

The Authors would like to thank the reviewer for their constructive comments. Specific replies to each comment, and associated changes to the manuscript, are presented here.

Reviewer 1: General comments: In several sections, “strong relationships” or “strong function of” are mentioned between two parameters, but it is not necessarily clear in the figures. For instance, on the top of page 6, the authors discuss a relationship between aerosol number and wind. What are the correlations between these two? When looking at both years in Figure 1, it is not immediately clear that this relationship exists until one examines the figure closely, when looking at the 2-minute data. Providing some sort of correlation coefficient would literally strengthen these statements.

Authors: The Pearson correlation coefficient for total aerosol number (from the EPC) to wind speed have been added to the manuscript as per the reviewer’s suggestion.

New text: *“Fig. 1b shows two facets of Antarctic aerosols: first, aerosol number concentrations are a function of wind speed (Pearson correlation value of 0.32) and, second, there is a steady-state aerosol concentration during calm and low-wind periods.”*

Reviewer 1: Presenting more than simply the sulfur species would make the case stronger that sulfate is the major contributor to the AMS aerosol population. What was the percentage of sulfate relative to total AMS particle mass? Specifically, on page 6, line 10, what percentage of the particles measured by the AMS were combustion-derived OA? On page 7, lines 3-4, showing the size distribution of the other AMS types would support the authors’ statement here. Without showing the other species, this leaves one to wonder if other aerosol types were relatively high at any point in time (i.e., no graphical evidence provided) in addition to how much of the aerosol were actually sulfate. For context, it would be helpful to provide data on the total aerosol population, perhaps as a time series and size distribution of the relative aerosol types for each season, even if it would be placed in the supporting information.

Authors: We agree with the reviewer that a full discussion of the overall particle composition is important, and we have a manuscript in preparation discussing this in detail. We feel the focus of this paper on sulfate is warranted for the following reasons: 1.) historically the sulfate aerosol population has been of specific scientific interest with regards to the Antarctic aerosol population (e.g. understanding the variability of non-sea-salt sulfate), and 2.) in terms of the aerosol number (not mass) population sulfate aerosol is a key contributor. The open questions regarding the sources, transport, and processing of sulfate over Antarctica are important enough to warrant a paper dedicated to those questions.

We agree with the reviewer that some information contextualizing the sulfate aerosols in terms of the total aerosol is important. Sulfate is the third most abundant species after Cl and Na which is consistent with the literature (approximately 60-80% Na and Cl, 5-30% sulfate depending on wind regimes). Combustion-derived OA was generally not observed except in certain low-wind circumstances and those local emission events have been filtered from this analysis. These details have been added to the text as per the reviewer’s suggestion.

New Text: *“While aerosol sulfate is the main focus of this manuscript, it is not the only aerosol component and the relative amount of sulfate measured by the AMS should be contextualized. Over both field seasons, sulfate generally makes up more than 50% of the total mass of the traditionally reported*

non-refractory species (organics, sulfate, nitrate, and ammonium). Both the absolute amount and relative percentage of total mass of sulfate is higher in 2014 than 2015. Ammonium, organics, and nitrate, in that order, make up the rest of the non-refractory species measured by the AMS. When adding measurements of refractory Na and Cl to the non-refractory species, sulfate is the third most abundant species at 5-30% of the total sub-micron aerosol mass.”

Reviewer 1: Several conclusions of the general seasonality of Antarctic aerosol are built upon the observations here, which only span a month or two during two consecutive years. How do the authors know if what they observed was typical or anomalous? For instance, the bottom of page 8 presents broader conclusions based on the intensive measurements presented. These statements would be more convincing if the same month or transition season was measured at least twice, for instance, if both time periods were measured in 2014 and 2015, which obviously cannot be done at this point. Although the observations are very intriguing, the authors should take care in how they interpret the results and try to steer away from making such bold conclusions of what the typical behavior of the aerosol would be this time a year. This could be alleviated by either referring to the observations from the 2014 or 2015 sampling of the transition seasons (versus the transition season in general) or providing more background on previous measurements that would corroborate their observations.

Authors: A note about the limited duration of the measurements has been added and the transition season has been noted as our “observed” transitional season. Unfortunately, this is the first observation of the transitional phase and previous corroborating measurements do not exist in the literature. The lack of previous observations is due to this being the first deployment of a high-resolution, high-sensitivity aerosol instrument to the continent. These measurements provide evidence of when and where future campaigns should look for non-sulfate particle formation sources and mechanisms.

