Anonymous Referee #1
Received and published: 12 November 2018

This study concerns the impact of Arctic DMS emission on sulfate aerosol concentrations and cloud formation under present and future sea-ice conditions. The authors use a model nudged to winds and temperature for year 2000 and 2050 with various assumptions for DMS concentrations. DMS missions are highly uncertain and modeling studies of the impact of DMS emissions are still few, and I think this study makes a useful contribution to the field. The perturbations of DMS concentrations are quite high, but the authors argue well that such high perturbations are needed. The conclusion about the negative feedback loop along with figure 8 are interesting findings. I would recommend this manuscript for publication after some clarifications given below sorted by line number:

Response: We are very thankful to the reviewer for reviewing our paper and providing very useful comments that lead to the overall improvement of the manuscript. Replies to the referee's comments are given below following the individual comment.

L40: Could you give examples of such favorable conditions in the atmosphere and ocean?

Response: This includes all processes enhancing turbulence at the air-water interface: wave break (in turn depending on wave types), bubbles, surface films, friction velocity, humidity and temperature gradients, convection, and ice shear (Jahne et al., 1987; Merlivat and Memery, 1983; Memery and Merlivat, 1985; Monahan and Spillane, 1984; Wanninkhof et al., 2009; Loose et al., 2014; van der Loeff et al., 2014). Please see our edits in the manuscript.

Although gas exchange is generally parameterized as a function of wind speed for convenience, this approach is clearly an oversimplifcation.

L51: Could you add why there is little evidence under present climate conditions?

Response: A review of several years of observations, laboratory experiments and modelling studies was reported by Quinn and Bates (2011) who argued that the sources of cloud condensation nuclei in marine boundary layer are numerous and the response of clouds to changes in aerosols are more complex than previously thought at the time of CLAW hypothesis. This is now reflected in the manuscript.

L52: Which important feedback loops exists in the Arctic?

Response: As found by Leaitch et al. (2013), DMS is an important source of new particle formation during cleaner Arctic summer. These newly formed particles influence cloud albedo through cloud microphysical processes and thus can influence local radiation budgets and feedback mechanisms.
Please also see our edits at lines: 58-64

L57: Again, why would the emissions be enhanced particularly in the Arctic?

Response: The loss of Arctic sea-ice allows further penetration of sunlight into surface ocean water (e.g. Nicolaus et al., 2012) that can increase net production of algae and phytoplankton (e.g. Arrigo and van Dijken, 2015). In addition, when sea-ice is melted the surface ocean water is more prone to wind stress that can enhance air-fluxes of trace gases (e.g. Bates et al., 2006; Ardyna et al., 2014). However, as discussed in the introduction section of the paper, future changes in net primary production and surface ocean water DMS concentrations are uncertain. Please see our changes in the manuscript. Lines: 68-74.

L63: How is this shown to be problematic?

Response: This has been elaborated in the subsequent lines which read as follows:

"*The small number of measurements from other locations in the Arctic is problematic, as recent research in the NETCARE network (Abbatt et al., 2018) has shown. 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). Gali et al. (2018) argue that biases in the climatology by Lana et al. arise from the application of objective interpolation procedures to a limited amount of measurements. Consequently, Arctic DMS concentrations based on Lana et al. (2011) differ substantially from those of an earlier climatology (Kettle and Andreae, 2000), ocean biogeochemical models, and DMS parameterizations (Tesdal et al., 2016a), indicating large uncertainties in estimates of surface seawater DMS concentrations*."

L71: How do the Arctic DMS concentrations from Lara et al. 2011 differ from previous studies?

Response: Annual mean concentration differences between Kettle and Andreae (2000) and Lana et al. (2011) include zonal mean higher concentrations in Kettle and Andreae (2000) in polar regions of both hemispheres. For the Arctic region (>60N) the emission fluxes are 25% less in Lana et al. (2011) using parameterization of Nightingale et al. (2000).

L111: I am a little confused about these sensitivity tests: are they linked to your study or are they different? If the latter; what did they show and how is your study different from them?

Response: Our study is completely independent of previous studies in terms of model used for experiment, the study region, and different perturbation scenarios/experiments. While our study focusses on the Arctic region, Fiddes et al. (2018) focussed on southern hemisphere and Grandey and Wang (2015) performed experiments relevant to artificial cooling effects as a potential geo-engineering solution to global warming. In our study we analyzed the sensitivity of sulfate aerosol concentrations and changes in cloud microphysical properties using different scenarios of ocean DMS concentrations. As mentioned in the introduction, we used a state-of-the-art climate model and the experiments are described in detail in the following sections.

