An ice-core chemistry record from Snøfjellafonna, northwestern Spitsbergen

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ABSTRACT. A 2.2 m deep pit and the top 42.5 m of an ice core recovered at Snøfjellafonna, northwestern Spitsbergen, were continuously analyzed for Na^+, Cl^-, NO_3^-, SO_4^{2-} and pH. Seasonal variations in ionic concentrations seem to have remained in the pit and the core, in spite of the relatively severe summer melting. We dated the core by counting annual peaks of Na^+ and made an adjustment with the use of a tritium peak in 1963 as a reference horizon. It turned out that the depth of 42.5 m went back to the early 1930s or late 1920s. The 60–70 year record of snow chemistry showed that the concentrations of both NO_3^- and SO_4^{2-} had increased in the 1950s and had decreased in the late 1970s and the 1980s. The increase would be explained in terms of anthropogenic inputs from the industrial areas. The later decrease of the same ions may have been caused by a combination of the reduction of the atmospheric precursors due to pollution controls and the meltwater-associated processes.

INTRODUCTION

Knowledge of the climatic and environmental history of Svalbard is expected to be very helpful in understanding the changes of climate and environment in the Arctic. However, very few detailed ice-core studies have been carried out in Svalbard. Although workers from the U.S.S.R. excavated several ice cores from Spitsbergen (Gordiyenko and others, 1980; Zagorodnov, 1988) and from Nordauslandet (Vaikmäe and others, 1984; Yevseyev and Korzun, 1985; Arkhipov and others, 1986), very few detailed chemistry data have been published. Studies of chemistry have concentrated on ice cores from Greenland and Antarctica. Ice-core investigators have paid much less attention to glaciers in Spitsbergen, since most of these suffer rather severe melting even in their highest parts (e.g. Hagen and Liestøl, 1990). Recent studies (e.g. Thompson and others, 1984; Nakawo and others, 1990; Aristarain and Delmas, 1993), however, suggest that ice-core data from ice caps with relatively heavy melting could still provide us with important climatic and environmental information.

We drilled an 84 m deep ice core at one of the highest ice fields in northwestern Spitsbergen to reconstruct the past climate and environment. Preliminary results from borehole-temperature, density and electrical conductivity measurements as well as stratigraphic observations were reported in an earlier paper (Kameda and others, 1993). This paper reports the results from chemical analyses of the top 42.5 m of the core.

FIELD-WORK

An 84 m ice core was retrieved at site A, Snøfjellafonna (79°08'10" N, 13°17'30" E, 1190 m a.s.l.; see Fig.1), from 4 to 14 August 1992 with an electro-mechanical drill (Kameda and others, 1993; Takahashi and others, 1993; Watanabe and others, 1993). Site A, the saddle point of Snøfjellafonna, was selected as the coring site, since we had hit a water table at a depth of 22 m when we first drilled at site B (79°08'10" N, 13°19'00" E, 1160 m a.s.l.), which is located on a flat area 530 m downslope from site A (Kameda and others, 1993), prior to the
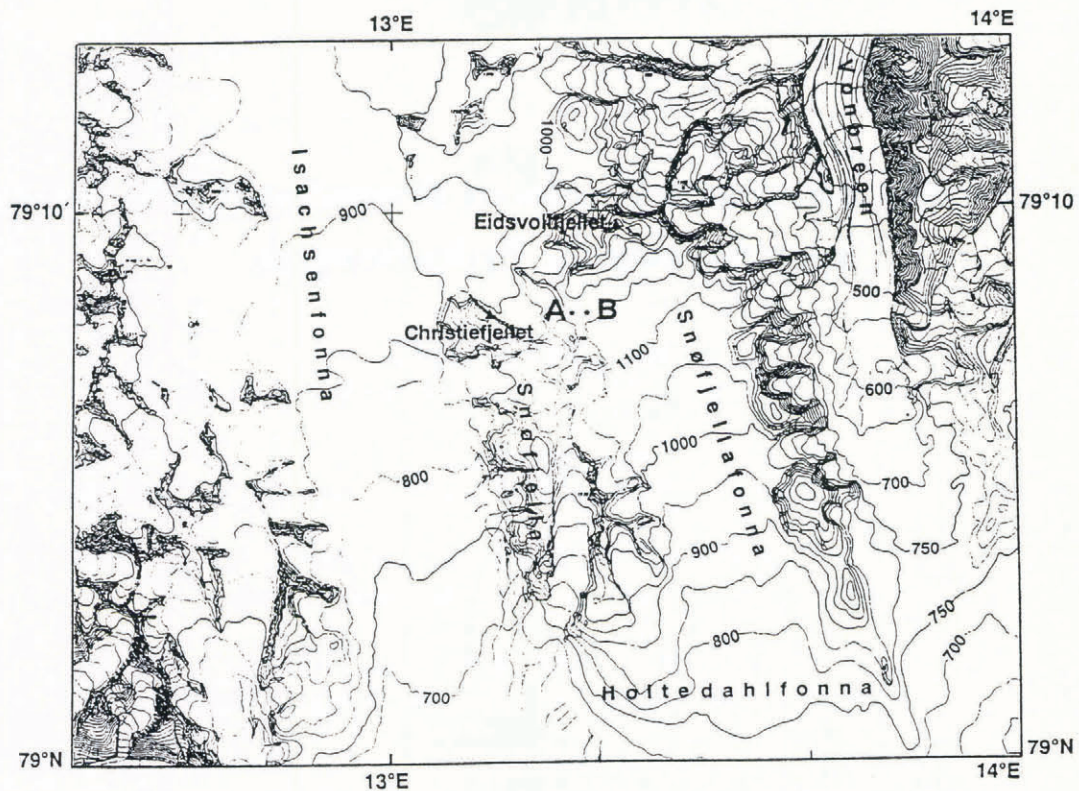
Fig. 1. Study sites (A and B) on Snøfjellafonna.

