

Regional characteristics of chemical constituents in surface snow, Arctic cryosphere

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Abstract: Surface snow observations were carried out on many glaciers in the Arctic. The observational sites were in Svalbard, Severnaya Zemlya, Greenland and arctic Canada. The seasonal variations of chemical constituents in snowfall remained in the surface snow on the central Greenland ice sheet where the effect of snowmelt was weak. Conversely, the chemical constituents in the snow layer changed with infiltration of melt water on the Svalbard glacier where strong snowmelt occurred, even in high latitudes. But in general, specific regional characteristics remained in all the glaciers. The concentrations of ions in snow were significantly decreased, because of percolation of melt water. But the ratio of NO₃ to Cl still had unique regional characteristic values.

1. Introduction

For the purpose of clarification of the climatic and environmental changes for the past 100 years in the Arctic, the Japanese Arctic Glacier Expedition conducted a large number of ice coring exercises at a number of glaciers from 1987 to the present (Watanabe and Fujii, 1988, 1990; Watanabe *et al.*, 1993, 2000; Watanabe, 1996; Kamiyama *et al.*, 2001; Motoyama *et al.*, 2001). During the same period, surface snow pit observations were carried out near ice coring sites to reveal the regional characteristics of environmental conditions. The geographical characteristics, such as the latitude, altitude and distance from the coast varied among the sites. The surfaces of the glaciers in Svalbard and Severnaya Zemlya melt in summer, chemical substances are washed out with percolated melt water, and the water is frozen again. Therefore, the profiles of chemical constituents change in quality (Davis *et al.*, 1982; Goto-Azuma *et al.*, 1993, 1995; Rabin and Theakstone, 1994; Motoyama *et al.*, 2000; Iizuka *et al.*, 2000). Jonsson and Hansson (1990) identified of annual layers in superimposed ice.

The seasonal variations of chemical constituents in snowfall were preserved in the surface snow on the central Greenland ice sheet where the effect of snowmelt is weak. As a result, the seasonal variation of the stable isotope ratio, strength of ECM, and ion concentrations could be used for dating of the ice core (Hammer, 1989). The en-

vironmental regime and climatic change have also been studied using various seasonal signals in the snow layer (Shuman *et al.*, 1998).

Thus, when we extract the past environmental signal from an ice core and discuss the environmental regime and climatic change, it is necessary to make clear the process of transition from snow to ice on the glacier.

Nitrate decreases between the surface and several meters depth in the low precipitation area in inland Antarctica where there is no influence of snow melt (Nakamura *et al.*, 2000; Wagon *et al.*, 1999), and there are also a few reports in the Arctic area (De Angelis and Legrand, 1995).

In this study, we attempt to make clear the regional chemical characteristics of snowfall and its change in quality after deposition.

2. Observations

We studied glaciers in Svalbard, Greenland, Severnaya Zemlya and arctic Canada. (Fig. 1 and Table 1). The basic methods of snow surface observation were as follows: An area free of contamination, with snow still in its deposited condition, was selected for the observation site. The snow pit was dug vertically. The chemistry samples were collected from the pit wall on the side of the windward to prevent contamination. Immediately after the pit wall was established, its vertical temperature profile was measured. The stratigraphy of the snow pit wall (*i.e.* kind of snow, boundary of snow

