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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The Importance of Blowing Snow to Antarctic Aerosols: Number Distribution and more than Source-Dependent Composition – results from the 2ODIAC campaign" *by* Michael R. Giordano et al.

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Response to Reviewer #1: The authors thank Reviewer #1 for their thoughtful and constructive comments on the manuscript. The reviewer's comments have been implemented as below:

Reviewer - General Comments: There seems to be a shift of focus between the title/introduction and conclusions. While the manuscript puts emphasis on the influence of wind speed on the aerosol population the conclusion mainly focuses on the halogen

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cycle. A section in the introduction on the halogen cycle is needed. For example, the two last result subsections use the reference Legrand et al. (2017) heavily, while he is not mentioned in the introduction.

Authors - A section on the halogen cycle as it relates to Antarctic aerosols has been added to the introduction:

"Halogen-containing aerosols in the Antarctic are connected to the depletion of near-surface tropospheric ozone, a greenhouse gas, (Kalnajs et al., 2013) and may also contribution to new particle formation (Sipilä et al., 2016). Numerous remote sensing measurements of halogen oxides in the boundary layer (Saiz-Lopez et al., 2007; Simpson et al., 2015; Prados-Roman et al., 2018) show active bromine and iodine chemistry in the Antarctic troposphere. Chemical ionization mass spectrometry measurements of Br2, BrCl, and BrO confirmed the coupling of bromine and chlorine chemistry in the springtime Antarctic troposphere (Buys et al., 2013). Reactions with non-methane hydrocarbons (e.g. Ramacher et al., 1999), elemental mercury (Saiz-Lopez et al., 2008), and DMS (von Glasow et al., 2004; Read et al., 2008) can all have large impacts on tropospheric composition over the poles. Additionally, some work has also been done examining iodine oxides' role in forming ultrafine aerosols (O'Dowd et al., 1998; McFiggans et al., 2004; Saunders and Plane 2005). Despite significant progress in our understanding of atmospheric halogen gases, a lack of observational data capturing the interactions between gas-phase halogens, aerosols, and snow persists, limiting our understanding of these multiphase processes. "

Reviewer - The title is cryptic: "more than Source-Dependent Composition" is not a description of this work's content that will inform the reader directly of what this manuscript is about. What about: The importance of blowing snow to aerosol and the halogen cycle in coastal Antarctica: Influence of Source region versus wind speed.

Authors - The authors agree with the reviewer and have changed the title to incorporate the suggestion of the reviewer: new title "The importance of blowing snow to halogen-

containing aerosol in coastal Antarctica: Influence of source region versus wind speed".

Reviewer - The notation of the chemical species throughout the manuscript is confusing: e.g., chloride, Cl, Cl-, Chl. I suggest to introduce a notation that makes clear with which instrument the species was measured, e.g., Cl- for IC results, Chl for AMS results. Please check the manuscript throughout, sometimes the ionic charge is not provided (forgotten). Below are some hints, but I might not have spotted all.

Authors - The authors thank the reviewer for pointing out that the nomenclature is not 100% consistent. A sentence has been added at the end of Section 3 denoting which notation implies which measurement. A few parenthetical reminders have also been added throughout the rest of the text due to the length of the manuscript.

"In this manuscript, 3 notations are presented for chemical identities and correspond to either instrument used to quantify or to their presence regardless of molecular structure. 1) For any ion measured with the IC system, ionic charge is noted (e.g. Cl-, Na+). 2) For compounds measured with the AMS, notation in the text follows AMS standard notation (e.g. Chl for chloride, SO4 for sulfate, NO3 for nitrate) with the exception of sodium measured by the AMS and corrected through the process described in S2.3 – NaAMS. 3) For general and/or hypothetical discussions without implication to measurement or molecular compounding, elements are referred to simply by their periodic table notation or written out (e.g. chloride, Cl, sodium, Na)."

Authors - Some exceptions are left in the text (e.g. "chloride salts" in S2.3) to avoid having to add another notation (e.g. XCl) but the meaning in these few cases should be obvious for all readers.

Specific Comments

Reviewer - Aerosol size distribution information based on AMS measurements are not provided in this manuscript. Therefore the description of size calibrations could be omitted to make the manuscript shorter.

Authors - The sentences describing the AMS calibrations have been replaced with: “The AMS was calibrated as described in Giordano et al., 2017”

Reviewer - In section 2.3, p. 7, second paragraph: Please include a brief description of the model by Salcedo et al. (2010) so the reader does not have to look up another publication to understand how the authors arrived from the e-folding time of 40 to the modelled Chl and Na concentrations as shown in Fig. 1. p. 9, l. 21: It is not clear whether those data points are shown as well. Please make this evident in the text.

