Melt-induced relocation of ions in glaciers and in a seasonal snowpack

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Abstract We have investigated the relocation of ions upon melting of snow/firn deposited onto subpolar glaciers and a polar ice sheet, as well as that of a seasonal snowpack, by analysing the melt-affected vertical profiles of ionic concentrations at these different sites. Layers with significantly high ionic concentrations were found at snow/ice boundaries on two of the subpolar glaciers. The production of these layers was explained in terms of the concentrating of ions by superimposed ice formation. We suggest that re-thaw of such layers could significantly affect the quality of runoff water even in the late stage of the melt season. At a polar ice sheet, occasional very high concentrations of NO_3^- and SO_4^{2-} were associated with melt layers which had been created by slight surface melting. On the other hand, no concentrating of ions was observed at the ice layers in the seasonal snowpack. The occurrence of concentrated layers strongly depended on the refreezing processes of meltwaters. Data from the different sites studied in this paper suggested that NO_3^- and SO_4^{2-} were preferentially eluted from snow/firn compared to Cl⁻, regardless of the ionic composition of the snow/firn. The preferential elution of NO_3^- and SO_4^{2-} compared with Cl⁻ resulted in either a decrease or increase in the pH of meltwaters, depending on the chemical composition of the snow/firn.

INTRODUCTION

Melting of snow causes migration of soluble and insoluble impurities. Since the solute composition of meltwater from a seasonal snowpack significantly affects the water quality of snow-fed streams and lakes, intensive work on the chemistry of seasonal

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snowpacks and meltwaters has been conducted. It is well known that most of the soluble ions stored within a snowpack are removed by the first fractions of snowmelt, and that ionic concentrations of a snowpack and runoff water generally decrease as melting proceeds (e.g. Johannessen & Henriksen, 1978; Colbeck, 1981; Suzuki, 1982). The chemical changes of a snowpack and runoff can be influenced by chemical composition of the snow and refreezing processes of the meltwater (Colbeck, 1981; Davies *et al.*, 1987; Bales *et al.*, 1990).

We studied the melt-induced redistribution of soluble ions in the surface layers of subpolar glaciers and a polar ice sheet along with that in a seasonal snowpack. At these study sites, the chemical composition of snow and melt-refreeze processes were different. We discuss how these factors affect the relocation of ionic species and the quality of discharge water, by comparing the vertical ionic concentration profiles from the different sites.

MELT-AFFECTED VERTICAL PROFILES OF IONIC CONCENTRATIONS AT DIFFERENT STUDY SITES

Study sites are listed in Table 1. More details of the sites and sample collection have been described in the references cited in Table 1.

Profiles of ionic concentrations, pH (measured at 25°C) and the stratigraphy of the surface layers of three subpolar glaciers are presented in Fig. 1, Fig. 2 and Fig. 3. The profile of solid particles greater than 12 μ m at Glacier No. 1 is also shown in Fig. 3.

The top 2 m layers of Isachsenfonna were all wet on the sampling date, and mainly consisted of granular snow. It was clear that melting had occurred within these layers. Concentration profiles of different ionic species from the 2.1 m deep pit samples at Isachsenfonna (Fig. 1) were all similar. The concentrations showed a general tendency

Site	Location	Site description	Type of samples
Isachsenfonna, western Spitsbergen	79.1°N, 12.9°E, 850 m a.s.l.	Accumulation area at a subpolar glacier	Pit samples collected in the mid melt season (Goto- Azuma <i>et al.</i> , 1993a)
Austre Brøggerbreen, western Spitsbergen	78.9°N, 11.8°E, 440 m a.s.l.	23 m above mean equilibrium line elevation during 1967- 1988 (Hagen & Liestøl, 1990) at a subpolar glacier	nPit samples and an ice core, dug in the mid melt season (Goto-Azuma <i>et al.</i> , 1993a)
Glacier No. 1 at the headwaters of Ürümqi River, Tianshan, China	43.1°N, 86.8°E, 4050 m a.s.l.	Close to mean equilibrium line elevation during 1958- 1988 (Liu <i>et al.</i> , 1991) at a subpolar glacier	Pit samples and a firn/ice core, excavated just after the melt season (Goto-Azuma <i>et al.</i> , 1994)
18C, Greenland (32 km southwest of Dye 3)	65.0°N, 44.6°W, 2600 m a.s.l.	Accumulation area at a polar ice sheet	A firn core (Clausen & Langway, 1989)
Sasagamine, Niigata, Japan	36.9°N, 138.1°E, 1300 m a.s.l.	Seasonal snowpack	Pit samples collected in the early melt season (Goto-Azuma <i>et al.</i> , 1993b)

Table 1 Study sites.

