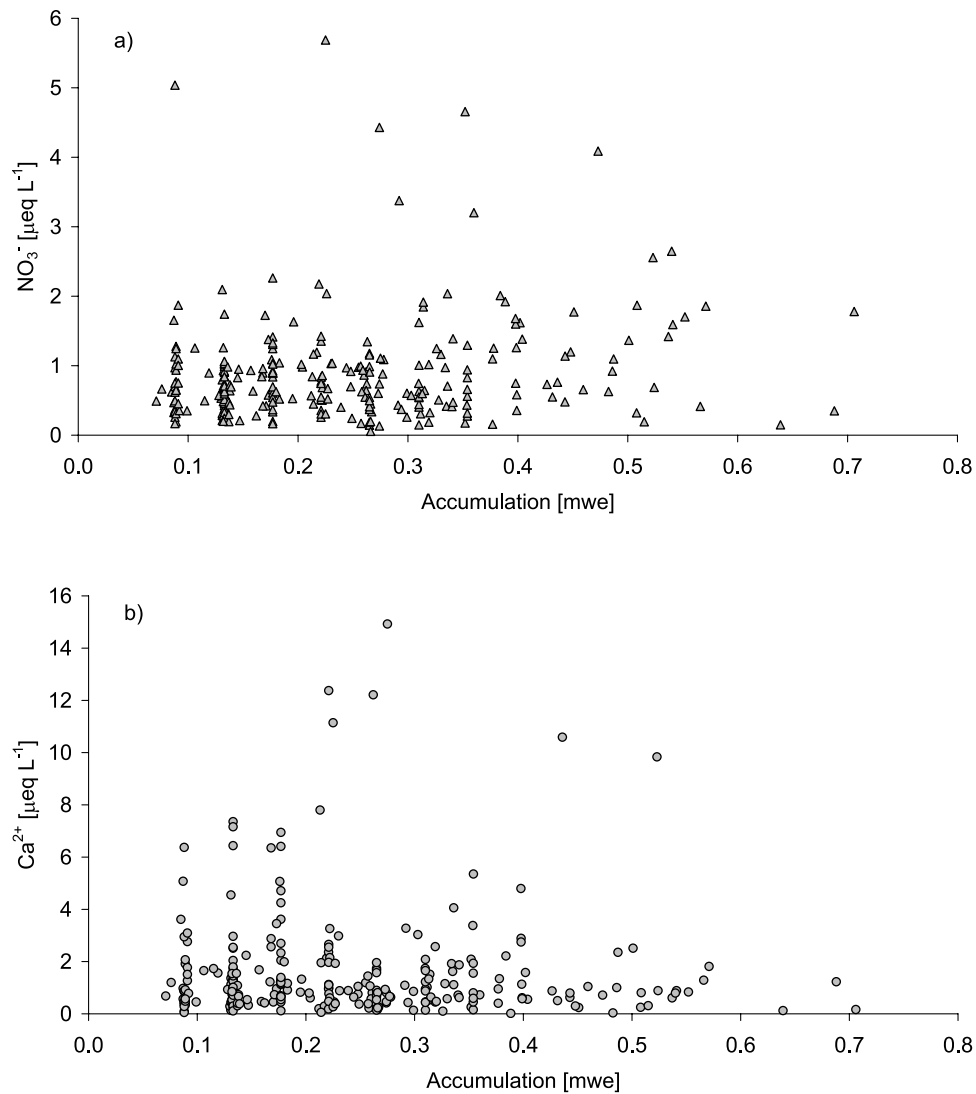
Figure 3. (a) Nitrate and (b) calcium concentrations as a function of accumulation rate. Concentrations are annual averages.

concentrations before 1783, and the Austfonna (Figure 1) shows high sea-salt concentrations between 1920 and 1963 [*Watanabe et al.*, 2001]. Sodium in our ice core shows low concentrations from mid eighteenth to mid nineteenth century, and then concentrations increase until the mid twentieth century. *Isaksson et al.* [2003] explain some of the differences between the records from Nordaustlandet and Spitsbergen as being due to the lower altitude

Table 1. Average Concentrations in Different Time Periods[a]

