

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 reflect the key findings of the analysis. The first half is rather well presented and written in a comprehensible 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?

Response: First we would like to thank the reviewer for providing a detailed evaluation of our work which lead to an overall improvement in the documentation of the results. All referee comments are answered below following original comments. For the referee questions regarding different simulations for model evaluation and sensitivity analysis we provide explanations in the following two points:

1) We keep model evaluation section (i.e. Section 3) separate from sensitivity simulations (i.e. Section 4) in order to avoid confusion. The long-term simulations discussed in section 3 (i.e. "hisCon" and "hisNoDMS") were necessary in order to perform a comprehensive evaluation of model simulations of sulfate aerosol and contribution of DMS to total sulfate aerosol over global ocean regions. Comprehensive shipboard observations of sulfate aerosols over global ocean regions are available from 1991 to 2003, which we consider to be important for model evaluation. Therefore, in order to provide an estimate of DMS contribution to total sulfate aerosols, in hisNoDMS simulations DMS was set to zero over global oceans.

2) For the model sensitivity simulations, we focused on the Arctic region by changing DMS only in the region north of 60N. These simulations were run for four years using present-day conditions (i.e. 1998-2001) and potential future conditions (i.e. 2048-2051) with a total of 50 (four year) simulations. Due to such a large of number of model simulations, needed to better understand influence of different DMS scenarios under present-day and future conditions, it was necessary to limit the simulation time period to a total of 4 years for each of the ensemble members for present-day and future conditions. The simulation time period for present-day conditions overlaps with the time period of the hisCon and hisNoDMS simulations. Since we did not change any physical parameters or boundary conditions between model evaluation simulations (discussed in section 3) and the sensitivity simulations (discussed in section 4), the simulated aerosol concentrations are fully consistent with each other in the different types of simulations.

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 CO<sub>2</sub> 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?

Response: For model boundary conditions for 2050 we used mean simulated sea ice and sea surface temperatures from a large 50-member ensemble which was conducted using the coupled version of the model (CanESM2), performed for CMIP5. Given the very large size of this ensemble, impacts of simulated natural variability on mean simulated sea ice and sea surface temperatures are negligible (Sigmond and Fyfe, 2016). The exact same greenhouse gas concentrations and emissions are specified for each individual ensemble member according to the RCP8.5 scenario. In order to further minimize the impact of natural variability in atmospheric and aerosol microphysical processes in simulations with CanAM, we use mean results from 5 different CanAM ensemble members with the exact same boundary conditions and baseline emissions. Ensemble members were generated by introducing random perturbations in radiative flux calculations which leads to small differences in meteorological conditions for each ensemble member. Similar to the approach used in comparable aerosol modelling studies using CMIP5 data (e.g. Ekman, 2014), our method ensures that ensemble mean results are robust and consistent with the boundary conditions and emissions that were used in the simulations. Please note the quantification of statistical uncertainty of the method in Figures 5-8. Based on these results we believe that we used a sufficiently large number of simulations in order to reliably address Arctic climate impacts of DMS emissions in our study.

Regarding the comment about spin-up period: We mention in the manuscript that the model spin-up years (1998 for 2000 and 2048 for 2050) were not included in 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?

Response: In grid cells that are affected by clouds, CanAM4.3 accounts for cloud albedo and lifetime effects (1st and 2nd aerosol indirect effects) as well the semi-direct effect. Parameterizations of droplet evaporation in the model do not account for aerosol effects, similar

to CMIP5 climate models. We agree that the representation of aerosol/cloud interactions is uncertain in climate models and would refer to the IPCC WG1 assessment for a summary of the scientific understanding of the impact of these processes on global climate. Although aerosol indirect effects are very difficult to constrain, some studies based on observations and cloud-resolving modelling indicate that cloud microphysical processes may produce negative or positive radiative forcings, depending on the meteorological situation and nature of the clouds (Stevens and Feingold, 2009). However, indirect effects in climate models are consistently associated with negative radiative forcings. A review of this topic is outside the scope of the current study.

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).

