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**Sea salt in snow on
Arctic sea ice and in
coastal regions**

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The origin of sea salt in snow on Arctic sea ice and in coastal regions

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Abstract

Snow, through its trace constituents, can have a major impact on lower tropospheric chemistry, as evidenced by ozone depletion events (ODEs) in oceanic polar areas. These ODEs are caused by the chemistry of bromine compounds, that originate from sea salt bromide. According to current ideas, bromide may be supplied to the snow surface either by upward migration from sea ice or by frost flowers being wind-blown to the snow surface. We investigate here the relative importance of both these processes by analyzing mineral ions in snow samples collected near Alert and Ny-Ålesund (Canadian and European high Arctic) in winter and spring. Vertical ionic profiles in the snowpack on sea ice are measured to test upward migration of sea salt ions and to seek evidence for ion fractionation processes. Time series of the ionic composition of surface snow layers are investigated to quantify wind-transported ions. Upward migration of unfractionated sea salt, to heights of at least 17 cm, was observed in snow sampled in winter, at temperatures near -30°C , leading to Cl^{-} concentration of several hundred μM . Upward migration thus has the potential to supply ions to surface snow layers. Time series show that wind can deposit aerosols to the top few cm of the snow, leading also to Cl^{-} concentrations of several hundred μM , so that both migration from sea ice and wind transport can significantly contribute ions to snow. At Ny-Ålesund, sea salt transported by wind was unfractionated, implying that it does not come from frost flowers. In the Arctic, frost flowers thus do not appear necessary to lead to large sea salt concentrations in surface snow, and to supply the bromide needed for ODEs. The data obtained also indicate that ODEs lead to significant deposition of Br^{-} to snow. We speculate that this can also take place in coastal regions and contribute to propagate ODEs inland. Finally, we stress the need to measure snow physical parameters such as permeability and specific surface area, to understand quantitatively changes in snow chemistry.

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1. Introduction

Interactions between the snowpack and the atmosphere lead to important modifications of atmospheric composition (Domine and Shepson, 2002), and the most dramatic example is perhaps the complete destruction of ozone from the ground up to altitudes greater than 1000 m, observed in the Arctic (Bottenheim et al., 2002a) and in Antarctica (Tarasick and Bottenheim, 2002) in the spring, when the frozen sea and the ground are almost entirely snow-covered. It is now reasonably certain that brominated species, derived from sea salt bromide, are key species in the chemistry of this ozone destruction (Barrie et al., 1988; Fan and Jacob, 1992; Tang and McConnell, 1996).

However, many aspects of this chemistry are not fully understood and prevent the detailed modeling, let alone the prediction, of these ozone depletion events (Bottenheim et al., 2002a). Among these, the processes by which sea salt and bromide reach the snow surface to become available for activation into brominated compounds is unclear. Two major processes can be envisaged.

The first process involves frost flowers, that often form on the refrozen surface of open leads and have a salinity around 100 g/kg (hereafter practical salinity units : psu) (Martin et al., 1995). Frost flowers are fragile structures that can be readily windblown and redistributed to the snow surface, where they can constitute an available source of sea salt (Wolff et al., 2003). Precipitation of mirabilite ($\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$) from the brine at the surface of refrozen ice often accompanies frost flower formation (Rankin et al., 2002), with the result that sea salt coming from wind-blown frost flowers is depleted in sodium and sulphate. Because Antarctic sea salt aerosol is often depleted in sodium and sulphate, especially in winter when the extent of sea ice is largest (Wagenbach et al., 1998), it has been concluded that sea ice, by the formation of fragile frost flowers, was the main source of sea salt to snow on the Antarctic continent and on surface marine snow (Wolff et al., 2003). Regarding the Arctic, fewer detailed studies have been performed and measurements are more delicate to interpret, as for example the sulphate signal is more obscured by anthropogenic contributions.

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The second process is the upward migration of a sea salt enriched brine from the sea ice to the snowpack. Brines are often present at the sea ice-snow interface and can travel up the snow by capillary forces. The wettability of snow crystal surfaces by the brine is then the driving force for this process. Numerous studies in Antarctica (Eicken et al., 1994; Massom et al., 2002) have shown that salinities up to 5 psu could often be found in snow at a height of 20 cm above the sea ice. More limited studies in the Arctic have also shown that a slush layer was present at the surface of Arctic ice (Garrity, 1992) from which the capillary motion of sea salt can be inferred. Direct salinity measurements (Eicken et al., 2002) have also shown that high salinities could be found in snow on pack ice. However, these measurements were done in the summer, where the warmer temperatures favor the presence of a liquid phase, while ozone depletion episodes mostly take place around early spring. It is thus not clear whether surface snow, that is more susceptible to interact with the atmosphere, can be directly affected by sea salt migrating up from the sea ice in the spring.

Both these processes, that can bring bromide from sea salt into the surface layers of the marine snowpack, are illustrated in Fig. 1. The open sea can also be a source of sea salt, through wind action that generate sea salt aerosols (Buat-Ménard et al., 1974). However, Wolff et al. (2003) have determined that, at their measurement sites in Antarctica, sea ice is a much more efficient source of sea salt aerosol than the open sea, so we will not consider this latter source unless it appears necessary.

Understanding and modeling the flux of brominated compounds to the atmosphere through bromide activation requires, among many other aspects, the determination of which of these two processes is predominant. The frequency of both processes may also evolve with climate change, and determining current processes is a first step towards predicting the future availability of bromide for activation in surface snow.

To help shed some light on this question, we took advantage of 3 expeditions to the Arctic to sample snow on the sea ice, where the direct influence of the ocean would be detectable, and on land near the coast, where the influence of marine air masses would be felt, but not that of migration from sea ice. These expeditions were the winter

and spring intensive campaigns of Alert 2000 (Bottenheim et al., 2002b) and the spring intensive of the NICE campaign in Ny-Ålesund, Spitsbergen, in April/May 2001 (Beine et al., 2003).

Our approach was to analyze mineral ions in snow by ion chromatography. Ion speciation yields useful data, compared to salinity measurements. Indeed, fractionation processes that can take place then become detectable, and this helps determine which process took place. For example, sodium and sulphate depletion due to mirabilite precipitation are expected if snow is affected by frost flowers deposited by wind (Rankin et al., 2002). Other ions can show fractionation due to the precipitation of other phases. In particular, Koop et al. (2000) calculated that Br^- could be enriched up to 100 times relative to sea water, at 230 K, after the successive precipitation of several salts. Mulvaney et al. (1993) observed sea salt fractionation in Antarctica, that may be interpreted by the precipitation of a salt having an MgCl_2 stoichiometry.

We present here vertical profiles and time series of ion compositions in several Arctic locations. Our initial objective in the first sampling campaigns was not to answer the title question of this paper, and the first samplings were thus not perfectly targeted. Subsequent samplings became more adequately focused. Although the present data do not fully answer our question, we feel that they do bring a useful contribution to our understanding of the mechanism of transfer of sea salt and bromide to surface snow layers.

2. Experimental methods

2.1. Sites

Around Alert, (North coast of Ellesmere island, Canadian high Arctic) the sampling site on land was within 400 m of the FTX building ($82^\circ 27.28' \text{ N}$, $62^\circ 29.69' \text{ W}$, Domine et al., 2002), about 5.4 km from the coast. On the sea ice, the winter sampling site was on Joliffe bay ($82^\circ 31.07' \text{ N}$, $62^\circ 42.05' \text{ W}$) and the spring sampling site was at $82^\circ 31.74' \text{ N}$,

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62°44.39' W, near Williams island. Sampling was also done on the Northern Arctic ocean on 25 April 2000, at 84°56' N, 67°28' W.

Around Ny-Ålesund (Svalbard archipelago, Spitsbergen island, European high Arctic), sampling was done near the Ny-Ålesund village (78°55.45 N, 11°55.67 E) by the Amundsen mast, about 30 m from the shoreline. Sampling on the sea ice was in one of the rare locations where sea ice was present around Ny-Ålesund in May 2001, at 78°49' N, 11°40' E.

2.2. Snow sampling

Snow was sampled in a manner essentially similar to that described by Domine et al. (2002) for physical measurements. Polyethylene gloves were used to avoid contamination, and snow was sampled into polyethylene sampling vials. For each snow layer, triplicate samples were taken for anions and another set of triplicates was taken for cations. Samples were usually melted and analyzed on site by ion chromatography within 12 h of sampling, and in a few cases were stored frozen for up to 48 h before analysis.

2.3. Ion chromatography analyses

Melted snow was analyzed by ion chromatography (Dionex IC mod. DX120 and DX100) using Dionex AS12 and CS12 columns for anions and cations, respectively, as detailed in Table 1. Detection was by conductimetry coupled with post-column suppression of eluent ions. The use of self-regenerating suppressors that enhance analyte conductivity while suppressing eluent conductivity allowed parts-per-billion (ppb) detection limits for anions and cations without preconcentration.

To optimize detection limits and to ensure quality data, special care was taken in the field and in the laboratory to the quality control/assurance procedures (Allegrini et al., 1999; Perrino et al., 2001). The lower detection limits (L.O.D.) of the analytical technique (Table 2) were calculated as the analyte signal which is three times the

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standard deviation (3σ) of a near-zero concentration measurement. The precision and accuracy of ion chromatography measurements were determined from the standard deviation and mean of repeated analyses of the laboratory standard. Typically, a multi-point calibration (seven standards in the range 5–1000 ppb) was used. The correlation coefficient from the regression equation of the average values was always greater than 0.999. The IC response was linear up to $20\ \mu\text{M}$ for each chemical species analyzed, and dilutions were performed for more concentrated samples. For chromatographic analyses, control samples, consisting of calibration solutions of anions (Cl^- , Br^- , NO_3^- , NO_2^- , SO_4^{2-}) and cations (Na^+ , NH_4^+ , K^+ , Mg^{2+} , Ca^{2+}), were analyzed every twenty samples in order to re-calibrate the ion chromatograph. Calibration solutions were prepared every two weeks from 1000 ppm standard solutions of salts (MERCK).