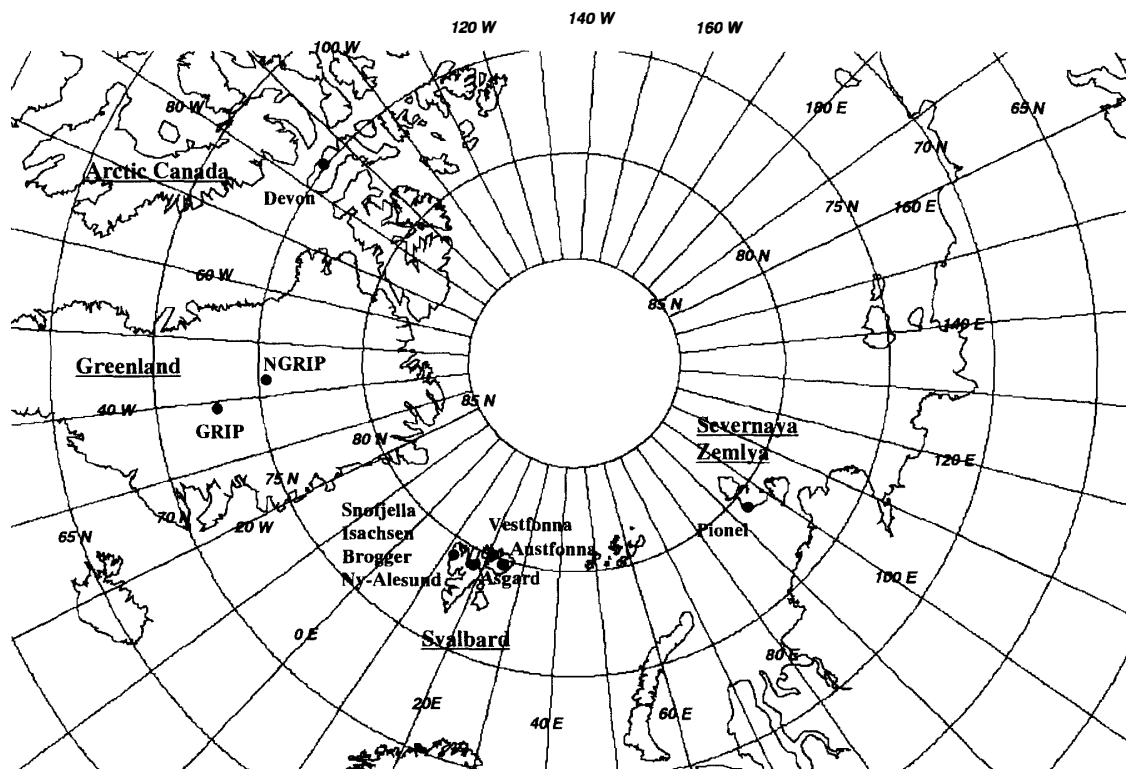


Fig. 1. Map of surface snow observation sites in Arctic.

Table 1. Surface snow observation information in the Arctic, including observation periods, site names, positions, altitudes, maximum pit depths and data sources.

Year	Date	Region	Site	Group	Position	Altitude	Max depth (cm)	Snow condition	References
1991	Aug. 6	Svalbard	Brogger	Bro	78.9 N, 11.8 E	440 m	61	wet	Goto-Azuma <i>et al.</i> 1993
1991	Aug. 12	Svalbard	Isachsen	Isa&Sno	79.1 N, 12.9 E	850 m	210	wet	Goto-Azuma <i>et al.</i> 1993
1992	Feb. 20	Svalbard	Brogger	Bro	78.9 N, 11.9 E	470 m	115	dry	Goto-Azuma <i>et al.</i> 1993
1992	Feb. 20	Svalbard	Brogger	Bro	78.9 N, 11.9 E	235 m	115	dry/wet	Goto-Azuma <i>et al.</i> 1993
1992	Aug.10-13	Svalbard	Snofjella	Isa&Sno	79.1 N, 13.3 E	1190 m	165	wet	Goto-Azuma <i>et al.</i> 1993
1992	Aug.14-15	Svalbard	Snofjella	Isa&Sno	79.1 N, 13.3 E	1160 m	249	wet	Goto-Azuma <i>et al.</i> 1993
1993	Jul.11-13	Svalbard	Asgard	Asg	79.5 N, 16.7 E	1140 m	157	wet	unpublish
1994	Sep. 10	Svalbard	Brogger	Bro	78.9 N, 11.9 E	445 m	(950) ^{*4}	wet	Motoyama <i>et al.</i> 2000
1995	Mar.8	Svalbard	Brogger	Bro	78.9 N, 11.9 E	445 m	163	dry/wet	Motoyama <i>et al.</i> 2000
1995	Jun.3	Svalbard	Vestfonna	Ves	80.0 N, 21.0 E	600 m	231	dry/wet	unpublish
1998	Mar.25	Svalbard	Austfonna	Aus	79.8 N, 24.0 E (Aus98A)	750 m	103	dry/wet	Iizuka <i>et al.</i> 2000
1998	Mar.26	Svalbard	Austfonna	Aus	SW800m from Aus98A	750 m	121	dry/wet	Iizuka <i>et al.</i> 2000
1998	Apr.10	Svalbard	Austfonna	Aus	NW250m from Aus98A	750 m	180	dry/wet	Iizuka <i>et al.</i> 2000
1999	Apr.28-29	Svalbard	Austfonna	Aus	79.8 N, 24.0 E (Aus99A) ^{*1}	750 m	187	dry/wet	unpublish
1999	Mar.17	Svalbard	Austfonna	Aus	NW2200m from Aus99A	750 m	146	dry/wet	unpublish
1999	May.22	Svalbard	Austfonna	Aus	S100m from Aus99A	750 m	175	dry/wet	unpublish
1996	Jul.13-14	Severnaya Zemlya	Pionel	Pio	79.9 N, 93.0 E	382 m	197	dry/wet	unpublish
1998	May 9-11	Arctic Canada	Devon	Dev	75.4 N, 82.5 W	2239 m	246	dry	unpublish
1991	July	Greenland	GRIP	Gri&Ngr	74.6 N, 37.6 W	3238 m	300	dry	unpublish
1997	Jun.8	Greenland	NGRIP	Gri&Ngr	75.1 N, 42.4 W ^{*2}	2959 m	165	dry	unpublish
1999	Jul.13	Greenland	NGRIP	Gri&Ngr	75.1 N, 42.4 W ^{*3}	2959 m	200	dry	unpublish

^{*1} 3km north of Aus98A, ^{*2} 3km west of main camp, ^{*3} 4km northwest of main camp, ^{*4} ice core drilling

layer, diameter of snow particles, hardness of snow) was measured. The sampling interval for chemical analysis and density measurement was within 10 cm. The sampled snow was melted *in situ* and was kept in pre-cleaned 100 cc bottles. The pH and electrical conductivity (EC) of the rest of the melt water were measured *in situ* by a pH meter (Horiba D13) and conductivity meter (Horiba ES12), respectively. These samples were transported to Japan in a cooled condition. They were stored frozen until we measured them. The chemical analysis of H_2O_2 (by fluorescence spectrometer, Kimoto Electric Co. Ltd. (see Kamiyama and Nakayama, 1992), major soluble ions Cl^- , NO_3^- , SO_4^{2-} , Na^+ , K^+ , Mg^{2+} , Ca^{2+} (by ion chromatography, Dionex 2000i/sp, see Igarashi *et al.*, 1998) and the stable oxygen isotope ratio ($\delta^{18}\text{O}$) (by mass spectrometer, Finnigan mat delta-E, measurement precision $\pm 6\%$) was performed at the laboratory of the National Institute of Polar Research.