coring at site A. No water table existed down to 84 m depth at site A. Moreover, borehole temperatures there were below 0°C throughout the borehole, and the minimum borehole temperature was about 2°C lower than at site B (Kameda and others, 1993). These facts suggest that site A was less affected by meltwater percolation.

Meteorological observations were made at site B between 28 July and 15 August 1992. Daily maximum air temperatures were above 0°C on eight days, and surface snow melting was observed at both sites during the observation period. Results of the meteorological observations will be published elsewhere.

Together with ice-coring, a 2.2 m deep pit was dug at site A on 10 August 1992. After stratigraphic observations and density measurements were made, 41 samples were taken from a pit wall. The samples were carefully collected with a stainless-steel knife and put into pre-cleaned plastic bottles.

The ice-core and the pit-wall samples from site A were transported frozen to Ny-Ålesund, about 40 km southwest of site A, by helicopter. The top 42.5 m of the core was cut in a cold room in Ny-Ålesund using a pre-cleaned microtome and disposable plastic gloves. The average vertical length of the core samples was 15 cm. The core samples were put into clean plastic bags.

The pit and core samples were melted at room temperature in Ny-Ålesund. While the pit samples were kept in the original bottles, the core samples melted in plastic bags were decanted into pre-cleaned plastic bottles for transportation to Japan. All the samples were shipped to Japan in the liquid phase. Immediately after they arrived at Nagaoka Institute of Snow and Ice Studies in mid-December 1992, they were refrozen in a cold room

and kept frozen until the chemical analyses were made.

CHEMICAL ANALYSES OF SAMPLES

The pit and core samples from site A were analyzed at Nagaoka Institute of Snow and Ice Studies: for NO_3^-, SO_4^{2-} and Cl^- with a Dionex Dx-100 ion chromatograph; for Na^+ with a Seiko SAS 7500 atomic absorption spectrophotometer equipped with a flame atomizer; and for pH with a TOA HM-40V pH meter.

The pit and core samples from the depth intervals 1.5–7.5 m and 35.3–41.1 m were analyzed for $\delta^{18}O$ with a Finigan MAT δ E mass spectrometer at the National Institute of Polar Research, Tokyo.

