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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A missing source of aerosols in Antarctica – beyond long-range transport, phytoplankton, and photochemistry" *by* Michael R. Giordano et al.

Anonymous Referee #2

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Giordano and co-authors provide a very detailed study of sulfur-containing aerosol from a sea-ice site in East Antarctica covering Austral winter to early summer. In two field seasons, a number of state of the art instruments were deployed to study aerosol number concentrations, size distribution and chemical composition primarily with online methods. The results presented are discussed in great detail and novel conclusions on the provenance of Antarctic aerosol are provided.

This study presents very valuable data from an understudied region and can contribute significantly to what is known about Antarctic aerosol. Nevertheless, before publication several points need to be addressed. Among those are a lack of methodological information, a number of vague conclusions and implicit argumentations, as well as the

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need to present more details of the positive matrix factorization results.

General comments:

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.

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?

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.

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.

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.

Specific comments:

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.

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.

l. 19: not clear whether you refer to the size or spatial distribution

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.

p. 3, l. 7: replace generally by “mostly”. Sea spray aerosol does have a submicron fraction.

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

p. 4, l. 23: How did you identify the data? What thresholds did you apply, for example? How many % of the data were that?

p. 5, l. 2f: What is the lower cutoff of the EPC?

L3: if the SP AMS is also high resolution that should be mentioned here explicitly.

l. 6: it is not clear how heating the inlet prevent the collection of wind blown snow. A more precise description is needed.

In the methods sections information is missing on how you verified that the CPCs counted correctly and are comparable for both field seasons. There are also no details on the AMS calibrations and how well did the quantification work? This is important since you compare the sulfate size distribution scaled to the mass measured by the AMS with the size distributions from other instruments to quantify the fraction of sulfate particle to total particle number. How accurate is this. There is no discussion on the potential uncertainties of this comparison (Fig. 4).

l. 32: The text says 15-minute while the figure caption says 1-hour. Please check.

p. 6, l. 6ff: Is the minimum of 50 p/ccm an absolute minimum or did you define concentrations below a percentile as minimum / background? I suggest to work with explicitly percentiles to make your statement more robust. Why is the background concentration higher in spring (125 p/ccm) than in summer (50 p/ccm)?

l. 13f: “Aerosol sulfate mirrors the total aerosol counts. . .” this statement is too strong. If it “mirrored” the total counts the time series would co-vary more closely or the actual correlation would be higher.

l. 14 ff: provide the months you refer to.

l. 24f: I do not understand the logic of the argument. Please make it explicit what you mean. What is the threshold you refer to? What is it based on? How does it relate to other observations?

l. 28: Why would any enhancement have to be a separate aerosol population? For example, if a major source of aerosol is wave breaking why would stronger winds not lead to an enhancement of the same aerosol population. This argument needs to be revised or clarified.

l. 30f: Which sulfate species are you referring to? Inorganic sulfate or do you include MSA as well? Do you refer to background or enhanced concentrations?

Replace the expression “PToF size” by “vacuum aerodynamic diameter”.

p. 7, l. 1: remove “remarkably” here and elsewhere. This is a value judgment.

l. 1f: provide the modal diameters from the Zorn et al. and Schmale et al. references. Which diameter was used in the Jourdain and Legrand publication? Is it comparable with your AMS data?

l. 18: it is not true that in “the rest of the world” external aerosol does not play a role. Further down on this page you describe yourself that external aerosol mixtures occur in urban aerosol. Revise this section here.

p. 8, l. 1f: Explain what you mean by special case? Do you mean that normally there are enough compounds in the atmosphere that condense on pre-existing particles to form internally mixed aerosol at locations where long-range transport is the major source of aerosol? In addition, you make a very general statement about Antarctic aerosol, but you measured on sea-ice actually not very far from the coast. I suggest making your conclusion more relative.

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.

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.

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

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

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

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

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.

l. 32: replace “is generating” by “may generate”

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?

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.

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.

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.

l. 30 Do you mean spring rather than fall?

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

Figures:

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

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

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?

Fig. 4: How did you smooth? Rename the y-axis to: “sulfate particle number ratio”

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.

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

Technical comments:

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

l. 15: to answer the question about “the chemical composition of” Antarctic aerosols.

l. 16: replace populations by those

l. 18 remove populations

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

l. 22 “and its evolution in Austral Spring”

be consistent with capitalizing the seasons

l. 23: remove to rest of the aerosol population

l. 26: what are highly aged sulfate particles?

l. 27 & 28 replace population by mass

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. . .”

l. 10, remove pathways

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

l. 27: remove “of the aerosol population”

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

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

p. 3, l. 9: remove population

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

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

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

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

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

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

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

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

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

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

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

p. 12, l. 7: data are

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

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

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

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