The Authors would like to thank the reviewer for their constructive comments and highly detailed proof reading of the manuscript. Specific replies to each comment and associated changes to the manuscript are outlined in this document.

General comments:

Reviewer 2: It is not clear why the authors do not show other species measured by the AMS than sulfate. The discussion on the lower particulate sulfate contribution to the total particle number throughout section 3.2 and more specifically related to Fig. 4 would be much more informative if the authors provided the mass fraction of sulfate measured by the AMS in relation to ammonium, nitrate and organics. I suggest including a figure showing either the time series of these species' masses or their fractional contribution. It seems that the authors plan another publication with a detailed discussion on these data, however this manuscript will strongly benefit from showing the general AMS-related results.

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 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 2: With regards to the PMF analysis, using only sulfur containing fragments as input is novel. For that reason, the methodology and the results need to be shown in more detail. Key diagnostics as outlined in Zhang et al. (2011) should be provided in the SI to show the robustness of the solution. The 3 factor solution is not very convincing as it is currently presented. Factors 1 and 2 are very similar and it is not clear why the authors concluded that these factors are not artificially split, especially since an explanation for the aged biogenic source is missing. Stronger evidence must be provided to justify the 3

factor solution. Also, how stable is the instrument's fragmentation at m/z 48 and 64? Is it stable enough not to introduce variability that's picked up by the PMF analysis?

Authors: The diagnostics as outlined in Zhang et al. (2011) have been included in the SI as per the reviewer's suggestion. It should be noted that the 2014 data has higher residuals than the 2015 data. The authors believe this is primarily a factor of how the combined solution was constructed: 2014 and 2015 data sets were run separately and similar (though not completely identical) factor solutions were obtained with reasonable residuals. Because the individual solutions appeared reasonable, the 2 data sets were combined and PMF run on the combined data set. In the combined set, over all factors and fpeaks, the residuals for the summer (2014) data are much larger in comparison to the 2015 data. We believe this is due to the instrument performance in 2015 vs 2014. Small changes in instrument background and sensitivity will impact the associated error of the instrument which goes into the PMF solution. In a low-signal environment such as Antarctica, this may cause the observed differences in residual, and influence the mass spectra identified by PMF.

Additional discussion and contextualization of the PMF has also been added to the text as per the reviewer's suggestion.

New Text:

Despite the minimal contribution of the aged biogenic factor, the three factor solution was chosen over the two factor solution for two reasons. The primary reason is the inadequacy of the 2 factor PMF solution with regard to MSA, which based on previous measurements in the Southern Ocean and the presence of a marker ion ($CH_3SO_2^+$ at m/z 79) should make up some of the sulfur contribution. 2-factor PMF results either apportioned m/z 79 to 2 factors with 48:64 ratios that did not resemble any known substance (e.g. things tested in a lab setting included ammonium sulfate, pure MSA, diluted H_2SO_4, and southern ocean sea water) or apportioned majority of m/z 79 to a factor that was not temporally consistent with the $CH_3SO_2^+$ fragment in the dataset. The secondary reason for choosing the three factor solution is that three factors was consistently the number where diminishing returns in Q/Q_{exp} began to occur. The attribution of the MSA marker ion to the aged biogenic and biogenic/MSA factor indicates that both factors are likely representative of either MSA directly or of "biologically influenced" aerosols. Comparison to direct atomization of MSA into the AMS (see SI) suggests that the biogenic factor is made up of more than just MSA contributions since PMF did not find a "pure" MSA factor mass spectra for this dataset. The ratios of $CH_3SO_2^+$ to the major sulfate peaks (SO^+, SO_2^+, HSO_3^+, SO_4^+) in the two biogenic factors differ from pure MSA measured by the AMS in the laboratory.

Reviewer 2: The contribution of new particle formation (NPF) to the observed aerosol needs a clearer discussion. On p. 11, l. 16 it says that no new particle formation events where observed. Does this mean that you conclude only from the literature that NPF is a potential source? If so, this needs be made explicit and less weight should be given to this conclusion as in that case no direct evidence is be available. If you have evidence for NPF, this is not clearly present in the manuscript currently

Authors: The reviewer has noted that NPF is used to mean two different things in this manuscript: local observable particle growth and regional (unobserved growth) particle formation. In the case of the former, there were no observed local NPF events. In the case of the latter, section 3.2 goes into detail

about why we believe (regional) NPF and transport to our site is a major factor in the Phase (2) aerosol population. NPF has been clarified in the text, in conjunction with Reviewer 1's comments, to refer to "newly formed particles" when the latter case is meant to avoid confusion.

