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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 #2

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The study reports on SP-HR-AMS measurements conducted at Villum Research Station in the north of Greenland from February to May 2015. The authors investigate the concentrations and evolution of refractory black carbon (rBC), particulate sulfate (SO4) and organic aerosol (OA). The first half of the manuscript focuses on rBC, the second on OA that was further investigated by conducting positive matrix factorization (PMF). Three factors were identified: hydrocarbon-like OA (HOA) with the smallest contribution, Arctic haze OA (AOA) with the largest contribution and marine OA (MOA).

Detailed measurements of rBC and OA in the high Arctic are rare, especially outside of the summer season. The real strength of this study are the real-time observations during the transition period from winter to spring when sunlight returns and Arctic haze conditions fade. While the authors make this point, they also “dilute” their message

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by putting emphasis on reporting average concentrations for the entire study period, which do not address the environmental change. Generally, this study provides valuable insights into the aerosol chemical composition in the high Arctic and should be published with major revisions as suggested below. General and specific comments are mentioned below, all other comments are highlighted in the attachment.

General comments:

A shortcoming of the study is that it underexplores the HR-AMS data. There is no reporting of hetero-atoms such as nitrogen or sulfur in the OA. The contribution of those as a function of time could reveal more details about the sources of MOA in particular. At the moment only O:C ratios are provided. I suggest exploring also the N- and S-containing contributions to OA. In particular the contribution of MSA should be quantified. MSA is discussed in the manuscript (l. 437ff), but rather superficially. See also respective comment in the manuscript.

The authors mention often the average concentrations of the constituents during the campaign. As mentioned above the real strength of the observations lies in having captured the transition periods and the transition cannot be described by campaign average but should rather be discussed as gradients are differences. How long does the transition take, which markers change first, which ones later, or all simultaneously? I suggest changing the emphasis to transition characterization throughout the whole manuscript. For example: l. 345: here an average BC concentration is mentioned; l. 367: a slope or gradient for the SO4 concentration would make more sense here;

I suggest renaming the title to “Biogenic and Anthropogenic sources of Arctic Aerosols at Villum Research Station”. “Arctic Aerosols” alone is misleading, because the measurements reflect the unique environment of VRS in northern Greenland. That is very different from the Canadian archipelago or Svalbard as the authors write themselves. Along the same line is the inaccuracy with which the authors cite literature in the introduction:

- L. 37: How do the authors define the “Arctic summer aerosols”? Do they mean the high Arctic, so basically the Arctic Ocean? Or do they include terrestrial parts of the Arctic. This makes a fundamental difference for the composition and other properties of aerosols.

- L. 79: This information is incomplete. The paper also states that SO4 decreased significantly in Alert and Zeppelin and that the lack of a trend at Barrow is likely due to the limited data coverage. This information needs to be added.

- L. 86: This article is focused on the Canadian Arctic mostly. Use literature that is more relevant to the entire Arctic. Furthermore, the article has been published in 2019 in ACP.

- L. 112 “DMS emissions in the Arctic have increased by 30 %...” Is this true for the entire Arctic or the Canadian sector? It is important to provide a differentiated picture of what is happening, otherwise false impressions are created.

- L. 114: “demonstrated” is an overstatement, the paper infers. The authors show the relationship but do not provide an explanation.

- L. 115: MSA does not nucleate or form new particles, it rather condenses and grows particles.

- L. 116: It is not only believed that ammonia comes from sea bird colonies, this has been shown multiple times. There are global inventories for ammonia seabird emissions even.

Specific comments:

L. 23: unclear whether the particulate sulfate or PM1 amounted to 2.3 ug / m3

l. 40: Why is it urgently needed to elucidate the chemical components? The authors probably mean that modeling the future of the Arctic requires process understanding. Just because climate is changing doesn’t mean we need highly time resolved aerosol

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data.

l. 45: consider referring to the special IPCC report on 1.5 °C and the AMAP 2015 report on BC and ozone in the Arctic.

l. 52: ice does not condense onto particles

l. 63: Is it truly "visible"? Strong haze events might be visible by eye, but the typical Arctic Haze is still orders of magnitude lower in mass concentrations as the visible urban air pollution, as is somehow inferred by this sentence.

l. 67: As it is written it contradicts above statement that says that Arctic Haze sources are located within in the polar dome. This needs some clarification or more exact formulation.

l. 87: why should vegetation fires not be considerable? It's a question of whether their emissions are transported to the high Arctic.

l. 93: Consider referring also to Chang et al., 2011, ACP doi:10.5194/acp-11-10619-2011They characterize PM1 aerosol measured with an AMS and PMF in the central Arctic during the ASCOS campaign. Also Willis et al., 2018, 10.1029/2018RG000602 provide and overview of what we know about Arctic aerosol and it's detailed composition.

l. 98 ff: This seems to be more a concluding statement which should be placed later. It is a bit awkward after the OA discussion.

l. 108: the explanation why the role is important is missing.

l. 110: Unclear where MSA is increasing.

l. 123: revise the sentence, it is grammatically incorrect and does not list the two disadvantages.

l. 126: delete “and trends”. Trends are longer term changes.