RESULTS AND DISCUSSION

Seasonal trends in snow chemistry of the pit

The stratigraphy, $\delta^{18}O$ and concentrations of ions for the pit are illustrated in Figure 2. The stratigraphy of the pit, with mainly granular snow layers and ice layers sandwiched in between them, implied that meltwater had percolated through the pit. The seasonality of $\delta^{18}O$, however, seems to have been preserved. We dated the pit according to the $\delta^{18}O$ profile. Besides the summer layer of 1992 just below the surface of the pit, another summer layer was identified just above the bottom of the pit. We did not count a small shoulder at about 1.2 m depth as an individual summer layer, since depth-hoar layers, which are generally formed in fall in this region (Goto-Azuma and others, 1993a), were observed at about this depth.
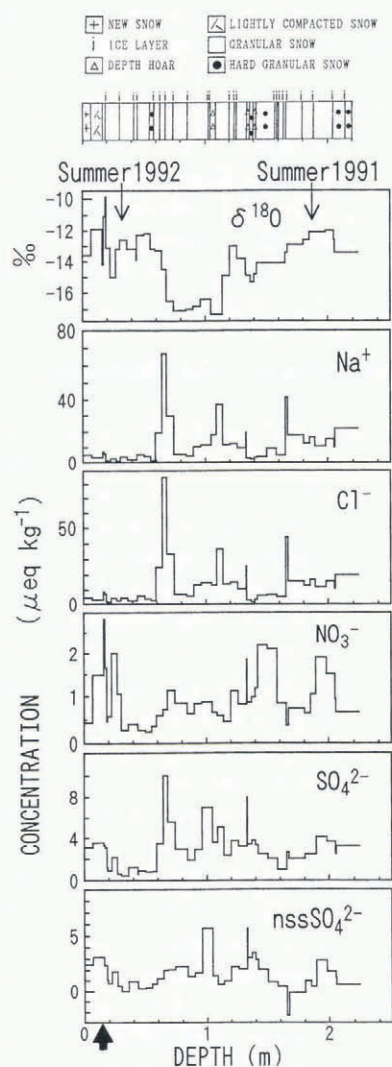
Fig. 2. Stratigraphy, $\delta^{18}O$ and ion concentrations of the pit. The horizontal axis shows real depth. Depth 0 m represents the surface of 10 August 1992. The heavy solid arrow shows the surface snow level of 4 August 1992 when ice coring was started.

The concentration of snow algae in the same pit also showed two peaks at depths of 0.36 and 1.9 m (personal communication from S. Kohshima and others, 1994). This confirms our dating, since the number of snow algae is considered to be high in summer layers.

Regression analysis gave an Na^+/Cl^- ratio of 0.87, which is close to the marine ratio 0.86. Accordingly, Na^+ and Cl^- were mainly of sea-salt origin. Na^+ and Cl^- showed high values between depths of 0.6 and 1.3 m (Fig. 2). This depth interval corresponds to the winter half of the year. This agrees with the fact that Na^+ and Cl^- generally peak during fall to early spring in Greenland (e.g. Finkel and others, 1986; Davidson and others, 1987) and in the Canadian Arctic.

NO_3^- showed high concentrations in the early summer to mid-summer of 1991, late summer of 1991, winter of 1991/92 and mid-summer of 1992. This may suggest that NO_3^- in recent snow on Snøfjellafonna shows anthropogenic winter to early-spring peaks in addition to natural summer peaks, as was suggested for central Greenland (Whitlow and others, 1992).

SO_4^{2-} has both sea-salt and non-sea-salt components. We calculated so-called nss (non-sea-salt) SO_4^{2-} (in $\mu eq\,kg^{-1}$) using the equation $[nss\,SO_4^{2-}] = [SO_4^{2-}] - 0.12 \times [Na^+]$ to estimate the non-sea-salt component. The results of this calculation indicated that the major part of SO_4^{2-} had originated from nss SO_4^{2-}. Concentrations of nss SO_4^{2-} in the pit were high in the early summer to mid-summer of 1991, fall of 1991, winter of 1991/92, and late summer of 1992. This may suggest that nss SO_4^{2-}, as well as NO_3^-, in recent Snøfjellafonna snow shows two peaks each year. We have to take into account, however, that the seasonality of nss SO_4^{2-} could have been disturbed by the eruption of Mt Pinatubo in June 1991. More evidence is needed to clarify the seasonalities of NO_3^- and SO_4^{2-}, since NO_3^- and SO_4^{2-} concentration profiles are more easily altered by melt-water percolation than Na^+ and Cl^- (e.g. Davies and others, 1982; Goto-Azuma and others, 1993b).