Melt-induced relocation of ions in glaciers



Fig. 1 Vertical profiles of ionic concentrations, pH and the stratigraphy at Isachsenfonna on 12 August 1991.

to increase from the surface to the bottom of the pit. These facts indicate that the leaching of ions had occurred at the top 2.1 m of the glacier snow.

When a snow pit was dug to the boundary between snow and superimposed ice at Austre Brøggerbreen, meltwater continuously emerged from the bottom of the pit. The entire snowpack (0.68 m in thickness) above the superimposed ice was wet. At Austre Brøggerbreen, concentration levels of ions in the snow and the superimposed ice below the snow (Fig. 2) were one order of magnitude lower than those in the winter snow cover (Goto-Azuma *et al.*, 1993a). However, high concentrations were found just below the boundary between the snow layer and the ice layer. These facts indicate that most of the ionic load, released from the snow above the superimposed ice, had moved downward and stopped at the snow/ice boundary.

Vertical ionic profiles similar to those of Austre Brøggerbreen were also observed at Glacier No. 1 (Fig. 3). Ionic concentrations of the layers above the snow/ice boundary I were much less (c. 1/10-1/2) compared to those of the winter snowpack (Wake *et al.*, 1992). Concentration profiles of all ionic species were similar; this is in contrast with the spring snowpack in which Na⁺ was not correlated with Ca²⁺, Mg²⁺, NO₃⁻ and SO₄²⁻ (Williams *et al.*, 1992). An extraordinary peak (peak A) was observed at the boundary I in concentration profiles of all the ionic species. It was obvious that heavy melting had taken place at the drilling site (Goto-Azuma *et al.*, 1994) and that most of the ions had







Fig. 3 Vertical profiles of ionic concentrations, solid particle (> $12 \mu m$) content, pH and the stratigraphy at Glacier No. 1 in October 1990. I, II, III are three of the boundaries between ice and snow (ice on top of snow).

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been removed from the layers above boundary I and had been concentrated at boundary I.

Figure 4 shows detailed profiles of anion concentrations together with melt features at site 18C, located about 32 km upstream from Dye 3, for the depth interval of 16-22 m. At 18C, summer melting is common, but deep percolation is minimal. Thin melt features (thickness less than 5 cm) were frequently found in the depth interval of 16-22 m. No major decrease of ionic concentrations such as those at subpolar glaciers were observed. However, the very high NO₃⁻ and SO₄²⁻ peaks at 18.6 m were associated with a 1 cm ice layer at that depth. A continuous record of anion concentrations for the past 220 years at 18C (Langway *et al.*, in preparation) revealed that SO₄²⁻ as well as NO₃⁻ concentrations were not associated with melt features in the 18C core.

Vertical concentration profiles of ions in a seasonal snowpack at Sasagamine on three different dates are illustrated in Fig. 5 along with stratigraphy. The major portion of the snowpack at Sasagamine originated from dry snowfall. On 28 February the snow temperature was lower than 0°C in the snow layers below the upper 5 cm, and most of the compacted-snow layers within the snowpack showed little melt. 9 March and 22 March were in the early snowmelt season. On 9 March the temperature of the entire snowpack was 0°C, and on 22 March the temperature of all the snow except that in the top 40 cm was 0°C. Between 28 February and 22 March, complicated behaviour of ions was observed. For example, the mean concentrations of NO_3^- and SO_4^{2-} decreased in layers 7 and 11 and increased in layers 6 and 9 between 9 and 22 March, while concentrations in layers 5, 8 and 10 did not change as much. The decrease in NO_3^- and SO_4^{2-} loads in layer 7 is too small to account for the increase in layer 6. The deficiency can be explained in terms of the input from the upper layer 11, since the increase in NO_3^- and SO_4^{2-} loads in layer 9 is much smaller than the decrease in layer 11.

