

Giordano et al. have done a fine job revising the manuscript based on the reviewer comments. Although they provided adequate responses to the comments, some responses did not result in a change to the manuscript, and there are a few additional issues that need to be addressed prior to publication in ACP.

General comments:

The main concern I have is regarding the AMS results, namely determining the size of the sulfate aerosol. I am no expert when it comes to AMS, but doesn't the AMS only measure single particles when there is sufficient mass in each particle? Jayne et al. (2000) report on the AMS, "...the current sensitivity of this AMS for single particle counting with 100% efficiency is $\sim 2 \times 10^{-14}$ gm (~ 300 nm for a pure component particle) and the sensitivity for the signal averaging mode is $\sim 0.25 \mu\text{g m}^{-3}$ for several minutes of signal integration." Considering the authors report sulfate aerosol sizes of < 300 nm (~ 250 nm), wouldn't the AMS need to integrate and average > 1 particle? Further, according to Onashe et al. (2012), "The SP-AMS, like other AMS instruments, provides average mass spectrometric measurements of the ensemble of sampled aerosols rather than measurements on a particle-by-particle basis." In light of this information, the authors should be clear on how the size of specifically sulfate aerosol was delineated from the measurements. Was sufficient mass present in each particle to actually measure the size *and* composition of single particles? If a bulk sample was measured, how can the authors be sure the sizes measured were purely sulfate particles? Further, the authors focus on differentiating internal versus external mixtures, which is indeed important. Although I am happy to see this distinguished, because the methods regarding how single particles were measured are not described in the level of detailed needed, it is not clear how the authors can differentiate external mixtures. If AMS integration and averaging for > 1 particle was done, how can external versus internal mixing state be determined? Perhaps this concern originates from my naïve knowledge of the AMS, but the authors should at least clarify how they came up with the size and composition of single particles to delineate sulfate aerosols.

T. B. Onasch , A. Trimborn , E. C. Fortner , J. T. Jayne , G. L. Kok , L. R. Williams , P. Davidovits & D. R. Worsnop (2012) Soot Particle Aerosol Mass Spectrometer: Development, Validation, and Initial Application, *Aerosol Science and Technology*, 46:7, 804-817, DOI: 10.1080/02786826.2012.663948.

J. T. Jayne , D. C. Leard , X. Zhang , P. Davidovits , K. A. Smith , C. E. Kolb & D. R. Worsnop (2000) Development of an Aerosol Mass Spectrometer for Size and Composition Analysis of Submicron Particles, *Aerosol Science and Technology*, 33:1-2, 49-70, DOI: 10.1080/027868200410840.

It doesn't appear that the results support the main conclusion that sulfate is the dominant Antarctic aerosol, at least, not at this level of breadth. Figure A1 shows the percent of sulfate to total AMS mass, but the percentage is $< 40\%$ most of the time, and doesn't seem to equate to the average value the authors present in the text on P 718-9, "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)." Further, the authors affirm that this value is even less when considering refractory Na and Cl (5-30%). This also does not include other non-refractory species like mineral dust. By number, I can see how this conclusion holds true (i.e., based on Fig. 4). A few things here: (1) Where did the Na and Cl come from? How were these measured? (2) Based on the concern regarding how sulfate number was derived, how can this hold true? (3) If sulfate particles were indeed measured on a particle-by-particle basis, thus size could be measured by AMS, the authors should

provide a more explicit conclusion that is corroborated by the results presented, i.e., sulfate is the dominant *non-refractory* Antarctic aerosol *by number*.

Specific comments:

P 2, l 8: How an aerosol affects the radiative balance *and* cloud microphysics.

P 5, l 10: In the responses, the authors state the extra vaporizing laser that measures BC was not used during ZODIAC. This needs to be clarified here in the methods.

P 5, l 15: Please be more specific here, since composition can include both the AMS and filters. Provide a few details of what will be “future work”. Publications on the filter samples? That way it is clear to the reader what will and will not be presented.

P 5, l 17: I am guessing this temperature is Celsius, but be sure to provide the units.

