The authors thank reviewer 1 for their further comments. It seems the reviewer is uncertain about how the AMS was operated, and how the processing of the data was done. We will address that below, but are unsure as to why the reviewer thought single particle data was used. A word search for single particle in the manuscript returned no results. Below, the reviewer comments are given in **red**, with responses in **black.**

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 ~ 2 x 10^{-14} gm (~ 300 nm for a pure component particle) and the sensitivity for the signal averaging mode is ~ 0.25 µg 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 7 l 8-9, "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***.

The AMS measurement principle is generally based on averaged particle data either bulk non-size resolved mass concentrations of various species or size resolved composition using what we call particle time of flight (PToF) mode. This mode is how we determine the size distribution of sulfate for this manuscript, and it relies on averaged data (***not*** single particle data). The various modes of the time-of-flight based AMS instruments is described in DeCarlo et al. (2006). We realize these details are not as well understood by non-AMS users and have added that clarification to the text.

"The PToF mode in the AMS is a mode of operation in which the size-resolved aerosol composition is determined by averaging many particles during the PToF operation time (2 minutes in this study). The PToF mode does not represent single particles but rather the size distribution of particles in which ions, such as sulfate ions, are present in."

We also note that the current data acquisition card used on our SP-AMS is capable of true "single particle" measurements but that data is not presented here, as it is a relatively new capability, and we are still analyzing that data.

Further, the reviewer should note that the Jayne et al. (2000) paper was using a quadrupole MS whereas the SP-AMS here was using the HR-ToF MS. The HR-ToF is much more sensitive than the quad MS with one-minute detection limits <0.04 µg m^{-3} (DeCarlo et al., 2006) though this is somewhat variable on an instrument-by-instrument and deployment-to-deployment basis. The detection limit for the Antarctica deployments of this instrument was calculated at 10 and 12 ng m^{-3} for the 2014 and 2015 campaigns, respectively. The detection limit values have been added to the text.

"The detection limit of sulfate in the AMS was calculated to be 10 and 12 ng m-3 for the 2014 and 2015 deployments, respectively (Jayne et al., 2000)."

With that background and explanation we can respond to some of the reviewers other comments:

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?

As mentioned above, we do not present single particle data here, nor do we mean to imply it. This has again been clarified in the text (see below). In regards to the reviewer's second point: the conclusion that the particles measured in the AMS at the 250nm distribution are primarily sulfate is based off of several pieces of information:
1) comparison to the sizing instruments (Fig. 3, note that Fig. 3 is averaged over long integration times to smooth out curves and that there are transmission and collection efficiency differences between the instruments),
2) that no other species besides sulfate exhibit a well-defined peak at 250nm in the AMS PToF, and
3) Rapid changes in meteorological conditions lead to abrupt shifts in other chemical species, but not aerosol sulfate. If particles were internally mixed then all particles would be impacted similarly. This has been clarified in section 3.1 of the text:

"Taken together, the fact that neither total aerosol counts nor aerosol sulfate fall below a minimum value suggests that the background Antarctic aerosol number population may be primarily composed of sulfate. The relatively constant mass concentration of sulfate, independent of wind speed, wind direction and air mass origin, would indicate that this background aerosol is relatively temporally and geographically invariant. Other chemical species showed strong dependence on meteorological conditions in contrast to sulfate. If the background aerosol is primarily composed of sulfate species, then any enhancements in total aerosol counts would have to be a separate aerosol number population. An independent, externally mixed sulfate mode would therefore be expected in the AMS particle time-of-flight (PToF) mode. A large-scale, externally mixed sulfate mode would be expected to maintain a consistent size, during variations in wind speed and direction. In both 2014 and 2015, the (inorganic) sulfate species in the AMS exhibited such a wind-independent mass distribution. Figure 2 shows the sulfate vacuum aerodynamic diameter distributions from the AMS for the 2014 field season as a function of wind speed and direction. Regardless of where an air mass originated from, the sulfate aerosol as measured by the AMS exhibited a well-distributed mode centered at a vacuum aerodynamic diameter of approximately 250nm. This result is consistent with previous ocean based size measurements of sub-Antarctic MSA containing aerosol (250 and 370nm, from Zorn et al., 2008 and Schmale et al., 2013, respectively) as well as off-line filter integrated measurements in coastal Antarctica (200-350nm aerodynamic diameter impactor stage, Jourdain and Legrand, 2001). None of the other species measured in the AMS showed a well-defined size mode. "

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.

This has been addressed in the above response. Note again that the data presented is not based on single particle data, but rather PTOF or averaged data.

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 7 l 8-9, "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%).