In layer 11 on 22 March, NO_3^- and SO_4^{2-} in sublayers of granular snow showed



Fig. 4 Detailed profiles of anion concentrations at 18C together with melt features. Melt features are qualitatively classified as: IL, ice layer; IW, ice wedge; DIM, definite indication of melting; SIM, slight indication of melting. The large peaks in NO $\frac{3}{3}$ and SO $\frac{2}{4}$ – profiles (the open arrows) were associated with a 1 cm thick ice layer (the solid arrow).



Fig. 5 Stratigraphy and ionic concentration profiles of a seasonal snow cover at Sasagamine, Niigata, Japan. Observations were made on 28 February, 9 March and 22 March in 1992. Layers 1-13 are numbered both in right-hand and left-hand figures. The notation used in ionic concentration profiles is as follows: G, granular snow layer; C, compacted snow layer; I, ice layer; •, concentration level of a duplicate sample from the water channel at the same depth.

lower concentrations than those in sublayers of compacted snow. Although concentration levels of Cl^- , NO_3^- and SO_4^{2-} in a duplicate sample from the water channel (region of preferential flow) in layer 11 on 9 March were lower than those in compacted snow at the same depth, concentration levels of all the ions in a duplicate sample from the water channel in layer 3 on 22 March were slightly higher than those in the compacted-snow layer at the same depth. On the other hand, concentration levels of Cl^- , NO_3^- and SO_4^{2-}

in thin sublayers of ice (e.g. ice layers in layer 2 on 22 March) were similar to those in the adjacent sublayers of granular snow. Thus, no clear dependence of ionic concentrations on the physical structure of the snow was seen in the seasonal snowpack at Sasagamine.

CONCENTRATING OF IONS BY MELT-FREEZE PROCESSES

As has been shown above, the relocation of ions at Austre Brøggerbreen and Glacier No. 1 occurred in a similar manner. At both sites, very high ionic concentrations were observed at a boundary between snow and ice; this suggests a mechanism common to both glaciers may be responsible for the redistribution of ions at the snow/ice boundary. At Glacier No. 1, a striking peak A' similar to the ionic peak A was observed in solid particle concentration, but at a deeper depth than ionic peak A by 0.6 m (Fig. 3). Formation of these remarkably high peaks of ions and solid particles can be explained by the following mechanism. Figure 6 shows diagrammatically the simple hypothesis behind the mechanism. When the snowpack on the glacial ice (Fig. 6(a)) starts to melt, meltwater enriched in ions moves downwards through water channels (Fig. 6(b), e.g. Suzuki, 1982). The movement of the meltwater is prevented at the snow/ice boundary BO since ice is impermeable. If further melting takes place, a thick layer L1 becomes soaked with meltwater (Fig. 6(c)). Solid particles sink to the boundary BO as they are heavier than water. Thus the layer just above the boundary BO becomes enriched in solid particles. When the meltwater refreezes from the bottom, particles are trapped in this layer and thus a layer with high particle concentration is produced just above the old boundary BO (Fig. 6 (d)). On the contrary, ions are rejected from the ice layer L2. Hence ions get more concentrated in layer L3. When layer L3 finally freezes, a layer with very high ionic concentrations is created at a new snow/ice boundary BO' (Fig. 6(e)).

Wakahama *et al.* (1976) examined the formation of superimposed ice, and proposed a model, which assumed a gradual and continuous supply of meltwater. Their model,





however, cannot explain the difference between the peak depth for particles and that for ions by 0.6 m (Fig. 5). The present hypothesis requires that a snow layer of significant thickness (0.6 m) becomes soaked with meltwater rather quickly on top of the ice in the early stage of the formation of superimposed ice. In effect, the formation of such a superimposed ice layer can be expected from the meteorological data obtained at a station near Glacier No. 1 (Goto-Azuma *et al.*, 1994).

Smaller ionic peaks than peak A (peaks B and C) were found to be common to all the ionic concentration profiles at snow/ice boundaries II and III. At depths below peak A, concentrations of all the ions except for those around boundaries II and III were again generally lower than those in the winter snowpack. In addition to the ionic peaks B and C, a peak in particle concentration profile (peak B') was seen at a slightly deeper site than boundary II. Peaks B, C, and B' were probably created by the same processes as peaks A and A'.

