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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Biogenic and Anthropogenic sources of Arctic Aerosols" *by* Ingeborg E. Nielsen et al.

Anonymous Referee #1

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This paper represents a significant contribution to furthering our understanding of Arctic aerosol, and how its chemistry evolves over the transition from dark winter to light spring. This work reports on three months (February - May) of high time resolution measurements of PM1 aerosol composition at Villum Research Station, at Station Nord, Greenland. In particular, this paper provides some of the most chemically detailed measurements available of the evolution of organic aerosol chemistry in the High Arctic during the transition between winter and spring. These results are in line with, and add to, our current understanding of Arctic organic aerosol. In the specific comments I have made some suggestions for additional data analysis, and while these suggestions may improve the paper I do not consider them required for publication. The comments and suggestions below are meant to help further improve an already very interesting and thorough paper.

General Comments: (1) The large amount of data and time span of these measurements from winter to spring mean that a number of interesting conclusions and observations are presented in this paper. The paper may benefit from sharpening the focus on what the authors feel are the most valuable or interesting conclusions. The main findings are somewhat obscured in the abstract and conclusions in the current version of the paper. For example, the abstract begins by highlighting the importance of BC aerosol in the Arctic, but by the end of the conclusions it is fairly clear that results related to the evolution of organic aerosol may be a larger outcome of this work. I suggest the authors revise some information in the abstract and conclusions to best highlight the largest conclusions of this work.

(2) The title is currently very general, and does not highlight the main findings of the work. I suggest the authors revise toward a more declarative title that helps highlight their specific findings.

(3) Throughout the paper the text appears to suggest that secondary organic aerosol and marine emissions are mutually exclusive (e.g., L37-38). If by marine emissions the authors mean only sea spray, then I suggest they state this clearly. Marine organic aerosol can arise from both primary and secondary processes, and it is extremely difficult to distinguish unambiguously between primary and secondary marine aerosol. With the evidence presented in this paper, the authors cannot reliably determine if the marine organic aerosol factor they measure at VRS is primary or secondary. I suggest that the authors can acknowledge these challenges while still highlighting the evidence that they do have for each mechanism (e.g., the presence of MSA in the MOA factor showing that secondary chemistry contributes to MOA).

Specific Comments:

L22-27 (and elsewhere): Given that the authors present novel measurements of aerosol chemistry over the period of transition from a dominance of long range transported Arctic Haze to a cleaner regime more dominated by inner-Arctic sources, it may not be particularly useful to quote average concentrations and mass fractions over the whole study period. One perspective is that the most important aspect of these measurements is that they cover this period of transition from anthropogenic to biogenic sources. Further, these mean values are likely dominated by the larger amount of data covering the Arctic haze period, and so are more reflective of the average composition in only on regime of the Arctic atmosphere.

L26: “Arctic Haze leveling-off” may not reflect our current understanding of how source regions and removal changes over this period

L63-64: While it is true that distinct layers of aerosol are often associated with Arctic Haze, it is also true that elevated levels of aerosol pervade through the depth of the troposphere, at least within the polar dome, during this time.

L89: What “this” is could be clarified

L94-95: Another reference that could be included here: Leaitch et al., ACP, 2018 https://doi.org/10.5194/acp-18-3269-2018. The author’s work is an extremely useful follow on from the lower time resolution work on OA at Alert.

L103-104: Revise “marine aerosols is a source of inorganic and organic aerosols” for clarity

L105: Are the “other mechanisms” worth elaborating here?

L108: Other work about sea salt in Arctic regions (e.g., Huang 2017 https://www.atmos-chem-phys.net/17/3699/2017/) may be worth including here

L188-189: The NH4 RIE can often differ significantly from 4, what value do the authors obtain when they calculate NH4 RIE from their NH4NO3 calibrations?

L191-192: While I agree that the Cl family of peaks likely comes from non-refractory chloride, Ovadnevaite et al (https://doi.org/10.1029/2011JD017379) have shown that

the signal for NaCl+ can be used as an indication for the presence of sea salt. Do the authors observe significant NaCl+ in their data set?

L197-199: Why not constrain C1+ to the expected ratio from Regal Black (or other material) and fit C2+ through C5+?

L229-232: The apparent RIE for rBC is a convolution of the true RIE and collection efficiency. Also, uncertainty in the MAC applied to the MAAP data could contribute to this discrepancy. While I don’t dispute the choice the authors have made in scaling SP-AMS rBC data to the MAAP, we know that all these approaches to measuring BC carry uncertainty and the MAAP may not provide the most accurate measurement in the case of an aged, highly internally mixed black carbon containing aerosol. If available, a direct comparison between the SP-AMS and the MAAP during calibration with representative BC materials may be useful here.

L245: How much of an effect does scaling SP-AMS rBC to the MAAP have on the comparison between AMS total PM1 and SMPS PM1?

L247-250: During the beginning few days of the study the SMPS concentration is higher than the SP-AMS concentration, and this is the only period where this appears to be the case. SP-AMS chloride was elevated during this time; was any measurable signal for NaCl+ present at that time?

