

Interactive comment on “High Summertime Aerosol Organic Functional Group Concentrations from Marine and Seabird Sources at Ross Island, Antarctica, during AWARE” by Jun Liu et al.

Anonymous Referee #1

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General Comments:

This manuscript provides an overview of one year of measurements of aerosol number concentrations, cloud condensation nuclei, and organic mass and functional groups (by FT-IR) made near McMurdo Station at the southern end of Ross Island in Antarctica from November 2015 to December 2016. The authors characterize the sources of organic aerosol, and are able to establish some evidence for the contribution of both primary marine organic aerosol and the formation of secondary organic aerosol. Both the seasonal coverage of these measurements and the general lack of measurements in this region makes these measurements very valuable and deserving of publication

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in ACP if the following issues can be addressed.

Major Comments:

1) This manuscript generally lacks clarity in the text and is difficult to follow. Many paragraphs contain a number of seemingly disparate thoughts or concepts. I suggest that the authors give this manuscript a very thorough revision with particular attention to the structure of the text.

2) The scientific questions being addressed in this work need some clarification. I agree completely with the author's statement on P3L2-4 that this is most thorough yearlong aerosol data set from Antarctica. However, the questions that are addressed with this unique dataset are not clear, especially at the end of the introduction. After reaching the end of the manuscript it was more clear that the authors aimed to (1) characterize the sources of aerosol across seasons in Antarctica - especially of organic aerosol - and (2) establish evidence for the contribution of primary versus secondary formation of this organic aerosol. This could be stated more clearly at the end of the introduction.

3) Related to point (2), above, the authors spend a considerable amount of the text describing how the local pollution aerosol was removed from the data set. Once this aerosol signal has been removed (only from the total CN measurements, as far as I can tell) the authors then state (P5) that the remaining aerosol is from natural sources. It is very difficult to make this statement without first walking the reader through all the evidence you have for this point (e.g., how can you be sure that the “background” concentrations you measure are not impacted by local pollution). Further, the authors use a “despike” algorithm to remove high values of CN from the total particle concentrations, but do not filter any of the other data (i.e., CCN concentrations). Do you obtain similar results if you use a wind-sector and wind-speed based filter to remove contamination from local pollution sources? How does a wind-sector filter impact CCN concentrations?

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4) The Results and Discussion sections (Section 3 and 4) seem to provide only very cursory discussions of the results. These sections also contain some topics (e.g., PMF details including seeds and Fpeaks) that are better placed in the methods section. Further, the authors seem to assume a significant amount of prior knowledge about the methods and measurement location, all information needed to understand and interpret these measurements should appear in the paper or the supplement.

5) The attribution of the “Marine and Seabird” PMF factor is not very well discussed. I do not dispute the claim, as it does appear likely given the proximity of large penguin colonies. Something as simple as a wind direction analysis relating high concentrations of the M&S factor with winds from the direction of penguin colonies would help your argument. Providing an analysis of back trajectories is also advisable.

Minor Comments:

P1 L30-31: This line reads as though the authors are implicating aerosol to some extent in the melting of the Antarctic ice sheet. I do not dispute that there can be a connection between atmospheric composition and ice dynamics (e.g., through cloud processes), but these processes are not nearly as well understood as the authors seem to suggest here.

P2 L1-21: This paragraph reads as a list of what has been done by others in this region. I agree this such a summary is an important part of this manuscript, but the introduction generally lacks a take-away message. For example, do the authors believe that we can say from past measurements that regional marine biological emissions are an important contributor to Antarctic aerosol? Do we know more about the biological sources of sulfate than organic aerosol? How does this help to motivate your study?

P2 L27 (and Table S1): What do we learn from this collection of amino acid measurements?

P3 L 11-20: Rather than referring to a web link for further information on the sampling

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site, and pertinent information should be summarized here. Information about inlets? Length, flow rates, transmission efficiency as a function of size?

P4 L3: The detection limit needs to be defined in order for the statement “above the detection limit” to convey any information.

P4 L29: CN has not been defined at this point - I assume this is the CPC measurement?

P5 L1-22: The topic of this paragraph is not clear. There is a mid-paragraph shift from discussing pollution aerosol and its removal from the data set to discussing “background” aerosol.

P5 L3: This “despike” algorithm need to be described in more detail in the methods section. Since the goal of this paper is likely not to demonstrate the effectiveness of this “despike” algorithm, this isn’t really a result but rather a part of data quality control that is applied before any interpretation can be made.

P5 L5: SLCE needs to be defined.

P5 L5-6: What does “accounted for” mean in this context? You need to provide a more quantitative assessment to make this statement. Further it is unclear that is meant by “occurred 19% of the time.”

P5 L7: A mean \pm standard deviation doesn’t really tell you that the change was rapid in time, a rate of change might be a better metric.

P5 L11: Is this the author’s hypothesis? If so, it should be stated as such. At this point in the analysis, it seems difficult to make this statement. For example, the remaining CN could be from background pollution not immediately associated with plumes.

P5 L13: “...correlated with...”

P5 L19 (Figure S1): Actually plotting CCN/CN would be informative, rather than asking the reader to estimate this ratio by looking at separate plots of CCN and CN

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P5 L19-22: The meaning in these two sentences is difficult to discern. My sense is that the authors are trying to make a statement about changing CCN activation diameters, but it is unclear (i.e., what “decrease in particle diameter”? Associated with what phenomena?). Could the change in CCN/CN ratio result from either a change in particle diameter in summer or a change in composition that results in a change in the activation diameter?