While we agree and have shown that sulfate is not the dominant mass contribution to Antarctic aerosol especially when including refractory Na and Cl signals (to be discussed in a forthcoming manuscript). However, due to the smaller size in comparison to NaCl particles, sulfate particles are predominant contributors to the aerosol number population during our measurement campaigns. For clarity we have retitled section 3.2:

Sulfate as a Predominant Component of the Background Antarctic Aerosol Number Population

This also does not include other non-refractory species like mineral dust.

The reviewer is correct in that this study does not include mineral dust. However, both the literature and our own measurements suggest that mineral dust loadings were minor to non-existent during the 2ODIAC campaign, and more importantly would not be large contributions to the aerosol number population given their larger size.

Since submission we have obtained additional filter based results for Mg and Ca from the filters (to be discussed in an upcoming manuscript) suggest that both Mg:Cl and Ca:Cl are indicative of sea-spray derived aerosols, not crustal material derived aerosols. This is not surprising since our wind fetch was purposely selected to avoid some of the potential local dust sources (e.g Ross Island and the Dry Valleys).

Additionally, the 2 closest studies in the literature both suggest extremely low dust loadings over Antarctica: a South Pole study suggested mineral dust derived aerosol Al and Fe concentrations are <1 ng m^{-3} and a coastal site (Georg von Neumayer) showed Mn concentrations at 0-30 ng SCM^{-1}. These two studies appear to also be in agreement with snow and ice-pack studies that suggest extremely low (1ppb) loadings of crustal material in Antarctica.

Cunningham, W. and Zoller, W.: The Chemical Composition of Remote Area aerosols, J. Aerosol Sci., 12(4), 367-384, 1981.
Wagenbach, D. et al., Coastal Antarctic aerosol: the seasonal pattern of its chemical composition and radionuclide content, Tellus B., 40B(5), 426-436, 1988.
Delmonte, B., J. R. Petit, K. K. Andersen, I. Basile-Doelsch, V. Maggi, *and* V. Lipenkov *(*2004*b),* Dust size evidence for opposite regional atmospheric circulation changes over East Antarctica during the last climatic transition*,* Clim. Dyn.*,* 23*,* 427–438*.*
Jung-Ho Kang, Heejin Hwang, Sang Bum Hong, Soon Do Hur*, Particle Size Distribution Analysis of Mineral Dust in Polar Snow Using a Coulter Counter,* Ocean and Polar Research*,* 2014*,* 36*, 4, 319*
Lambert, F.; Bigler, M.; Steffensen, J. P.; Hutterli, M.; Fischer, H.. 2012 Centennial mineral dust variability in high-resolution ice core data from Dome C, Antarctica. *Climate of the Past*, 8 (2). 609-623. 10.5194/cp-8-609-2012

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?
The literature is clear that coastal Antarctic aerosols are heavily influenced by the ocean and thus NaCl is *primarily* of oceanic origin and likely sea spray. Some Na and Cl is always measurable in the AMS but full quantification of it is difficult and has been the subject of focused laboratory studies to be discussed in an upcoming manuscript. We now provide sulfate contribution to non-refractory species and refractory Na and Cl plus non-refractory species. This gives the reader all of the values in proper context for comparison to the existing literature.

(2) Based on the concern regarding how sulfate number was derived, how can this hold true?
From the above discussion, and points below, the reviewer's point is taken that the manuscript should be more explicit in its statement. We have included the following text in section 3.2 to clarify the manner in which we calculated the sulfate number value:

"This process assumes sphericity of particles takes the mass distribution measured in vacuum aerodynamic diameter space, and using the density value transforms it to volume vs volume equivalent diameter for that sulfate mode, which can then be converted into a number distribution and integrated to find the total number of particles from the measured sulfate mass distribution. "

(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***.
Again, the AMS is not reporting single particle measurement in the context of this paper, but rather an averaged mass distribution as a function of vacuum aerodynamic diameter. This is converted into a

number population as described above. The fraction of the particle population is not limited to non-refractory particles, since the EPC used in the denominator does not discriminate between refractory and non-refractory particles. So we agree that sulfate is the dominant Antarctic aerosol by number, we do not need to add "non-refractory" to the statement as a qualifier.

Specific comments:

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

The reviewer's suggestion has been added to the text.

New text: "…on the global 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 2ODIAC. This needs to be clarified here in the methods.

The text now reads:

"…The soot vaporizing laser was not used in this study."

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.

The text now reads:

"…other parts of the instrumentation suite, including the particle filters and snow samples…"

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

"C" has been added to the text, the text now reads:

"…3°C and 3% accuracy, respectively…"

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.

