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Atmospheric
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Interactive comment on "The origin of sea salt in snow on Arctic sea ice and in coastal regions" *by* F. Domine et al.

Anonymous Referee #0

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This paper uses measurements of ionic concentrations in snow on sea ice to attempt to determine the sources of high sea salt concentrations; the ultimate goal is to further understanding of the origin of bromide responsible for ozone depletion in the troposphere. This is certainly a useful aim, and the measurements here provide some interesting case studies that build up evidence for the situations that might occur. However, they are a few, rather-limited and unsystematic samplings. This is unavoidable, but the result is that all the authors can do is present their findings and then try and rationalise them with individual explanations. It is not at all clear how widely the results can be generalised: for example to the main body of the ice pack where most BrO/ODEs must be generated. At the end of it, we have not really got any closer to deriving rules that allow us to answer the question that was posed. Indeed, the question itself is posed in a rather false way, as discussed below. But basically, this is interesting information

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that can add to our knowledge; the authors do come to some conclusions about how far upwards migration might be effective in transporting salt, and they do have some intriguing evidence that ODEs might themselves lead to enhanced bromide concentrations in snow: these ideas warrant further investigation. In conclusion, I feel that the paper warrants publication, but at shorter length, and in less detail.

One aspect that should be corrected is the way the question is posed. The authors take as a starting point the proposal of Rankin et al that sea salt in coastal Antarctic air and snow derives mainly from the sea ice surface (with frost flowers as a major element of that). They then appear to imply that any finding of upward migration of salt to the snow-on-sea-ice surface would oppose this. (For example, near the end "in contrast to Wolff et al/Rankin et alĚupward migration may contribute"). But Wolff et al and Rankin et al ONLY addressed the origin of salt in snow and aerosol over land ice and ice shelves. They did not even discuss snow on sea ice, and nothing in their papers would deny a possible role for either mechanism (both of which incidentally would lead to fractionation of mirabilite if the temperature was cold enough). This paper, in contrast, only discusses snow on sea ice. Although the papers are clearly related, this one should not be set up in opposition to the other, but rather as an extension to the important question of snow on sea ice.

For halogen activation, the idea that has been discussed most in this area is that the frost flowers themselves (or perhaps the aerosol derived from them) are the source of the bromide (as well as the Rankin papers referred to, see also the recent paper by Kaleschke et al., Kaleschke, L., A. Richter, A.M. Rankin, J. Burrows, J. Hollwedel, H.K. Roscoe, O. Afe, T. Wagner, H.W. Jacobi, G. Heygster, and J. Notholt, Frost flowers on sea ice as a source of sea salt and their influence on tropospheric halogen chemistry, Geophysical Research Letters, 31 (16), L16114, doi: 10.1029/2004GL020655, 2004.). The current paper, in contrast, is considering whether snow on sea ice, with some bromide blown in from frost flowers, or lifted from the sea ice interface, is the place where the halogens come from. This is certainly worth looking at. But it should also be borne in mind that the attraction of frost flowers as a source is their high surface area combined with salinities of 100 psu; the snow described in this paper certainly has high surface area, but the highest salinities mentioned are more than 4 orders of magnitude lower than this. I think the authors should make a clearer distinction between the primary source (frost flowers or the brine on sea ice, at very high salinities), and this much more dilute secondary source.

Another issue on which this paper is rather confusing is that of fractionation. The main fractionation referred to in papers on frost flowers to date has been the loss of sulfate due to mirabilite deposition. Sodium is also lost in this process, but the relative proportion of sodium lost is small, so it is doubtful it could be used as a marker. The present paper refers frequently to fractionation without being clear what fractionation it is looking for. The issue of Br fractionation and the paper of Koop et al also needs to be handled carefully. As I recall, Koop's point was that eventually NaCl precipitates, leaving NaBr still in the liquid phase. Just as with the mirabilite issue, this can lead to a fractionation if the liquid phase can move while the solid one is fixed. In the ideas of Rankin et al, the liquid phase moves from the brine layer up the frost flowers, so that if the loss of mirabilite occurs in the brine layer (requiring the brine layer to be below -8 degrees), fractionation occurs. To get bromide fractionation by this mechanism requires that the brine below growing frost flowers is below -23 degrees: less likely to be true. Thus the issue of whether we expect Br enhancement is highly dependent on temperature and on whether a physical separation of the precipitated and liquid fractions can occur. This is not discussed clearly in this text.

However, there are some interesting indications and ideas in the text. The Alert measurements seem to show that upward movement occurs but is limited, and that windblown material is also important. The suggestion that ODEs can lead to enhancement of bromide in surface layers is intriguing, and deserves at least a quantitative calculation to see if it is realistic (i.e. how much ozone depletion is needed to give a significant enhancement of surface Br in snow?).

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On the other hand, the Ny Alesund data where analytical problems occurred are really not very helpful. (Indeed, the high temperatures at Ny Alesund make it impossible for fractionation to occur, and highly improbable that frost flowers occur: it is really not a good location for testing these hypotheses). The description of the times series data is extremely confusing and hard to follow. I would prefer to see these sections simplified and shortened.

Section 3.2.3 is not at all convincing. With only 3 datapoints nothing is really proved. During a wind storm I would expect the main process to be redistribution of snow (blowing snow), and the measurement of aerosol concentration becomes very difficult at such times. Although the conclusion might easily be correct, there is simply insufficient data to support it here. I would remove this section and figure.

The abstract is mainly fine, but I question the sentence “In the Arctic, frost flowers thus do not appear necessary to lead to large sea salt concentrationsĚ, and to supply the bromide needed for ODEs”. Firstly, I make the same point as earlier that the concentrations here are not at all large compared to those in frost flowers. But I am not sure the second half of the sentence is justified: as far as I can see you have presented no evidence that ODEs occurred in association with these sea salt concentrations at Ny Alesund.

Finally the paper is just too long for the limited conclusions. One obvious reduction would be in tables and figures: the relevant data all appear in tables and figures - the authors could choose one or the other. I don’t see the value of Figure 10 (result can simply be stated in the text) and, as stated above, I am not convinced by figure 11.

Technical notes and corrections: Page 4756, line 2. Note that some information on size distribution of fractionated aerosol has been presented in Rankin, A.M., and E.W. Wolff, A year-round record of size-segregated aerosol composition at Halley, Antarctica, Journal of Geophysical Research, 108 (24), 4775, doi:10.1029/2003JD003993, 2003. Table 3 and 4: Arctic Ocean, 85N, add date Table 3 and 4: Time series, Alert,

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put something in top line to say what is the difference between these two sets (I assume one is 3 Feb, one 7 Feb) Table 4 caption: clearer to say molar ionic ratios (referenced to Na) for snow samples. Figure 1, etc: is there any significance to the different symbols on the snow layers? Figs 2,3,5: spurious lines have appeared between the symbols for the samplings.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 4737, 2004.

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