As described above, the layers with strikingly concentrated ions at glaciers Austre Brøggerbreen and Glacier No. 1 were related to the formation of superimposed ice. Similar very high ionic peaks at the temperate Folgefonni Ice Cap (60.1°N, $6.4^{\circ}E$, 1625 m a.s.1.) have been reported by Davies *et al.* (1982, 1987), although the plausible mechanism of ice formation at Folgefonni Ice Cap is the compression of wet snow (e.g. Colbeck *et al.*, 1978), not superimposed ice formation. The high ionic peak at a temperate glacier could be formed by a different ice formation mechanism.

High concentrations of NO_3^- and SO_4^{2-} were seen in some of the melt layers at 18C. These concentrations, however, did not occur necessarily in all of the ice layers in the 18C core. Also, in contrast to Bales *et al.* (1990), who found that ions were concentrated in ice layers in a Wyoming snowpack, we did not observe the concentrating of ions in ice layers within the snowpack at Sasagamine. These facts could be explained by the following process: the meltwater created either at the ice sheet surface or at a seasonal snowpack, which is rich in ions, flows down through the layers underneath. When the water refreezes at an impermeable layer, ions remaining in concentrated brine may be allowed to flow further downwards, since the freezing point of the brine is lower than that of pure water. The ice layer formed at this stage is not rich in ions. If the water with more concentrated ions finally refreezes in a lower layer where there is a layer which is impermeable to the concentrated brine, a melt layer with concentrated ions is created just above the impermeable layer.

The degree of concentration would depend not only on the amount of melting but also on the "efficiency" of an impermeable layer. A layer with a low temperature and a relatively thick ice layer could work as an efficient impermeable layer to highly concentrated brine. At a site on a polar ice sheet such as 18C, the subsurface temperature is well below 0°C even when surface melting is taking place. Therefore, a lower layer can refreeze highly concentrated brine. At a glacier site with superimposed ice formation, such as Austre Brøggerbreen and Glacier No. 1, the thick continuous ice layer below the melting snowpack can work as an efficient impermeable layer. Also, at a site on a temperate glacier such as the Folgefonni Ice Cap, continuous ice below firn can work as an efficient impermeable layer. In contrast, a seasonal snowpack on relatively warm ground is less likely to form efficient impermeable layers within the pack, because of the geothermal heat flux at the snow/soil interface. For this reason, the concentration of ions in ice layers of seasonal snowpacks would not be as enhanced as that at snow/ice boundaries in glaciers or polar ice sheets. Remarkably high ionic peaks such as those found in glaciers could not be created in seasonal snowpacks on warm ground, even if the amount of melt were the same.

THE EFFECT OF A LAYER WITH HIGHLY CONCENTRATED IONS ON RUNOFF CHEMISTRY

The layer with highly concentrated ions existing at the boundary between snow and superimposed ice, such as those observed at Austre Brøggerbreen and Glacier No. 1, can be exposed at the glacier surface if further melting takes place in the later stage of the melt season. If such a layer melts, the ion content in supra-glacial meltwater would be high again in the later stage of the melt season. Concentrations could be even higher than those in the meltwater released at the initial stage of the melt season, as ions are much more concentrated in such a layer than in the winter snowpack. In fact, at Austre Brøggerbreen, supra-glacial meltwaters with very high ionic concentrations were found at lower elevations where the winter snowpack had just been melted away (Goto-Azuma *et al.*, 1993a). As for the Folgefonni Ice Cap, the layer with extremely high ionic peaks existed only temporarily, and it disappeared by the end of the melt season (Davies *et al.*, 1982, 1987). This suggests that a temperate glacier with a water table could also release ion-rich meltwater in the later stage of melt season, when a layer with high ionic concentrations is re-thawed. When we study the chemical aspects of glacier hydrology, we have to take into account the existence of such layers.

PREFERENTIAL ELUTION OF IONS

It is well known that some ions are removed more rapidly than others upon melt, although the elution sequence has not been well understood (e.g. Davies *et al.*, 1982; Brimblecombe *et al.*, 1985; Tsiouris *et al.*, 1985; Davies *et al.*, 1987). This phenomenon is called "preferential elution".