L158: Testdal/Tesdal reference
Response: Thank you, this is corrected.

L160: Would the nudging to temperature and winds influence how the perturbed DMS emissions impact clouds compared to running with free meteorology?

Response: No, this is not the case. We previously investigated the impact of different nudging strategies on simulated radiative forcings and compared with results from simulations without nudging. Based on these tests, we are confident that the simulated radiative forcings are meaningful. Only emissions of DMS are perturbed, which affects sulfate aerosol concentrations but does not lead to significant changes in concentrations of other radiative forcing agents in the model. Changes in sulfate concentrations lead to changes in cloud albedo via the first indirect effect in the simulations. Atmospheric temperatures and winds in the simulations are already strongly constrained by the use of specified SSTs and sea ice fraction in the model, which allows us to study the impact of DMS emissions for specified climate conditions for present-day and in the future. We do not apply any nudging to the simulated specific humidity and cloud water content so cloud radiative forcings are not directly affected by nudging. Diagnosed radiative effects from our study provide a foundation and benchmark for future studies with fully coupled Earth System Models which can be used to simulate feedbacks in sea ice and global climate to changes in DMS emissions.

L185: 'good agreement': a bit vague; could you add a number here?

Response: This sentence has been removed. The numbers for model and observation comparisons are provided in the same paragraph and the two subsequent paragraphs.

L231: Why did you choose this number (16.9nM)?

Response: This number is based on grid-point average of 10*Lana etal. Climatology – please see Figure 3 for reference. The number is arbitrary and provides an additional scenario for testing the sensitivity of forcings under uniform surface seawater concentrations of DMS and also provides a test of linearity of such responses.
Please also see our changes in the manuscript at lines: 262-265

L247: Can you also add that you are using the sulfur emissions from RCP8.5 as well

for year 2050? (if that is what you are using?)

Response: As described in lines 141-143, the aerosols and precursor emissions are from RCP4.5 for future simulations.

Figure 4-8: it is a bit difficult to separate the lines for the different runs in the panel which shows the zonal means. Also; why do you show 60N-90N for fig 4, and NH for the others? As far as I can see, you don't discuss the results south of the Arctic, so I suggest only showing the Arctic latitudes -and make the plots larger, the lines thicker and/or different colors to make it easier to distinguish the lines.

Response: We Agree and appreciate the referee for these suggestions. The figures have been revised accordingly.

L274: Can you add the numbers for increased precipitation and wet removal?

Response: This is included in Table S1 of the supplementary materials file.

L281: How large are the reductions?

Response: These are summarized in Table S1.

L290: Do you think you would get a significant signal if the run was longer?

Response: Given the large variability in CDNC in the simulations it seems unlikely that we would get a statistically significant difference in CDNC even if we extended the simulation by several decades. Although it would be possible to increase the significance of the results by extending the runs, this is not feasible from a practical point of view given the considerable costs of running the model. Also, this may not yield much new information since the differences in CDNC are likely to be small given that changes in DMS emissions are small. Furthermore, as we show in Fig. 8, we are able to obtain a robust relationship between cloud radiative forcing and DMS emissions despite relatively large variability in the results for near-surface CDNC.

L307: Could you remind us of what the condensation sink is in the model?

Response: Sulfuric acid gas condenses efficiently at the surface of aerosol particles owing to the low volatility of the gas. The rate of gas-to-particle transfer by condensation is determined by rates of diffusion and surface accommodation. If the rate of condensation of sulfuric acid is reduced, e.g. in an environment with low aerosol surface area, the formation of new aerosol particles is increased according to binary homogeneous nucleation theory (if nothing else is changed). We added a brief explanation in the text.

L382: 'However, in the future': change to 'for year 2050 simulations' or similar?

Response: This is changed as suggested, “for 2050 simulations”. Thank you.

L399: Can you add a couple of sentences about the main uncertainties in your model + set-up and link this to the last sentence?

Response: Agree. The last paragraph of the manuscript is revised to incorporate model uncertainties, the paragraphs reads as:

“*The model used in the current study is not interactively coupled with ocean and sea ice DMS and therefore rely on specified surface seawater DMS concentrations. The increases in sulfuric acid condensation sink due to increased emissions of sea salt and organic aerosols from the open ocean are not accounted for in the current version of the model, 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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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 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?