The time resolution for each instrument has been added in the instrumentation section. The hourly interval is only included in Fig.1B to aid in readability in the figure and is not included in any analysis. The standard deviation for the 2-minute average of Fig 1A and 1B has been included in the figure caption. This has been calculated by averaging the standard deviation for each 2-minute averaged period of 1-second resolution data since the reviewer's question seems to deal with the variability within each 2-minute averaged period and not the record as a whole.

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 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 m s^{-1} were excluded later in the paper but then also discuss the trends during these high WS time periods earlier on.

The reviewer is correct in pointing out that high wind speed conclusions are not germane to this manuscript. The correlation between wind speed and particle number concentrations is not as relevant to this manuscript as it is to the forthcoming blowing snow manuscript so this statement has been removed from the text. The main focus of this section of the manuscript is to draw attention to the minimum particle number concentration so the average loadings as a function of wind speed will also be relegated to a future publication.

With regards to the reviewer's comments on other mentions of high wind speeds, these have been clarified in the text with the statement:

"…since high wind speeds and particle counts are strongly correlated, to be discussed in a forthcoming manuscript"

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.

The authors feel that the first paragraph sets the hypothesis that sulfate is a key background aerosol, by stating the observation that neither sulfate or particle counts every drop to 0. The remainder of the paragraph examines this hypothesis in more detail offering additional observations to strengthen this hypothesis.

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}?

We feel that this inclusion is important to provide additional importance for the observation of an external mixture to a broader audience. While we do not explicity calculate radiative forcing, we are are mentioning the work of others (e.g. Myhre et al.) who, based on our work, correctly make this assumption.

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

The transmission efficiency in the methods is of the aerosol inlet to the inlet of the AMS, not the AMS's transmission efficiency. P5 L10-14:

"The inlet for the aerosol sampling line was covered and heated to prevent sampling of wind blown snow and to prevent riming, respectively. At the flow conditions and geometry of the sampling inlet, transmission of <1µm particles to the AMS was >95%. Overall, the system had a 50% transmission efficiency at 5µm and 0% transmission efficiency of particles >9µm. All transmission efficiency values are as calculated by a particle loss calculator using the specific geometry of the setup (von der Weiden et al., 2009)."

P8 L17 explicitly states "aerodynamic lens of the AMS" but the text on P5 has been changed to:

"…Overall, the aerosol inlet system had a 50% transmission efficiency at 5µm and 0% transmission efficiency of particles >9µm. …"

Header to section 3.2: By mass? By number? By both? Please be more specific.
Section 3.2 has been renamed to: Sulfate as a Predominant Component of the Background Aerosol Number Population. See below.

P 10, l 26: Define SEMS.
SEMS has already been previously defined in Section 2.2.

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.
The reviewer is citing the aerodynamic lens range transmission but what matters to the statement on P10,L26-27 is the mass loading in PToF mode. In PToF mode, the mass loading of particles is greatly reduced as opposed to "open" mode. In the transitional period, the total aerosol number is low (200-400 #/cc) which, even at low detection limits in the AMS, translates to extremely small mass loadings at particle diameters of 40-150nm. Note that the AMS is sensitive to mass not number, so a 50 nm particle has 125 times less mass than a 250 nm particle. The fact that there is a measurable sulfate distribution on the lower end of the distribution curve is testament to how sensitive the AMS is given how little sulfate there is in Antarctic aerosol compared to other environments. The statement has been changed to clarify this point:

"Unfortunately, given the instrumentation deployed, it is impossible to determine the composition of these very small particles (e.g. less than 100 nm) due in part to the low mass loadings inherent in Antarctica and the effects that low mass loadings have on AMS PToF data."

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.
As mentioned before we have retitled section 3.2 to read:
"Sulfate as a Predominant Component of the Background Antarctic Aerosol Number Population"

For the reviewer's second point, we are candid about the fact that these measurements are the first of their kind and are limited because this is the only observation of this transitional period in the record. We do not mean to suggest, nor do we believe we write, that the transitional period is important to global climate. Rather, we point to it as potential evidence of a new particle formation mechanism over Antarctica that should be closely examined in future studies.
Our solar irradiance measurements are semi-quantitative, and are not be shown in the figures due to the potential for these values to be taken as more accurate than they are. Solar irradiance in the manuscript is mentioned only to serve as a guide and a relative measurement.

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.
During the review process we were asked to place this information in the appendix, which is why there is both an appendix and an SI. Given the usefulness of the figures for interpreting the

manuscript we will continue to have the appendicies included in the main paper, and the supplementary information (less necessary for interpretation of the manuscript) separate.