Years	Na^+	Cl^-	K^+	Mg^{2+}	Ca^{2+}	SO_4^{2-}	$CH_3SO_3^-$	NO_3^-	NH_4^+	n
Whole core	7.2 (0.2)	7.9 (0.2)	0.17 (0.01)	1.6 (0.1)	1.5 (0.1)	3.3 (0.1)	0.10 (0.00)	0.79 (0.02)	1.1 (0.0)	1028
1123–1400	6.4 (0.2)	6.8 (0.2)	0.12 (0.01)	1.0 (0.1)	0.45 (0.04)	1.5 (0.1)	0.03 (0.00)	0.34 (0.02)	0.95 (0.04)	74
1400–1550	8.1 (0.2)	8.3 (0.2)	0.19 (0.01)	1.7 (0.1)	1.1 (0.1)	2.3 (0.1)	0.10 (0.01)	0.59 (0.02)	1.1 (0.0)	98
1550–1700	7.6 (0.3)	7.9 (0.3)	0.17 (0.01)	1.7 (0.1)	1.5 (0.1)	2.3 (0.1)	0.11 (0.00)	0.69 (0.02)	0.90 (0.04)	171
1700–1750	7.3 (0.4)	4.2 (0.5)	0.21 (0.01)	1.7 (0.1)	1.3 (0.1)	2.5 (0.2)	0.10 (0.01)	0.72 (0.05)	0.89 (0.06)	76
1750–1800[b]	6.2 (0.3)	7.2 (0.4)	0.17 (0.01)	1.6 (0.1)	2.0 (0.3)	3.1 (0.3)	0.11 (0.01)	0.68 (0.03)	0.92 (0.05)	106
1800–1850	6.4 (0.4)	7.2 (0.4)	0.20 (0.02)	1.7 (0.1)	1.5 (0.2)	2.4 (0.2)	0.12 (0.01)	0.75 (0.05)	1.2 (0.1)	127
1850–1900	7.7 (0.5)	8.8 (0.5)	0.24 (0.02)	1.8 (0.2)	2.2 (0.2)	3.2 (0.3)	0.12 (0.01)	0.91 (0.07)	1.3 (0.1)	151
1900–1920	9.3 (0.7)	11 (0.8)	0.20 (0.02)	2.2 (0.2)	1.9 (0.2)	4.0 (0.4)	0.13 (0.03)	0.76 (0.05)	1.1 (0.1)	77
1920–1950	9.0 (0.7)	9.1 (0.6)	0.11 (0.01)	1.8 (0.2)	2.6 (0.4)	5.2 (0.6)	0.06 (0.01)	0.71 (0.06)	0.80 (0.06)	88
1950–1970	6.9 (0.6)	7.5 (0.7)	0.15 (0.03)	1.5 (0.3)	1.0 (0.1)	7.5 (1.0)	0.08 (0.01)	1.5 (0.1)	1.5 (0.3)	63
1970–1997	4.5 (0.4)	5.8 (0.5)	0.07 (0.01)	0.70 (0.08)	0.88 (0.15)	4.0 (0.4)	0.05 (0.01)	1.2 (0.1)	1.2 (0.1)	97