Response: We agree that both sea salt and primary organic particles could be important sources of CCN. However, there is no consensus on the CCN activity of sea spray aerosols (including primary organic aerosols and sea salts) (Neukermans et al., 2018). Based on historical shipboard observations, Quinn et al. (2017) concluded that a small fraction of marine cloud condensation nuclei are made up of sea spray aerosol especially in regions north of 60N. Leaitch et al. (2016), based on recent observations in the Arctic region, also found that small particles (up to 20nm) are activated in summer. Similarly, Collins et al. (2017) reported frequent occurrence of activation of ultrafine particles in the Canadian Arctic Archipelago.

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.

Response: Agreed, this is now mentioned in the text which reads as "*Willis et al. (2016) found that gaseous MSA may also play an important role in the initial growth of new particle formation.*" Further, please note that MSA is treated as sulfuric acid in model for simplicity, as indicated in the manuscript. Hence, we may be able to account for some of the impacts of MSA.

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

Response: Thanks for noticing the omission. We changed the sentence to clarify that the reference is to surface seawater DMS concentrations here. We believe that our current notation

of referring to surface seawater DMS concentrations (instead of DMS(aq)) is sufficiently concise and we would prefer to keep this approach.

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

Response: The questions posed here are well beyond the scope of our paper. We are well aware of the uncertainty surrounding estimates of future DMS emission in the Arctic, and this is why we tackle the problem using a sensitivity analysis with a wide range in sea-surface DMS concentrations (see next question). This being said, we will briefly reply the reviewers' questions below.

First of all, the reviewer should note that DMSP is not explicitly represented in our model. Rather, sea-surface DMS fields are prescribed from either a climatology or in situ measurements (Lana et al. 2011) or satellite-based estimates (Galí et al. 2018). The latter satellite algorithm does estimate DMSP from phytoplankton biomass, and subsequently DMS. As shown by Galí et al. (2015), this is possible because DMS concentrations adjust rapidly to changes in the plankton community, due to the short turnover time of DMS (1-2 days).

Regarding cell sinking: It is very unlikely that this process would significantly impact DMS(P) budgets due to the following two reasons. First, because high DMSP producers are generally small-celled and do not sink appreciably (except for colonial *Phaeocystis* blooms), i.e., they would sink at less than 1 m/d (in addition, they are motile!). In a mixed phytoplankton population, only diatom cells larger than about 30  $\mu\text{m}$  would sink at appreciable speeds of  $>2 \text{ m d}^{-1}$ , but diatoms generally are low DMSP producers and make a minor contribution to total DMSP stocks (McParland and Levine 2018). In consequence, DMSP turnover due to cell sinking out of the surface layer has a much longer turnover time than DMS production from DMSP degradation, which is typically 1-2 days (specific rates of 50% to 100%  $\text{d}^{-1}$ ). Lizotte et al. (2008) found that DMSP turnover due to sinking was  $<2\% \text{ d}^{-1}$  in a North Atlantic diatom bloom.

Regarding grazing pressure: again, this cannot be explicitly accounted for in our study. We refer the reviewer to the study of Galí et al. (2015), which showed how sea-surface DMS can be estimated from environmental variables (chiefly light, as done in the Galí et al. 2018 algorithm). In this approach, food-web interactions are not explicitly represented, but are partly accounted for in an implicit way through the light-mediated seasonal changes in the plankton community structure.

Regarding the last question: Future changes in the dominance of haptophytes vs. diatoms are difficult to predict. Yet, there is evidence for Atlantification of some Arctic sectors (Barents Sea) with northward propagation of coccolithophore (*Emiliania huxleyi*) blooms following polar front (e.g. Neukermans et al. 2018). (see next question).

- 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?

Response: We did address this issue in detail in the introduction section of the manuscript. We understand that it is difficult to be certain about future Arctic seawater DMS concentrations. Some of the discussion that is included in the introduction section is given here:

*“Surface seawater DMS concentrations measured in the Canadian Arctic in July and August of 2014 and 2016 were substantially higher than those used by Lana et al. for July and August (e.g., NETCARE median concentrations of 4.4 nmol/L and 7.3 nmol/L, Martine Lizotte, personal communication; median concentration range from 0.5 to 4.4 nmol/L for Lana et al., <https://saga.pmel.noaa.gov/dms/>). Furthermore, melt ponds on sea ice represent a yet missing source of DMS in studies of the Arctic (Mungall et al., 2016; Ghahremaninezhad et al., 2016; Gourdal et al. 2018; Abbatt et al., 2018).*