L291-293: It may be worth clarifying this. Long range transport suggests to me sources far outside the Arctic, but the authors suggest that this is likely not the case after the transition to cleaner conditions. Or, do the authors suspect that the MOA was transported from more southerly marine regions? It may be useful to provide some general indication of the meteorological regime or air mass histories, for example, for before and after the Arctic Haze decreases substantially at Villum.

L300-301: Long range transport from marine regions can mean that a portion of springtime Arctic sulphate is from DMS oxidation (https://www.atmos-chem-phys.net/17/8757/2017/)

L313: Is the contribution of MSA subtracted from reported organics and sulphate?

L315: Is this a statement based only on the measurement period presented in this study. Measurements of MSA at Alert would suggest that DMS peaks later in the summer (Leaitch et al., Elementa, 2013: https://www.elementascience.org/articles/10.12952/journal.elementa.000017/)

L331: Biomass burning may be a larger source to the Arctic than farms

L342: Current evidence suggests that frost flowers may not be an important source of aerosol (see for example, Huang 2017 https://www.atmos-chem-phys.net/17/3699/2017/ and references within)

L351-353: This type of information may belong more in the introduction, rather than the results and discussion

L364-366: The authors need to screen the data based on wind direction, or another measured parameter, before reporting and interpreting R2 values here.

L385-395: A mean pToF size distribution for the Arctic Haze period and the more biogenically influenced period could help with this interpretation of mixing state (and would be very interesting!).

L406: Secondary or highly aged primary organic aerosol, it is difficult to interpret a mechanism based solely on CO2+ contribution alone. I suggest the authors elaborate on how they reached this conclusion

L412: It might be useful to indicate the significance of these correlations in Table 2 in some way (e.g., bold R2 values)

L441-442: It is really difficult to attribute primary or secondary sources from the mass spectrum alone. Marine OA observed at Mace Head is likely a combination of primary and secondary OA. Ovadnevaite et al., GRL 2011

(https://doi.org/10.1029/2010GL046083) state (paragraph 9): “The relatively high amount of oxygenated organics typically indicates a chemical aging of the aerosol [Jimenez et al., 2009] with possible contributions from both oxidation of primary aerosol organics and SOA (secondary organic aerosol) formation.”

L 454-455: “In April, the highest OA concentrations is observed where AOA accounts for around 70% of OA (Figure 4). In May, MOA becomes the dominating OA while AOA nearly disappears.” This seems to be an important point of the paper, which could be further highlighted in the abstract/conclusions and results & discussion sections.

L456-457: While comparing to this Alert study is valuable, Narukawa use a very different method and their data represent measurements from 15 years prior to this study. A direct comparison is difficult to make, but I agree it is interesting despite these differences.

L461-462: Some additional references related to DMS would be useful here. The marginal ice zone is also important for DMS

L465-471: Some more detailed information about the source regions impacting Villum during winter and spring might help this discussion and interpretation.

L473-474: Why speculate about the emergence of a continental factor?

L474: Reporting an overall average MOA fraction here is a bit confusing, since the previous discussion demonstrates its much higher contribution once the AOA decreases.

L475-477: In addition, and perhaps more importantly MOA dominates the organic aerosol mass when the overall concentrations are very low, particle numbers are low, and so cloud condensation nuclei concentrations can be low.

L480-481: Oxidation of DMS and other VOCs would be considered secondary. The wording of this sentence is a bit confusing

L482-483: In Croft ACP 2019, secondary OA accounted for up to half of the summer-time OA, and primary marine OA also contributed. The authors may want to be more clear in their usage of marine OA, primary OA and secondary OA. Marine OA can come from both primary and secondary processes.

L484-487: This introductory information may fit better within the introduction.

L493-495: Comparing to Alert may also be warranted, given its proximity to VRS

L495-500: The authors' clear observations of changing OA character and sources over the winter to spring to late spring transition may be a more important conclusion that that these organic species can be mixed with rBC.

L506-507: The observations presented here cannot unambiguously determine whether AOA and MOA is primary or secondary in origin. The mass spectrum similar to Ovadnevaite 2011, only suggests that the aerosol is marine in origin. More information would be needed to suggest a dominant formation process. While the correlation of AOA with sulphate may suggest secondary processes, this aerosol is also transported over very long distances and so aerosol from somewhat different formation processes may co-vary in time at such a remote location.

L512-514: I agree in general with this statement, but some more information about source regions impacting Villum would go a long way in this interpretation. Further, do the authors have access to CO data that could potentially help to demonstrate the increase in deposition mentioned here? (e.g., see Garrett et al., GRL, 2011 doi:10.1029/2011GL048221)

Figure 3: That the authors observe a distinct HOA factor in Arctic haze that co-varies in time more closely with rBC than with AOA or sulphate is interesting. Intuitively I would expect Arctic haze aerosol to be overall extremely oxidized, though the prevalence of HOA in the dark winter suggests not. Do the authors have specific evidence to show that the HOA was not more regionally sourced than the AOA? Do polar plots of wind direction/speed and PNF factor intensity shed any light on differences in source

regions?

Figure 4: Does the MSA-to-sulphate ratio, and organic-to-sulphate ratio, increase in a similar manner to MOA on a monthly basis?

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