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 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?

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.
Please see our edits at lines 280-289.

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.

Please also see our edits at lines: 171-177.

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.
Please also see our edits at lines: 446-453

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 is 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 of 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 µm would sink at appreciable speeds of >2 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% d^{-1}). Lizotte et al. (2008) found that DMSP turnover due to sinking was <2% 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 hux.) 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.

Please also see our edits at lines: 146-148 in the manuscript.

- 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^{-3} s^{-1}), 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.
Please also see our edits at lines: 233-237.

- 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 "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.

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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Sensitivity of Arctic sulfate aerosol and clouds to changes in future surface seawater dimethylsulfide concentrations

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Abstract. Dimethylsulfide (DMS), outgassed from ocean waters, plays an important role in the climate system, as it oxidizes to methane sulfonic acid (MSA) and sulfur dioxide (SO_2), which can lead to the formation of sulfate aerosol. Newly formed sulfate aerosol resulting from DMS oxidation may grow by condensation of gases, in-cloud oxidation, and coagulation to sizes where they may act as cloud condensation nuclei (CCN) and influence cloud properties. Under future global warming conditions, sea-ice in the Arctic region is expected to decline significantly, which may lead to increased emissions of DMS from the open ocean and changes in cloud regimes. In this study we evaluate impacts of DMS on Arctic sulfate aerosol budget, changes in cloud droplet number concentration (CDNC), and cloud radiative forcing in the Arctic region under current and future sea ice conditions using an atmospheric global climate model. Given that future DMS concentrations are highly uncertain, several simulations with different surface seawater DMS concentrations and spatial distributions in the Arctic were performed in order to determine the sensitivity of sulfate aerosol budgets, CDNC, and cloud radiative forcing to Arctic surface seawater DMS concentrations. For any given amount and distribution of Arctic surface seawater DMS, similar amounts of sulfate are produced by oxidation of DMS in 2000 and 2050 despite large increases in DMS emission in the latter period due to sea ice retreat in the simulations. This relatively low sensitivity of sulfate burden is related to enhanced sulfate wet removal by precipitation in 2050. However simulated aerosol nucleation rates are higher in 2050, which results in an overall increase in CDNC and substantially more negative cloud radiative forcing. Thus potential future reductions in sea ice extent may cause cloud albedos to increase, resulting in a negative climate feedback on radiative forcing in the Arctic associated with ocean DMS emissions.

1 Introduction

Dimethylsulfide is produced in the surface ocean by biological processes that involve phytoplankton, zooplankton, and bacteria (Simó 2001; Stefels et al. 2007). A fraction of the surface seawater DMS is vented to the atmosphere, depending on ~~prevailing atmospheric conditions and properties of the surface ocean including wind speed and~~ turbulence at the air-

water interface (generally parameterized as a function of wind speed; Wanninkhof et al., 2009; see also ~~, wave types, wave break, bubbles, surface films, friction velocity, humidity and temperature gradients (e.g.~~ Jahne et al., 1987; Merlivat and Memery, 1983; Memery and Merlivat, 1985; Monahan and Spillane, 1984) and the depth of the oceanic upper mixed layer that exchanges with the atmosphere (Galí and Simó, 2015). Atmospheric DMS is subsequently oxidized to MSA and SO_2. The latter is further oxidized to sulfuric acid (H_2SO_4), which can cause formation of new aerosols or condense on pre-existing aerosols. These aerosols may then act as CCN and affect cloud microphysical properties, especially in remote marine environments where concentrations of other types of CCN are low (Clarke et al., 1998; Leaitch et al., 2013; Dall'Osto et al., 2017, Collins et al., 2017). Willis et al. (2016) found that gaseous MSA may also play an important role in the initial growth of new particle formation.