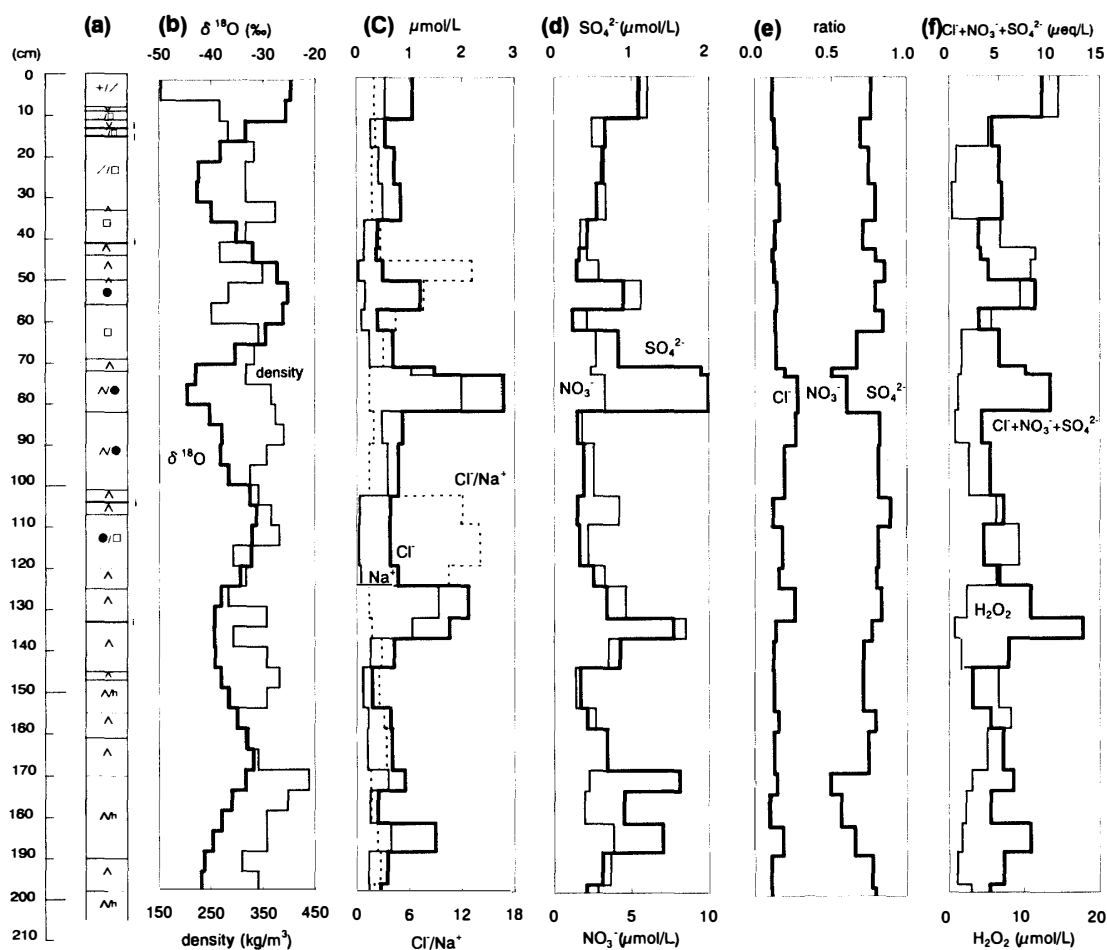


Fig. 2. Result of surface snow observations at NGRIP, Greenland in 1999. (a) Snow stratigraphy. Classification of snow was based on Colbeck *et al.* (1990). (b) Oxygen isotope ratio ($\delta^{18}\text{O}$, bold line) and snow density (thin line). (c) Concentrations of Cl^- (bold line), Na^+ (thin line) and ratio of Cl^- and Na^+ concentrations (broken line). (d) Concentrations of SO_4^{2-} (bold line) and NO_3^- (thin line). (e) ratio of Cl^- (left), NO_3^- (center) and SO_4^{2-} (right) to their total. (f) Concentrations of $\text{Cl}^- + \text{NO}_3^- + \text{SO}_4^{2-}$ (bold line) and H_2O_2 (thin line).