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.
"Higher" in this context is relative to what was measured in the early summer of 2ODIAC 2014. The sentence suggests that in mid-late summer, MSA should increase as compared to what was measured in early summer. This statement is verified in that there is virtually no MSA observed in winter-spring and it only begins to appear in early summer. We explicitly state this in the precedeing sentences of the paragraph.
The location of the measurements is mentioned because it is assumed that proximity to source is important in the interpretation of concentration measurements. From our observations and other literature (e.g. Minikin et al., 1998; Preunkert et al., 2007; Preunkert et al., 2008; Read et al., 2008;), it seems clear that MSA concentrations are both temporally and spatially dependent.

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.
The following has been added to the text to clarify why the factors were named as they were:

New text: The naming of the factors, Sulfate, Biogenic/MSA, and aged biogenic, for Factors 3, 2, and 1, respectively, was chosen based on two main aspects of the resultant PMF spectra. The sulfate factor was named due to its lack of ions containing both Carbon and Sulfur atoms, and the fact that it bears some resemblance to the mass spectra obtained by atomizing diluted sulfuric acid into the AMS. The Biogenic and Aged Biogenic factors were distinguished primarily by the contribution of the CxHySxOp ions appearing in the Biogenic factor and to a lesser extent in the Aged Biogenic factor. If this assumption is not correct then the naming schema of the last factor may need to be adjusted in the future. Again, the three factor solution was chosen due to it minimizing the Q/Q_{exp} of the PMF solution without creating obviously extraneous factors (see SI Fig.S4).

Factors 1 and 2, while similar, exhibit clear differences in their mass spectra. There are large differences in the CxHySzOp ions contributions and times series of the factors. The CxHySzOp ions appear to a greater extent in Factor 2, as well as the ratios of the major sulfate peaks to each other (e.g. SO^+:SO_2^+, SO_2^+:SO_3^+, etc.) and to their hydrogenated compounds (HSO_2^+, HSO_3^+, $H_2SO_4^+$). In the AMS, these ratios are consistent and robust for specific chemical species. Factors 1 and 3 are more closely related than 1:2 or 2:3 but this is due to the fact that *m/z* 48, 64, and 80 (major sulfate peaks) make up the majority of both factors. Even so, Factor 3 still has CxHySzOp peaks that Factor 1 does not and has differing sulfate:HxSOy ratios from Factor 1. The R_tseries vs R_profiles output from the PMF analysis (Ulbrich et al., 2009) can be seen in SI Fig. S4.D for interested readers.

New text: Factors 1 and 2 are differentiated in both their percent contributions of the CxHySxOp ions as well as the ratios of major sulfate peaks (e.g. SO^+:SO_2^+).

PMF has almost exclusively been applied solely to organic masses from the AMS. The few studies that have elucidated MSA factors out of marine aerosol do not apply PMF to the sulfates but only the CxHySxOp ions so there are no comparable studies to compare to. The laboratory measured MSA spectra is included in SI for comparison but, as already mentioned in the text, suggests that the Biogenic/MSA factor is not solely composed of MSA due to differences in the principle ratios discussed above.

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

The factor numbering was preserved from the PMF output and should not be taken to be indicative of any valuation on the factors themselves. The following has been added to the text to reflect this point:

New text: It should be noted that the numbering of the factors is preserved from the PMF solution and is not indicative of any valuation on the factors themselves.

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

"Non-refractory" has been clarified in multiple places in the text.

P 13, l25: Define "Q/Q_{exp}".

Q/Qexp has been defined in the text at the beginning of S3.3.1.

New Text: PMF analysis uses as input any number of mass spectra as a function of time, measured in the AMS, to pick out patterns in both temporal and mass spectral phases of the data. PMF determines "factors" that contribute to the time-series and mass spectral series by minimizing a "quality of fit" parameter (the sum of the residual of a given input not fit by the algorithm scaled by estimated error in the time and mass-spectral series). The PMF algorithm is run over multiple starting points in the rotations of the matrices and limited to given numbers of factors. Q/Q_{exp} is used as a metric to determine how well the error model assumed in PMF is represented by the dataset. In general, the lowest values of Q/Qexp are the best solution. This metric is described in detail in Ulbrich et al. (2009).

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.

As discussed above, no single particle data is used in this paper. PMF analysis uses mass spectra (of the ensemble aerosol mass, not size resolved) at given times to elucidate patterns in the MS and time series data. The input into PMF in this analysis consisted of over 35,000 individual mass spectra. PMF analyzes all of the spectra to pick out patterns that are "Factors". This has been clarified in the text:

New text: PMF analysis uses as input any number of mass spectra as a function of time, measured in the AMS, to pick out patterns in both temporal and mass spectral phases of the data…In this study, over 33,000 individual mass spectra were used as input to PMF.

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

The figure has been changed as per the reviewer's suggestion.