According to the so-called CLAW hypothesis (Charlson et al., 1987), a negative feedback loop operates between ocean ecosystems and the Earth's climate. In particular, formation of new aerosol particles from ocean DMS emissions to the atmosphere leads to increased cloud albedo and reduced surface ocean temperature and/or incident irradiance, which then suppresses production of DMS in the ocean and emission to the atmosphere. Recent studies concluded that there is little evidence to support CLAW under present day climate conditions (Woodhouse et al., 2010; Quinn and Bates, 2011; Browse et al., 2014) since the sources of cloud condensation nuclei in marine boundary layer are numerous and the response of clouds to changes in aerosols are more complex than previously thought at the time of CLAW hypothesis (Quinn and Bates, 2011). However, marine DMS emission may still drive submicron aerosol populations over much of the remote marine atmosphere (Quinn et al., 2017). I~~i~~mportant local impacts of DMS on climate~~climate feedbacks~~ may still exist in the Arctic where summertime aerosol and clouds are strongly influenced by DMS (Leaitch et al., 2013). Newly formed particles influence cloud albedo through cloud microphysical processes and thus can influence local radiation budgets and feedback mechanisms. Considerable concentrations of ultrafine particles and DMS, which are likely involved in naturally occurring aerosol/climate interactions, have been observed in the Arctic in summer (Willis et al., 2016; Ghahremaninezhad et al., 2016; Burkart et al., 2017). Furthermore, Grandey and Wang (2015) found that artificially enhanced DMS emissions in different latitude bands could potentially offset greenhouse gas induced warming across most of the world and especially in the Arctic region. The loss of Arctic sea-ice allows further penetration of sunlight into surface ocean water (e.g. Nicolaus et al., 2012) that can increase net production of algae and phytoplankton (e.g. Arrigo and van Dijken, 2015). In addition, when sea-ice is melted the surface ocean water is more prone to wind stress (Rainville et al., 2011; Martin et al., 2014) that can enhance air-fluxes of DMS and other gases (e.g. Bates et al. 2006). Possible increases in DMS emissions and sulfate aerosol concentrations are particularly important in the Arctic where changes in aerosol radiative forcings are amplified by powerful Arctic feedback processes, including the surface albedo feedback (Pithan and Mauritsen~~et al.~~, 2014; Gagné et al. 2015).

Arguably, uncertainties in surface seawater DMS concentrations and parameterizations of DMS emission fluxes limit scientific progress on climate effects associated with CLAW. For instance, the widely used climatology by Lana et al. (2011) is based on a compilation of data sets from different Arctic field campaigns that took place during the time period from 1985 to 2008. Only measurements from the warm season were used, mainly from the Atlantic portion of the Arctic. The small number of measurements from other locations in the Arctic is problematic, as recent research in the

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NETCARE network (Abbatt et al., 2019~~98, in preparations~~) has shown. 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., 2019~~98, in preparations~~). Galí~~i~~ et al. (2018) argue that biases in the climatology by Lana et al. arise from the application of objective interpolation procedures to a limited amount of measurements. Consequently, Arctic surface seawater DMS concentrations based on Lana et al. (2011) differ substantially from those of an earlier climatology (Kettle and Andreae, 2000), ocean biogeochemical models, and DMS parameterizations (Tesdal et al., 2016a), indicating large uncertainties in estimates of surface seawater DMS concentrations.

Over the last few decades, Arctic temperature has increased at a rate much faster than in other parts of the world (ACIA, 2005~~.~~; AMAP, 2017). Enhanced Arctic warming is largely caused by sea-ice-albedo feedbacks and it is expected that Arctic summer sea-ice may completely disappear well before the end of this century if warming continues at rates simulated by current climate models (Stroeve et al., 2012). Browse et al. (2014) found a weak response of CCN concentrations to enhanced Arctic DMS emissions from complete loss in summer sea-ice due to efficient scavenging of aerosol by drizzle associated with stratocumulus clouds. They did not find evidence for climate feedbacks through changes in cloud properties from enhanced aerosol sources in an ice-free summertime Arctic. However, Browse et al. used an atmospheric chemical transport model with specified meteorological conditions, which excludes responses of Arctic clouds and precipitation to changes in sea ice conditions. In a subsequent study, Ridley et al. (2016) performed fully interactive simulations of sea-ice, ocean biology (with DMS estimated using an embedded empirical algorithm; Simó and Dachs, 2002), aerosols, and clouds with the HadGEM2-ES model. They found a two- to five-fold increase in DMS emissions, an increase in sulfate CCN concentration, and an associated 1 W m^{-2} reduction in simulated summer cloud shortwave radiative forcing in the Arctic.

