Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC2
https://doi.org/10.5194/acp-2020-1270-RC2, 2021
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Comment on acp-2020-1270

Anonymous Referee #1

Referee comment on "Large seasonal and interannual variations of biogenic sulfur compounds in the Arctic atmosphere (Svalbard; 78.9°□N, 11.9°□E)" by Sehyun Jang et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-1270-RC2, 2021

General comments

This manuscript presents a 5-year time series of aerosol properties in spring and summer in the Atlantic Arctic (Gruvebadet station), focussing on the relative importance of natural and anthropogenic sources of sulfate and MSA in aerosols. The study presents an interesting dataset for the understanding of natural aerosol sources during the Arctic summer, which play an important climatic role. The article is generally well written and concise and the measurements reported look technically sound. However the study does not present, to my understanding, relevant conceptual or methodological innovations. In particular, the factors that drive the transition between the Arctic haze season, when anthropogenic pollution dominates sulfate aerosols, and the "clean" summer season, when local biological sources play a more important role, have been known for a long time (as demonstrated by many of the references cited by the authors). This pattern results from the interplay between the seasonal changes in atmospheric circulation, ocean activity and atmospheric photochemistry and condensation sink.

To make the article less incremental and more interesting, in my view the authors should place more emphasis on the intriguing interannual variations, and less on the well-known seasonal shift from anthropogenic to natural aerosols. Interannual changes are, in my view, the most interesting aspect of the dataset, but the authors fail at explaining their causes. A more complete analysis of satellite data, including the use of recently developed satellite algorithms for marine sulfur compounds, combined with air mass back trajectories, could help explain interannual differences. Satellite and air-mass datasets are presented but not fully exploited. In addition, I suggest a more careful consideration of differential sources and sinks of MSA and SO4 in the Discussion, because both sources and sinks modulate the MSA/Bio-aerosol ratio. Less importantly, I prompt the authors to word more carefully some sentences on marine and sea-ice biological activity.

Specific comments

Abstract

L26: Please tone down. Replace “obviously” by something more neutral. Can sea-ice DMS sources by completely ruled out?

Introduction

L42: Acidification appears out of the blue here and breaks the flow. If the authors want to elaborate on the potential impact of acidification on marine DMS emission (for which there is inconclusive evidence), as I suspect, this needs to be better introduced.

L50: Does this conform with more up-to-date references on MSA? Veres et al. 2020; Hoffmann et al. 2016; Dawson et al. 2012 (the three in PNAS). Please review references on MSA chemistry in the rest of the Introduction.

L61: please mention sea ice, where DMS production and emission does also occur. Levasseur et al. 2013 (NatGeo); Park et al. 2019 (ESPI); etc.

L83: Is “Bio-aerosol” an appropriate expression? It seems to disregard non-sulfur biogenic aerosol sources, like VOCs other than DMS and primary organic aerosol. I suggest using a more precise expression.

Results

L168: “a” or “the”?

L171: For clarity, please add “during previous studies” before “Svalbard”.

L190: This is not that surprising. Norman et al. (1999, JGR) already showed a dominant contribution of Anth SO4 all year round at Alert (high Canadian Arctic), with the lowest monthly Anth contribution in August with about 55% (roughly corresponding to 45% DMS contribution). The authors may also want to check Mahmood et al. 2019 (ACP), which compared that dataset to model outputs which showed agreement.

L201: please replace “nearly absent” by something more objective, like a Chl a concentration range.

Discussion

L252: “absence of biological activity”: Even if back-trajectories do not support a sea-ice source, this information needs to be corrected because sea ice can host extremely active microbes (Leu et al. 2015, PiO) which can produce DMS in significant amounts, e.g. Levasseur et al. 2013 (NatGeo); Hayashida et al. 2020 (GBC).

L280: The conclusions of the study of Park et al. 2018, quoted here, relied on a satellite proxy for DMSP-producing phytoplankton. Given that the satellite algorithm was consistent with atmospheric measurements, why not using it again, in combination with air mass back-trajectories, to understand DMS source regions in the current study?

L340: Moffett et al. 2020 (JGR-A) suggested MSA condensation on anthropogenic (fossil fuel combustion) particles. Please revise if needed.

L370: Doesn’t this conclusion contradict previous paragraphs? (eg L340).

L375: please check Moffett et al. 2020 (JGR-A) for time series of MSA and nss SO4 in the Pacific sector of the Arctic (UtqiaÄivik, station formerly known as Barrow).

L398: is the use of a “single ratio” common practice in atmospheric chemistry modelling studies? Please provide references.

Conclusions

L408: can we really assume that the MSA / Bio-aerosol ratio is equal to the branching ratio, without knowing the differences in the sinks? Concentrations in aerosols results from both sources and sinks, which are very likely different for each compound. Please revise.

L420: please check Gali et al. 2019 (PNAS), which seems a relevant reference to support this point.

Technical corrections and typos

L266: “snow”, not plural.