[a]Standard errors are shown in parentheses. Concentrations are in μeq L^{-1}; n is number of samples.
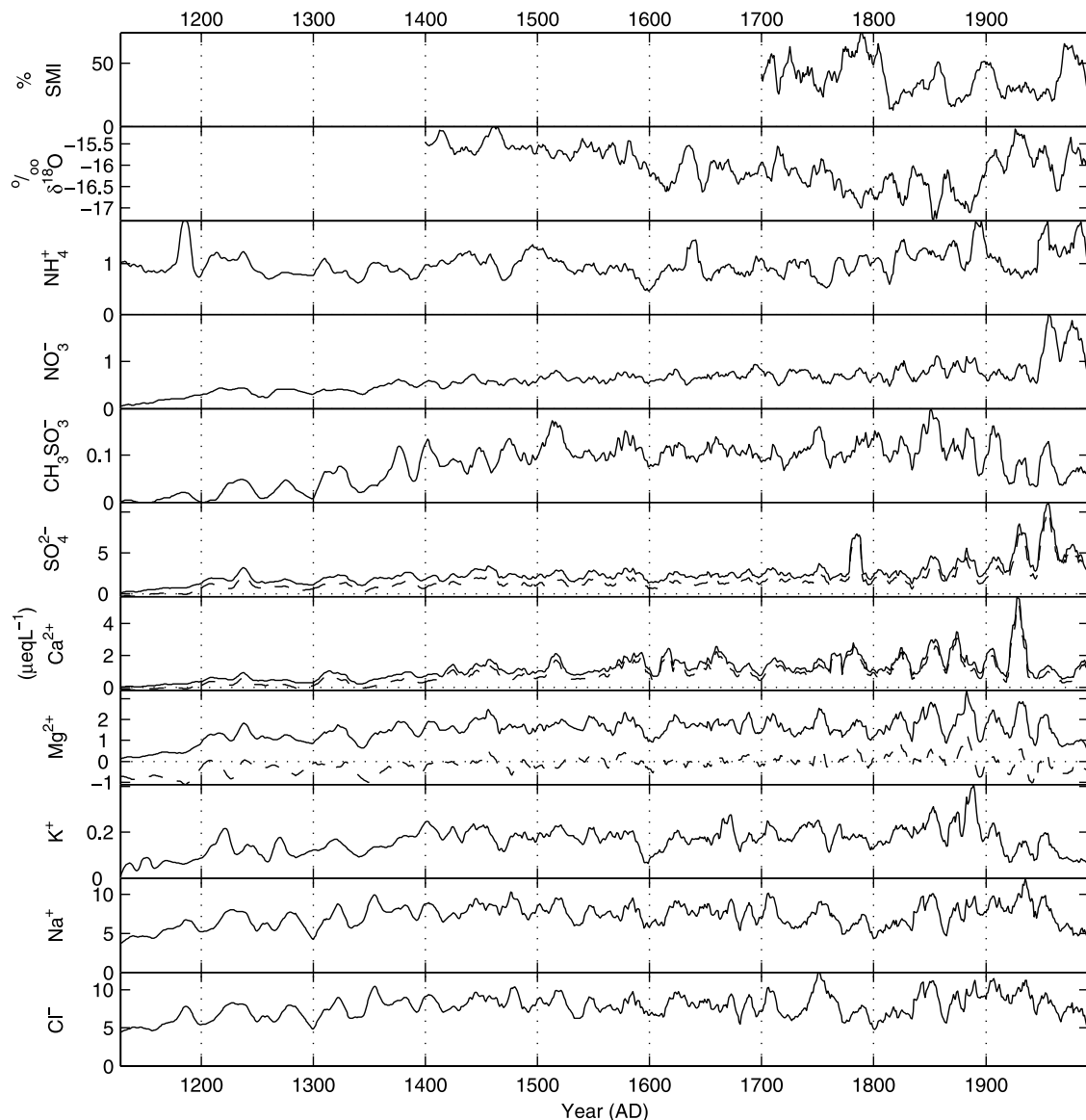[b]Without sulfate concentrations during Laki volcanic eruption (two samples).

Figure 4. Time series of water-soluble ion concentrations (in μeq L^{-1}). The original 5–10 cm resolution data were resampled to yearly values using the model shown in Figure 2, and 10 year running means were taken. Non-sea-salt (nss) magnesium, calcium, and sulfate are marked by dashed lines; the $\delta^{18}O$ profile (‰) is adapted from *Isaksson et al.* [2003].

Nordaustlandet sites being more influenced by changes in storm tracks than the relatively high altitude Lomonosov-fonna site. However, in general, sodium and chloride concentration are rather variable with no clear association with climatic variations. Between 1920 and 1940 sodium displays a very spiky profile with many high values. This is known as a time period when NAO and AO were fairly uncorrelated and the atmospheric circulation patterns were reorganizing after the end of the LIA [*Jevrejeva and Moore*, 2001].

[17] All sea-salt ion concentrations (Na^+, Cl^-, K^+ and Mg^{2+}) start to decrease at the beginning of the twentieth century, when rapid warming of climate occurred, but after 1950 average concentrations are even lower. Sodium, chloride, potassium and magnesium concentrations are 24%, 18%, 43% and 35% lower, respectively, after 1950 compared with whole core mean concentrations (Table 1). Melting and runoff could be a possible explanation for the decrease of these ions, with potassium and magnesium more easily lost from the core than sodium, as has been observed elsewhere [*Iizuka et al.*, 2002; *Davies et al.*, 1982]; however, chloride is also expected to be lost more easily than sodium. If runoff is the reason for low ion concentrations, then the larger decrease for sodium than chloride suggests an additional chloride source to the core, presumably HCl in Arctic haze [*Heintzenberg*, 1989].