Reviewer 2: Generally, to provide an impression of the geographical sources of the sampled aerosol and with that the potential source region for NPF, a back trajectory analysis throughout the measurement periods would be very helpful. This could also address the question of how and why there is a transition period and how source regions might change between seasons.

Authors: In addition to the clarification regarding NPF above, we generally agree with the reviewer. However, accuracy of back trajectory calculations are highly dependent on the meteorological data that feeds them. In the case of McMurdo Antarctica, the available meteorology comes from the GDAS 0.5° x 0.5° record. The one-half degree resolution of the data is insufficient to resolve local orography effects on the air flows that arise from the complex topography of the Ross Island Region.

Understanding this limitation, we have performed HYSPLIT back trajectory analyses over the whole of the 2ODIAC campaign. Generally speaking, the majority of air masses were subject to long-range transport over the continent though some air masses did originate over the Southern Ocean. However, without resolving orography, it is impossible to tell if an air mass was exposed to open ocean immediately prior to sampling or originated completely inland.

Beyond being confident that the data presented in this manuscript are not contaminated by anthropogenic sources (McMurdo or Scott Base, 2ODIAC generators, etc), it is not currently possible to identify with any certainty the local source regions observed during the field campaign. This is the reason we are careful to discuss all of the known and suspected particle formation mechanisms relevant to Antarctica (see p.12).

Reviewer 2: How representative are the two field seasons? It would be very helpful if at least more long-term meteorological data from the nearby station could be presented to show whether wind direction, wind speed, temperatures, solar irradiance etc. are comparable for the intensive observational periods. This is needed to back up the conclusions of the paper regarding the general background aerosol characteristics, the evolution of aerosol characteristics between winter and summer and the potential contribution of NPF.

Authors: Both the bimodal wind direction distribution and higher late-winter/early-spring wind speeds are typical of the region, and this has been noted in the manuscript. Including a detailed climatology for the region is beyond the scope of this work, however a reference to an analysis of the prevailing meteorology of the Ross Island region has been added to the manuscript.

New Text: *"…These meteorological patterns and seasonal differences are not unusual for this region (Seefeldt et al., 2003)*." (p6, l.15)

Specific comments:

Reviewer 2: The term 'aerosol population' is used very often. Mostly it is unclear whether the authors refer to mass, number, size distribution or chemical composition. Consider replacing the term by more precise terminology.

Authors: "Aerosol population" has been changed to identify aerosol number or aerosol mass specifically throughout the paper. This has been clarified early in the text to reflect this as per the reviewer's suggestion.

Reviewer 2: p. 2, l. 15 consider including information from Hamilton et al. (2014) that the Southern Ocean is one of the few places left on Earth to sample pristine aerosol.

Hamilton, D. S., Lee, L. A., Pringle, K. J., Reddington, C. L., Spracklen, D. V., and Carslaw, K. S.: Occurrence of pristine aerosol environments on a polluted planet, Proceedings of the National Academy of Sciences, 111, 18466-18471, 2014.

Authors: This has been added to the text as per the reviewer's suggestion.

New Text: "*…Measurements in Antarctica, provide insight into one of the more pristine environments and can be useful in the understanding of preindustrial background aerosol (e.g. Hamilton et al., 2014). However, the ability to sample pristine aerosols is directly related to an areas inaccessibility…*"

Reviewer 2: l. 33: it is not necessary to discuss volcanic eruptions to such detail. It is sufficient if you state that volcanic eruptions are not important for Antarctic aerosol except for a few instances. Also, while volcanic eruptions can inject large amounts of sulfur species into the atmosphere and are therefore a temporarily limited major source of aerosol, also anthropogenic emissions need to be considered as they are a constant important source.