Despite substantial research activities (e.g. Gabric et al., 2005, Thomas et al., 2010, Woodhouse et al., 2010; Browse et al., 2014; Ridley et al., 2016) it is still very challenging to estimate DMS emissions and even more so how sea-ice reductions may affect DMS emissions in the Arctic in the future. Results of several modeling studies indicated that global warming related sea-ice loss would result in enhanced DMS emission fluxes in the Arctic region (Bopp et al., 2003; Gabric et al., 2005; Levasseur, 2013; Browse et al., 2014; Galí et al., *submitted*). Other studies suggest that the changes in DMS emission flux may be negative in sign due to potentially enhanced ocean acidification (e.g. Six et al., 2013; Schwinger et al., 2017). Phytoplankton species composition is a controlling factor for DMS concentrations given the wide range of cellular dimethylsulfoniopropionate (DMSP) quota among different phytoplankton species (Stefels et al. 2007). For example, the cellular DMSP quota of haptophytes is greater than that of diatoms by a factor of >10 (Stefels et al. 2007). 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.

Given large uncertainties in present-day surface seawater DMS concentrations and potentially large increases in future concentrations, the approach in the current study is to consider a wide range in concentrations in the Arctic for present-day and future conditions. The lower bound of the concentration range is provided by Lana et al. (2011), and the upper bound is obtained by scaling these concentrations by a factor of 10. A number of scenarios for DMS concentrations within this range are considered. These will be used to determine relationships between surface seawater DMS concentrations and climate variables in the Arctic. By selecting widely different scenarios, the robustness of the relationships can be tested without making a priori assumptions that only apply to specific DMS scenarios. Model sensitivity tests with similarly enhanced DMS concentrations have previously been performed (e.g. Grandy and Wang, 2015; Fiddes et al., 2018). Sensitivities of sulfate aerosols, CDNC and cloud radiative forcing to different DMS emission scenarios are investigated using a state-of-the-art atmospheric global climate model. Based on the sensitivity simulations we provide an assessment of Arctic annual mean changes in sulfate aerosol budget and cloud microphysical properties in relation to the mean DMS concentration in the Arctic. Note that an evaluation of surface seawater DMS data sets is outside the scope of the study.

2 Summary of model features

We used version 4.3 of the Canadian Atmospheric Model (CanAM4.3) which is an improved version of CanAM4 (von Salzen et al., 2013). The improvements to version 4.3 compared to version 4 include a higher vertical resolution, improved parameterizations for land surface and snow processes, DMS emissions, and clear-sky radiative transfer. CanAM4.3 has 49 vertical levels extending up to 1 hPa with a resolution of approximately 100 meters near the surface. Model simulations are performed using a spectral resolution of T63 which is equivalent to the horizontal resolution of approximately 2.8 × 2.8 degrees. The model uses separate parameterizations for layer and convective clouds. Aerosol microphysical processes are based on the Piecewise Lognormal Approximation (von Salzen, 2006; Ma et al., 2008; Peng et al., 2012; Mahmood et al., 2016; AMAP, 2015). 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). A detailed description of parameterizations of ocean DMS flux to atmosphere, oxidation and removal processes is provided in Tesdal et al. (2016a). Briefly, surface seawater DMS is ventilated to the atmosphere based on modeled wind speed and the piston velocity parameterization of Nightingale et al. (2000). There are no DMS emissions from sea ice. Furthermore, the model does not account for emissions of organic aerosol species from the ocean.

In the atmosphere, DMS is oxidized to MSA and SO_2 by hydroxyl (OH) radicals during day time and nitrate radical (NO_3) during night, with further oxidation of SO_2 to sulfuric acid (H_2SO_4) by OH in clear-sky conditions. MSA is treated as sulfuric acid in model for simplicity. Binary homogeneous nucleation of H_2SO_4 and water vapour may cause

formation of new aerosol particles, depending on temperature and relative humidity (Kulmala et al., 1998; von Salzen et al., 2000). In-cloud production of sulfate requires ozone (O_3) and hydrogen peroxide (H_2O_2) as oxidants (von Salzen et al., 2000), with oxidant (OH, NO_3, H_2O_2, O_3) concentrations specified according as climatological results from the Model for Ozone and Related Chemical Tracers (MOZART, Brasseur et al., 1998). Dry deposition of aerosol depends on concentrations of aerosols in the near surface model layer (Zhang et al., 2001). Wet deposition includes in-cloud scavenging in both convective clouds and layer clouds, and below-cloud scavenging. Emissions of non-DMS biogenic and anthropogenic aerosol precursors and primary aerosols for the time period up to year 2000 are specified according to Lamarque et al. (2010) and after that according to the IPCC RCP4.5 scenario (Moss et al., 2010).