3. Results and discussion

3.1. Examples of surface snow observations

The stratigraphy and profile of major soluble constituents at three different sites are shown in Figs. 2–4.

a) NGRIP 1999 in Greenland

The result of the pit observation at NGRIP in 1999 is shown in Fig. 2. The depth hoar was developed, well under 40 cm thick, by dry metamorphism. The vertical profile of $\delta^{18}\text{O}$ showed seasonal variation clearly. Usually the snow layer with a maximum peak of $\delta^{18}\text{O}$ corresponds to summer snow, and the snow layer with a minimum peak of $\delta^{18}\text{O}$ corresponds to winter snow (Hammer, 1989). This means that the snow cover from surface to 200 cm depth consisted of 3.5 years' snowfall. Winter snow density was larger than summer snow density as Narita (2000) mentioned. Concentrations of Cl^- and SO_4^{2-} increased in winter and spring (or summer). NO_3^- increased in spring and summer. The ratio of Cl^- to Na^+ was larger than that of sea water, which is 1.18 (Clausen and Langway, 1989). It became particularly high in summer because Cl^- was supplied from somewhere except the sea, as for example HCl.

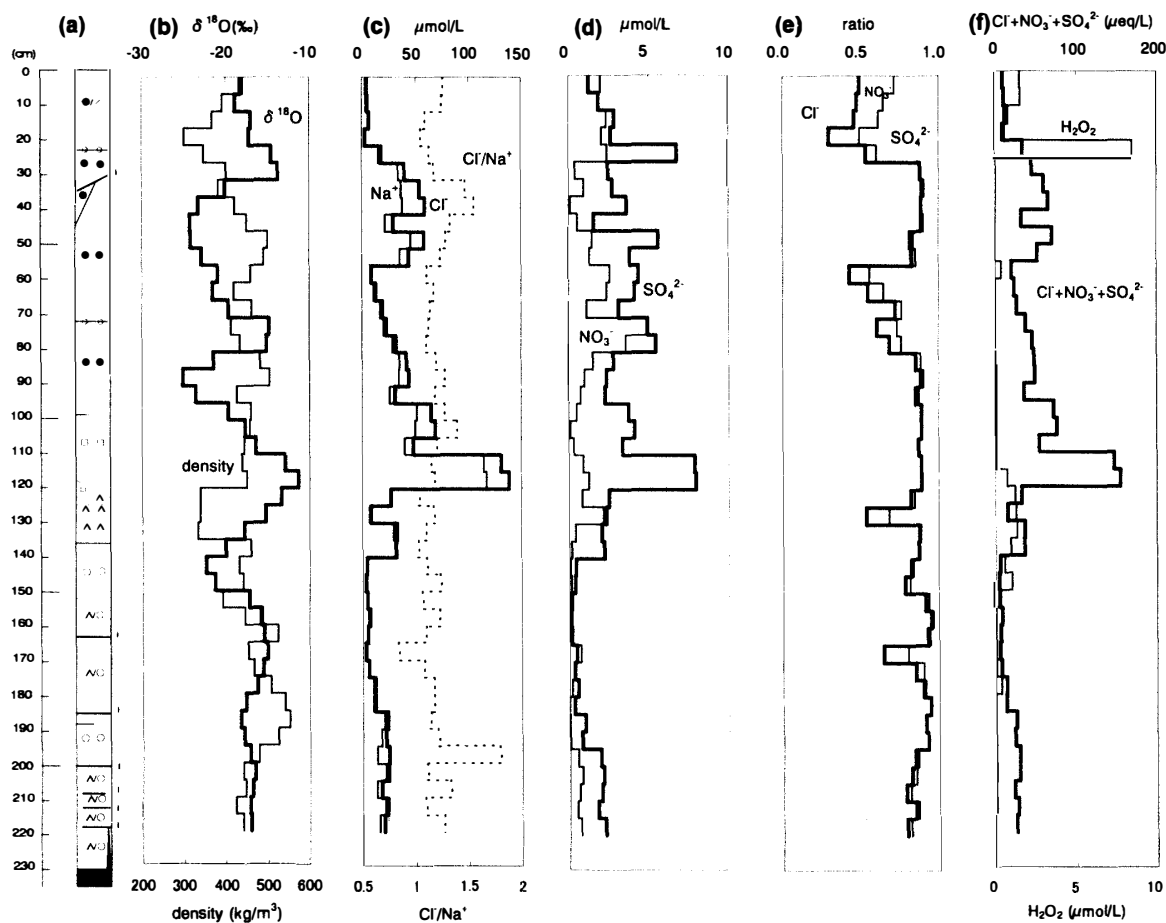


Fig. 3. Result of surface snow observations at Vestfonna, Svalbard in 1995.