Authors: Given the proximity of the field site to an active volcano that is constantly emitting aerosols and SO_2, we believe that the three sentences explaining why Mt. Erebus is not likely impacting the results presented here are warranted. The mass spectral fingerprint of Mt. Erebus is also more difficult to distinguish from background (without isotopic analysis), unlike the anthropogenic sulfur sources near the field site (e.g. McMurdo, Scott Base, diesel powered energy/transportation).

Reviewer 2: p. 3, l. 7: replace generally by "mostly". Sea spray aerosol does have a submicron fraction.

Authors: This has been changed in the text as suggested.

New Text: "*…Sea-spray aerosols are mostly supermicron in size and production is a strong function of wind speed…*"

Reviewer 2: l. 30 an appropriate reference would also be Petters and Kreidenweis, 2007.

Authors: This has been added to the text as suggested.

New Text: *"…Determining the CCN spectrum of a given aerosol population is possible once the size distribution, size-resolved composition, and mixing state of the aerosol population is known (Petters and Kreidenweis, 2007…*"

Reviewer 2: l. 3 – 5: It is unclear to me what you try to say with this and I do not understand what you base your arguments on.

Authors: The major point is that the extent of external mixtures tends to decrease as distance from emission source increases. This does not appear to be the case for the measurements presented here. The text has been modified to better reflect the meaning.

New Text: "*…Because Antarctic aerosols seem to primarily be composed of sulfates and salts, the effect of the mixing state on cloud forming predictions may be minimized over the continent itself but overestimated as continental air masses flow out over the Southern Ocean and gain organic components…*"

Reviewer 2: l. 3.2: Why do you say "If the externally mixed sulfate . . . is, . . ., the primary component. . .". As mentioned in the general comments, you have information on other species like ammonium, nitrate and organics from the AMS measurements. You can also estimate the fraction of seasalt that you see with the AMS, see Ovadnevaite et al. 2012. Ovadnevaite, J., Ceburnis, D., Canagaratna, M., Berresheim, H., Bialek, J., Martucci, G., Worsnop, D. R., and O'Dowd, C.: On the effect of wind speed on submicron sea salt mass concentratio and source fluxes, J. Geophys. Res., 117, 2012.

Authors: As noted earlier, the mass fraction of sulfate has been added to the text. Of the "traditional" AMS species, sulfate makes up the majority of the aerosol measured. Additionally, an estimation of the refractory Na and Cl has been performed (similar to Salcedo et al., 2010) and is the subject of an upcoming manuscript and is beyond the scope of this paper. Even upon including the "non-traditional" species, i.e. refractory sea salts, sulfate is still the third most common species behind Cl and Na.

Reviewer 2: p.9, l. 2: not clear if you mean sea-salt sulfate or non-sea salt suflate with "not nss-"

Authors: This has been clarified in the text.

New Text: "*…persistent aerosol sulfate component (total, i.e. not nss-) seen multiple times over the continent in the winter…"*

Reviewer 2: p. 10, l. 3f: I do not understand the logic of the argument. Make it explicit. Why would the observed behavior be opposite?

Authors: The text has been clarified with the sentence: *"The non-sulfate particles would have to be the same size or larger than the sulfate particles or there could be no observed changed in measured total mass."*

Reviewer 2: l. 11: What do you mean by inlet dynamics?

Authors: Dynamics has been changed to geometry.

Reviewer 2: p. 11, l. 11: remove important, and make the statement more relative: the correlation is weak, this needs to be reflected in the conclusion.

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

New Text: "*…These correlation values have two implications: first, that the change…."*

Reviewer 2: l. 16: Do you mean that NPF were not observed at all, or that you did not observe any local events but rather already grown particles from NPF further away? As indicated above, the observations and conclusions regarding NPF are not clear.

Authors: As per the previous comment, NPF has been clarified in the next to read as "newly formed particles" where "regional NPF" was meant in the text.

Reviewer 2: l. 32: replace "is generating" by "may generate"

Authors: This has been changed in the text.

Reviewer 2: l. 33: rephrase to "and it is possible that contributions from" and it is not clear what you mean by non-sulfate aerosol formation mechanisms?

Authors: This sentence has been modified in the text. Non-sulfate mechanisms refers to the possibility that DMS does not play a role in this aerosol. This has been clarified in the text as well.