3. Results and discussion

3.1. Vertical profiles on pack ice

The snow pack was studied in Joliffe Bay, near Alert, on 11 and 17 February 2000. The ice is essentially permanent there, and the probability that our sampling was on multi-year ice is then high. The snow stratigraphy was very variable, as detailed by Domine et al. (2002), with snow depth varying between 0 and 50 cm. The aspect of the snow bank that was sampled is shown in Fig. 2, along with sampling levels. During sampling, no liquid water could be detected anywhere, and the temperature in the snow near the surface of the snow pack was -40°C , increasing to -27°C 5 cm above the sea ice. The Na^+ , Cl^- , and Br^- profiles are also shown in Fig. 2. The ionic composition of the snow samples are detailed in Table 3, where the snow salinity has also been calculated from these analyses. Ionic ratios in the snow samples are shown in Table 4, where they can be compared to those of sea water. The maximum concentrations, with $295\ \mu\text{M}$ of Cl^- are found in the windpack closest to the sea ice, and the minimum concentrations ($30\ \mu\text{M}$ of Cl^-) are found in the depth hoar. Cl^-/Na^+ molar ratios vary from 1.25 (lowest

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sampled layer) to 1.79 (depth hoar), i.e. somewhat higher than the sea salt value of 1.165 (Holland, 1978). This difference is not significant for the lowest layer sampled. Br^- concentrations are below detection limit (b.d.l.) in two cases. Cl^-/Br^- values are 631 in the lower windpack, essentially identical to the sea water value of 647 (Holland, 1978), while it is 126 for the uppermost layer, i.e. enriched in Br^- by a factor of 5 relative to sea water. Subsequent samplings at the same levels on 17 February yielded similar results. Interestingly, the lowest layer sampled had ratios of Cl^- , Br^- , Mg^{2+} , and K^+ to Na^+ were all within 10% of those of sea water (Table 4). $\text{Ca}^{2+}/\text{Na}^+$ was 10 times the ocean value, but this can be explained by soil dust (Toom Saunry and Barrie, 2002), whose ubiquitous presence in the lowermost snow layers gives the snow a darker color. Furthermore, the highest ionic concentrations were also found in this layer, and it appears reasonable to suggest that unfractionated sea salt was present 17 cm from the ice/snow interface, and that it reached this snow by migration from the sea ice. Unfortunately, no detailed sampling was done in the vicinity of the sea ice to confirm the presence of a gradient and to evidence higher concentrations near the ice surface.

Migration from the sea ice may not explain all the compositions observed. $[\text{Cl}^-]$ in the uppermost layer is $49.8 \mu\text{M}$, while that of the depth hoar is $30.1 \mu\text{M}$. This may seem to rule out migration from the base of the snowpack. However, if migration of ions is by capillary forces and therefore takes place on the surface of snow crystals, it is possible that the ionic concentrations are proportional to the specific surface area (SSA) of the snow. Domine et al. (2002) mention that the depth hoar, the top windpack, and the recent surface snow had SSAs of 200, 240, and $680 \text{ cm}^2/\text{g}$, respectively. The higher SSA of the higher layers imply that a significant part of their ion content could have been caused by upward migration. However, an examination of the ionic ratios (Table 4) show that the snow composition is different from that of sea salt. For example, in the top snow layer, $\text{Mg}^{2+}/\text{Na}^+$ and K^+/Na^+ are 2.1 and 1.7 times the sea salt values. We then conclude that, even if upward migration reached surface snow, wet or dry deposition made significant contribution to the ionic content of the surface snow layers.

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Data obtained on the Arctic Ocean site (84°56' N, 67°28' W) on 25 April 2000 tend to confirm this conclusion. The sampling site was in the vicinity of a recently refrozen lead, on an ice plate surrounded by pressure ridges. Judging from the extensive depth hoar development, the ice must have formed in the previous fall, and may even have been multi-year ice. The snow stratigraphy and chemistry are shown in Fig. 3. Large concentrations of sodium (202 μM) and chloride (356 μM) were seen near the surface, while much lower values (1.4 and 2.7 μM) were observed in the lowermost layer sampled, 13 cm above the ice. This clearly rules out upward capillary migration to explain the high near-surface concentrations. Ionic ratios are also very different from sea water values. Cl^-/Na^+ values in the 4 layers sampled are, from top to bottom, 2.75, 1.76, 2.06, 1.96, all much higher than the 1.165 ocean value. Only two Br^- concentrations are available and the Cl^-/Br^- ratios are 25 and 32 in the top and bottom layers, much lower than the 647 ocean value, indicating bromide enrichment by a factor over 20. $\text{Mg}^{2+}/\text{Na}^+$ and K^+/Na^+ ratios are also greater than in sea water, by factors up to 5. This vertical profile thus clearly indicates that, although sea salt has to be invoked to explain the high sodium and chloride concentrations observed, it clearly did not come directly upwards from the sea ice. The low Cl^-/Br^- ratio may be due to the active bromine chemistry involved in ozone destruction, that would produce large atmospheric HBr concentrations which could be taken up by the snow. Indeed, complete ozone destruction was observed at Alert in the morning of 26 April (Bottenheim et al., 2002a) and the air mass was coming from the Arctic ocean, where active ozone destruction chemistry must have been taking place during and before sampling. Chloride is also activated by this chemistry, resulting in large HCl concentrations, and this may explain the high Cl^-/Na^+ ratios. Another possible explanation for the low Cl^-/Br^- ratio is that ions were supplied by wind-blown frost flowers, where enhanced Br^- concentrations can be suggested from the calculations of Koop et al. (2000). However, the hypotheses underlying those calculations may be questioned, based on one set of measurements by Rankin et al. (2002) of frost flowers in Antarctica, that found Br^-/Na^+ ratios similar to that of sea water. The high $\text{Mg}^{2+}/\text{Na}^+$ and K^+/Na^+ ratios can be interpreted as a contribution from

other sources, such as soil dust (as confirmed by high Ca^{2+} values) and anthropogenic emissions (as confirmed by high sulphate values).

These first two examples show that both upward migration and deposition from the atmosphere can lead to snow of salinity 0.02 psu (Na^+ content around $200 \mu\text{M}$), about 20 cm above the ice surface. However, snow compositions in both these cases are very different, and this makes it possible to conclude that upward migration took place near Alert, while wind deposition took place on the northern Arctic ocean.

A further example of sea salt ions brought by wet or dry deposition comes from a study of the snowpack on sea ice near Alert on 23 April 2000 (Fig. 4). Again, only the top layers were analyzed. Cl^- and Na^+ concentrations are fairly high, showing that sea salt contributed to snow chemistry. The shapes of the vertical profiles do not appear to be caused by upward transport, although windpacks usually have SSAs greater than depth hoar (Legagneux et al., 2002) and the process cannot be ruled out from the profiles only. Ionic ratios are different from those of sea salt (Table 4): Cl^-/Na^+ values in the 4 layers sampled are, from top to bottom, 2.55, 1.92, 2.20, 3.28. $\text{Mg}^{2+}/\text{Na}^+$ and K^+/Na^+ ratios are 3 to 5 times higher than in ocean water, again indicating multiple sources. Interestingly, the Cl^-/Br^- ratio is less different from the ocean values than in the preceding case. The top layer has a value of 137, i.e. enriched by a factor of 5 relative to the ocean, but the other 3 layers have values of 515, 610 and 432, i.e. barely significantly different from the ocean values. We note that sampling took place before active ozone depletion chemistry (Bottenheim et al., 2002a), and the comparison between these last 2 vertical profiles suggests that ozone depletion chemistry may lead to the deposition of large concentrations of bromide to surface snow.

The importance of upward motion of salt from the sea ice was tested in the spring near Ny-Ålesund. Figure 5 shows the stratigraphy and chemical composition of the snow pack on sea ice on 6 May 2001. Judging from the thin snowpack, the sea ice had formed recently, probably in late April. West Spitsbergen gets little sea ice, as it is under Gulf Stream influence. Moreover, 2001 was an anomalous year, in that the

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Fjord at Ny-Ålesund was only frozen for a few weeks in April. As shown in Fig. 5, the snow temperature was fairly high, around -5°C , while the air temperature on this totally overcast day was -5.4°C , 1.5 m above the surface. Significant amounts of liquid water were observed in the bottom layer, 2 cm thick. Figure 5 shows that huge Na^+ and Cl^- concentrations are found in the lowest layer: this snow has a salinity of about 5 psu. This snow did not appear to be water-saturated, and this is confirmed by its low density of 0.25. According to Colbeck (1982), this implies that the liquid water content was less than 14%, and the liquid was then rather a brine with a Cl^- content of at least 28 g/L. Unfortunately, an error in sample handling during the dilution procedure prevented the accurate quantification of all ions, and the values for this layer, (Tables 3 and 4) are then lower limits. The Cl^-/Na^+ ratio measured, 8.4, appears unrealistic and illustrates the problem encountered.