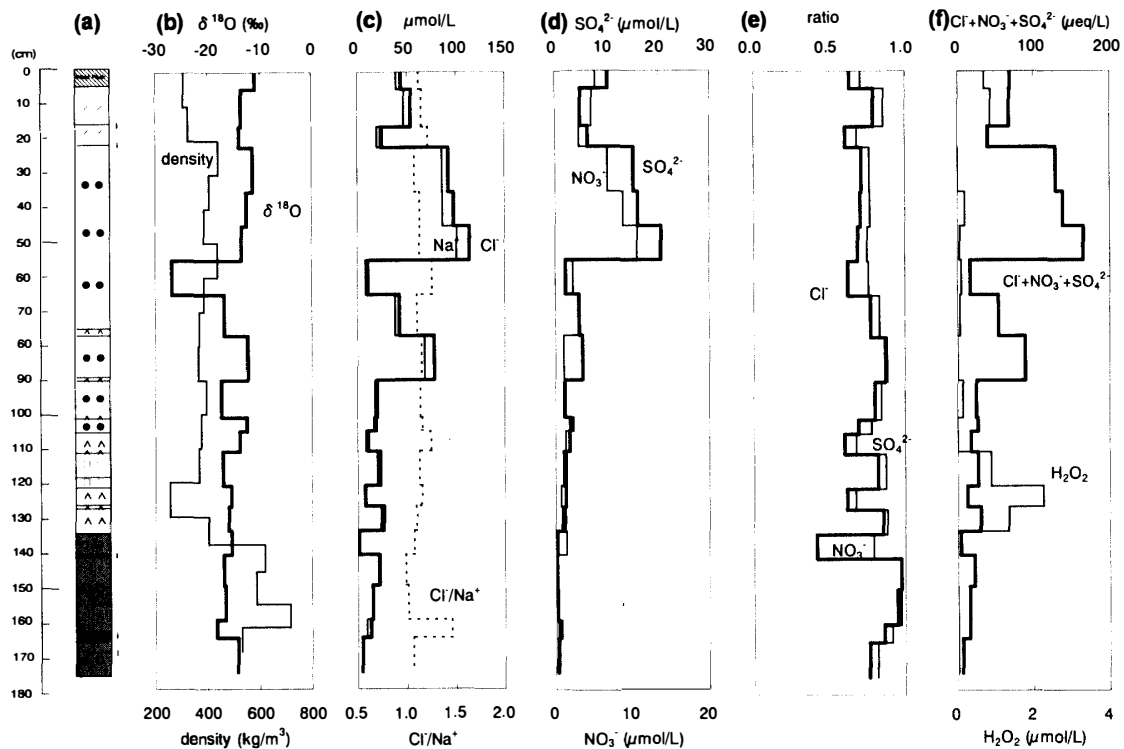


Fig. 4. Result of surface snow observations at Austfonna, Svalbard in 1999.

Considering the anion components, the ratio of Cl^- was almost stable. The ratio of SO_4^{2-} seemed to increase; NO_3^- decreased during winter and spring.

Concentrations of total anions ($\text{Cl}^- + \text{NO}_3^- + \text{SO}_4^{2-}$) increased in winter and spring. Concentration of H_2O_2 increased in summer and fall. Beer *et al.* (1991) also obtained a similar result with a phase shift between the H_2O_2 and $\delta^{18}\text{O}$ summer peaks in pits from Dye 3 in Greenland. Neftel (1996) described the reason why both species had a different transfer function from air to snow and/or from snow to firn. The result in NGRIP seemed to be similar to that of Dye 3. Peaks of H_2O_2 decreased with depth. This result was same as the pit profile from Dye 3 and shallow core profile from central Greenland (Beer *et al.*, 1991 and Sigg, 1990, respectively). De Angelis and Legrand (1995) reported that HCl and HNO_3 content of snow sample within a few days or a few weeks after the precipitation in central Greenland is significantly higher than concentrations measured in the aged firn layer. This result is not clear in our study (Fig. 2d).

b) Vestfonna 1995 in Svalbard

The result of the pit observation at Vestfonna in 1995 is shown in Fig. 3. Wet metamorphism was found below 135 cm depth. A large ice layer formed under 230 cm. All ions began to decrease from 135 cm depth. So that, this boundary layer at 135 cm was the summer/fall transition. The variation of $\delta^{18}\text{O}$ up to 135 cm had three peaks. But it may have been formed by fall and winter snows, which had different $\delta^{18}\text{O}$ values. Igarashi *et al.* (2001) reported that the seasonal variation of $\delta^{18}\text{O}$ in precipitation from Ny-Ålesund, northwestern Svalbard was indistinct. Snows which were produced water vapor of different origins fell. So that in appearance, $\delta^{18}\text{O}$ varied like seasonal change. Under 135 cm, the range of $\delta^{18}\text{O}$ variation was small. Na^+ and Cl^- showed the same

change. The ratios of Cl^- to Na^+ mostly fit well with that of sea water. Total anion ($\text{Cl}^- + \text{NO}_3^- + \text{SO}_4^{2-}$) also began to decrease from 135 cm (Fig. 3f). Considering the anion components, the ratios of NO_3^- and SO_4^{2-} decreased quickly (Fig. 3e). H_2O_2 was detected fragmentarily. It seems that concentration of H_2O_2 was changed by percolation of snowmelt water after accumulation. Snow with high concentration of ions from 110 to 120 cm was formed by warm snowfall which had rich concentrations of Na^+ , Cl^- and SO_4^{2-} ions.

c) Austfonna 1999 in Svalbard

The result of the pit observation at Austfonna in 1999 is shown in Fig. 4. The percolated snowmelt water was stored up in the layer which is deeper than 135 cm during the previous year, and it had refrozen. As a result, the snow density increased rapidly. The variation of $\delta^{18}\text{O}$ was small except around 60 cm in depth. A seasonal change seems not to exist in $\delta^{18}\text{O}$ in precipitation, as reported by Igarashi *et al.* (2001). The ratios of Cl^- to Na^+ mostly fit well with that of sea water. The concentrations of ions were decreased as it became deeper than the 55 cm (or 90 cm) depth. Consequently, the ions included in the layer between 55 cm and bottom of the pit dissolved with percolated snowmelt water. The ratios of NO_3^- and SO_4^{2-} to the total anion had some peaks. It looked as though it had been formed by depth hoar development. H_2O_2 was detected fragmentarily as at Vestfonna.