P5 L23-33: This paragraph begins by discussing aerosol growth factors and ends by discussing aerosol absorption. The connection the authors are trying to make, if any, is unclear.

P5 27-29: The meaning of this sentence is unclear (i.e., a much smaller change in what?).

P6 L2: Figure 2 is referred to in the text before Figure 1.

P6 L3-6: Is the sea salt measured at this site during winter transported from open water areas? What evidence do the authors have for this? The authors go on to suggest that frost flowers are the major source of sea salt at this site, but more discussion is warranted here. While, sodium to chloride or sodium to sulfate ratios provide some evidence, I do not see that this rules out other sources of aerosol, such as blowing snow, which has been observed in Antarctica (e.g., Jones et al., 2009 www.atmos-chem-phys.net/9/4639/2009/). Field observations and laboratory experiments seem to suggest that frost flowers are rigid and difficult to break, even at wind speeds up to 12 m/s (e.g., Yang et al., 2017 <https://doi.org/10.5194/acp-17-6291-2017>; Roscoe et al., 2011 [doi:10.1029/2010JD015144](https://doi.org/10.1029/2010JD015144)). A recent model study suggests that the frost flower source of sea salt aerosol cannot explain the seasonality of sea salt aerosol across several Arctic stations (Huang et al., 2017 [doi:10.5194/acp-17-3699-2017](https://doi.org/10.5194/acp-17-3699-2017)). A more thorough discussion of possible sources supported by meteorological variables and back trajectories is needed to draw a conclusion here, especially since upward migration of brine and incorporation of frost flowers can lead to depletion of the sulfate-

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to-sodium ratio relative to bulk sea water such that this chemical signature may not be unique to frost flowers.

P6 L11: Define what “baselined” means in this particular case. This should probably be discussed in the methods section.

P6 L13-15: These specifics belong in the methods section. In the Results and Discussion section I suggest you discuss why your choice of two factors is the physically most meaningful choice (some of this discussion exists, but could be expanded).

P6 L 24-25: The motivation for carrying out a PMF analysis and a k-means cluster analysis of the FTIR spectra is not adequately explained here. Presumably if these two approaches yield similar results, then this lends confidence to assignment of source types. This information needs to be stated clearly.

P6 L25-26: This needs to be actually shown in some way, even in the supplement.

P6 L32 (Table 1): In addition to stating the observation that the residual is much larger than the MS factor in winter, the authors should discuss what it means for interpretation of their results. This comment applies to all “ratios” provided in Table 1, the authors state that this provides a measure of uncertainty, how do the authors want the reader to interpret this? For example, does this result suggest that the PMF factors are robust across seasons? Only in specific seasons? This information should be clear.

P7 L11: Is the FTIR method sensitive to inorganic ammonium? If so, can a spectrum of e.g., ammonium sulfate be included to support this hypothesis.

P7 L15: “80 % hydroxyl group” - Is this a fraction of the total organic mass? How do you know that all organic mass is accounted for? Do you have to assume an average molecular weight for aerosol components? More description of how you arrive at these values is warranted (in the methods section).

P7 L15-16: Are these differences between Arctic and Antarctic summer aerosol significant? Do you expect a difference? More discussion is needed here, rather than just

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stating that there is a difference between these two, arguably rather different, regions.

P7 L19: What is your detection limit for total organic mass?

P7 L19: Is it not the other way around - high summer OM and low winter OM attributed to marine and seabird sources?

P7 L20: If the highest concentrations of salt aerosol are in the winter (as shown) then it makes sense that M&S OM is not correlated with sea salt. But, what about only in the summer months when M&S OM is elevated?

P7 L28: Do you mean that gas phase ammonia is neutralizing acidic particle phase species?

P7 L32: More discussion on what the "CHNO fragments" detected by Schmale et al. 2013 indicate is warranted here. How does it relate to your measurements?

P7 L32-33: More than simply the wind rose shown in the supplement is needed to make this statement. The authors can likely easily show that when these signals were high the wind was indeed bringing air from Cape Crozier.

P8 L3: It is not clear why a coastal source is suddenly implicated here. Is there evidence from back trajectories to show this? Is the proposed coastal source different from the penguin colonies?

P8 L13-14: It is not immediately clear that this is true.

P8 L26: What is meant by "emission concentrations?"

P8 L27: Is this the mean summer OM concentration?

P9 L11: The authors should provide a URL or, ideally, a DOI for this data.

P10-12: There are some issues with the reference formatting (e.g., missing DOI's, all caps titles). I believe that DOI's should be prefaced with "doi:" or shown as an active link (i.e., <http://dx.doi.org/xxxx...>). Please check through the references.

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Table 1: Is a table the best way to display this information? Box and whisker plots would facilitate a better visual comparison of the data across seasons.

Figure 1: Can inter-quartile ranges be shown here?

Figure 5: Barrow marine and Alert marine spectra from 1500 – 1800 cm^{-1} more closely resemble the FFC spectrum rather than the M&S spectrum. How do the authors explain this? What does this imply about the assignment of the FFC source? Also, where does particle phase ammonium appear in this spectrum?

Figure 6: Does this correlation necessarily say that this fraction of the OM is driven by secondary processes? Could it be that the source of particle phase species coincides in time with available solar radiation? Do the peaks indicating N-containing species correlated with solar irradiance? Do any other organic functional group correlate with solar irradiance?

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2017-1225>, 2018.

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