In the snow layer above, that appeared dry and that was made of small rounded grains 0.4 to 0.6 mm in diameter, the Cl^- content was $728\ \mu\text{M}$ about 3 cm above the ice, and it is reasonable to suggest that this high value was caused by upward capillary motion from the underlying brine. Again, analyses for this level was problematic and data for other ions are not reliable. In this same layer, but 6 cm above the ice, the Cl^- content was down to $82\ \mu\text{M}$, a value that we have observed, although rarely, in precipitating snow in coastal regions. The composition of this snow (Table 4) appears consistent with migration of sea salt, and additional atmospheric input of SO_4^{2-} and Ca^{++} . The Cl^-/Br^- ratio is half that of sea water, and this may be due to atmospheric input. 8 cm above the sea ice, the Cl^- content was $3\ \mu\text{M}$, which is fairly low even for precipitating snow. The ion ratios in this snow are not very different from those of sea water (Table 4). This suggests that its composition could be the result of upward migration, but this would require a very low ionic content of the precipitating snow, so that its original composition would not obscure the sea salt signature. Alternatively, this composition could be due to the atmospheric scavenging of mostly sea salt, which is quite possible in this marine environment. Finally, the Cl^- concentration from the top layer, $62\ \mu\text{M}$, cannot be due to transport from the ice at all, considering the low

concentration of the underlying layer.

Two conclusions can be derived from this vertical profile. First, upward transport of sea salt from the sea ice is evidenced in the presence of a brine. This clearly is not new, as numerous salinity studies of snow on sea ice have observed this (Eicken et al., 1994; Massom et al., 2001). However, the interesting observation is that, even at the warm temperatures observed, upward capillary motion leads to high sea salt concentrations only up to 6 to 7 cm, over a time scale that we estimate as a month at the most. Even in this thin 10 cm snowpack, the basal brine does not affect the top snow layer, at least over a time scale of about a month. Thus, in this case, the atmosphere is not in direct contact with snow that has been affected by sea ice.

To sum up this section, 2 of these 4 profiles indicate that upward capillary motion does take place up to a certain height in the snowpack. In the other 2 cases, this conclusion is not reached simply because the relevant measurements were not made, and the existence of this physical process cannot be ruled out. However, in all 4 cases we can safely say that the composition of the uppermost snow layer was not significantly affected by upward capillary motion. The chemical composition of these top layers indicate several contributing sources, and we may have detected the effect of ozone depletion chemistry on snow composition, as this process may explain high Br^- concentrations. Sea salt deposited from the atmosphere was clearly a contributor to the chemistry of the top snow layer. We cannot determine whether this “atmospheric” sea salt was supplied by frost flowers, as sulphate can have multiple sources, and the $\text{SO}_4^{2-}/\text{Na}^+$ ratio cannot be used for that purpose. In the case of the Spitsbergen profile, Cl^- depletion, possibly due to fractionation of atmospheric sea salt by sulphate (Legrand and Delmas, 1988) present in high concentrations, is visible in the top layer. In addition to these vertical profiles, time series of the chemistry of a given snow layer provide valuable insights into the processes responsible for snow composition.

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3.2. Time series

3.2.1. 3 and 7 February layers, Alert

The chemistry of the snowfalls of 3 and 7 February 2000 (Domine et al., 2002) was monitored on land at Alert, near the FTX building. The evolutions of the Cl^- and Br^- concentrations are shown in Fig. 6a. As detailed in Domine et al. (2002) both layers were deposited on a hard wind-packed layer. The 3 February layer fell under windy conditions, accumulated in wind-sheltered spots such as the lee of sastrugi and was discontinuous. The 7 February layer fell under calm conditions and was homogeneous. Its composition (Tables 3 and 4) indicate contributions from sea salt, long range transport of SO_4^{2-} and other ions, and a local source of several ions, essentially Ca^{2+} (Toom-Sauntry and Barrie, 2002). The Cl^- concentrations showed little variations until 22 February. The slow decrease of Cl^- in the 7 February layer is probably due to the dilution of this layer by surface hoar, that formed continuously (Domine et al., 2002) but in insufficient amounts to be sampled separately. To try to determine the chemistry of surface hoar, hoar frost that was growing on antenna guy wires was sampled, and showed a Cl^- content of $11.9 \mu\text{M}$ on 16 February and of $13.3 \mu\text{M}$ on 20 February, confirming the dilution suggestion. Until 20 February, Cl^- in the 3 February layer shows slight variations without a clear trend, ascribed, as in the case of its physical properties, to the spatially variable wind action during deposition (Domine et al., 2002). Br^- decreases slightly in the 7 February layer, while its decrease is dramatic in the 3 February layer, as it falls below detection limit on 20 February. A possible interpretation is that Br^- is depleted by dark reactions that activate bromide into Br_2 and BrCl , as detected by Foster et al. (2001) with an instrument located near our sampling site. The decrease is less in the 7 February layer because Br^- can be dry-deposited and because surface hoar contains 110 nM of Br^- , as suggested by hoar frost analyses.

As shown in Fig. 6b the Cl^-/Br^- ratio is fairly stable around 100 in the 7 February layer until 20 February, showing that Br^- is enriched relative to sea salt by a factor of 5 to 8. Regarding the 3 February layer, the ratio on 9 February is similar to that of

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the upper layer, but then rises rapidly to the sea water value on 15 February. On 20 February, the ratio clearly shows a large Br^- depletion relative to sea water, but we cannot quantify it.

On 22 February, a wind storm took place, with wind speed measured at the nearby GAW laboratory reaching a maximum of 7 m/s. This mixed both layers and accumulated them in wind-sheltered spots, with the probable addition of a few loose crystals from the windpack. Both Cl^- and Br^- concentrations increased dramatically. Ianniello et al. (2002) analyzed aerosols and gases at Alert between 9 and 21 February 2000, with a 48-h time resolution. They report mean HCl and HBr atmospheric concentrations of 11.3 and 16.7 $\text{ng}\cdot\text{m}^{-3}$, while Cl^- and Br^- concentration from aerosols were 73.9 and 5.2 $\text{ng}\cdot\text{m}^{-3}$. However, these values show much scatter with time, and to estimate a value for 22 February, we prefer to consider the value measured for the period from 19 to 21 February, which are undetectable for HCl, 23 $\text{ng}\cdot\text{m}^{-3}$ for HBr, and 31.8 and 10.5 $\text{ng}\cdot\text{m}^{-3}$ for Cl^- and Br^- . Both aerosol deposition and the scavenging of HCl and HBr may have contributed to the increases in snow concentrations. Judging from the mass of snow present on the ground before and after the wind storm, sublimation by wind transport cannot explain the increase. In any case, this wind storm proves that large amounts of Cl^- and Br^- can rapidly be deposited to snow by wind. Enrichment by windpack crystals can be safely ruled out, as the concentrations there were lower than in both February layers, and Br^- levels in particular were undetectable. Finally, while $(\text{Cl}^- + \text{HCl})/(\text{Br}^- + \text{HBr})$ in the atmosphere was 8.8 on a molar basis, the wind-blown snow took up about 50 μM of Cl^- and only about 0.3 μM of Br^- , indicating that Cl^- was transferred to the snow much more efficiently than Br^- . A quantitative comparison of ion uptake with other wind storms will be done after the description of those observed at Ny-Ålesund.

3.2.2. 27 and 29 April 2001 layer, Ny-Ålesund

The 27 April snow fall started around 7h00 until 11h00, while the air temperature (at 2.7 m height) was between -1 and -2°C (Fig. 7). It consisted of rimed dendritic crys-

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5 tals. Snow showers then occurred in the afternoon and evening, with snow crystals having various shapes: plates, dendritic crystals, and needles in the evening. The total thickness of the snowfall was about 3 cm (Fig. 8). The air temperature then rose to 0.5°C in the evening of both 27 and 28 under overcast conditions, initiating melting at the top of the layer. This produced a superficial melt-freeze crust about 5 mm thick, that was still permeable and never resembled an ice layer. No percolation was observed in the underlying part of the layer. This snow layer was covered on 29 April by a new snowfall, 4.5 cm thick, made up of small irregular crystals, that fell under an air temperatures between -1 and -3°C, between 6h00 and 23h00. A wind crust progressively formed on the 29 April layer, that reached a thickness of 1 cm on 4 May (Fig. 9). This new snowfall and the crust certainly hindered subsequent exchanges between the atmosphere and the 27 April layer.

15 At Ny-Ålesund, snow falls were very frequent, making conditions quite different from those at Alert. Solid precipitations were observed on 18, 20, 23 and 25 April, producing snow layers a few cm thick at the most. Also, while Alert is generally quite calm, there is always wind at Ny-Ålesund. As a result, some snow is frequently raised by wind, some layers are eroded and several different layers are exposed. A newly deposited layer can thus easily be contaminated by wind-raised snow, originating from different layers of potentially very different chemical composition. The snow surface is very irregular, with sastrugi about 5 to 20 cm high, of wavelength between 2 and 10 m, that produce irregular wind pumping and therefore variable dry deposition of atmospheric species, whose snow concentrations are bound to be spatially very variable. The result of these conditions is that snow composition was found to be spatially heterogeneous, and triplicate samples often showed a large variability. The analytical procedure was essentially the same as at Alert, and thus cannot be the cause of the moderate to poor analytical reproducibility. We attempted to minimize the effect of those spatial variations by taking triplicate samples as close as possible to each other, and by doing subsequent sampling close to previous ones.

25 The 29 April snow layer was sampled at two levels: at the very surface (top 1 cm) and

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about 3 cm below the surface. Figure 8 shows that initially, the 29 April snowfall was very poor in Cl^- : $0.5 \mu\text{M}$ for the snow sampled at 13h00, when accumulation was about 2 cm. Precipitating snow was also sampled at 18h30, when accumulation was about 4 cm, and had a very low Cl^- content: $1.9 \mu\text{M}$. Subsequently, the Cl^- content of the middle part of the layer remained essentially stable around $2 \mu\text{M}$, with one exception on 29 April when contamination by the underlying layer may have taken place, as suggested by the large standard deviation. On the contrary, the Cl^- content of the top part of the layer, which was developing a wind crust, increased to $70 \mu\text{M}$ on 30 April and remained stable around that concentration.