3.2. Regional characteristics of surface snow

In the following examination, the pit snow was classified as either non-melted snow (D) or snow with a melting history (W) based on the snow stratigraphy and the

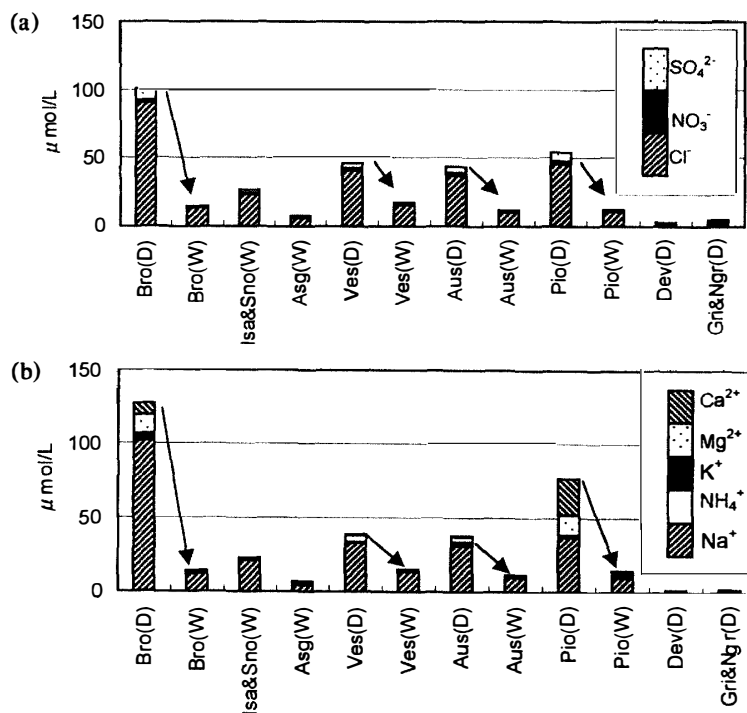


Fig. 5. Ion concentrations averaging for each surface layer and region. (a) major anion, (b) major cation. (D) means non-melted snow. (W) means the snow with a history of melting.

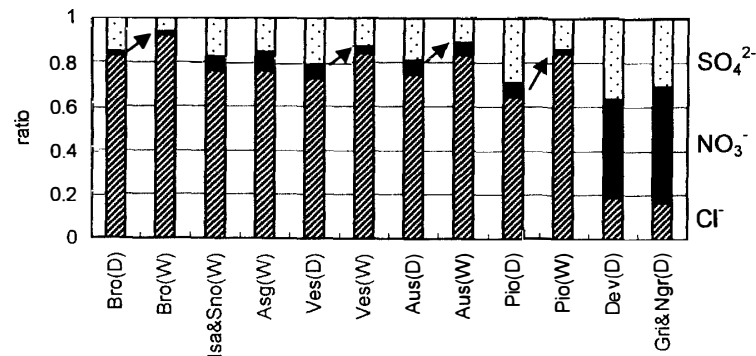


Fig. 6. Ratio of Cl^- , NO_3^- and SO_4^{2-} to their total averaging for each surface layer and region. (D) means non-melted snow. (W) means the snow with a history of melting.

distribution of ion concentration. The mean values were calculated in each region.

From the pit observations, these snow layers consisted of 1 to 4 years of buildup. Concentrations of ions averaged in the snow layer of the study glaciers are shown in Fig. 5. These snow layers contained snowfall from a 1 to 4 year period. The data could be divided into two groups: group (I) consisted of glaciers in Svalbard and Severnaya Zemlya where the concentrations of ions were large. The concentration of ions in non-melted snow (D) in these areas was much higher than snow with a melting history (W). It is suggested that the ions were flushed out from the snow pack during the thaw. The second group, group (II), consisted of glaciers in Greenland and arctic Canada where ion concentrations were small in non-melted snow. The ratios of anion composition are shown in Fig. 6. For group (I), the ratio of Cl^- ions was up to 60% and SO_4^{2-} was less than 30% in non-melted snow. On the other hand, the former was up to 70% and the latter was less than 20% in snow with a history of melting. Such a change of ratio like this seemed to indicate that the SO_4^{2-} dissolved in water more quickly than Cl^- , as some previous studies have reported (Davies *et al.*, 1982, 1987; Brimblecombe *et al.*, 1985). For group (II), the proportion of Cl^- was less than 20%, SO_4^{2-} was 30–40% and NO_3^- was 40–50%.