[18] Potassium and magnesium usually peak at the same time as sodium and are typically associated with a sea-salt source but magnesium certainly has terrestrial sources on Svalbard [*Teinilä et al.*, 2004]. Nss-magnesium actually has a slight negative excess sea-salt fraction over the whole core which means that some of magnesium ions are lost by
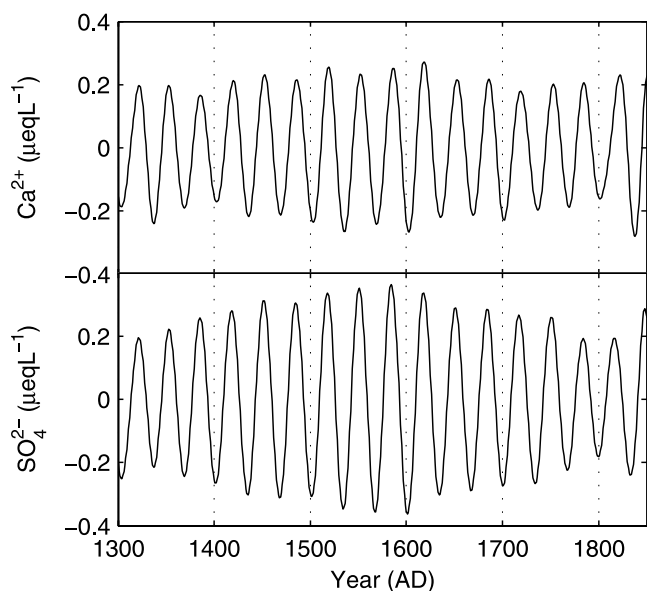
Figure 5. Singular spectrum analysis [e.g., *Jevrejeva and Moore*, 2001], with a 200 year window, components for calcium and sulfate showing 33 year phase-locked periodicities that account for 6% and 10%, respectively, of the variance in the concentration data during the preindustrial period. There is also similar behavior during the twentieth century but at 25 year periodicity. The components representing these periodicities are significant at the 95% level in Monte Carlo testing against a red noise model.

runoff from the snowpack (or were fractionated at their source) especially during the twentieth century and before the fifteenth century (Figure 4). In fact, there is an excellent agreement between nss-magnesium and broad features of the $\delta^{18}O$ profile, with generally low $\delta^{18}O$ values (cold) associated with positive nss-magnesium and high $\delta^{18}O$ (warm) with negative nss-magnesium. Nss-magnesium is of course directly related to magnesium sodium ratio, which *Iizuka et al.* [2002] showed was a good indicator of melt and hence summer temperatures.

4.3. Sulfate and Methanesulfonate

[19] Over the whole ice core sulfate is the next most abundant ion after sodium and chloride. Sulfate represents 14% of total ions (in μeq L^{-1}). Sulfate has many sources: volcanic, anthropogenic, biogenic, terrestrial and marine [e.g., *Legrand and Mayewski*, 1997]. Sea-salt and non-sea-salt fractions are shown in Figure 4, and clearly nss-sulfate is the dominant fraction.

[20] A clear feature in the sulfate profile is the large peak at a depth of 66.79 m. A volcanic ash particle identified using SEM-EDS matched the known chemical composition of tephra from the Icelandic volcanic eruption of Laki in 1783. This has also been identified as the most prominent volcanic eruption in other recent Svalbard and Canadian Arctic ice cores [*Watanabe et al.*, 2001; *Matoba et al.*, 2002; *Grumet et al.*, 1998]. In contrast to many ice core records, other volcanic signals are not clearly present in the Lomonosovfonna sulfate record. This is because of the maritime conditions around Svalbard leading to high fluxes of both sea-salt sulfate and marine biogenic sulfu-

ric acid, together with relatively high annual precipitation diluting volcanic sulfate input.

[21] Sulfate concentrations have increased 65% after 1950 compared with whole core average concentrations, which is in agreement with ice core data from other sites on Svalbard, Greenland and Canadian Arctic [e.g., *Matoba et al.*, 2002; *Yalcin and Wake*, 2001; *Goto-Azuma et al.*, 1995; *Mayewski et al.*, 1986, 1990; *Neftel et al.*, 1985]. Sulfate records from Greenland [*Bigler et al.*, 2002; *Fischer et al.*, 1998] show a probable anthropogenic rise beginning about 1890. Average concentrations (Table 1) in the Lomonosovfonna sulfate data suggest that there is also a significant rise by the end of the nineteenth century. Such an early increase is not observed in other Arctic ice cores outside Greenland [*Matoba et al.*, 2002; *Goto-Azuma and Koerner*, 2001].