On 30 April, wind was moderate, between 3 and 4.5 m/s for a large fraction of the day, and did not raise snow. This is when the Cl^- concentration increased rapidly to $70 \mu\text{M}$ in the top sublayer, suggesting that dry deposition by wind-pumping was operative. After that windy spell, conditions were calm, with wind speed staying essentially below 2 m/s until the last sampling on 2 May. The stability of the ionic concentrations in the top crust can be due to the low wind speed, or may also be attributed to the formation of the wind crust, with a much reduced permeability. The Cl^-/Na^+ ratio stayed stable in the top part, around 1.5 to 1.6 with error bars around 0.3. For the middle sublayers, error bars were around 1 or greater, and data are not meaningful. We then suggest that the increase in Cl^- in the top sublayer is due to the deposition of moderately depleted sea salt, with no detectable deposition of HCl.

The 27 April layers offers another excellent opportunity to evaluate the effect of dry deposition due to wind pumping. The storm of 8 May lasted 15 h (Fig. 7) with wind speeds reaching 12.5 m/s, that produced a strong erosion of the snow pack. The top layer was the wind crust of the 29 April layer, that was partly eroded (Fig. 9). The unprotected soft snow from the 29 April layer became also quickly eroded, thus exposing the 27 April melt/freeze crust in many places, but not everywhere. Figure 9 and Table 3 show that before the storm, the Cl^- concentration of the 27 April crust remained essentially constant, around $40 \mu\text{M}$, while its Na^+ content was around $35 \mu\text{M}$. On 8 May at 23h00, the 27 April crust was sampled, both in exposed parts and in parts where it

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was still sheltered by the 29 April layer. Figure 9 clearly shows that the ion concentrations of the unexposed portion was not affected by the wind storm. On the contrary, the exposed portion saw its chloride content jump to $450 \mu\text{M}$.

The error on the Cl^-/Na^+ ratio is large, because the error on each concentrations, taken as the standard deviation in triplicate samples, is large. Nevertheless, Figure 10 shows that the Cl^-/Na^+ ratio remains constant around 1.165, the sea water value. We thus conclude from this case study that (i) the Cl^- and Na^+ in the 27 April melt/freeze crust were probably unfractionated sea salt, and not scavenged HCl. Likewise, after exposure on 8 May, Cl^- and Na^+ were again present as sea salt. (ii) Dry deposition, enhanced by wind pumping, can deposit enormous quantities of sea salt onto surface snow in just 15 h. (iii) A few cm of snow are sufficient to shield underlying layers from sea salt deposition by wind pumping, at least with the surface snow structure that was observed. The depth of penetration of wind-pumped sea salt into snow then appears to be here only about 2 cm. A similar depth can be derived from our study of the 29 April snow layer. Of course, we realize that this may vary with the physical properties of snow, and in particular with its permeability (Albert and Schultz, 2002).

3.2.3. Efficiency of ion uptake

From the 3 time series discussed above, we now consider quantitatively the deposition of ions to snow by wind storms. The data detailed above indicate that, during wind storms, deposition of aerosols is more efficient than the uptake of gases. To a first approximation, we then expect the amount of ion deposition, Dep , to be proportional to wind speed, V , to the concentration of ions in the atmosphere in an aerosol form, C , and to the duration of the wind storm, t , so that:

$$Dep = AV Ct \quad (1)$$

where A is the proportionality factor. To test Eq. (1), Dep for Cl^- has been plotted as a function of the product VCt in Fig. 11. According to Teinilä et al. (2003), the Cl^- atmospheric concentration during the 30 April storm was 276 ng.m^{-3} , and it was

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649 ng.m⁻³ during the 8 May storm. For the 30 April storm, we use a mean wind speed of 3 m/s during 0.7 day, and a mean wind speed of 10 m/s during 0.5 day for the 8 May storm. We use a Cl⁻ value of 32 ng for the Alert storm, as detailed above, and a wind speed of 4 m/s during 0.35 day.

5 Figure 11 shows that Eq. (1) seems to hold when only the Ny-Ålesund data is considered: the proportionally factors for both events are within 10% of each other. However, it is 9 times higher for the Alert event. We suggest that this is because during the Alert event, the snow was wind-blown, resulting in a maximal exposure of the snow to atmospheric aerosols. At Ny-Ålesund, on the contrary, aerosol had to penetrate through
10 a wind crust or a melt freeze crust, and this extra resistance certainly limited aerosol penetration. We come to the conclusion that at least one factor is missing in Eq. (1), that should include the efficiency of air circulation in the snow. Obviously, it is maximal when the snow is airborne. For snow layers on the ground, we suggest testing the hypothesis that the inclusion of snow permeability in Eq. (1) would improve its applicability. Perhaps the snow SSA should also be included, as this may impact the efficiency
15 of capture of aerosol particles.

4. Conclusions

The objective of this paper was to test the importance of two processes potentially contributing to the ionic content of snow on sea ice: upward migration from the sea ice, and deposition by wind. Wolff et al. (2003) and Rankin et al. (2002) hypothesized
20 from Antarctic observations that wind-deposited ions may come essentially from frost flowers.

Our observations of Figs. 2 and 5 show that upward migration can indeed take place and be observed even in winter snow at temperatures near -30°C, and affect the snow-pack up to at least 17 cm from the ice surface, where a Cl⁻ concentration of 300 μM
25 was observed. The data of Fig. 2 and Table 3 show that sea salt in this case migrated upward without detectable fractionation. Domine et al. (2002) mentioned that snow

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thickness on Arctic sea ice near Alert was very variable, between 0 and 50 cm. Eicken et al. (1994) showed that snow thickness was very variable on the Weddell sea (Antarctica), with maximal frequencies around 5–25 cm. We then conclude that migration of ions from sea ice indeed has the potential to affect a significant fraction of the surface snow layers on sea ice. The sea salt reaching the surface of the snowpack can then be transported by wind and scattered over the entire marine snowpack.

However, in the four case studies of Figs. 2 to 5, we could never come to the conclusion that most of the ions in surface snow came from upward migration from sea ice. We believe that more numerous case studies are needed. Also, our selection of sampling site was biased towards thicker snow banks, subjectively viewed as more interesting. Furthermore, dry deposition of SO_4^{2-} and Ca^{2+} , ubiquitous in the Arctic troposphere, can rapidly obscure any sea salt chemical signature and make the detection of upward migration in surface layers difficult. We suggest that time series of vertical profiles on sea ice, over several weeks to months, would shed additional light on the actual impact of upward migration on surface snow composition.

The impact of wind deposition on snow composition was tested on land, to avoid interference from sea ice. We conclude that wind can rapidly increase ionic concentrations in snow, especially if the snow becomes airborne. Wind deposition was observed to lead to Cl^- concentrations of $450 \mu\text{M}$ at Ny-Ålesund, similar to those produced by upward migration on sea ice, 17 cm up the snow. This process was found to affect only the top 2 cm of the snowpack, but wind or melt/freeze crusts were always present, and this may have limited the depth of penetration of wind-borne aerosols. In the Ny-Ålesund case, the deposited ions had clearly a sea salt signature without fractionation. If future measurements confirm that frost flowers are always fractionated, then we conclude that frost flowers were not the source of these ions and are thus not necessary to produce very large ionic concentrations in snow. Since the sea was essentially ice free around Ny-Ålesund, we observe, in contrast to observations made in Antarctica by Wolff et al. (2003) and Rankin et al. (2002) that the ice-free ocean can also be an efficient contributor of sea salt to the continental snowpack. The impact of this conclusion

should be minimized, however, because our sampling site was right on the coast, and we believe that the size distribution of sea salt aerosols generated by open water and by frost flowers should be compared to evaluate the distance over which they can be transported.

5 To sum up our conclusions on the origin of sea salt in Arctic snow, we find, in contrast to Wolff et al. (2003) and Rankin et al. (2002) that upward migration from the sea ice and transport by wind from the open sea can significantly contribute to the sea salt loading of snow. Of course, the question of the spatial and temporal representativity of our few measurements remain. But it is clear that different processes may dominate
10 in the Arctic and the Antarctic. The Antarctic seas are warmer than the Arctic ocean, the ice is mostly less than one year old, and open leads are more likely to open and produce frost flowers. Furthermore, the Halley Bay and Neumayer sites are close to frequent open waters, and may be particularly subjected to frost flowers influence. In any case, more measurement campaigns over long durations and at many different
15 types of sites will have to be performed before the sources of sea salt to polar snow can be assessed.

Regarding the Br^- content of snow, our data from Alert and the northern Arctic Ocean suggest that ozone depletion chemistry can significantly enhance the Br^- content of snow. This suggests a mechanism to transport Br^- over continents, and propagate bromide-driven ozone destruction over land.
20

Despite this enhancement caused by essentially marine chemistry, the highest Br^- concentration observed was on land at Ny-Ålesund, in the wind-exposed 27 April layer, where its concentration reached $0.62 \mu\text{M}$. As mentioned above, this Br^- definitely came from unfractionated sea salt, implying that frost flowers are not necessary to produce
25 large Br^- concentrations in snow.

Our discussions of ion migration up the snow pack and of wind deposition of ions indicate that physical studies of the snow are necessary to quantify ion transfer to snow. In particular, we suggest that measurements of the permeability and specific surface area are indispensable in future studies.

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Table 1. Operating conditions of the IC analyses during all campaigns.