The values of pH, Cl^-/Na^+ , $\delta^{18}\text{O}$ and $\text{Mg}^{2+}/\text{Na}^+$ in the surface snow of the study regions are shown in Fig. 7. The values of pH were scattered and did not depend on the region. The ratios of Cl^-/Na^+ are mostly similar to that of sea water except for those in Greenland and arctic Canada. Surplus Cl^- in Greenland seemed to be supplied by HCl (De Angelis and Legrand, 1995). The $\delta^{18}\text{O}$ values were similar in the Svalbard glaciers regardless of altitude. They were low in Greenland because of high altitude and the long distance from the coast. Iizuka *et al.* (2000) reported that the ratio of $\text{Mg}^{2+}/\text{Na}^+$ represented the index of the percolated melt water effect. When the melt water percolated through the snow layer in summer, the Mg^{2+} which contained snow particles dissolved in water more quickly than the Na^+ . Because it is clearly shown by arrows in Fig. 7b, the surface snow in Svalbard had a large dissolving during summer.

The relationships between Cl^- and $\text{Na}^+/\text{SO}_4^{2-}/\text{NO}_3^-$ are shown in Figs. 8–10, respectively. The circle diameters correspond to certain ion concentrations. The Cl^- and Na^+ relationships are the same as that of sea water except for that in Greenland and arctic Canada (Fig. 8). As in non-melted snow at Brogger, a lack of Cl^- was found.

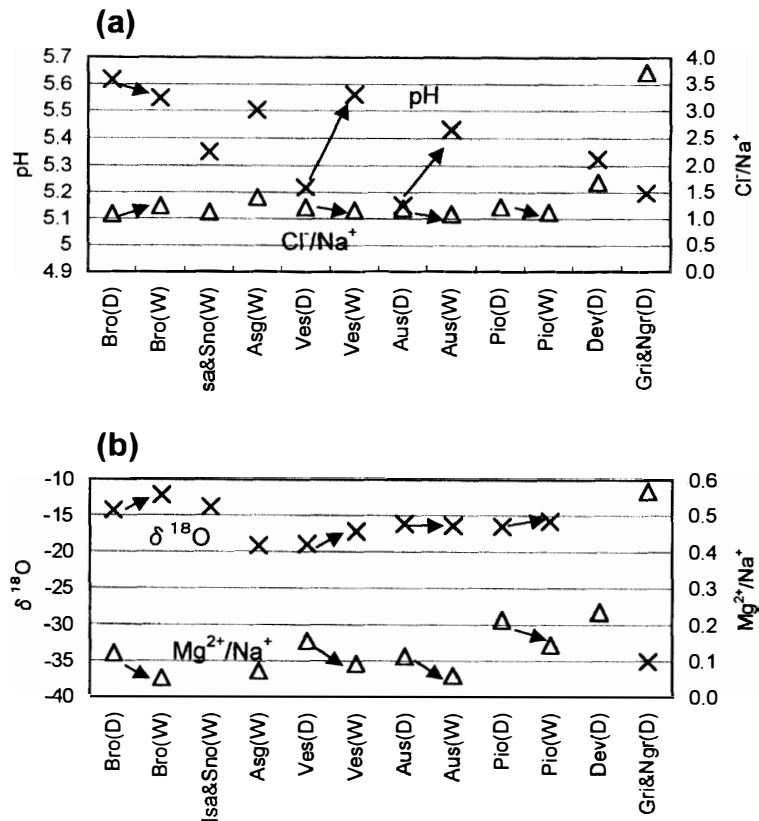


Fig. 7. Typical constituent averaging for each surface layer and region. (a) pH (cross) and ratio of Cl^- and Na^+ concentrations (triangle). (b) Oxygen isotope ratio ($\delta^{18}\text{O}$, cross) and ratio of Mg^{2+} and Na^+ concentrations (triangle). (D) means non-melted snow. (W) means the snow with a history of melting.

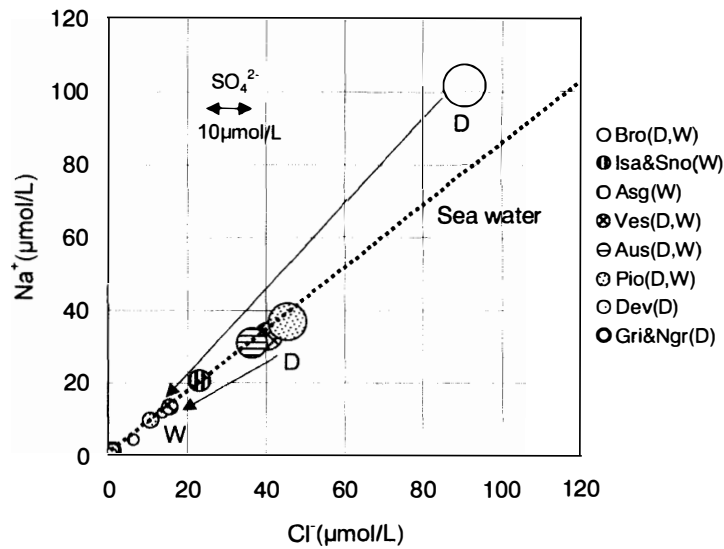


Fig. 8. Relation of Cl^- and Na^+ concentration averaging for each surface layer and region. Circle diameters are in proportion to SO_4^{2-} concentration. The solid line shows the ratio of Cl^- and Na^+ ions in sea water. (D) means non-melted snow. (W) means the snow with a history of melting.