Methods section: Be sure to provide the time resolution for each of the instruments. I see they are provided in the R&D for some measurements, but not all. Additionally, in the beginning of the R&D, the authors express that the data are averaged every 2-minutes (and some are averaged hourly as shown in the figures). What are the standard deviations for these? The authors might want to consider showing these in the figures, or at least the SI.

P 6, l 18: With a Pearson’s of 0.32, this statement still seems like an over interpretation of the results. Perhaps tone down this deduction, or at least explain that periods with high WS typically had lower aerosol number but higher sulfate mass (at least this is what it looks like in Fig. 1). It would be good to provide the average particle number and sulfate mass concentrations during high WS and low WS time periods at some threshold; that will demonstrate what the authors are trying to conclude. Also, with the issue of blowing snow that the authors bring up, shouldn’t the time periods with $WS > 8 \text{ m s}^{-1}$ be eliminated (based on their statement on P 22, l 31), thus that would change the correlation coefficient? In general with linking the WS to particle number and sulfate mass, the authors should take care in clearly stating if the data were or were not eliminated and why. As is, they state data $> 8 \text{ m s}^{-1}$ were excluded later in the paper but then also discuss the trends during these high WS time periods earlier on.

Section 3.1: This section contains information that seems out of order. First, the first paragraph basically concludes that sulfate is omnipresent and the main contributor to Antarctic aerosol, thus should be placed in the following section (3.2). Second, the authors do a nice job of highlighting the climate relevance of external mixtures, but the remaining paragraphs seem more like a “broader implications” section that should be placed before the conclusions. If this section were broken up and placed in the relevant locations, the order and flow would make more sense.

P 8, l 14-15: Without calculating the radiative forcing for the current work, linking to previous estimates does not connect. How does this work support the RF estimate from previous work, do the authors instead mean this work supports the fact that sulfate is a major contributor to Antarctic aerosol based on previous work, which could then impact the RF by -1 W m^{-2} ?

P 8, l 17: The methods state that the system had a 50% transmission efficiency at $5 \mu\text{m}$ and a 0% at $> 9 \mu\text{m}$. The aerodynamic lens permits submicron particles, so where did the information in the methods come from?

Header to section 3.2: By mass? By number? By both? Please be more specific.

P 10, l 26: Define SEMS.

P 10, l 26-27: According to the Aerodyne website, the SP-AMS measures particles from 40 nm to 1 μm , thus this statement is incorrect. The authors have the 40 – 250 nm particles to work with to elucidate the composition of these transitional aerosol.

P 11, l 4-6: This statement contradicts the main conclusion that sulfate is the dominant aerosol. What percentage of the time during the studies were these conditions met? Perhaps it was for a short time period, thus not influencing the overall composition that much. On a similar note, the authors discuss the solar irradiance measurements several times; it would help to show a solar irradiance trace in Fig. 1.

P 11, l 8: This is in the appendix, not the SI. Along these lines, what is the purpose of having an appendix and a supporting information? It would be simpler if all the appendix information were placed in the supporting information, i.e., all in one referenced location.

P 12, l 23-24: Higher relative to what, to the Austral spring as presented in the current study? The authors have the data during an Austral summer to support this claim (i.e., the 2014 field season), why not validate this statement in the context of the reconstructed MSA concentrations? Although, the concentrations are low relative to previous studies...perhaps this is solely location-dependent and not on the season as the authors state. Some clarification could be used here.

P 13, l 5-10: Even though this PMF method is novel, the reasoning behind classifying Factor 3 as sulfate, Factor 2 as aged biogenic, and Factor 3 as biogenic/MSA must be based on something. How exactly were these classified? Factors 2 and 1 are very close, what is the distinguishing factor? Are there any previous studies that had factors that at least support how the current ones were classified? Or were these based on known substances measured in a laboratory setting? More explanation and clarification on how these types were classified is needed.

P 13, l 5: Why is the most abundant the last Factor?

P 13, l 12: **non-refractory** mass. Please take care in clarifying here and throughout the manuscript.

P 13, l 25: Define " Q/Q_{exp} ".

Fig. 6: Are these based on an average of single-particles or a bulk sample? How many spectra contributed to each factor? Even though the authors did add more information on PMF from the first draft, more information is still needed.

Fig. A1: Label the axes similar to the previous figures, i.e., with Austral spring and summer.