	IC DX 100 cation analysis	IC DX 120 anion analysis
Column	CS12A (4×250 mm)	AS12A (4×200 mm)
Pre-column	CG12A (4×50 mm)	AG12A (4×50 mm)
Suppressor	CSRS-II (4 mm)	ASRS-II (4 mm)
Operative Flow Rate	1 ml/min	1.5 ml/min
Loop	150 μ l	300 μ l
Elution procedure	Isocratic	isocratic
Dionex Eluent	Methanesulphonic acid, MSA 20 mM	Buffer solution of Na ₂ CO ₃ 2.1 mM, NaHCO ₃ 0.8 mM
Analyte species	Na ⁺ , K ⁺ , Ca ²⁺ , Mg ²⁺ , NH ₄ ⁺	SO ₄ ²⁻ , NO ₃ ⁻ , Cl ⁻ , NO ₂ ⁻ , Br ⁻
Detector	Conductivity cell	Conductivity cell

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Table 2. Laboratory precision and detection limits during all campaigns.

Measured species by IC	Precision ¹ [%]	3 σ L.O.D. [ppb]
Cl ⁻	3.33	2
NO ₂ ⁻	3.29	0.6
Br ⁻	1.40	0.8
NO ₃ ⁻	1.36	0.6
SO ₄ ²⁻	2.18	2
Na ⁺	2.99	1
NH ₄ ⁺	3.63	0.3
K ⁺	2.29	0.3
Mg ²⁺	2.46	2
Ca ²⁺	1.42	10

¹ The laboratory precision is estimated on the basis of repeated analyzed carried out at a concentration level of 100 ng/ml typical of samples collected in polar sites.

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Table 3. Ionic composition of snow samples, in units of μM ; bdl: below detection limit; na: not available.

Layer or date	Cl ⁻	Br ⁻	NO ₃ ⁻	SO ₄ ⁻	Na ⁺	NH ₄ ⁺	K ⁺	Mg ⁺⁺	Ca ⁺⁺	Salinity,psu
Vertical profiles										
Alert, February 2000										
7 February Fall	49.8	0.39	4.8	5.6	33.5	1.2	1.2	7.9	36.4	0.0051
Upper windpack	40.0	bdl	3.6	3.4	29.9	0.6	1.0	5.3	12.7	0.0033
Depth hoar	30.2	bdl	2.6	3.0	16.8	0.8	0.4	2.4	7.9	0.0023
Lower windpack	294.8	0.47	1.2	9.5	235.1	0.0	5.7	29.1	55.2	0.0200
Arctic Ocean, 85°N										
Surface hoar	4.0	0.16	9.3	1.7	1.5	0.5	0.2	0.5	2.2	0.0010
Crust	356.1	bdl	2.9	6.2	202.5	1.9	9.0	49.0	8.4	0.0200
Windpack	65.0	bdl	1.0	3.1	31.5	0.5	1.0	3.8	1.3	0.0036
Depth hoar	2.7	0.08	1.1	0.8	1.4	0.2	0.2	0.3	0.3	0.0003
Alert, April 2000										
Recent snow	15.5	0.11	9.3	3.2	6.0	0.7	0.4	2.5	5.8	0.0019
Windpack, top	36.2	0.07	4.5	2.8	18.8	0.6	1.5	8.3	5.7	0.0028
Windpack, bott.	39.0	0.06	4.5	2.6	17.8	0.6	1.4	8.5	5.4	0.0028
Depth hoar	34.7	0.08	5.0	2.4	10.5	0.7	1.1	5.8	2.7	0.0023
Spitsbergen, May 2001										
Surface crust	62.3	0.17	6.8	18.6	160.2	3.0	3.8	20.9	8.4	0.0092
Decomp. grains	3.1	bdl	3.7	2.0	3.3	1.0	0.1	0.5	0.6	0.0007
Round grains, top	82.2	0.29	4.7	13.0	75.9	3.9	2.4	12.5	4.8	0.0069
Round grains, bot.	728.7	bdl	0.0	24.6	102.9	0.0	1.5	7.9	1.6	0.0309
Wet snow	110757.0	bdl	0.0	3157.7	13194.7	0.0	0.4	1.4	0.3	4.5334
Time series										
Alert, February 2000										
09/2/2000	19.6	0.23	2.2	3.5	na	na	na	na	na	na
10/2/2000	19.5	0.24	2.1	3.7	11.5	1.2	0.8	3.8	5.3	0.0018
15/2/2000	17.5	0.22	3.0	5.9	11.2	2.0	0.6	2.9	5.6	0.0020
20/2/2000	15.7	0.13	3.0	6.2	9.7	2.0	0.5	2.7	7.0	0.0020
22/2/2000	66.4	0.38	1.8	3.3	na	na	na	na	na	na
Alert, February 2000										
09/2/2000	13.9	0.16	4.5	2.8	na	na	na	na	na	na
10/2/2000	15.5	0.10	2.6	1.5	6.6	0.7	0.5	2.3	3.4	0.0012
15/2/2000	18.0	0.03	0.7	2.0	8.6	0.3	0.3	1.7	2.2	0.0012
20/2/2000	13.2	bdl	1.0	2.4	9.2	0.5	0.4	2.0	3.1	0.0012
22/2/2000	66.4	0.38	1.8	3.3	na	na	na	na	na	na
Ny-Alesund Spring 2001, 27 April layer, sheltered										
27/4/2001	48.0	0.11	8.4	11.2	39.4	10.2	1.0	5.0	3.9	0.0047
02/5/2001	63.7	0.20	4.2	12.2	61.6	6.4	2.3	12.6	5.9	0.0059
04/5/2001	38.2	0.16	9.4	12.6	32.3	12.1	0.9	4.5	3.6	0.0044
07/5/2001	33.9	bdl	7.5	7.0	30.7	3.1	0.9	4.4	3.8	0.0034
08/5/2001	44.8	bdl	10.4	9.0	37.3	4.6	0.9	5.1	4.6	0.0044
Ny-Alesund Spring 2001, 27 April layer, exposed										
08/5/2001	449.4	0.62	10.0	31.7	378.2	4.5	8.1	44.4	15.9	0.0305
09/5/2001	358.4	0.53	9.8	27.6	300.1	4.6	6.9	37.4	14.6	0.0248

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Table 3. Continued.

Layer or date	Cl ⁻	Br ⁻	NO ₃ ⁻	SO ₄ ⁻	Na ⁺	NH ₄ ⁺	K ⁺	Mg ⁺⁺	Ca ⁺⁺	Salinity.psu
Ny-Alesund Spring 2001, 29 April layer, middle										
29/4/01 13:00	0.5	bdl	2.3	0.3	0.7	0.3	0.1	0.1	0.2	0.0002
29/4/01 18:30	4.7	bdl	3.5	1.2	4.1	0.8	0.2	0.6	0.6	0.0006
30/4/01 10:30	1.4	bdl	1.9	0.5	1.6	0.4	0.2	0.2	0.2	0.0003
30/4/01 22:30	2.5	bdl	2.3	1.3	3.2	0.6	0.5	0.4	0.4	0.0005
01/5/01 10:45	1.8	bdl	2.2	1.1	1.8	0.3	0.0	0.2	0.2	0.0004
01/5/01 21:00	1.8	bdl	2.4	1.6	1.8	0.3	0.1	0.2	0.2	0.0004
02/5/01 14:30	0.7	bdl	2.4	0.8	0.8	0.4	0.0	0.1	0.1	0.0003
Ny-Alesund Spring 2001, 29 April layer, top										
29/4/01 18:30	1.9	bdl	1.8	0.7	2.0	0.6	0.2	0.3	0.5	0.0003
30/4/01 10:30	25.5	0.10	2.6	3.5	23.5	1.3	0.6	2.6	1.0	0.0021
30/4/01 14:00	36.7	0.26	3.0	4.2	33.5	1.6	0.9	3.7	1.4	0.0029
30/4/01 15:00	72.5	0.22	2.9	6.9	72.6	2.8	1.7	8.5	2.2	0.0055
30/4/01 22:30	64.2	0.15	2.7	6.7	62.6	2.2	1.5	6.9	1.9	0.0049
01/5/01 10:45	62.5	0.17	3.0	6.7	60.2	2.3	1.3	6.7	1.7	0.0048
01/5/01 21:00	70.1	0.19	3.1	8.3	70.5	2.7	1.5	8.2	2.2	0.0055
02/5/01 14:30	64.9	0.15	3.2	7.6	63.3	2.6	1.3	7.3	1.8	0.0050

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Table 4. Molar ionic composition of snow samples, normalized to Na⁺. Sea water composition is shown for comparison.

