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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Sensitivity of Arctic sulfate aerosol and clouds to changes in future surface seawater dimethylsulfide concentrations" *by* Rashed Mahmood et al.

Anonymous Referee #2

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The presented study uses an atmospheric global climate model to determine the impacts of increased oceanic DMS emissions on the Arctic sulfate aerosol budget, cloud droplet number concentration (CDNC) and cloud radiative forcing. The main finding is that increased wet scavenging of sulfate in 2050 compensates for any increased aerosol production due to higher DMS emissions. Furthermore, significantly higher CDNC and more negative cloud radiative forcing are found in 2050 because of higher nucleation rates. The first half of the paper describes the comparison of a historical simulation (1991-2003) with observations from ship-based campaigns and at Alert in the Canadian Arctic. The second half of the paper reports results from a simulation experiment for 2000 and 2050. The illustrations and visualization of results adequately

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Discussion paper

reflect the key findings of the analysis. The first half is rather well presented and written in a comprehendible way. The second half (Sect. 4) is very difficult to evaluate because of a lack of information and a confusing description of the simulations. The manuscript should have a section (for example after Sect. 2) that provides an overview of all details and motivation behind the simulations in one place. Moreover, it is unclear why the historical simulation was not used as present-day reference simulation for the future runs, but instead a new one, “2000”, integrated over four years, is introduced on page 7. How can we be sure that “2000” has the same robustness as the historical simulation?

Scientifically, the study is questionable due to three major issues:

1) The future state of the atmosphere in 2050 is derived from integrated simulations of four years, 2048-2051, which seems to be too short. It appears that the climate simulation was meant to follow the RCP8.5 emission scenario. Have atmospheric concentrations of CO2 and other trace gases as well as the global air temperatures in 2050 reached levels that are comparable to multi-decadal climate simulations for RCP8.5? In case this has been achieved during the 4-year run: has the output from the spin-up period been excluded from the analysis?

2) Since the increase of CDNC due to increased emissions of DMS in 2050 compared to 2000 was significant, I would expect that the cloud microphysics are likewise significantly influenced by the higher number of cloud condensation nuclei (CCN), as this has been demonstrated for the summertime Arctic (Leaitch et al., 2013). If a cloud forms on a higher number of CCN the condensed water will be distributed over many small droplets rather than over a few large ones, given that the available amount of water is the same. An increase in CCN concentration results in faster evaporation rates owing to smaller cloud droplets. The faster evaporation rate leads to enhanced entrainment of sub-saturated air surrounding the cloud and a decrease in cloud fraction, in turn lowering the aerosol effect on cloud albedo (Zuidema et al., 2008). Have such aerosol-induced changes on the cloud macrophysics be considered?

3) Sea-salt particles and primary organic particles could be much more efficient CCN than particles derived from DMS (Quinn and Bates, 2011). If the sea salt or organic particle was already sufficiently large to serve as a CCN, the addition of DMS-derived sulfur to the particle will not increase the number of CCN. The increase of the primary sea-spray emissions with retreating ice, would not just be compensated by increased wet scavenging, but might outcompete DMS as precursor for CCNs. Clearly, increased emission of sea-salt aerosol will inhibit the development of precipitation. It will also cause more large CCNs, which can efficiently suppress activation of some of the smaller (sulfate) particles (O'Dowd et al. 1999).

Specific Points:

- P.1 line 41: Mention that gaseous MSA also nucleates and plays an important role in the initial growth of new particles. Importantly, a recent study in the Canadian Arctic Archipelago by Willis et al. (2016) presents observational evidence that the growth of nucleation mode aerosol in the summertime Arctic is correlated with the presence of particulate MSA and organic species.

- P.2 line 71: DMS in water or in air. Suggest to denote seawater DMS as DMS(aq) and gaseous DMS as DMS(g).

- P.3 line 98: How much of the produced DMSP is transferred to sediments? Does is not depend on grazing pressure how much DMS is actually produced? How sensitive are diatoms and haptophytes to seawater temperature changes?

- P.3 line 108: Please explain why a factor of 10 is used. Are there any projections about future DMS emission (from the same water column, not due to ice loss) that justify this order of magnitude increase?

- P.4 line 127: How well does Piecewise Lognormal Approximation cope with newly introduced particles from nucleation?

- P.4 line 130: In high latitude regions - characterized by low temperatures and high

wind speeds - estimated DMS transfer velocity from wind speed parameterizations will be biased high if only the Schmidt number normalization is used.

- P.4 line 131: Does the model include oceanic emissions of sea-salt particles and primary organic particles? If not, that must be stated here.

- P.4 line 134 - 135: "MSA is treated as sulfuric acid in model for simplicity" - this is problematic in multiple ways. MSA forms in one step from the oxidation of DMS. Given it nucleates as sulfuric acid, then the atmospheric nucleation would be much too efficient. Please provide the average nucleation rates of Table 2 with literature data from the Arctic, e.g. Karl et al. (2012) and Leaitch et al. (2013). Although MSA is much less efficient than sulfuric acid in forming particles with water molecules, several laboratory studies and computational studies confirmed that MSA forms particles with alkylamines (Dawson et al., 2012; Chen et al., 2015; Xu et al., 2018). The presence of water seems to control the new particle formation in this system.

- P. 6, line 190 - 194: What explains the high modelled biogenic sulfate in June in Fig. 2c? Can it be related to the DMS seawater concentrations?

- P. 6, line 205: Please explain the occurrence of the October peak in the annual cycle.

- P. 6, line 217: Please provide more details about the radiative flux calculations in Sect. 2. Which parameters were perturbed? A table with parameters and perturbation values for all 25 simulations would be very helpful.

- P. 7, line 241: Explain better what "corresponding simulation" means here.

- P. 7, line 242-244: An explanation is missing here, why a new simulation "2000" and not the historical simulation "hisCont" was used as present-day reference.

- P. 8 line 286: There could be two reasons why wet deposition increased, growth of particles to larger sizes by condensation of DMS oxidation products which makes them more accessible to wet scavenging or the increase of precipitation rates. Please provide information on the average precipitation rates in the simulations in Table 2.

Also give the average liquid water content of clouds in Table 2.

- P. 8 line 295: Is “10ˆ7 mˆ-3” the number change or the absolute number of CDNC? Please set the value in relation to average CDNC in the present-day simulation.

- P. 11 line 375: How much does in-cloud sulfate production increase? More details on aqueous-phase production of sulfate in the model and about treatment of in-cloud scavenging should be provided in Sect. 2.

- P. 11 line 390 - 395: Could you elaborate on the expected feedbacks due to increased SST? Warmer water would be less favorable for diatoms but the solubility of DMS would be lower.

- P. 11 line 396-397: Steady-state atmospheric oxidant concentrations are not the only additional uncertainty. The assumptions on nucleation rates and in-cloud scavenging of sulfate seem to be critical for the conclusion of this study.

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