Dating of the ice core

Kameda and others (1994) determined the 1963 summer layer of the ice core by a tritium concentration peak at 20.49–20.635 m depth. They assigned this peak to the fall-out from the 1961–62 atmospheric nuclear bomb tests. In the same core, Pinglot and others (1994) found the maximum concentration of ^{137}Cs at about the same depth as the tritium peak, and they determined this as the 1962–63 layer. Since redistribution of ^{137}Cs by meltwater percolation was minimal (Pinglot and others, 1994), we used this layer of ^{137}Cs and tritium peaks as a reference horizon in dating the core.

Ion concentrations and pH of the core are presented in Figure 3. Based on the stratigraphy and the depth–density curve, Kameda and others (1993) considered that meltwater had played an important role in the densification processes. Under such circumstances, seasonal variations of some ions and $\delta^{18}O$ tend to be destroyed. But we considered that seasonal variations of Na^+ and Cl^- concentrations as observed in the pit (Fig. 2) still remained in the core, for the following reasons. There were 29 ± 4 major peaks in the Na^+ profile between 1.5 and 20.5 m depth; this agrees with the number of years determined by the 1963 layer. Also, the Na^+ profile for the depth interval of 1.5–7.5 m seems to show anti-phase variations with respect to $\delta^{18}O$ profile (Fig. 4), as seen in the pit. Although amplitudes of $\delta^{18}O$ variations in Figure 4 were not large enough to identify annual layers, this was partly due to low resolution of the sampling interval of 3–5 samples year^{-1}. We therefore assigned Na^+ peaks to winters and $\delta^{18}O$ peaks to summers. The concentration profile of Cl^- is similar to that of Na^+, while seasonality is a little less clear compared to Na^+. A relatively high annual accumulation rate of 0.48 m w.e. year^{-1} (Kameda and others, 1994; Pinglot and others, 1994) was a favorable factor in preserving the seasonal variations.

We dated the core on the basis of the seasonal variation in Na^+ concentration profile, with the use of the 1963 layer as a reference horizon. The results of our dating are given in Figure 3. The depth of 42.5 m corresponded to the early 1930s. The dating error is ± 2–3 years down to a depth of 20.5 m, and may be greater below this depth, where there were no reference horizons.
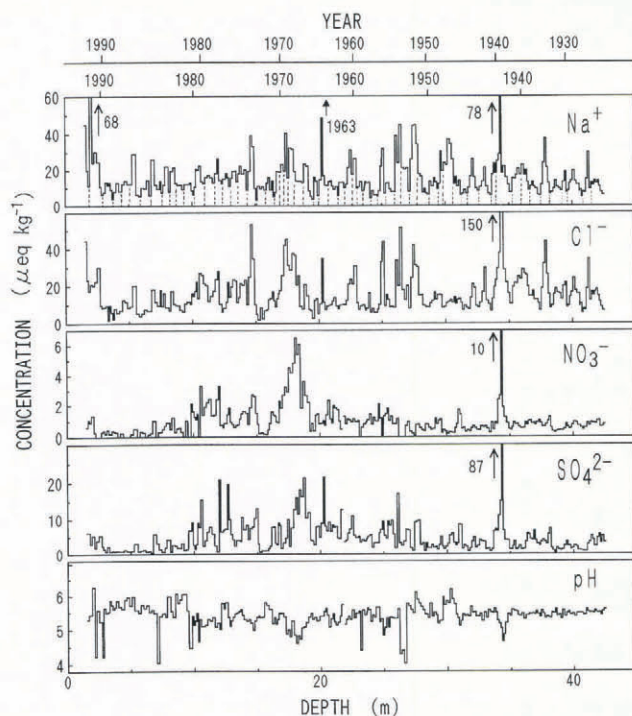
Fig. 3. Ion concentrations and pH of the core. The horizontal axis represents real depth. Depth 0 m shows the surface of 4 August 1992. The upper year axis shows results of dating based on a constant net accumulation rate. The lower year axis shows results of dating based on seasonal variation of Na⁺, adjusted by the 1963 layer as a reference horizon. The solid arrow shows the 1963 summer determined by the tritium concentration profile. Dotted lines show the annual layer boundaries (i.e. summer layers) estimated by seasonal variation of Na⁺.