Authors - A brief description of the Salcedo model has been added to the text: “The model employed by Salcedo et al. (2010) assumes an incoming mass flux of a slow-vaporizing species which impacts the vaporizer, sticks to it, and then desorbs at a rate proportional to the amount on the vaporizer at any given time. This assumption results in an explicit ordinary differential equation.”

A figure showing the data fit to obtain the e-folding time has been added to the supplement for this work (SI Fig. 2).

Reviewer - p. 9, l. 27: “chloride losses” Do they refer as relative to Na+ or to the total chloride concentration?

Authors - The text has been changed to reflect the cited manuscript: “Virkkula et al. (2006) noted chloride losses (relative to Na+) linked with. . .”

Reviewer - p. 10., l. 3: How can winter blizzard conditions be more consistent with the 2ODIAC campaign which happened later in the season?

Authors - The text states “blizzard and strong wind conditions”, where “strong wind conditions” is consistent with what was measured in the Spring (2015) campaign (over 60% of the total time of that deployment seeing winds over 8ms-1). “Blizzard” is left in because that is part of how Hara et al., 2004 describes their results.

Reviewer - p. 12, l. 30: Are those the total numbers of trajectories or just at one release height? Please specify in the manuscript.

Authors - A parenthetical addition has been made to make the sentence clearer: “(37 and 36 days, respectively, at 3 hour intervals for each release height)”.

Reviewer - Figure 5: It would be helpful to make all data points grey and overlay them by wind speed binned averages with standard deviations.

Authors - The figure has been modified as recommended.

Reviewer - p. 14, l. 15f: What was the size range of particles that Hara et al. (2014) measured? From the text it is not evident that the comparison makes sense.

Authors - “primarily in the 500nm - 2μm size ranges” has been added to the text. This is on the upper range of what would be captured by the AMS but should be well encompassed by the Lighthouse OPC used.

Reviewer - Section 3.7: Could the vertical displacement of the back trajectories be an indicator of possible processes? The authors looked at source regions which do not provide a clear hint, but potential influence of high tropospheric or stratospheric air masses could be important.

Authors - The HYSPLIT trajectories examined often showed a lot of movement altitudinally over their lifetimes. However, analyzing how the vertical mobility of an air mass over its lifetime affects the halogen and/or aerosol content is outside of the scope of this manuscript. However, the reviewer brings up an excellent point. The following has been added to the text in S3.7 to make the reviewer’s point known and to point out potential future work: “The results presented here suggest that source regions for air masses do not significantly affect aerosol halogen concentrations in Antarctica. However, we do not here assess what, if any, impact that vertical displacement during transport may have on the halogen concentrations in the aerosol phase. Due to the low concentrations of aerosol-phase halogens measured, the uncertainty in the HYSPLIT runs, and the fact that the air masses examined in most of the HYSPLIT runs had large altitude changes during their back trajectory lifetimes, assessing the effect of vertical

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displacement of an air mass on halogen or aerosol concentrations or compositions is outside of the scope of this manuscript. However, it should be noted that models have shown there to be significant differences in the vertical distribution of gas-phase halogens in the boundary layer (Lehrer et al., 2004; Saiz-Lopez et al., 2008). Any vertical movement of an air mass could therefore have significant impacts on the halogen cycle and halogen-aerosol interactions."

Reviewer - Conclusions: p. 22, l. 8: The mixing state of particles has not been mentioned before in the manuscript. If there is a strong argument for external halogen mixture, this needs to be included in the respective results section. It will also be highly informative to include a couple of sentences what more information of the halogen cycle in Antarctica will be important for.

Authors - The statement about mixing state has been removed since no evidence for it has been presented in the text. The following has been added to demonstrate the importance of understanding the halogen cycle: "A clearer understanding of the halogen cycle over Antarctica will result in better modelling capabilities of the oxidative capacity of the polar troposphere which impacts both current climate (e.g. DMS oxidation) and paleoclimate reconstructive abilities."

Technical Comments:

Reviewer - (Grammatical/Spelling comments:) p. 5, l. 26: "an" instead of "An" p. 7, l. 28: "and sodium my occur in the data." p. 8, l. 19: delete "while in the field and subsequent data analysis" p. 9, l. 6: "it" p. 9, l. 27: "was" instead of "is not clear." p. 12, l. 15: "differences that" p. 13, l. 19: "Two" instead of "Several" p. 13, l. 25: "which in turn is a function: : :" p. 14, l. 14: "These results are: : :" p. 16, l. 9f and elsewhere: The hyphens turn out very long and spaces are missing. p. 17, l. 2: "region" instead of "point". The sources are rather regions that specific points. p. 17, l. 5: Delete "it was quickly noted that" p. 18, l. 5: " that the increased chloride concentrations as measured by the AMS are not: : :" p. 19, l. 13 and elsewhere: if this is the AMS derived ratio use

Chl:Na p. 20, l. 14: “with the AMS” instead of “in the AMS” p. 20, l. 20: “by Maffezzoli” according to the sentence structure p. 21, l. 21: “summer”, l. 26: delete “overall” p. 21, l. 29: Based on AMS measurements

Authors - The above grammatical/spelling comments have all been implemented into the manuscript.