Reviewer 2: p. 13: more accurate would be : " in regions where the origin of particulate sulfate was dominate by open . . ." since there were other major local sources of aerosol as well.

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

New text: "*…Both of the previous measurements took place in regions where the origin of particulate sulfate was dominated by open ocean source regions and took place in the austral summer and fall…"*

Reviewer 2: l. 9-11: This sounds like a contradiction to me: the observed concentrations of MSA in the literature are higher than observations in this study while the argument is the other way around.

Authors: Lower concentrations in 2ODIAC, not the previous campaigns. This has been clarified in the text.

New Text: *"…Lower MSA and sulfate concentrations during 2ODIAC are therefore not surprising given the differences in season and location as compared to the previous studies…*"

Reviewer 2: l. 30 f: More discussion on the origin of the aged biogenic factor is needed. How can it occur at the earliest time in the season that was observed? What can be the source during winter, when it is dark and more sea-ice is covering the ocean? Also provide number on how many % each factor contributes.

Authors: The PMF discussion section has been expanded, including discussion on the origin of the aged biogenic factor. During the winter, the most likely source is long-range transport from areas of the Southern Ocean that are not ice-covered and in perpetual darkness. However, it should be noted that our measurements took place at the extreme end of winter/early spring. There was measureable (~50 W/m2) sunlight for 4-5 hours during the first few days of the 2015 campaign. The sun came up quickly after that. The actual distance one has to travel north from Ross Island to reach "normal" daylight in early September is not that far.

The percent contribution for each factor has been included as well.

Reviewer 2: l. 30 Do you mean spring rather than fall?

Authors: The reviewer is correct, fall has been changed to spring.

Reviewer 2:Conclusions: p. 15, l. 10: Also exploring NFP over the Southern Ocean is important.

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

New Text: "*This work further underscores the need to closely examine new particle formation over Antarctica, and the Southern Ocean, in the early Austral spring.*"

Figures:

Reviewer 2:I suggest including a map or preferably a satellite image showing your measurement sites and the sea-ice extent during the field seasons.

Authors: A satellite image (Landsat 8 SLI, retrieval date 10/14/15) has been added to SI. The 2014 and 2015 sea ice edges and field sites have been marked.

Reviewer 2:Fig. 1b: There are points below the minimum line. How can that be?

Authors: Some points are filter periods that were not removed correctly, the minimum is defined as the 99^{th} percentile. The filter periods have now been removed.

Reviewer 2:Fig. 2: One needs to guess which line is ESE/NW, low wind and high wind. Consider using a different line type. Why are ESE, Med. Wind and NW, high wind so smooth compared to the other lines?

Authors: The line types have been changed to address the reviewer's concern.

ESE_MW is approximately the same "smoothness" as ESE_HW. These 2 wind regimes, along with NW_HW, had higher mass loadings than the low wind speed regimes which results in a "smoother" trace (as signal:noise is improved at higher mass loadings). NW_MW had similar mass loadings but had a much reduced sampling time as compared to the other three (high signal) regimes. This exacerbates the noise in the NW_MW trace as well.

Reviewer 2:Fig. 4: How did you smooth? Rename the y-axis to: "sulfate particle number ratio"

Authors: "Boxcar smoothing" has been included in the text. The figure axis has been changed as per the reviewer's request.

Reviewer 2:Fig. 5: The colors of the symbols from Zorn et al. and Schmale et al. are misleading. As far as I understand the color is not related to the color code. However the colors are part of the range of colors in the code. Either chose different colors for the literature data or use simply open symbols with black margins.

Authors: The Zorn and Schmale data symbols have been changed to black and grey to resolve the issue the reviewer points out.

Reviewer 2Reviewer 2: Fig. A3: are the read lines in the lower panel real data?

Authors: The red lines were an AMS IE calibrations/size calibrations that did not get removed from the figure. They are now removed. As per reviewer 1, the generator contamination has also been removed from the figure.

Technical comments:

Reviewer 2: p.1, l. 14: low "temporal" resolution and remove the expression in parenthesis which is not needed for the abstract

This has been changed in the text as suggested.