According to this dating, mean net annual accumulation rates (w.e.) for the 1980s, 1970s, 1960s, 1950s, 1940s and
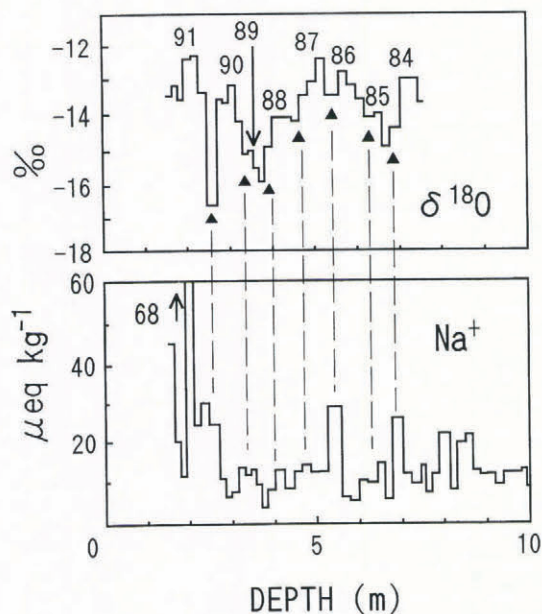

Fig. 4. $\delta^{18}O$ and Na⁺ concentrations for the top 10 m of the core. The horizontal axis represents real depth. Years are assigned to summer layers. Triangles denote winter layers.

1930s are 0.44, 0.52, 0.49, 0.50, 0.63 and 0.72 m, respectively. Relatively large variation could be partly due to the dating error. If we use a constant net accumulation rate of 0.48 m w.e. (i.e. the average for the period 1992–63), the depth 42.5 m corresponds to the year 1925. Results of dating by this method are also shown in Figure 3. We plan to analyze the deeper part of the core (42.5–84 m) to find more reference horizons such as volcanic signals so that we can date the core more accurately for the depth below 20.5 m.

Temporal trends in ice-core chemistry

As seen in Figure 3, NO_3^- and SO_4^{2-} concentration levels started to increase about 1950. This was accompanied by pH decrease. Both NO_3^- and SO_4^{2-} had increased by a factor of 3–4 by the late 1960s. In the late 1970s, however, NO_3^- and SO_4^{2-} started to decrease and pH started to increase. In the 1980s NO_3^- and SO_4^{2-} concentrations dropped to values lower than those before 1950. In contrast to NO_3^- and SO_4^{2-}, Na⁺ and Cl⁻ do not show drastic changes in concentration levels throughout the last 60–70 years except that the concentration of Cl⁻ seem to have been somewhat lower in the 1980s compared to the rest of the period. Amplitudes of seasonal variations of Na⁺ and Cl⁻ were greater before 1950 than for the rest of the period.

The rise in NO_3^- and SO_4^{2-} concentrations about 1950 probably reflects the recent increase of these ions by anthropogenic inputs, as was found in Greenland (Neftel and others, 1985; Finkel and others, 1986; Mayewski and others, 1986) and in the Canadian high Arctic (Koerner and Fisher, 1982; Barrie and others, 1985). Whereas NO_3^- clearly began to increase later than SO_4^{2-} at Dye 3, Greenland (Finkel and others, 1986; Mayewski and others, 1986; Clausen and Langway, 1989), NO_3^- and SO_4^{2-} began to increase at the same time at Snøfjella-fonna.

The decrease of NO_3^- and SO_4^{2-} in the late 1970s to 1980s could be due to strict pollution controls on NO_x and SO_2 set in the late 1970s (French, 1990). In fact, NO_3^- and SO_4^{2-} concentrations in snow in the Swiss Alps (Wagenbach and others, 1988) seem to have dropped since about 1980. However, in the Swiss Alps both NO_3^- and SO_4^{2-} levels in the 1980s were higher than those before 1950. The 1980s NO_3^- and SO_4^{2-} levels at Snøfjellafonna, which were lower than those before 1950, are probably too low to be fully explained by the decrease in source inputs. The remarkable decrease of NO_3^- and SO_4^{2-} at Snøfjellafonna could be attributed to increased leaching of ions by increased summer melting. We plotted Na⁺/Cl⁻ ratios for the core in Figure 5 to evaluate the ions washed out from site A. Since Cl⁻ is preferentially eluted from melting snow compared to Na⁺ (Davies and others, 1982), the Na⁺/Cl⁻ ratio can be used as an indicator of the extent of leaching of ions (Aristarain and Delmas, 1993). During the period from the 1950s to the 1980s, Na⁺/Cl⁻ values were generally much higher than the marine reference value 0.86 and exhibited more marked variations compared to earlier years. In the 1980s, departure of the Na⁺/Cl⁻ ratio was most remarkable and Cl⁻ levels were somewhat lower than in the other periods. Before the 1950s, Na⁺/Cl⁻ ratios were
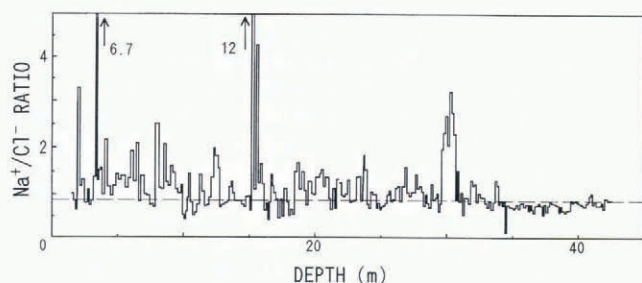
Fig. 5. Ratios of Na^+/Cl^- in the core. The horizontal axis represents real depth. The horizontal broken line shows the marine ratio 0.86. Above the line, departure of the ratio from the marine ratio indicates increased removal of Cl^- due to increased melt.