Layer or date	Cl ⁻	Br ⁻	SO ₄ ²⁻	Na ⁺	K ⁺	Mg ²⁺	Ca ²⁺
Vertical profiles							
Sea water	1.165	0.0018	0.0597	1	0.0218	0.1133	0.0219
Alert, February 2000							
7 February Fall	1.487	0.0118	0.1673	1	0.0370	0.2376	1.0884
Upper windpack	1.339	bdl	0.1144	1	0.0318	0.1786	0.4245
Depth hoar	1.795	bdl	0.1771	1	0.0247	0.1443	0.4716
Lower windpack	1.254	0.0020	0.0405	1	0.0241	0.1238	0.2349
Arctic Ocean, 85°N							
Surface hoar	2.749	0.1116	1.1373	1	0.1388	0.3584	1.4704
Crust	1.758	bdl	0.0304	1	0.0444	0.2421	0.0417
Windpack	2.062	bdl	0.0989	1	0.0303	0.1195	0.0398
Depth hoar	1.905	0.0589	0.5453	1	0.1302	0.1792	0.2404
Alert, April 2000							
Recent snow	2.557	0.0186	0.5261	1	0.0602	0.4177	0.9612
Windpack, top	1.922	0.0037	0.1511	1	0.0777	0.4424	0.3004
Windpack, bott.	2.199	0.0036	0.1475	1	0.0801	0.4770	0.3061
Depth hoar	3.287	0.0076	0.2247	1	0.1001	0.5484	0.2591
Spitsbergen, May 2001							
Surface crust	0.389	0.0010	0.1162	1	0.0236	0.1305	0.0525
Decomp. grains	0.942	bdl	0.6031	1	0.0235	0.1387	0.1797
Round grains, top	1.084	0.0038	0.1710	1	0.0312	0.1641	0.0638
Round grains, bot.	7.078	bdl	0.2393	1	0.0150	0.0766	0.0157
Wet snow	8.394	bdl	0.2393	1	0.0000	0.0001	0.0000
Time series							
Alert, February 2000							
09/2/2000	na	na	na	na	na	na	na
10/2/2000	1.697	0.0207	0.3238	1	0.0711	0.3279	0.4621
15/2/2000	1.570	0.0199	0.5299	1	0.0547	0.2552	0.4978
20/2/2000	1.628	0.0134	0.6381	1	0.0496	0.2766	0.7223
22/2/2000	na	na	na	na	na	na	na
Alert, February 2000							
09/2/2000	na	na	na	na	na	na	na
10/2/2000	2.364	0.0150	0.2266	1	0.0721	0.3453	0.5234
15/2/2000	2.093	0.0036	0.2283	1	0.0345	0.1990	0.2601
20/2/2000	1.444	bdl	0.2642	1	0.0397	0.2138	0.3370
22/2/2000	na	na	na	na	na	na	na
Ny-Alesund Spring 2001, 27 April layer, sheltered							
27/4/2001	1.219	0.0029	0.2840	1	0.0245	0.1261	0.0985
02/5/2001	1.035	0.0032	0.1986	1	0.0379	0.2054	0.0966
04/5/2001	1.183	0.0050	0.3883	1	0.0264	0.1388	0.1122
07/5/2001	1.103	bdl	0.2293	1	0.0280	0.1419	0.1232
08/5/2001	1.200	bdl	0.2419	1	0.0254	0.1367	0.1233
Ny-Alesund Spring 2001, 27 April layer, exposed							
08/5/2001	1.188	0.0016	0.0838	1	0.0213	0.1173	0.0420
09/5/2001	1.194	0.0018	0.0919	1	0.0230	0.1245	0.0486

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Table 4. Continued.

Layer or date	Cl ⁻	Br ⁻	SO ₄ ²⁻	Na ⁺	K ⁺	Mg ²⁺	Ca ²⁺
Ny-Ålesund Spring 2001, 29 April layer, middle							
29/4/2001 13:00	0.675	bdl	0.4640	1	0.1920	0.1158	0.3512
29/4/2001 18:30	1.126	bdl	0.2847	1	0.0536	0.1361	0.1449
30/4/2001 10:30	0.901	bdl	0.3368	1	0.0980	0.1226	0.1487
30/4/2001 22:30	0.765	bdl	0.4183	1	0.1695	0.1150	0.1189
01/5/2001 10:45	0.988	bdl	0.5964	1	0.0237	0.1068	0.0879
01/5/2001 21:00	0.993	bdl	0.8642	1	0.0280	0.1201	0.1275
02/5/2001 14:30	0.935	bdl	1.0125	1	0.0000	0.1455	0.1765
Ny-Ålesund Spring 2001, 29 April layer, top							
29/4/2001 18:30	0.963	-	0.3468	1	0.0895	0.1439	0.2328
30/4/2001 10:30	1.086	0.0044	0.1475	1	0.0243	0.1120	0.0418
30/4/2001 14:00	1.097	0.0078	0.1246	1	0.0275	0.1094	0.0410
30/4/2001 15:00	0.999	0.0030	0.0949	1	0.0228	0.1169	0.0305
30/4/2001 22:30	1.026	0.0025	0.1073	1	0.0240	0.1108	0.0300
01/5/2001 10:45	1.038	0.0028	0.1119	1	0.0208	0.1108	0.0282
01/5/2001 21:00	0.994	0.0027	0.1174	1	0.0212	0.1157	0.0316
02/5/2001 14:30	1.025	0.0024	0.1206	1	0.0209	0.1155	0.0283

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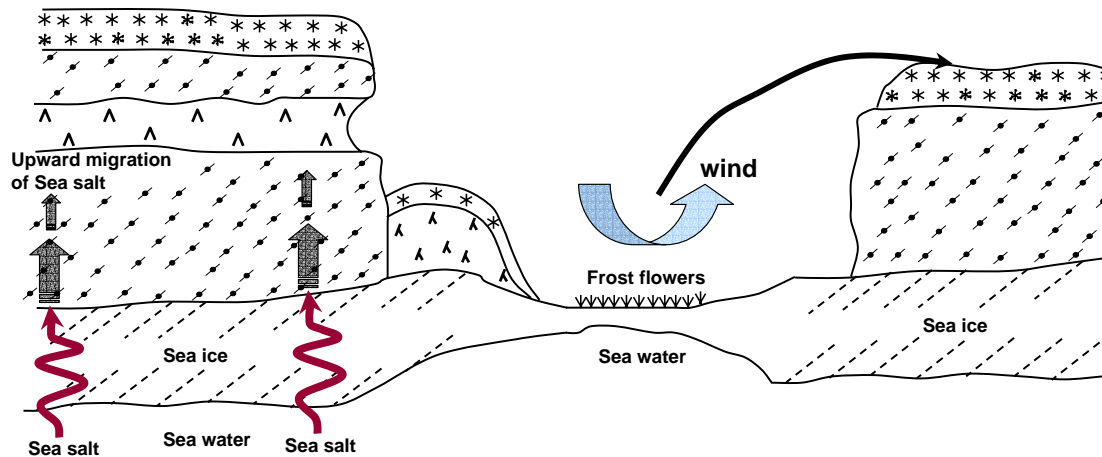


Fig. 1. Illustration of the main two processes suspected of supplying sea salt ions to snow: upward migration from sea ice, and wind-blown frost flowers.

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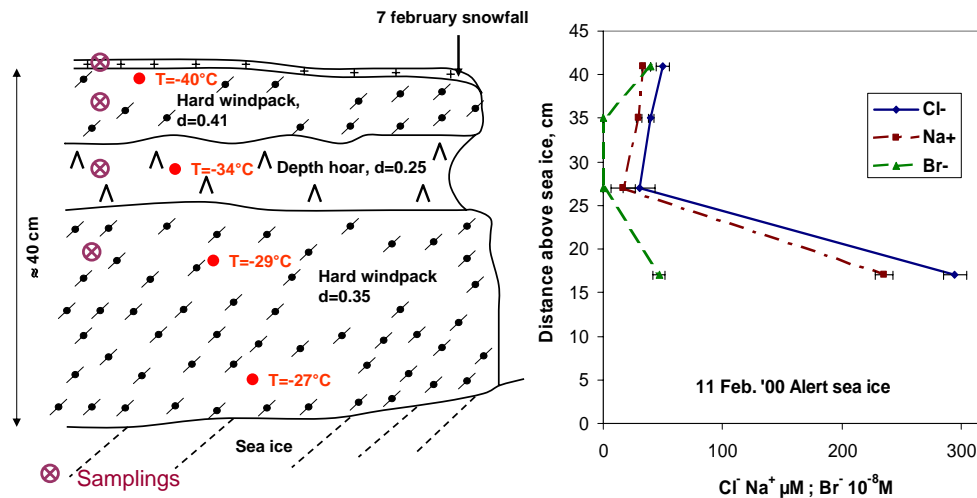


Fig. 2. Stratigraphy and concentrations of Cl^- , Na^+ and Br^- in a snow bank on sea ice near Alert, on 11 February 2000. Density (d) and temperature (T) values of the snow are indicated.

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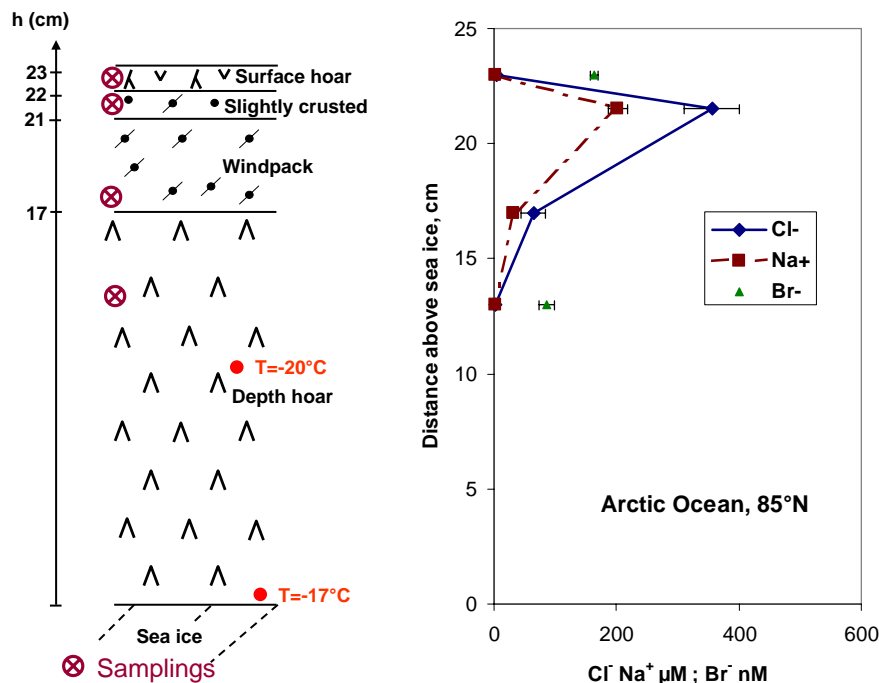


Fig. 3. Stratigraphy and concentrations of Cl⁻, Na⁺ and Br⁻ in a snow bank on the Northern Arctic Ocean (84°56' N, 67°28' W) on 25 April 2000.