Reviewer 2: l. 15: to answer the question about "the chemical composition of" Antarctic aerosols.

This has been changed in the text as suggested.

Reviewer 2: l. 16: replace populations by those

"Populations" keeps the sentence completely unambiguous. "Those" could conceivably refer to "seasonal cycles". This has not been changed to prevent ambiguity.

Reviewer 2: l. 18 remove populations

This has been changed in the text as suggested.

Reviewer 2: l. 20 the abbreviation SP-AMS does not follow from high resolution. . .

The SP-AMS is an upgrade to the HR-ToF-AMS (). This has been clarified in the instrumentation section.

Reviewer 2: l. 22 "and its evolution in Austral Spring" be consistent with capitalizing the seasons

The capitalization of the seasons has been made consistent across the entirety of the manuscript.

Reviewer 2: l. 23: remove to rest of the aerosol population

This has been changed in the text as suggested.

Reviewer 2: l. 26: what are highly aged sulfate particles?

"Aged sulfate" is defined in the text, specifically the PMF section.

Reviewer 2: l. 27 & 28 replace population by mass

The first instance has been changed as suggested, the second instance has been left as population.

Reviewer 2: p. 2 l. 8: "climate impacts depend on their effects on the radiative balance which are a function of the aerosol hygroscopicity, chemical composition and physical optical properties. . ."

Reviewer 2: l. 10, remove pathways

This has been changed in the text as suggested.

Reviewer 2: l. 21f: remove the sentence starting with “Aerosol measurement. . .”

This has been changed in the text as suggested.

Reviewer 2: l. 27: remove “of the aerosol population”

The sentence has been reworded to “…component of that aerosol, especially…” This prevents the ambiguity and improves flow of the sentence.

Reviewer 2: l. 28: “the sulfate aerosol mass which has long. . .”

This has been changed in the text as suggested.

Reviewer 2: l. 30: shouldn’t it be “Kulmala” et al. 2002?

The authors thank the reviewer for catching this typo.

Reviewer 2: p. 3, l. 9: remove population

This has been changed in the text as suggested.

Reviewer 2: l. 25: “of aerosol physical properties. . .”

This has been changed in the text as suggested.

Reviewer 2: l. 28: introduce the abbreviation CCN and use it in the next line.

This has been changed in the text as suggested.

Reviewer 2: p. 4, l. 3: replace exaggerated by overestimated

This has been changed in the text as suggested.

Reviewer 2: l. 7f: “this manuscript focuses on the. . ..”

This has been changed in the text as suggested.

Reviewer 2: l. 16: remove the sentence “Cracks in the . . .” the context is already well enough explained.

This has been changed in the text as suggested.

Reviewer 2: P5 l. 22: what does down sampled mean? Is it averaged?

Down sampled has been changed to averaged in the text.

Reviewer 2: l. 18: remove “but the same order of magnitude as”

This has been changed in the text as suggested.

Reviewer 2: p. 8, l. 14: what do you mean by “middle ground”?

This has been changed in the text to average value.

Reviewer 2: p.9, l. 11: replace “ is drowned” by “decreases”

This has been changed in the text as suggested.

Reviewer 2: l. 13: a subject and verb are missing in this sentence

“From Fig. 1 during Phase (2), both the total counts on the EPC and the sulfate mass in the AMS trend upward but total counts increases faster than the mass captured in the AMS.”

To Trend is the verb in this sentence, EPC counts and sulfate mass is the subject. “But total counts…” has been revised to a second sentence.

Reviewer 2: p. 10., l. 2f: Delete the first sentence, it does not provide any new information.

This section has been revised completely as per Reviewer 1’s suggestions.

Reviewer 2: p. 12, l. 7: data are

This has been changed in the text as suggested.

Reviewer 2: l. 18f: delete the first sentence of the sub section, it does not provide any new information.

This section has been revised completely as per Reviewer 1’s suggestions.

Reviewer 2: l. 21: what do you mean by “mirabalite fractionation”?

Mirabalite is defined in the introduction as is the fact that sodium fractionates during its formation.

Reviewer 2: Fig. 1A: replace “fraction” by “percent”

This has been changed in the figure as suggested assuming the reviewer means Fig. A1.