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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Frost flowers and sea-salt aerosols over seasonal sea-ice areas in north-western Greenland during winter–spring" *by* Keiichiro Hara et al.

Anonymous Referee #3

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This manuscript describes measurements of frost flowers and sea salt aerosol particles during winter/spring 2014 on the coast of Greenland. The goal of simultaneous measurement of potential sea salt aerosol sources (e.g. frost flowers) and aerosol is commendable. However some potential sources of aerosol (e.g. surface snowpack) do not appear to have been measured, and the data presentation and argumentation in the manuscript is not very clear. The manuscript does show some results that appear significant. The enrichment of sea salts in frost flowers compared to sea water and depletion of sulfate in frost flowers compared to sea water ratios are both observed in agreement with past literature. However, the subsequent points of the abstract are not well argued for or are not clear.

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Discussion paper

Major points

The manuscript lacks an error analysis that would allow one to determine the significance of results. Words like "significant" are used, but that is not clearly related to a rigorous statistical definition or simply a qualitative word. To make statements about composition being different from sea water ratios, the authors would need to discuss error analysis more rigorously. For example, figures 4, 5, and 6 should all have error bars. Particularly in Figure 6, the lack of error bars makes it challenging to tell if deviations from the sea water composition is significant or not. The text seems to indicate that sea water was sampled, which could be used to assess random and systematic errors in analysis of these ionic species, but no such analysis is shown. This is particularly concerning for species that are analyzed by different methods (e.g. I- and Br- are analyzed by different methods than Cl-, and cations in different runs from Cl-).

The manuscript contains many technical errors and is difficult to read due to non-standard use of English. An example of this problem is on page 6, line 19, where the text says "Figure 3b presents a close up photograph of frost flowers...". Figure 3b is a photo of people on sea ice and houses on the shore. Many other examples of technical errors are in the text, another being page 15, line 18 indicates A, B, and C are on a diagram, but that diagram actually has A through D. The use of ternary plots is also not clear. Normally ternary plots are useful when the sum of the three components is 100%. However, in Figure 10, Cl, S, and Na are plotted as atomic percentages. These samples also have other atoms in their composition (e.g. oxygen that is a part of SO4, Mg, etc.), so I guess that the plots show atomic percentage of Cl, S, and Na atoms? Some points are then very strange, such as on Figure 10b, three course points have >80% Cl and little Na. Something must charge balance the Cl-, but that is not clear on the plot.

The paper often describes frost flowers as "fragile", while laboratory studies of frost flowers in a wind tunnel failed to produce aerosol, and field studies often show frost flowers get buried under blowing snow (e.g. snow blows while frost flowers remain

intact). Therefore, there is not clarity in the literature that frost flowers are the only source of sea salt aerosol, and to the contrary, blowing snow and/or aerosol production from open water are often discussed in the literature. This manuscript doesn't describe the chemical composition of snow, which could be relevant to the production of aerosol, nor does it consider nearby open water and potential of aerosol production from that source.

The manuscript claims in the abstract that "Aerosol number concentrations, particularly in coarse mode, were increased considerably by release from sea-ice surface under strong wind conditions." However, the figures and text really do not back up that claim. Figure 9 is presumably the data for this claim, but the authors do not indicate what periods to look at on that figure to see they effect they claim. In general, I see high coarse-mode aerosol on about DOY 12-18, 30, and 50-55. Those periods often have some winds, but not peak winds. Two points at peak wind periods (DOY 41 and 59) do show spikes in aerosol, but other high wind periods don't have spikes (e.g. DOY 36 and 38). Overall, the authors need to make a clearer argument to their claim.

The claim that bromide is being released from frost flowers made at the bottom of page 9 and page 10 would seem to imply a large release of bromine from frost flowers to the atmosphere. The authors should do a mass balance argument to indicate how much bromine would be released from this proposed release and compare the value to observations of atmospheric bromine (e.g. BrO). If that calculation led to unreasonably large BrO concentrations, then it would be evidence against this hypothesized direct halogen release. The lack of error analysis also makes it challenging to tell what is significant on these plots. Lastly, field evidence (Pratt et al., 2013) and multiple laboratory studies indicate that the pH of surfaces should be acidic for efficient halogen release, while highly saline samples were not efficient at releasing halogens.

The referencing of the paper is not accurate. An example of this is on line 3-4 of page 3, which the authors say "Reportedly, Br- enrichment occurs in frost flowers at Barrow, Alaska (Douglas et al., 2012)...". However, the text of that citation says "There is no

enhancement in bromide to chloride ratios in the frost flowers compared to brine or seawater".

Overall

This paper needs major revisions to be acceptable for publication. There is a great deal of interesting data in the publication, but it is not presented in a form that gains scientific understanding from the data.

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