Added text: *“The results presented here, although limited in seasonal coverage and duration of sampling, suggest that radiative forcing models for Antarctica should continue to treat the sulfate population as an external mixture.”*

Reviewer 1: It is great that the authors provided such a detailed explanation on the possible sources of uncertainty or limitations in the measurements that could lead to what was observed (i.e., section 3.2), however, this lengthy discussion draws away from the focus on the uniqueness of the observations. Instead, the authors could condense this section to a paragraph or two (and put some or all of the “A” figures in the supporting information), and focus more on bolstering what was observed, particularly the chemistry measurements. Present each of the three explanations separately and more directly, but focus more on the observation itself than what could be wrong with it. As is, when the three possible explanations in the beginning of the section are posed, I thought to myself, they have the data to prove this. Then, the data would be discussed much later. The section is presented more as a thought process to understand the results than a results and discussion section. Also, this section initially is focused on phase 2, but during the explanations, all time periods are discussed. Overall, the section could use some restructuring and condensing, which would provide clarity as well.

Authors: The authors thank the reviewer for this comment and have taken the reviewer’s advice. The bulk of the discussion has been moved to an Appendix (Appendix B) and Section 3.2 has been condensed to contain only the conclusions of the section. We believe this enhances the readability of the

manuscript as a whole without losing any detail for those readers who wish to delve into the minutia of the reasoning behind the conclusions presented.

Reviewer 1: Perhaps the biggest issue in this manuscript revolves around the new particle formation discussion: The authors provide contradicting evidence that new particle formation is a large contributor to the aerosol number. This is concluded in the abstract, and several locations throughout the manuscript (e.g., page 11, lines 31-32), yet on page 11, lines 16-17, the authors directly state no new particle formation events were captured during ZODIAC. Please be clear throughout on if new particle formation was a major source. It is difficult to discern any “banana plots” in Figure A3, so where did the conclusion that new particle formation is the major source of aerosol during this time period originate from? Perhaps zooming in on some of those growth events towards the end of 2014 would elucidate if these were indeed new particle formation events or simply emission of small, primary particles.

Authors: The reviewer makes the salient point that “new particle formation” has been used in two different ways: first, to mean local observable particle growth, and second, to refer to the population of unknown composition (newly formed particles) that appears during the transitional period (phase 2). This has been clarified in the text by changing the terminology to refer to “newly formed particles”.

New Text: *“...Phase (2) is consistent with measuring newly formed particles that have been transported to our measurement location during a transitional period during the extended Antarctic sunrise...”*

As per the reviewer’s comments on Fig. A3: the growth events that are observed (e.g. 3 or 4 times in 2014) are strongly indicated to be contamination from the diesel generators running the field site (as mentioned in the caption of Fig.A3). These periods have been eliminated from the rest of the data presented in the paper and will be removed from this figure as well to prevent any reader confusion.

Reviewer 1: More explanation and background is warranted in the PMF section. Are these typical AMS particle classifications that have been previously used or are universal? What are some previous studies that have classified AMS particle types like these? More supporting evidence is needed regarding the classifications for what the particle types were. Labeling the peak fragments in Figure 6 would help as well.

The authors briefly mention the collection of filters for offline analyses in the methods. If the analyses, whatever they might have been, were conducted, those results could provide significant supporting evidence to the conclusions drawn based on the AMS and number concentration measurements that are discussed. Of course, this is also limited by the filter pore size, which was not mentioned. If chemical analytical techniques were applied to the filters, that information could fill in quite a few gaps throughout the manuscript and would potentially provide explanation for much of section 3.2.

Authors: Additional discussion expanding on the PMF section has been added as per the reviewer’s suggestion. The use of only sulfur compounds in PMF is novel and comparison to other studies is not possible. Still, some text contextualizing these results has been added.

New Text: *“...Since using only the sulfur containing ions in PMF analysis is novel, it is difficult to compare these PMF results to previously published results. The closest related study is Schmale et al. (2013) which measured Antarctic/Southern Ocean air masses. In both the results presented here and in Schmale et al. (2013), the percent contribution of MSA to the total aerosol burden increases as sunlight (phytoplankton activity) increases over the Southern Ocean. Additionally, the MSA associated factor in that study is*

postulated to contribute significantly to the total sulfate signal, although it is not measured explicitly, which agrees with the results here....”

Secondly, the filters have recently been analyzed using an IC and the results will be discussed in an upcoming manuscript. Broadly speaking, the filter results are consistent with the AMS data, although direct comparison with the AMS data is not possible due to differences in size cuts on the filters (which include supermicron particles) to the submicron measurements of the AMS. The supermicron mass signal from the filters is dominant and they integrate over far longer time scales. For these reasons the filter data would not add significantly to this manuscript, and could distract from the central focus.

Specific comments:

Reviewer 1: Page 5, line 4: Although there are a couple references provided, a few sentences on the specifications and operating principles for the SP-AMS is needed. Especially considering the authors discuss the instrument limitations later on in the manuscript.

Both reviewers have noted that it is unclear that the SP-AMS is largely the same instrument as HR-ToF-AMS with the addition of an extra laser to measure black carbon. This extra laser was not utilized during this campaign, making the SP-AMS identical to the HR-ToF-AMS in terms of operating principle and performance. This has been noted in the text to prevent confusion by readers.