*Long-term observational studies provide evidence that high DMSP-producing haptophytes are becoming more prevalent in the Arctic in the last decade (Winter et al., 2013; Nöthig et al., 2015; Soltwedel et al., 2016). Furthermore, Arrigo et al. (2008) suggest that primary productivity may increase more than 3 times compared to 1998-2002, if Arctic sea ice loss continues. A combination of a shift in the species composition and an increase in primary productivity (e.g. Yool et al., 2005; Vancoppenolle et al., 2013) could imply a multiplicative increase in surface seawater DMS concentrations in future climate.”*

Furthermore, projections based on extrapolation of satellite-based estimates suggest a 2-3 fold increase in Arctic DMS emission (north of 70N) for an ice free Arctic summer (May to August). This is quite uncertain and does not factor in changes in species distribution (Gali et al., 2019, submitted).”

We are not aware of any projections or other studies that would provide evidence for lower uncertainties in surface seawater DMS concentrations. In general, information about uncertainties in future Arctic surface seawater DMS concentrations is very limited in the available literature.

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

Response: The model simulates binary homogeneous nucleation of sulfuric acid and water vapour. Newly formed particles grow by condensation and coagulation. The numerical treatment of these processes is highly accurate and compares well with other methods (von Salzen, 2006). Unfortunately, we are not aware of any measurements that would allow us to validate the representation of newly formed particles from nucleation in the Arctic. However, simulated concentrations of CN, CCN, and CDNC are realistic for the small number of available observations in the Arctic. We believe that further analysis is required in order to validate simulations of nucleation mode aerosol in the model, which is beyond the scope of this study.

- 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.

Response: Tesdal et al. (2016) considered the impact of different gas transfer velocity parameterizations and found that differences in these parameterizations lead to considerable uncertainties in global DMS fluxes. Based on the results of Tesdal et al. (2016), we selected a parameterization that seems to produce realistic fluxes of DMS in combination with the Lana et al. surface seawater DMS climatology. For instance, comparisons with observed sulfate concentrations in Fig. 1 and 2 produce reasonable agreement. Unfortunately, we are not aware of any additional measurements that we could use to directly validate the simulated DMS fluxes in the model.

We are not aware of any particular biases in transfer velocities in the Arctic. The Arctic summer seems to be characterized by relatively moderate to low wind speeds (see e.g. Hughes and Cassano, 2015) compared to other regions of the ocean. Regarding temperature, the Schmidt number ( $Sc$ ) already includes temperature effects on gas diffusivity ( $Sc$  is defined as the ratio between the kinematic viscosity of seawater and DMS diffusivity) (Wanninkhof et al., 2009). Indeed, this results in low sea-air gas transfer coefficients in the Arctic, particularly in seasonally ice-covered waters which have low temperatures during most of the summer.

Finally, note also that the effects of ice shear on interfacial turbulence might also alter sea-air transfer  $k$  in the Arctic, causing departures from relationships based on wind speed. However, contradicting results have been reported, such that gas exchange was found to be either lower (Van der Loeff et al., 2014) or higher (Loose et al., 2014) than expected based on a linear scaling to percent ice cover. Therefore, it is reasonable to scale  $k$  by ice fraction.

- 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.

Response: Yes, the model includes oceanic emissions of sea-salt particles but there are no emissions of organic aerosol species from the ocean. We added this information to the manuscript at the end of this paragraph.

- 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.

Response: The nucleation rates given in our Table 2, seem to be comparable with Karl et al. (2012, with values 0.04 to 0.1 cm<sup>-3</sup> s<sup>-1</sup>), however we only have vertically integrated nucleation rates available from the model while Karl et al. reports near-surface values based on shipboard observations. We agree that the treatment of MSA is very simple. We currently don't have the capability to simulate MSA and adopted this approach in order to account for an enhancement in nucleation rates due to a combination of binary homogeneous nucleation of MSA and water vapour (inefficient) and new particle formation in the alkylamine/water vapour system. Alternatively, omitting MSA would likely lead to nucleation rates that are too low.

- 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?

Response: There could be several explanations for this model bias. Generally, the model seems to overestimate biogenic source contributions for all months at this location, especially in June. This is consistent with biases at Alert according to Fig. 1.