[22] A detailed look at the nss-sulfate in comparison with the lower-elevation Vestfonna (600 m asl, Figure 1) shows that much less nss-sulfate species reach the Vestfonna ice cap than the Lomonosovfonna ice cap. In the Vestfonna ice cap, 45% of sulfate comes from non-sea-salt sources, while sodium and chloride concentration are also much higher than in our ice core [*Matoba et al.*, 2002]. In the Lomonosovfonna ice core 74% of sulfate is nss-sulfate. Since 1950 nss-sulfate is 88% of total sulfate. The higher concentrations of sodium and chloride (and lower nss-sulfate) at Vestfonna compared with the higher-elevation Lomonosovfonna are due to more effective deposition of sea-salt aerosols at lower altitude. Similar altitude differences were noticed in the aerosol samples at Ny-Ålesund by *Teinilä et al.* [2004], who reported that higher altitudes are influenced more by free tropospheric air than by marine air.

[23] Since about 1950, nss-sulfate accounts for 21%, and sodium and chloride together account for 52% of all ions in the core. Ny-Ålesund aerosol measurements in 2001 found nss-sulfate was 56% of all ions, while sodium and chloride together were 33% [*Teinilä et al.*, 2003]. This shows that nss-sulfate is much more abundant ion in the aerosol phase at Ny-Ålesund than in the ice core. Ny-Ålesund aerosol measurements also show that during warm periods the air masses originate from the west and have higher concentration of sea-salt aerosols, while during cold periods, the air is from the east and is enriched in sulfate [*Staebler et al.*, 1999; *Hara et al.*, 1997]. The lower nss-sulfate ratio in our core suggests that the air masses come more from the west to Lomonosovfonna than at Ny-Ålesund which is dominated more by air from the east. The Lomonosovfonna ice cap is dominated more by sulfate enriched air masses from the Barents Sea direction than Vestfonna ice cap but clearly less than Ny-Ålesund area. Another reason for lower nss-sulfate concentrations is that some of it is lost after deposition. Even though nss-sulfate has gaseous precursors, sulfate is not volatile after deposition, though clearly it is susceptible to runoff.

[24] A good terrestrial marker is calcium ion concentration (see section 4.5). In the second half of the eighteenth century, only sulfate and calcium show high concentrations (Table 1), and both ions exhibit 25–33 year quasi-coperiodicity (Figure 5) (T. Kekonen et al., Calcium and sodium carbonates in the Lomonosovfonna ice core, submitted to *Geochimica Cosmochimica Acta*, 2004, hereinafter referred to as Kekonen et al., submitted manuscript, 2004). This probably reflects a terrestrial calcium sulfate

source [cf. *Legrand and Mayewski*, 1997], and because both ions have high concentrations during the coldest period of the Little Ice Age, it indicates less snow cover or windier conditions in local regions allowing more efficient transport of dust to the ice cap. If all the non-sea-salt calcium were associated with sulfate (Figure 4 and Table 1), it would account for almost all the preindustrial non-sea-salt sulfate concentration. However, the covarying calcium sulfate concentration is about 0.4 μEq L^{-1} (Figure 5), while much of the remaining calcium is associated with carbonate, which is not directly measured, but shows up as a 2 μEq L^{-1} imbalance between anions and cations, (Kekonen et al., submitted manuscript, 2004); therefore much of the non-sea-salt sulfate must be associated with other sources.

[25] Biogenic sulfate input must be related to the methanesulfonate profile as both species originate from a common source [*Delmas*, 1992]. There are strong correlations over decadal periods (Figure 4), between the species such as the obvious peaks and troughs in the twentieth century. Methanesulfonate concentrations are high during the Little Ice Age relative to both before and after it (Figure 4). Methanesulfonate appears to have a local Barents Sea sources [*O'Dwyer et al.*, 2000]. The decrease after the end of the Little Ice Age reflects the reduction in sea ice around Svalbard in between 1900 and 1920 [*Vinje*, 2001], which certainly caused changes in the local biogenic production. Presumably, lower concentrations prior the fifteenth century also reflect less sea ice around Svalbard. From the fifteenth to the mid eighteenth centuries both sulfate and methanesulfonate concentrations are rather stable, so a constant flux of biogenic sulfate seems likely. The methanesulfonate record is discussed in more detail by *Isaksson et al.* [2005].

