Review of "Single particle investigation of summertime and wintertime Antarctic sea spray aerosols using low-Z particle EPMA, Raman microspectroscopy, and ATR-FTIR imaging techniques."

Overview

This manuscript is a very solid description of aerosols from a unique and understudied region (Antarctic). The study provides unique and valuable data regarding two samples (summer and winter) that have distinct differences in their composition. The combination of the three methods, particularly the Raman/ATR-FTIR comparison, is a powerful unified approach to explore the detailed physicochemical composition of these particles. The Ro group has really advanced the ATR-FTIR coupled with Raman approach that allows for insights regarding how sulfate is bound, for example. Overall, the data interpretation is thorough and solid. The authors do a nice job of placing their results in the context of other research in the field. A few small issues should be addressed in a revised manuscript and are indicated below. Overall, this manuscript has the potential to be a useful contribution to the field.

Major Comments

One major concern with the manuscript is the drawing of broad conclusions with limited samples. Specifically, connecting everything back to the biological content without knowledge of sample to sample variability during the different seasons. Given the samples collected this might not be possible, but some discussion of the degree of variability that would occur between two samples of the same season would help provide context as to how much these differences are due to real differences between seasons or differences that happen to occur between these two specific samples that could be due to other reasons.

The second concern is that, while the authors do go to great lengths to justify it, I remain not fully convinced that the organic spectra can be so definitively identified as alanine. Additionally, the confidence of assigning it to the Mg-hydrate salt seems not fully justified, given the challenge of distinguishing ions such as Ca^{2+} from Mg^{2+} in ambient vibrational spectra. I believe that alanine is likely one of a number of compounds contributing to the modes observed, along with the associated hydrate salts of different cations. The authors should soften that language a little to make clear that alanine is unlikely to be the sole contributor to those modes. The classification for fatty acids by contrast appears to be the appropriate level of specificity.

There is very little discussion of the nitrate observed in the samples, particularly in the sample S2. Given the distinctly different HYSPLITs between samples 1 and 2 the enrichment of NO_3^- in S2 that passes over Chile seems to provide evidence of heterogeneous processing through the classic $NaCl + HNO_3 \rightarrow HCl + NaNO_3$ reaction. This is barely discussed, but seems like an important point that should be discussed in more detail. Aging of SSA and organics from SSA is well established with EDX and Raman [*Adachi and Buseck*, 2015; *Ault et al.*, 2013; *Ault et al.*, 2014; *Laskin et al.*, 2012; *Liu et al.*, 2011; *Trueblood et al.*, 2016], but primarily from CAICE-style waveflume experiments or in field studies the mid-latitudes. Discussion of heterogeneous aging in the less-studied Antarctic in the context of the impact observed and modeled [*Bauer et al.*, 2007] in the midlatitudes would strengthen the impact of the paper.

Minor Comments

This is a stylistic point, but I believe the “a” in “Chlorophyll-a” should be italicized.

The uncertainty of the chl-a measurements should be discussed further. Specifically, what is the LOD? The uncertainty should be included when referenced in the abstract and experimental sections in the form of +/- after the values. How certain are the authors that the difference is really 19 time for summer versus winter. This should be addressed further.

On lines 74-75 it could be interpreted that two samples were collected total or that 2 samples from summer and 2 from winter were collected. Consider revising to make this clear.

Lines 102-107 As noted above, some further comparison of the differences between the Hysplit trajectories would be useful for data interpretation.

Further discussion of the organic overlay from Raman and with elemental overlay from EDX would be useful. It seems that the organic portions from the Raman are the thickest portions. This is not necessarily intuitive, though the 2800-3000 window seems like a logical choice.

The silicon-containing particles are interesting. Why would the enhanced sea ice noted on lines 264-265 specifically hinder Si species emission? Is there something about the winter time emissions that is somehow chemically selective? This was unclear and should be explained in more detail.

For the 1052 cm^{-1} peak noted on line 300, how are the authors able to distinguish $Mg(NO_3)_2$ the symmetric stretch of the aqueous nitrate ion observed around 1055 cm^{-1}? See [*Zangmeister and Pemberton*, 2001]and [*Ault et al.*, 2014] for reference. It seems unlikely that the two peaks could be distinguished cleanly and that even if one is present there may be contribution from the other mode.

The phrase “encountering frequency” is used frequently, but is an odd choice of wording. It is suggested that it be replaced with different terminology, as well as to clearly define the terminology used.

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