New Text: *“...aerosol composition was measured with a Soot Particle Aerosol Mass Spectrometer (Aerodyne Research Inc. Billerica, MA, SP-AMS, DeCarlo et al., 2006; Onasch et al. 2012). The SP-AMS is a combination of the Aerodyne High-Resolution Time-of-Flight aerosol mass spectrometer (HR-ToF-AMS) and a soot vaporizing laser (from Droplet Meas. Tech.).”*

Reviewer 1: On page 6, line 31, a PToF size is discussed but it is unknown up to this point that the SP-AMS contains a ToF mass spectrometer. Defining this in the methods would alleviate any confusion.

Authors: This has been clarified in the text via the addition pointed out in the previous comment.

New Text: *“...The SP-AMS is a combination of the Aerodyne High-Resolution Time-of-Flight aerosol mass spectrometer (HR-ToF-AMS) and a soot vaporizing laser (from Droplet Meas. Tech.).”*

Reviewer 1: Page 5, line 5: What offline analyses?

As the filter data are not directly relevant and are beyond the scope of this manuscript, this line has been removed from the text.

Reviewer 1: Page 5, lines 29-30: Is this typical and/or expected in this region?

Authors: Both the bimodal wind direction distribution and higher late-winter/early-spring wind speeds are typical of the region. A reference to an analysis of the prevailing meteorology of the Ross Island region has been added to the manuscript.

Reviewer 1: Page 5, line 32: The caption for the figure says 1-hour, not 15-minute.

Authors: The text line has been changed to reflect the (correct) figure caption.

New Text: *“Figure 1b shows the number concentration from the EPC over both field seasons. The figure shows the 2-minute average as well as a 1 hour average.”*

Reviewer 1: Page 7, line 16: Use the acronym for CCN when they are first discussed in the introduction and simply use the acronym here (it was spelled out twice in the introduction).

Authors: The text has been changed as per the reviewer’s suggestion.

New Text: *“...radiative forcing of Antarctic aerosol and in predicting CCN number concentrations (N_{CCN}) in the Antarctic troposphere.”*

Reviewer 1: Page 7, lines 20-22: This statement is highly speculative based on the data provided. Considering the limitations of the AMS (refractive aerosol, the size range), this conclusion is not fully supported by the available observations, especially since the measurements were not conducted during all seasons (Sep – Nov). A statement of this level would require a longer time period of measurements covering a wider range of aerosol types and sizes.

Authors: The text has been modified regarding the seasonality and limited nature of the 2ODIAC measurements.

New Text: *“The results presented here, although limited in seasonal coverage and duration of sampling, suggest that radiative forcing models for Antarctica should continue to treat the sulfate population as an external mixture. This work does support the assumptions of older estimates of radiative forcing for sulfate aerosols over Antarctica of approx. -0.1 Wm^{-2} (Myhre et al., 1998).”*

Reviewer 1: Page 10, line 30: Wind speed is all that is presented here, not all local meteorology. Simply stating wind speed would suffice.

Authors: The text has been changed as per the reviewer’s suggestion.

New Text: *“In fact, neither field season exhibits a strong dependence on wind speed...”*

Reviewer 1: Figures: I get why the authors are showing 2015 before 2014 in the figures, to enable the data to be presented in a seasonal versus chronological order. Perhaps labeling them as “Austral spring (2014)” and Austral summer (2015)” would make more sense if keeping the data in this order.

Authors: The figures have been modified as the reviewer suggests.

Reviewer 1: Figure 1: What are the time resolutions for wind direction/speed and AMS?

Authors: The figure caption has been modified to reflect the time resolutions.

New Text: *“Figure 1: For both the 2014 and 2015 field seasons, with 2015 leftmost: A) Wind direction record colored as a function of wind speed, displayed as a 2-minute average record B) 2-minute (light blue) and 1-hour (black) records of particle number concentration from the EPC, C) 2-minute records of sulfate concentration from the aerosol mass spectrometer. Dotted lines on (B) indicate the minimums in particle number concentrations (99th percentile) measured over the field seasons.”*

Reviewer 1: Figure 3: Why is the UHSAS so noisy? Why is only 2014 shown?

Authors: The UHSAS is a high speed and high resolution instrument, and for this campaign data was taken at 10 second resolution and then coherently averaged to longer times. During low and medium wind speeds, this instrument is operating close to the detection limit for the short sample times, which can introduce counting noise to the signal. Furthermore, there are some small artifacts introduced to the coherently averaged signal from the combined gain stages within the instrument, however the overall size distribution from the UHSAS is consistent with the data from the other particle-sizing instruments. Unfortunately, the UHSAS was damaged during shipment and was inoperable for the 2015 field season.

Reviewer 1: Figure 6: I see this is the combined time series from the different years, yet could cause some confusion since these data were not obtained from the same year. Be sure to label the year that corresponds to each data time period on this figure, similar to the previous figures. Also, labeling the peak fragments on the mass spectra would be helpful.

Authors: The figure has been modified as per the reviewer's suggestion.