4.4. Nitrate and Ammonium

[26] Nitrate represents 3% and ammonium 5% of total ions (in μeq L^{-1}). Nitrate and ammonium concentrations show more similar behavior than nitrate and sulfate even thought they all have anthropogenic sources (Table 1). *Kekonen et al.* [2002] show that there was an ammonium nitrate (NH_4NO_3) source until the mid-1980s, and *Teinilä et al.* [2003] reports covariation of nitrate and ammonium aerosols. Nitrate and ammonium concentrations have increased 65% and 26%, respectively, after 1950 compared with whole core average concentrations. Nitrate concentrations show the most dramatic increase a few decades later than sulfate, and nitrate also shows a large peak around 1980 when sulfate concentrations are relatively low (Figure 4).

[27] The troughs in nitrate concentrations around 1970 and the 1990 seem uncorrelated to published nitrate source changes, possibly suggesting loss of nitrate from the core. However, ammonium and $\delta^{18}O$ also show relatively low values around 1970 (Figure 4), and since ammonium is not lost easily at Lomonosovfonna [*Pohjola et al.*, 2002a], especially in relatively cool conditions, runoff appears an unlikely cause of the low nitrate concentration prior to 1990. Furthermore, most of the nitrate was bound on either sea-salt (sodium) or terrestrial (calcium) particles in aerosols [*Teinilä et al.*, 2003, 2004] that tend to stabilize against elution effects (see section 4.5). Nevertheless, the low nitrate concentrations are extremely low and almost certainly do indicate some runoff, but a fuller picture probably needs to consider changes in transport from distant sources of nitrate or reduced production from local sources.

4.5. Calcium

[28] Calcium account for 7% of total ions, and 80% of the calcium comes from non-sea-salt sources. In common with other ions shows a clear decrease in concentrations since 1950. This decrease corresponds to a general increase in accumulation rate both recorded in the ice core [*Pohjola et al.*, 2002b] and surrounding glaciers [*Pälli et al.*, 2002]. There is no indication of any direct influence of accumulation rate on calcium concentrations (Figure 3b). However, increased snow cover may have reduced wind erosion of local dust sources.

[29] Calcium has high concentrations and displays great variability from the mid eighteenth to the mid twentieth century, which includes the coldest part of the Little Ice Age according to the Lomonosovfonna $\delta^{18}O$ profile (Figure 4). As accumulation appears reasonably constant for this period [*Pohjola et al.*, 2002b], calcium variations probably reflect variation in windiness. This is expected during the coldest part of the Little Ice Age but also occurs in the post Little Ice Age period. This may be related to the large-scale atmospheric circulation shift associated with the change in the Icelandic low that occurred after 1920 or to local changes in winds caused by the retreat of the sea ice edge around western Svalbard [*Vinje*, 1999].

[30] Calcium and sulfate sometimes peak together independently of sodium, potassium and magnesium, for example, in the second half of the eighteenth century (Figure 4). Their high concentrations are due to calcium-containing dust particles that tend to be quite large [*Teinilä et al.*, 2003] and so must be mainly from local sources, and sensitive to wind speed and extent of snow free source area. However, the lack of any relationship with accumulation (Figure 3) suggests that wind speed may be more important. Calcium has larger concentrations in aerosols during summer than winter periods [*Teinilä et al.*, 2003]. This strongly supports the calcium source being mainly from local rock surfaces with seasonal snow cover. The geology in the area is consistent with this interpretation as it contains proterozoic-mesozoic sedimentary facies, including carbonate rocks. In later studies, *Teinilä et al.* [2004] also found evidence that calcium comes from both local and long-range origins in aerosol samples from Ny-Ålesund.

[31] Calcium has a clear association with nitrate [*Röthlisberger et al.*, 2000; *Kekonen et al.*, 2002] in ice core samples. *Röthlisberger et al.* [2000] suggested that calcium may be associated with nitrate via the stabilizing effect of dust particles. Aerosol samples in Ny-Ålesund show that nitrate is bound to terrestrial particles during the summertime [*Teinilä et al.*, 2003]. There is also a high correlation between nss-calcium and nitrate, and *Teinilä et al.* [2003] assumed that nitrate and calcium have a local source during the Arctic summer. This supports our earlier finding [*Kekonen et al.*, 2002], but now it seems that calcium and nitrate are bound together before deposition rather than as a result of postdepositional movement.