close to the marine value and were relatively constant. These facts suggest that some of the Cl^- ions were washed out after about 1950, with more wash-out having taken place in the 1980s. Hence, NO_3^- and SO_4^{2-}, which are much more easily eluted than Na^+ and Cl^- (Davies and others, 1982; Goto-Azuma and others, 1993b), would also have been removed from the snow deposited after about 1950. Leaching of NO_3^- and SO_4^{2-} might have been most pronounced during the 1980s. Increased leaching, as well as the decreased source strengths due to pollution controls, could have reduced the NO_3^- and SO_4^{2-} concentration levels markedly in the late 1970s and 1980s. On the other hand, high levels of NO_3^- and SO_4^{2-} concentrations during the 1950s to early 1970s suggest that the increase in source strengths has compensated the increase in wash-out. We excluded the possibility that both increase and decrease might have been produced simply by the movement of ions from the upper to the lower layers through meltwater percolation. Since temperatures were below 0°C throughout the borehole, meltwater created near the surface could not have percolated through as long a vertical distance as 10–20 m. We believe that meltwater percolated horizontally and away from site A when heavy melting occurred, because site A is located at the saddle point. Evidence of this possibility is the presence of the water table at site B, as mentioned in an earlier section.

No clear volcanic SO_4^{2-} signals were found in this work, probably because of increased background SO_4^{2-} concentration after 1950. It is more difficult to detect non-major volcanic SO_4^{2-} signals after 1950. Although SO_4^{2-} concentrations were extremely high around the 34 m depth, concentrations of all the other ions were high as well. This is a typical feature of ion concentration due to heavy melting and refreezing processes (Goto-Azuma and others, 1993a). High ion concentrations in multiple samples cut from about 34 m depth deny the possibility of contamination, because it is unlikely that multiple samples only from this depth interval would be contaminated through the same cutting procedures as for all the other samples. Broad peaks of Cl^-, NO_3^- and SO_4^{2-} concentrations at 17–18 m depth may also have been created by melt–freeze processes. Very low concentrations of these ions at 15–16 m depth suggest that these ions would have migrated from the upper 15–16 m layer to the lower 17–18 m layer.

CONCLUSIONS

Seasonal trends in snow chemistry still remained in the upper 42.5 m of the ice core retrieved at Snøfjellafonna, in spite of the relatively heavy summer melting. This, along with the reference layer of atmospheric nuclear bomb tests in 1961–62 enabled us to date the core. The 42.5 m turned out to cover the last 60–70 years. Temporal trends in snow chemistry, however, seem to have been somewhat altered by meltwater percolation. Post-depositional changes associated with meltwater seem to have been less pronounced before the 1950s.

Concentration levels of NO_3^- and SO_4^{2-} increased from the 1950s onwards and decreased in the late 1970s to 1980s, while those of Na^+ and Cl^- showed little change. The increase of NO_3^- and SO_4^{2-} may be due to the anthropogenic inputs. The decrease of the same ions could be due to both pollution controls and more leaching by heavier melting. The causes, however, could not be clarified in this study. A continuous $\delta^{18}O$ analysis and detailed chemical analyses of the rest of the core will be necessary to interpret the temporal trends found in this work.

ACKNOWLEDGEMENTS

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