We suspect that the biases may depend on the location of the comparison. Results in Fig. 1 indicate that sulfate concentrations agree better with observations in other regions of the ocean. As explained in the introduction and also shown by Tesdal et al. (2016), surface seawater DMS concentrations are particularly uncertain in the Arctic and therefore we cannot rule out biases in Arctic DMS emissions. In addition, the use of climatological oxidant concentrations may be problematic. Furthermore, deposition processes are uncertain in the Arctic which leads to large differences in simulated Arctic aerosol concentrations in different models (e.g. Mahmood et al., 2016). Further investigations are needed in order to understand the causes of the biases, which is beyond the scope of this paper.

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

Response: We are not aware of an explanation from earlier studies. Sharma et al. (2012) showed that MSA concentrations at Alert are anti-correlated with sea ice fraction. It is possible that the peak in October is related to increased fluxes of DMS into the Arctic atmosphere due to the minimum in sea ice fraction in September. It is also possible that DMS is transported to Alert from lower (subpolar) latitudes, where fall phytoplankton blooms are a dominant feature of the marine ecosystem (Ardyna et al., 2014). In any event, the focus of the study is on annual mean results and we have not investigated this.

- 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.

Response: Perturbations to model variables are used in order to introduce random variability in climate model results in order to assess natural atmospheric variability in model ensembles. Different techniques are used by the climate modelling community. Lifetimes of frontal system

and, more generally, time scales associated with conversion of heat and moisture in the atmosphere are typically much shorter than the model spinup time period. Consequently, any perturbation to initial conditions in the atmosphere, whether radiative fluxes, winds or temperatures are perturbed, will produce statistically indistinguishable variations in meteorological variables at the end of the model spin up time period. This is a consequence of the highly non-linear atmospheric system, which is well documented in the literature. A review of ensemble modelling techniques and details of the well-established perturbation method employed in our model does not seem appropriate to us in the context of our study.

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

Response: We have tried elaborating on this in the manuscript so that the relevant sentence now reads: *“Simulated horizontal wind and temperature in each individual member of an ensemble (i.e. 5 separate simulations) were nudged towards specified results from a corresponding simulation (i.e. separate free running model simulation) with CanAM4.3 using a nudging time scale of 6 h.”*

- 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.

Response: This has been answered in detail as response to the general comment above.

- 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.

Response: We did provide information on several parameters including precipitation, wet deposition and liquid water content of clouds in supplementary materials (see Tables S1-S3 and Figures S1-S2).

- P. 8 line 295: Is “ $10^7 \text{ m}^{-3}$ ” the number change or the absolute number of CDNC? Please set the value in relation to average CDNC in the present-day simulation.

Response: It represents maximum change in CDNC relevant to CNTRL run which is important to explain quantitative changes shown in Figure 6. Average CDNC in relation to control runs is already provided in Table 2 both for present-day and future simulations.

- 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.

Response: Information about in-cloud sulfate production was provided in supplementary materials (please see Fig. S3). For details regarding parameterizations of aerosol and cloud



interactions in the model please see von Salzen (2006) and von Salzen et al., (2013), which are also cited in the paper.

- 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.

Response: Lower DMS solubility enhances its flux to the atmosphere. However, this has a marginal effect on seawater DMS concentrations because the latter are set by the dynamic equilibrium between sources and sinks, and ventilation is generally <10% of DMS sinks in the upper mixed layer of the ocean (e.g. Galí et al. 2015). Therefore, ventilation generally does not control sea-surface DMS concentrations.

- 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.

Response: We agree, the final paragraph of the conclusion section is modified and now reads as: *“The model simulations used in the current study are not interactively coupled with ocean and sea ice DMS and therefore rely on specified surface seawater DMS concentrations. The current model version does not account for increases in sulfuric acid condensation sink due to increased emissions of sea salt and organic aerosols from the open ocean, which may lead to overestimates in nucleation rates in the simulations. Additional uncertainty in the strength of the feedback arises from the fact that atmospheric oxidant concentrations are assumed to be steady in our study. More comprehensive assessments of the strength and impacts of DMS/climate feedbacks in the Arctic will become possible once a new generation of Earth System Models with interactive ocean and sea ice DMS, chemistry, and climate processes becomes available.”*

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