4.6. Deepest 10 m of the Ice Core

[32] The deepest 10 m of the record has a very different character than the rest of the core. All the ions except

ammonium have anomalously low concentrations (Table 1). Explanations for the lower concentrations include migration of ions from the deepest and warmest ice to the bed or loss of ions in the original snowpack caused by warm conditions prior to about 1300. Calculations of likely flow rates to the bed suggest that the former explanation is unlikely. Snow pit analyses over recent seasons suggest that loss of ions by runoff from the original snowpack is the most likely explanation.

[33] Since 1997 we have performed regular snow pit studies showing that there is presently summer loss of ions by runoff from the original snowpack. If we take $\delta^{18}O$ values to be directly related to surface temperature, then we may expect the ice cap before 1400 to have suffered periods when runoff of ions occurred. Detailed stratigraphic analysis of the core shows very large interlocking crystals that do, however, preserve undisturbed layers of bubbly and clear ice crossing crystal boundaries. It seems unlikely that the ice cap experienced negative mass balance at any stage of the record, but it may have been growing in size for the early part of the record, possibly explaining some of the decreasing trend in the $\delta^{18}O$ profile.

5. Conclusions

[34] The Lomonosovfonna ice core spans the last 800 years and includes several different climatic periods. Changes in sources of soluble ions are clearly visible in different sections of the core. The two largest changes are the rapid warming of the climate in early twentieth century (determined by the retreat of sea ice cover around western Svalbard between 1900 and 1920) and the sharp rise in anthropogenic sources from the mid twentieth century.

[35] The core is dominated by marine ions. Sodium and chloride account for 63% of the whole ice core water-soluble ion concentrations. In addition to sodium and chloride, sea-salt ratios show potassium and magnesium are also marine in origin. Potassium and magnesium do not show any changes in average concentrations until the twentieth century. Only sodium and chloride concentrations are higher between the fifteenth and mid sixteenth centuries, which correspond to the warmest part of the Lomonosov-fonna $\delta^{18}O$ profile, and most probably reflect a relatively northerly sea ice edge and therefore more sea-salt aerosol transported to the ice cap. All sea-salt and terrestrial ion concentrations (Na^+, Cl^-, K^+, Mg^{2+} and Ca^{2+}) start to decrease in the mid twentieth century. This could reflect melting and runoff even though sulfate, nitrate and ammonium concentrations are high after the mid twentieth century. High sulfate and low sea-salt ion concentrations may also reflect increasing influence of easterly air masses. However, if melting has caused the decrease of marine and terrestrial ion concentrations, our results suggest an additional, and most probably anthropogenic, chloride source in the core.

[36] Clear anthropogenic impacts are found for sulfate, nitrate and ammonium after the mid twentieth century. Sulfate data also suggest that there is a significant rise by the end of the nineteenth century. Such an early increase is not observed in other Arctic ice cores outside Greenland. Sulfate and methanesulfonate profiles correlate over decadal periods, clearly noticeable in the twentieth century. Between the fifteenth and mid eighteenth centuries, sulfate and methanesulfonate concentrations are remarkably stable, suggesting constant biogenic sulfate input. The nss-sulfate ratio suggests that Lomonosovfonna ice cap is dominated more by sulfate enriched air masses from the Barents Sea direction than Vestfonna ice cap but clearly less than the Ny-Ålesund area.

[37] Sea-salt ratios show calcium is mostly a terrestrial ion (and aerosol data suggest primarily from local sources). Calcium has higher and very variable concentrations from mid eighteenth to the mid twentieth century and may be explained by higher wind speeds. About 1/4 of the calcium is associated with 0.4 μEq L^{-1} of sulfate during the preindustrial period, and both ions display significant multi-decadal periodicity in concentration that likely indicates variation in local wind speeds.

[38] The bottom 10 m of the core show anomalously low concentrations compared with the rest of the core, and this probably indicates loss of ions either during the original transformation of the snowpack to solid ice or by loss of ions form the ice to the bed over a much longer period. This is the focus of ongoing work and will be related to the paleothermometery of the stable isotope record from the ice core.

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