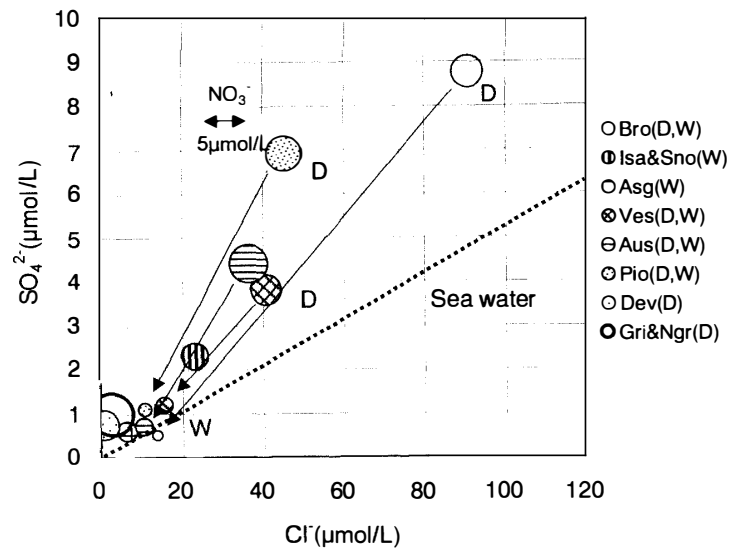


Fig. 9. Relation of Cl^- and SO_4^{2-} averaging for each surface layer and region. Circle diameters are in proportion to NO_3^- concentration. The solid line shows the ratio between Cl^- and SO_4^{2-} ions in sea water. (D) means non-melted snow. (W) means the snow with a history of melting.

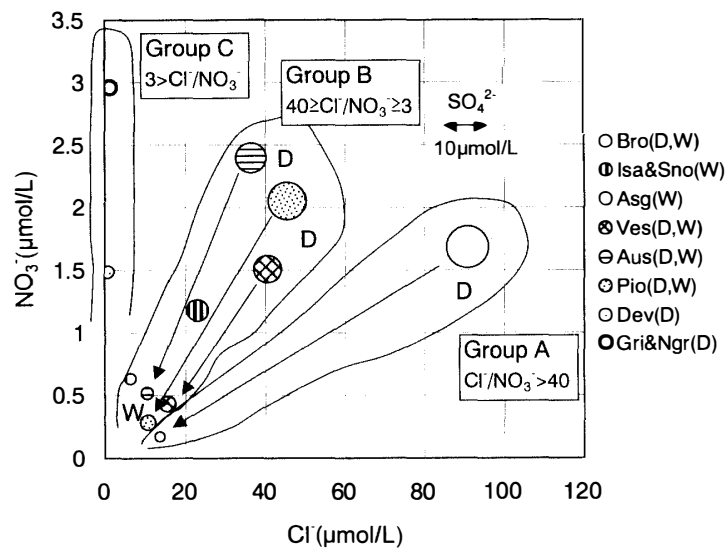


Fig. 10. Relation of Cl^- and NO_3^- concentrations averaged for each surface layer and region. Circle diameters are in proportion to SO_4^{2-} concentration. They are grouped into three categories: A, B and C. (D) means non-melted snow. (W) means the snow with a history of melting.

The SO_4^{2-} concentration appears to increase with Na^+ and Cl^- . In Fig. 9, non-sea salt SO_4^{2-} was present in large amount in non-melted snow of group (I). This rich non-sea salt SO_4^{2-} caused a lack of Cl^- , because the deficit of Cl^- relative to Na^+ was mainly due to the presence of Na_2SO_4 as a result of a sea-salt alteration reaction (Legrand and Delmas, 1988). As shown by the arrows, when the snow received the influence of melt water, the ratio of Cl^- and SO_4^{2-} approached the ratio in sea water, because the SO_4^{2-}

dissolved more quickly (Davies *et al.*, 1982, 1987; Brimblecombe *et al.*, 1985). The relationship between Cl^- and NO_3^- is shown in Fig. 10. Because Cl^- and NO_3^- are supplied from different sources, they can be grouped into three categories based on the ratio between them: Brogger (Group A), Svalbard without Brogger and Severnaya Zemlya (Group B), Greenland and arctic Canada (Group C).

Group A: close to the sea, low altitude ($\text{Cl}^-/\text{NO}_3^- > 40$).

Group B: the middle ratio of Group A and C ($40 \geq \text{Cl}^-/\text{NO}_3^- \geq 3$)

Group C: far from the sea, ($3 > \text{Cl}^-/\text{NO}_3^-$)

The regional characteristics of the snow still remained, in spite of the dissolving of the ions by snowmelt water percolation.

In the future, the flux of chemical components and their seasonal variation in various regions will need to be studied more precisely to understand arctic climatic and environmental systems. The study of the atmosphere-snow transfer function should be a main focus of attention.

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