Reviewer - p. 17, l. 33 and elsewhere: sometimes it is written Figure x or Fig. x or figure. Check the journal style.

Authors - All instances of “Figure” have been changed to “Fig.” unless they are at the start of a sentence for consistency.

Reviewer - p. 17, l. 16: replace “in the AMS” by “as measured by the AMS”. The original formulation, here and elsewhere, sound like the species are generated within the AMS.

Authors - There are a few instances where “in the AMS” is the correct usage in context (e.g. S2.3, other occurrences regarding vaporization) but the rest of the occurrences have been changed as suggested.

Reviewer - p. 3, l. 10: elaborate which elements are meant by “marine elements”

Authors - “(Li, K, Mg, Ca, Sr)” has been added to the text to represent what was measured (minus Na since it is a focus here) by Weller et al., 2008.

Reviewer - p. 7, l. 11: “Figure 1 shows how the wind direction and: : :” The wind direction is not shown, but can be added easily.

Authors - “. . .direction and. . .” referred to an older version of the figure and has been removed. Wind direction changes are not relevant to the decay period shown here.

Reviewer - p. 8, l. 1: “freezer in Antarctica.” Admitted, there is a limited total number of freezers in Antarctica, but more specificity is desirable, e.g., the research site’s freezer.

Authors - “in the Crary Lab” has been added to the text.

Reviewer - p. 8, l. 21f: It is not clear what is meant by “increasing temporal resolution patterns.”

Authors - The sentence has been removed because it does not add anything to the context of the rest of the paragraph.

Reviewer - p. 10, l. 24 / p. 11, l. 10-12: Do you mean “SO4 2-“?

Authors - The authors thank the reviewer for this catch; the instances on p.10 and p.11 have been corrected to “2-“

Reviewer - p. 11, l. 2f: The abbreviations have not been introduced.

Authors - The abbreviations have been removed and the sentence changed to read: “This is not in agreement with results from Eom et al. (2016) which measured higher inorganic salt concentrations in summer aerosols by Raman microspectrometry and attenuated total reflection Fourier transform infrared imaging techniques”

Reviewer - p. 11, l. 32: sulphate or sulfate?

Authors - “Sulphate” has been changed to “sulfate” to be consistent with the rest of the manuscript and abide by IUPAC recommendations.

Reviewer - p. 12, l. 29: SI or Supplemental Information?

Authors - “Supplemental Information” has been changed to “SI” for consistency throughout the manuscript.

Reviewer - p. 14, l. 33: It is unclear what is meant by “make up the aerosol to inland continental snow.”

Authors - The sentence has been changed to make the meaning clearer: “. . .may explain longer range transport of chemical species (found primarily in the aerosol phase) to inland continental snow.”

Reviewer - p. 15, l. 27: It is unclear what is meant by " by extension concentrations."

Authors - The sentence has been changed to make the meaning clearer: "...and, by extension, aerosol concentrations."

Reviewer - p. 18, l. 13 and elsewhere: Do you refer to the IC measured species Cl- and Na+? Or the AMS derived species Chl and Na?

Authors - This comment is addressed in the "General Comments" section above.

Reviewer - p. 19, l. 25: Do you mean at intermediate high wind speeds?

Authors - "than at high wind speeds" has been removed to make the sentence clearer. Sentence now reads: "This is near the same wind speed at which snow particle numbers increase more rapidly (7-13ms-1; Fig. 4)."

Reviewer - p. 21, l. 27f: the notation of the chemical species is not clear,

Authors - This comment is addressed in the "General Comments" section above.

âĂČ Response to Reviewer #2

General Comments

Reviewer - The first part of the Introduction section contains an overview of Antarctic aerosol studies, but mainly in instrumental aspect. Given the title/research focus of this manuscript is about blowing snow and relevant aerosol, a brief introduction is needed. For example, on page 3 lines 27-29, it reads 'Some studies in Antarctica have hypothesized that elevated wind speeds can cause sea salt concentration differences through unknown mechanisms', then which studies do you refer to? References should be supplied here. Yang et al. (2008) has proposed a mechanism of SSA production from blowing snow through sublimating saline wind-blown snow particles, which parameterisation has been implemented in global models to investigate high latitude SSA (e.g. Levine et al., 2014; Huang and Jaegle 2016; Rhode et al., 2017). Frost flowers are also thought to be a SSA source. More information can be found in e.g. Abbatt et al., (2012). Yang,