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l. 139: PMF cannot reveal source regions just source types.

l. 153: Where is the HVS data used? This is not evident in the manuscript. If they are used that needs to be stated and then more information like flowrate, sample duration etc. needs to be added, or a references to the supplement needs to be given.

l. 176: “inspected” "inspected" sounds like the flow rate was measured once. I hope it was checked several times during the campaign.

l. 176 if the size calibration was conducted with ammonium nitrate, a DMA must have been operated as well to select a range of sizes. This information is missing entirely.

l. 179: Why was there no determination of the relative ionization efficiency of sulfate with ammonium sulfate?

l. 191: The AMS also sees NaCl, see Ovadnevaite et al., 2012, doi:10.1029/2011JD017379. and other publications. The influence of NaCl needs to be considered as well.

l. 214: add manufacturer and model number of the SMPS.

l. 224: “majority”. Can the authors be more specific and provide the quantiles?

l. 253: the sentence is confusing.

l. 273: “chemical composition” instead of “chemistry”

l. 285: A comparison to other studies is missing that would reveal why the concentration can be perceived as relatively high.

l. 302: What is the role of light here?

l. 303: “at its source region” This should rather read: "in the vicinity of the source region, " SO2 oxidation does not happen immediately and normally SO2 has already been transported away some distance from the source before it is oxidized to SO4 2-

l. 305: Figure 3 is mentioned before Figure 2.

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l. 308: “originating from Siberia” Is this not a contradiction to the main wind direction from the south-west? How representative is the wind direction of the general atmospheric circulation around VRS?

l. 319: How do you define spring season? In my understanding mid-April and later is spring. So the sentence does not make sense to me.

l. 323: Would the pollution from the military not result in a separate PMF factor? Or is the HOA that is long-range transported so similar to the fresh HOA?

l. 331: Is this also true for winter? Are there birds all year around?

l. 335: Add a reference for the longer lifetime.

l. 336: Please correct Cl to Cl- throughout the manuscript.

l. 339: should be chloride and not chlorine

l. 361: Is this true that the sources are the same for the entire Arctic, for all seasons or the Haze period where you have long lifetimes and hence rather well mixed conditions?

l. 364 – 366: To me it doesn’t make sense to include local contamination periods for a general conclusion on rBC and SO4 correlation. I suggest removing the local influence first and then redoing the correlation analysis.

l. 407: “AOA is abundant during February to mid-April. . .” this is redundant. The sentences before that say the same.

l. 421: I cannot follow the argument. What is the contribution quantitatively and what would be expected from the literature? Is the literature appropriate for a comparison?

l. 433: Please be more specific in how far it resembles the Mace Head spectrum.

l. 443: How does the MOA factor resemble HR-AMS spectra from the Southern Ocean? doi:10.5194/acp-13-8669-2013 Can the authors discuss whether the MOA factor is more universal, i.e. VRS, Mace Head, other oceans?

l. 456:What is the lowest concentration of OA?

l. 456: What does “this” refer to? The concentration of OA or the 75 % MOA in the OA?

l. 475ff: This sentence is confusing. I do not understand the main message.

l. 480: “oxidation products of DMS and other VOCs” These are also secondary. The argument does not make sense like this.

l. 481: Âńprimary components including colloidal gels. . .Âż As far as I read the sentence MOA is the specific factor found by the authors using the HR-AMS. So the question is whether the primary compounds like gels would actually be seen in the MOA factor? To my knowledge they evaporate at temperatures higher than 600 °C. This means that generally marine organic aerosol can contain these compounds, but the MOA factor likely doesn’t due to instrumental limitations.

l. 487: enhancement through the lensing effect?

l. 494f: 75 + 3 + 12 + 12 is > 100 %.

l. 503 What does "reduced" mean? The least amount of oxygen?

Figure 2: I suggest to either make the axis logarithmic or but them off at 0.05 (with indicating the true extent of the big peaks) to make the pattern visible. The AOA and MOA spectra are not informative like they are now because on cannot see anything.

Figure 3: I suggest to move the rBC trace up. It’s not visible like this and hence not usefull.

Figures S3: the figures have very low resolution.

Figure S2: The y-axis could start at 0.5.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2019-130/acp-2019-130-RC3-

supplement.pdf

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