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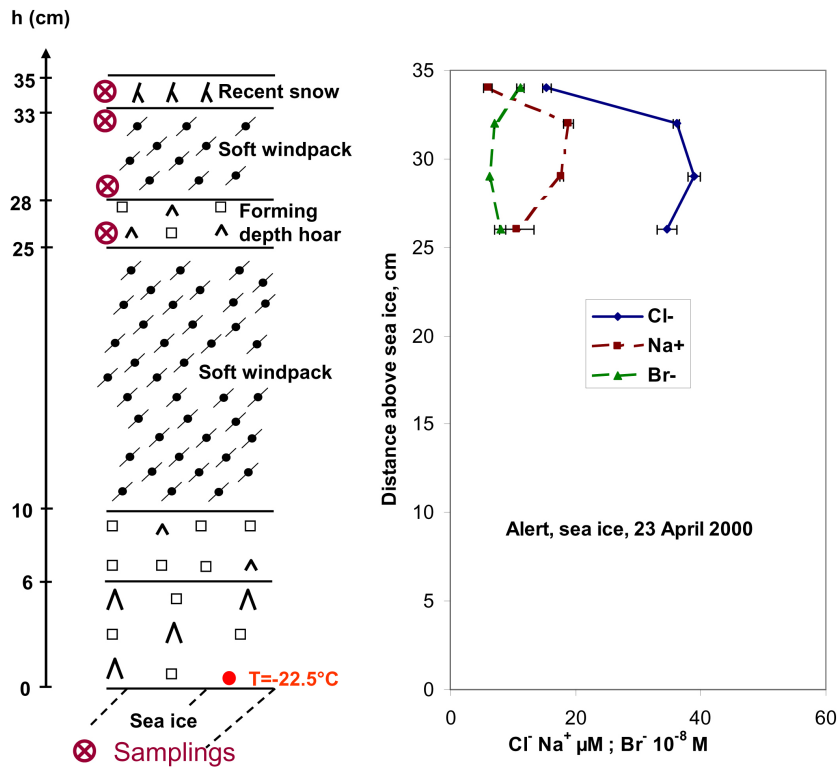


Fig. 4. Stratigraphy and concentrations of Cl^- , Na^+ and Br^- in a snow bank on sea ice near Alert, on 23 April 2000.

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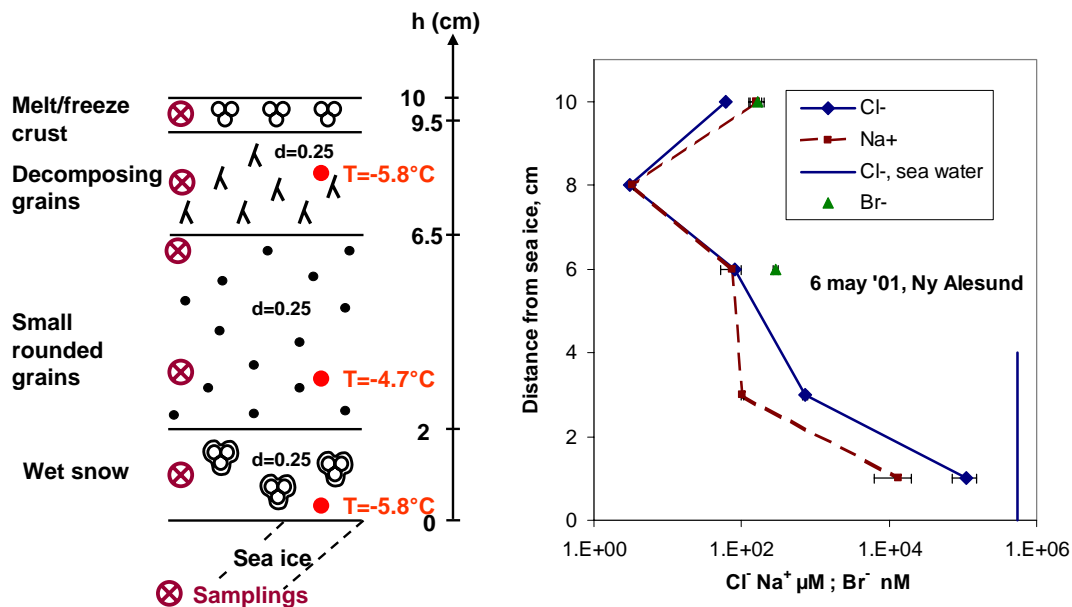


Fig. 5. Stratigraphy and concentrations of Cl⁻, Na⁺ and Br⁻ on sea ice near Ny-Ålesund, on 6 May 2001. Density (d) and temperature (T) values of the snow are indicated.

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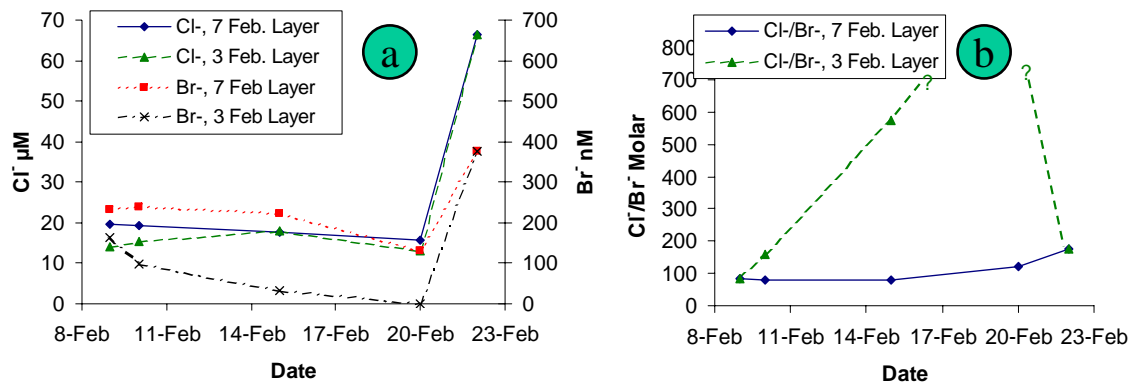


Fig. 6. (a) Time series of the Cl⁻ and Br⁻ concentrations of two snow layer on land, near Alert, in February 2000. Both snow layers were mixed together by a wind storm on 22 February, and were then sampled together. (b) Time series of the Cl⁻/Br⁻ ratio of these same layers.

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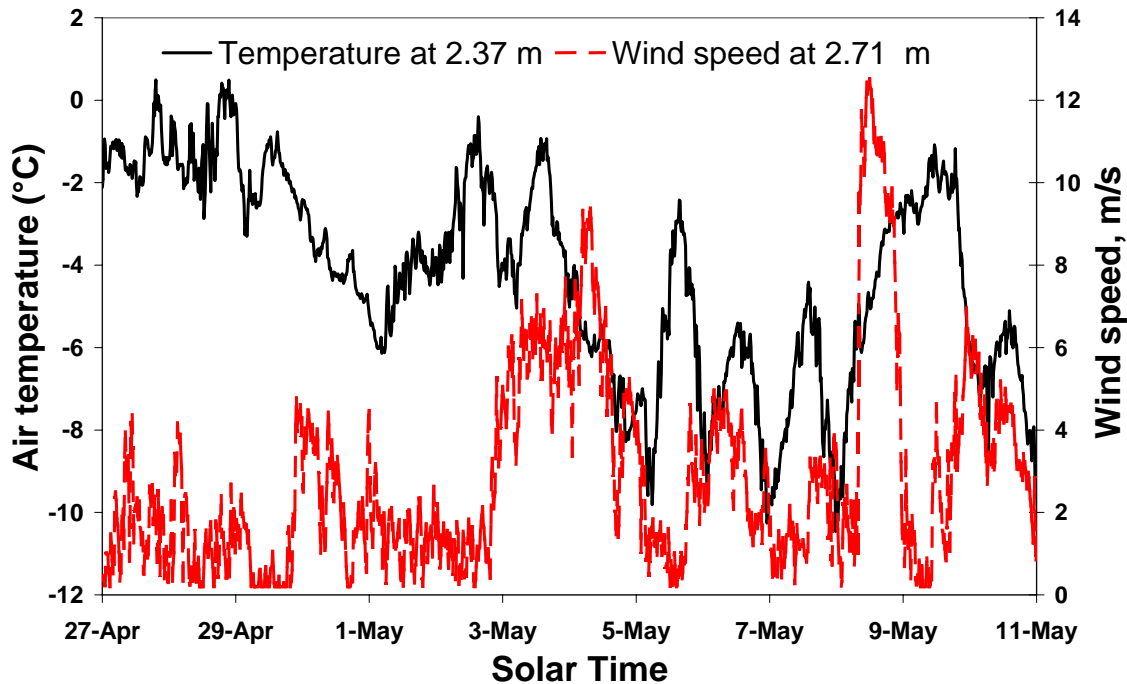


Fig. 7. Temperature and wind speed measured near Ny-Ålesund, within 50 m of our sampling site at the Amundsen mast, in spring 2001.