et al.: Sea salt aerosol production and bromine release: Role of snow on sea ice, Geophys. Res. Lett., 35 (L16815), doi:10.1029/2008gl034536, 2008. Huang J., Jaeglé, L., Wintertime enhancements of sea salt aerosol in polar regions consistent with a sea-ice source from blowing snow, ACP, doi:10.5194/acp-2016-972 (2016). Levine, et al.: Sea salt as an ice core proxy for past sea ice extent: A process-based model study, J. Geophys. Res. Atmos., 119, 5737–5756, doi:10.1002/2013JD020925 (2014). Rhodes, et al.: Sea ice as a source of sea salt aerosol to Greenland ice cores: a model-based study, Atmos. Chem. Phys., 17, 9417-9433, https://doi.org/10.5194/acp-17-9417-2017 (2017). Abbatt, et al: Halogen activation via interactions with environmental ice and snow in the polar lower troposphere and other regions, Atmos. Chem. Phys., 12, 6237-6271, https://doi.org/10.5194/acp-12-6237-2012, 2012.

Authors - The authors thank the reviewer for this insightful comment and have implemented it in the text. The text at the end of the introduction now reads: “Proposed mechanisms for enhanced sea salt aerosol concentrations include sublimating saline snow hydrometeors which has is hypothesized to produce sea salt aerosol at an order of magnitude higher production rate than over open ocean (Yang et al., 2008). This ‘blowing snow’ mechanism has been implemented in global models and various implementations have seen varying success in simulating sea salt concentrations (Levine et al., 2014; Huang and Jaeglé, 2016; Rhodes et al., 2017). However, there is the lack of experimental studies investigating the proposed mechanism of sea salt aerosol production from blowing snow, and the experimentally measured aerosol production rates. The results presented here, which vary in temporal resolution from low (days) to very high (minutes), suggest that coastal Antarctic aerosol composition cannot be explained without taking local meteorology (e.g. wind speed and direction) into account. Though the results presented here do not identify specific mechanisms, they strongly suggest that blowing snow may drive the composition dependence and aerosol enhancement observed as a function of wind speed, and further studies are necessary to investigate both production rates and mechanisms.”

Reviewer - Section 3.7 (about bromine): Sander et al.'s (2003) global sea spray dataset clearly shows a general bromine depletion in micron mode, however, in ultra-fine mode, bromine is largely enriched. A similar phenomenon is also seen in polar dataset (Legrand et al., 2016). Do you have any comments on this size-dependent behaviour and relevance to your data interpretation? Sander, et al.: Inorganic bromine compounds in the marine boundary layer: A critical review, Atmos. Chem. Phys., 3, 1301–1336, 2003.

Authors - The authors thank the reviewer for bringing up these points. The following has been added to the text: "As a bulk measurement, mass fractions in the filter measurements are strongly biased towards larger, supermicron particles, whereas the AMS measurements are only sensitive to submicron particles. The below LOD concentrations of Br- in the filters combined with measurable Br with the AMS is generally consistent with measurements of the cycling of inorganic bromine in the marine boundary layer showing depletions of bromine in supermicron aerosol and enrichments in submicron aerosol (Sander et al., 2003). These results are also consistent with measurements at another coastal site in Antarctica (Legrand et al., 2016). This apparent size dependence for bromine in the aerosol phase supports models such as Legrand et al.'s (2016) that implement size-dependent depletion factors for bromine from aerosols. Additionally, the AMS measured Br as a function of wind speed results support the hypothesis that blowing snow provides a source of bromine. The local maxima of submicron Br concentrations above the wind speed threshold for blowing snow may suggest that gas-phase bromine is being liberated and absorbing or heterogeneously reacting onto the submicron aerosol population. Unfortunately, the lack of concurrent gas-phase bromine measurements means these results are not conclusive evidence for these hypotheses but do point out the need for more measurements."

Specific Comments Reviewer - P5L26: 'An' should be 'an'

Authors - This has been corrected.

Reviewer - P6L1-2: ‘50m’ or ‘50mL’? A full stop is needed before the second ‘50 mL’ (?)

Authors - The sentence has been changed to read: “The snow samples were collected when there was adequate snow in 50mL polyethylene plastic centrifuge tubes for future ion chromatography analysis”

Reviewer - P7L28: delete the duplicated word ‘in’

Authors - This has been corrected.

Reviewer - SI Fig.S3: remove duplicated brackets in the caption.

Authors - This has been corrected.

Reviewer - SI Fig. S4: where are ‘squares’? which ‘solid’ line (red or coloured) do you refer to?

Authors - The caption has been corrected to more accurately reflect the figure.

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