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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Secondary Sulfate is Internally Mixed with Sea Spray Aerosol and Organic Aerosol in the Winter-Spring Arctic" *by* Rachel M. Kirpes et al.

Anonymous Referee #2

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The detailed particle analyses given in this paper offers important information about the nature of the Arctic aerosol during winter. Both the presence of sea salt associated with many particles and the demonstration of a potential Prudhoe Bay influence (Fig. 5) are interesting. The paper is well presented. Improvements can be made with some relatively simple additions/changes. Please consider the following comments.

1) January and February are winter months. There are no spring measurements given in the paper, and the title needs to be changed appropriately. There are other places in the paper where this work is mistakenly referred to as winter-spring.

2) While the authors have done a good job of representing previous work, there are

a few references that should be added. Particularly relevant to your paper is the recent work of Köllner et al. (https://doi.org/10.5194/acp-17-13747-2017) that discusses single particle measurements in the Arctic, including the distribution of sea salt particles relative to other components. In support of the discussion of aerosol influences on longwave radiation on page 2, you should include some reference to work by Tim Garrett (U. Utah).

3) In the abstract, please mention that there are seven days of sampling. As it stands now, the abstract implies there are two months of sampling. Also in the abstract, please add that the results are based on 24,847 particles analyzed by CCSEM-EDX, 300 by Raman microspectroscopy and 290 by STXM-NEXAFS.

4) On page 5, lines 23-24 you note that 290 particles from the Feb 26 night sample (0.10-0.18 um) were analysed by STXM-NEXAFS. Is that meant to indicate you analysed 290 particles in the 100-180 nm size range? In Fig. 4 you show an example of the STXM-NEXAFS analysis from that sample, which shows the analysis of particles much larger than 180 nm. Please clarify.

5) You state that 50-60% of the 0.1-0.5 um particles contain sea salt. Since a substantial fraction of the winter-spring Arctic aerosol number concentration resides in that size fraction, your result would indicate that about 50% of the winter Arctic number concentration is due to primary emissions of sea salt. As the other reviewer notes, that has strong implications for the modelling of the winter Arctic aerosol, and thus we need to know if your various analyses have the same efficiency for smaller particles as they do for larger particles. Are smaller particles more difficult to analyse, or is the fraction of analysed particles to sampled particles in the 200 nm size bin (bins as shown in Fig. 2) the same as the fraction of analysed to sampled particles in the 1000 nm size bin? I would like to see a plot of the number size distribution of analysed particles added to the paper or supplement.

6) On page 9, line 12 and elsewhere, you refer to "inorganic (sulfate) components".

Does ‘inorganic’ include sea salt? Please clarify.

7) How fresh is the “fresh SSA” in this case? You based the term ‘fresh’ on chemical transformation, but in the dark winter boundary layer of the Arctic, chemical transformations may be substantially reduced compared with such processes at lower latitudes (e.g. sulphuric acid and nitric acid production). In Fig. 5, you limit your trajectories to two days, which is relatively short compared to the lifetime of an aerosol particle in the Arctic (e.g. see Stohl, JGR, 2006). What were the sea ice conditions under the trajectories shown in Fig. 5 c and d? Were there any large areas of open water in the region during the sampling times? Do you think all the SSA was from leads (on page 6, you mention leads as a possible source)? Could there have been a contribution from frost flowers, as in Shaw et al. (2010)? Could the fresh SSA component have been an accumulation of the transport over much greater distances, while remaining less chemically processed? A paragraph added to your discussion is needed to provide some context for the potential origins of the SSA.

8) A follow-on to comment 7 - What were the wind speeds during the sampling? At the beginning of section 3.4, you mention there was no correlation of certain chemical ratios with wind speed, but sampling at some distance or time from marine sources will make wind speeds difficult to correlate with chemistry. If your sampling was done during times of particularly high sea salt influence due to regional winds, then your results may be skewed to the north coast of Alaska; although sea salt is prevalent in other areas of the winter Arctic that are more distant from open water (e.g. Alert, Leaitch et al., ACPD, 2017). If your source is purely regional, then that point needs to be made clear.

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