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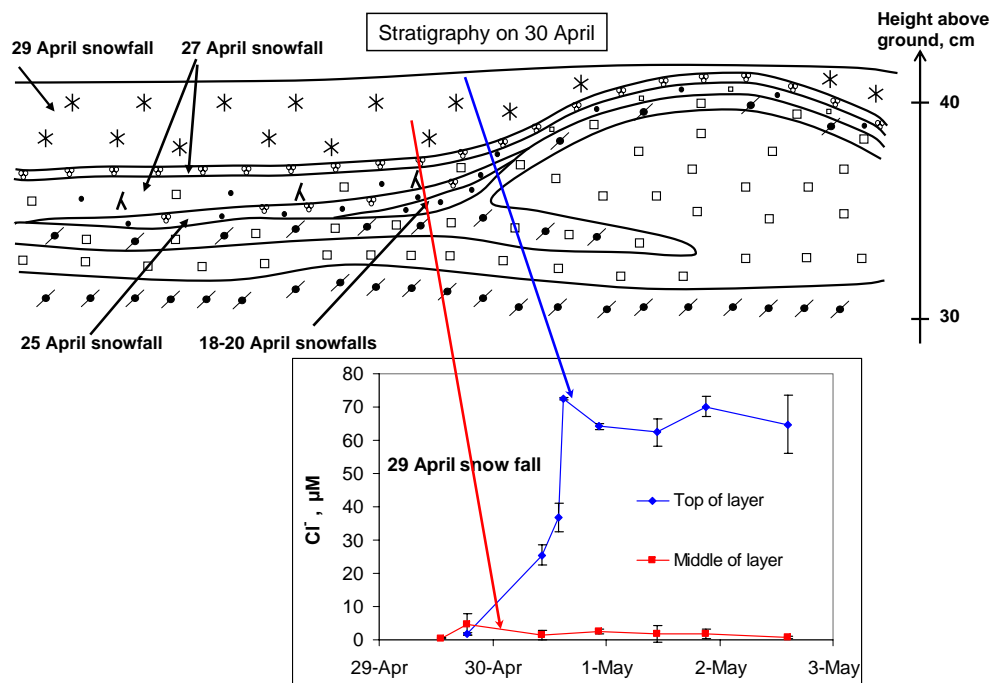


Fig. 8. Stratigraphy of the Ny-Ålesund snowpack on 30 April 2001, and time series of the Cl^- concentration in two levels of the 29 April layer.

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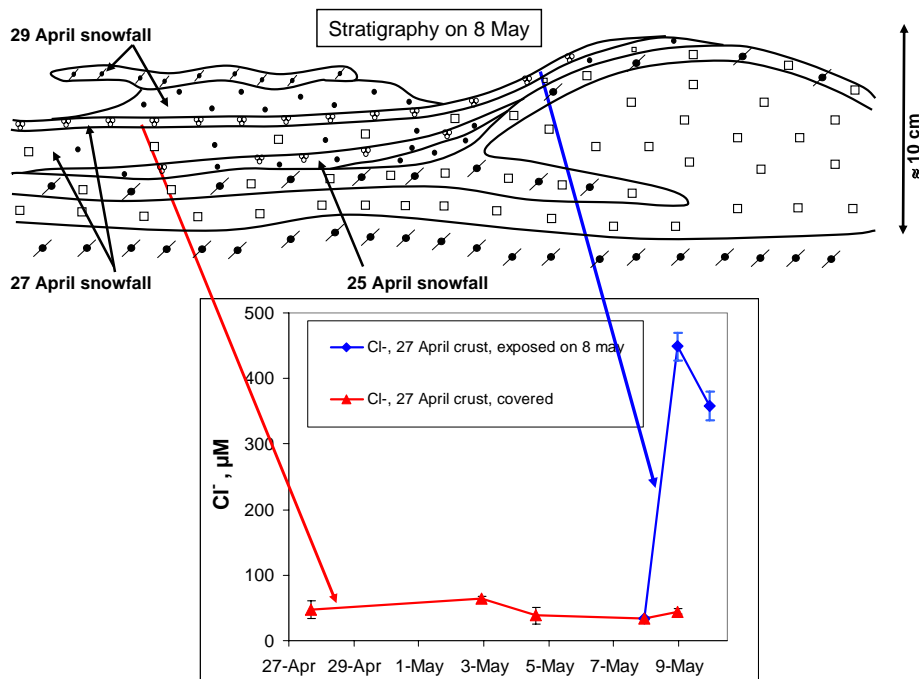


Fig. 9. Stratigraphy of the Ny-Ålesund snowpack on 8 May 2001, and time series of the Cl^- concentration in melt/freeze crust of the 27 April layer. On 8 May, a windstorm eroded the overlying 29 April layer, partially exposing the crust. The Cl^- concentration of the exposed crust is also shown starting on 8 May.

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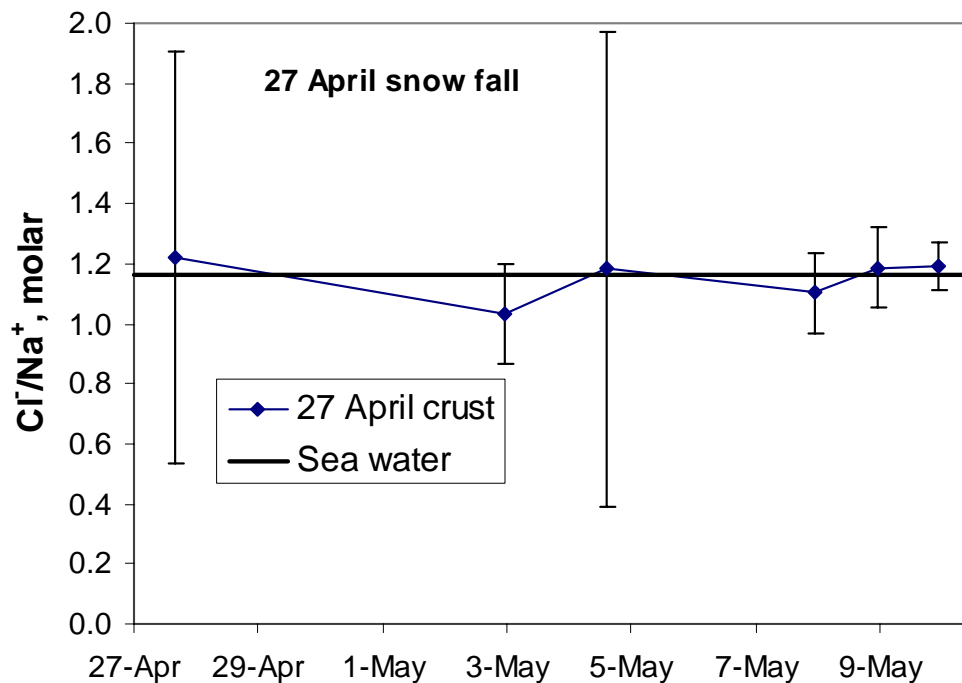


Fig. 10. Cl^-/Na^+ ratio of the melt/freeze crust of the 27 April snow layer. Starting on 8 May, values are those of the exposed crust (see Fig. 9).

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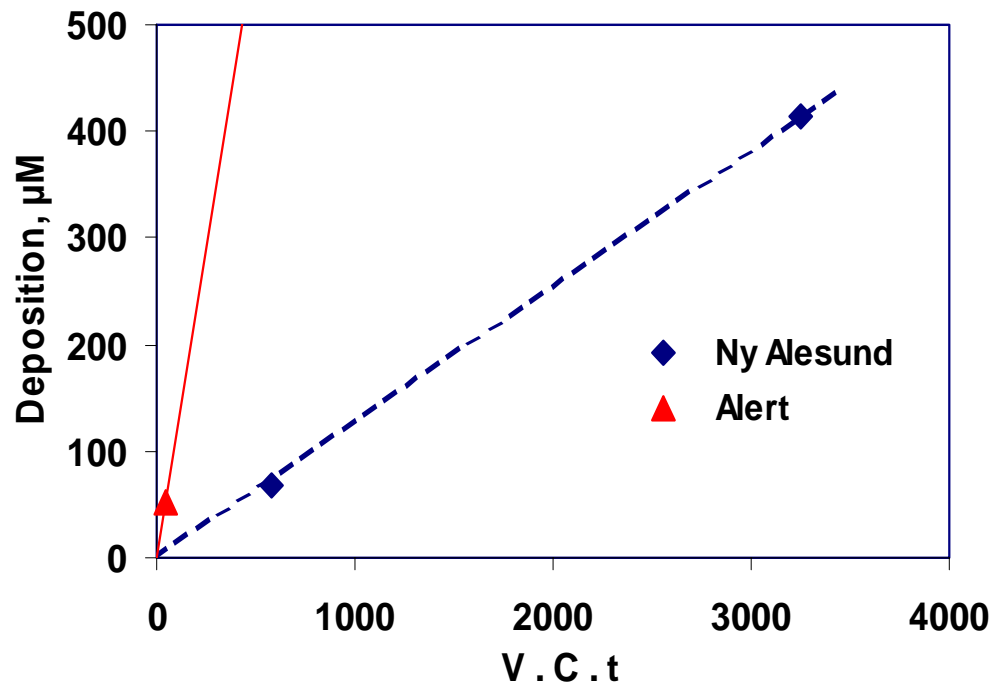


Fig. 11. Test of the validity of Eq. (1). The amount of Cl^- deposited by wind storms is plotted as a function of the product of wind speed, V , (m/s), atmospheric concentration of aerosol Cl^- , C , (ng m^{-3}) and duration of the wind storm, t (days). The dashed line shows the similarity of the proportionality factor A in Eq. (1) in both Ny-Ålesund case studies. The solid line shows that the proportionality factor A is much higher for the Alert case study, where snow became airborne.

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