At Austre Brøggerbreen, the ratio of the peak ionic concentration to the average ionic concentration in the entire depth except for the peak was greater for NO_3^- and SO_4^{2-} than for Cl⁻ and Na⁺ (Fig. 2). This may reflect the preferential elution of NO_3^- and SO_4^{2-} to Cl⁻ and Na⁺ from melting snow (e.g. Davies *et al.*, 1982, 1987). Since 40-90% of the total SO_4^{2-} in the winter snowpack at Austre Brøggerbreen was of sea-salt origin (Goto-Azuma *et al.*, 1993a), the residual 10-60% of SO_4^{2-} probably originated from H₂SO₄. The origin of NO_3^- was likely to be HNO₃. The preferential elution of acidic components might have resulted in the drop of pH in the layer just below the snow/ice boundary.

At glacier No. 1, the ratios of the ionic concentrations at peak A to the average ionic concentrations in the entire top layers above peak A were: NO_3^- , 106; SO_4^{2-} , 39; Cl⁻, 26; K⁺, 18; Mg²⁺, 63; Ca²⁺, 28; Na⁺, 37 (Fig. 3). The sequence of preferential elution from the layers above peak A can be deduced as follows:

$$NO_3^- > Mg^{2+} > SO_4^{2-}, Na^+ > Ca^{2+}, Cl^- > K^+$$

Since 80% of SO_4^{2-} in Tianshan snow was deposited in association with Ca^{2+} and Mg^{2+}

(Williams *et al.*, 1992), the major part of SO_4^{2-} is considered to have been deposited as salt, and not as acid. The preferential elution of NO_3^- and SO_4^{2-} to Cl^- thus leads to an increase of pH in the layer of peak A, while the preferential elution of the same ions leads to a decrease of pH at Austre Brøggerbreen and Isachsenfonna.

At Isachsenfonna, the chemical composition of winter snow might be similar to that at Austre Brøggerbreen. The decrease of pH with depth (Fig. 1) may reflect the preferential elution of H_2SO_4 and HNO_3 .

The slight melting at 18C affected NO₃⁻ and SO₄²⁻ concentrations but did not affect Cl⁻ concentrations significantly, while much heavier melting at the subpolar glaciers Austre Brøggerbreen and Glacier No. 1 strongly affected the concentrations of all ions. This is the result of the preferential elution of NO₃⁻ and SO₄²⁻ to Cl⁻ at 18C, where NO₃⁻ and SO₄²⁻ were mostly of acid origin (Finkel *et al.*, 1986). Moreover, melt features were more frequently associated with NO₃⁻ peaks than with SO₄²⁻ peaks at 18C: NO₃⁻ concentration was always high in a melt layer associated with high SO₄²⁻ concentration, but the opposite was not always true. This suggests the elution sequence of NO₃⁻ > SO₄²⁻.

The seasonal snowpack in the early phase of snowmelt at Sasagamine showed more enhanced relocation of NO₃⁻ and SO₄⁻ than Cl⁻ (Goto-Azuma *et al.*, 1993b). This also reflects the elution order of NO₃⁻, SO₄⁻ > Cl⁻ at Sasagamine, where SO₄²⁻ and NO₃⁻ were mainly of acid origin. Although the elution order of NO₃⁻ and SO₄²⁻ seemed to have varied from layer-to-layer and from time-to-time, NO₃⁻ was eluted more easily than SO₄²⁻ in most cases (Goto-Azuma *et al.*, 1993b).

The results show that NO_3^- and SO_4^{2-} seem to be eluted more easily from snow/firn than Cl⁻, regardless of the proportional ionic composition of the snow/firn. The preferential elution of NO_3^- and SO_4^{2-} from the snow at Isachsenfonna and Austre Brøggerbreen caused a decrease in the pH of meltwater, while the preferential elution of the same ions from the snow at Glacier No. 1 caused an increase on the pH of meltwater. Thus, the change of pH depended on the proportional ionic composition of the original snow. Although the elution sequence of NO_3^- and SO_4^{2-} was not definite, and may vary among different layers, our results favour the rank $NO_3^- > SO_4^{2-}$. This seems to be contradictory to the earlier investigations (e.g. Davies *et al.*, 1982, 1987; Brimblecombe *et al.*, 1985, 1987). Further investigations on preferential elution have to be carried out in association with the texture and the chemical composition of the layer.

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