Cloud droplet number concentrations are calculated based on the assumption of a parcel of air which ascends from the subcloud layer into the cloud layer with a characteristic vertical velocity (Peng et al., 2005), where the standard deviation of the subgrid-scale cloud vertical velocity probability distribution is parameterized using the approach by Ghan et al. (1997). Aerosol particles that are suspended in the parcel of air may activate and grow into cloud droplets by condensation of water vapour. A numerically efficient solution of the condensational droplet growth equation (e.g. Seinfeld and Pandis, 1998) is employed for this purpose. In grid cells that are affected by clouds, CanAM4.3 accounts for cloud albedo and lifetime effects (first and second~~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 Coupled Model Intercomparison Project phase 5 (CMIP5) climate models. 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).

3 Sulfate concentrations in CanAM4.3

We performed two sets of historical model simulations, one with the full model with all natural and anthropogenic aerosols and their precursors included ("hisCont") and one with zero surface seawater DMS ("hisNoDMS"). Monthly mean surface seawater DMS concentrations in hisCont are specified according to the climatology of Lana et al. (2011) [hereafter referred to as L10]. The global annual DMS emission flux in hisCont is 24.96 TgS/yr, which is very close to 25.3 TgS/yr reported in Test~~t~~dal et al. (2016a) and well within previously reported ranges (e.g. Lana et al., 2011; Tesdal et al., 2016a). Both simulations were integrated for the time period 1991 to 2003 during which extensive observations of sulfate are available. Wind and temperature in each simulation were nudged towards specified results from a common simulation with CanAM4.3 for this time period. According to Kooperman et al. (2012) nudging of meteorological model variables reduces the influence of natural variability and therefore improves estimates of differences in diagnosed aerosol indirect effects. Similarly, biases in simulated aerosol and CDNC concentrations between the simulations are also reduced according to a statistical analysis of CanAM model results (not shown). The contribution of DMS oxidation to total sulfate concentrations is determined by calculating the difference in simulated sulfate concentrations between these two simulations (i.e. hisCont - hisNoDMS), which is interpreted as biogenic sulfate in the following.

A slightly different version of CanAM has previously been evaluated (Eckhardt et al., 2015; Tesdal et al., 2016b). In a multi-model comparison, Eckhardt et al. (2015) compared model simulations of sulfate and black carbon aerosols with observations from different stations and aircraft campaigns and found that most models, except CanAM, significantly underestimated observed concentrations in the Arctic region. Mahmood et al. (2016) found that black carbon concentration differences in four models are related to differences in wet removal processes in the models.

For the current study we used observed data from various ship-based campaigns and observations from Alert in Canada to further validate simulations of sulfate, with particular emphasis on the role of biogenic emissions. Shipboard data from the National Oceanic and Atmospheric Administration, Pacific Marine Environmental Laboratory (NOAA PMEL) was obtained from cruises that fell within the period 1992-2002. Only non-sea-salt sulfate (nss-SO_4^{2-}) data was selected and summed for all available bin sizes. The gridded model data was matched to the nearest location of the observations, shown in Fig. 1a.

Figure 1 shows that simulated sulfate concentrations agree well with observations, especially in regions where modeled DMS contributions are relatively large. The mean value for all ship-based observations is 3.416±4.018 (μg/m^3) and the model mean value is 2.079±1.815 (μg/m^3), corresponding to a model underestimate of ~39%. Most of the underestimates in simulated mean concentrations are associated with locations where the model simulates a large contribution of fossil-fuel (non-biogenic) sulfate to total sulfate concentrations. ~~Overall, simulated sulfate concentrations are in good agreement with the observations in regions with large contributions of biogenic sulfate.~~

From the Alert data, with highly variable contributions of DMS, it is evident that the model overestimates the contribution of DMS to total sulfate concentrations at this location (Fig. 1b). Overall, CanAM4.3 is able to capture the sulfate annual cycle very well at Alert, with slight underestimation in winter and spring and overestimation in summer (Fig. 2). The correlation coefficient for the mean annual cycle between model and observations is 0.95. Mean observed and simulated sulfate concentrations at Alert are 0.475±0.413 and 0.419±0.228 μg/m^3 respectively, corresponding to a model underestimate of ~12%.

An analysis of isotopic data is available for Alert, which can be used to distinguish between contributions of biogenic and fossil-fuel sources to sulfate concentrations in the observations at this site (Norman et al., 1999). The ratio between sulfur isotopes ^{34}S and ^{32}S of an observed sample is compared with an international standard ratio based on sulfur isotopes in Vienna-Cañon Diablo Troilite (Beaudoin et al., 1994; Krouse and Grinenko, 1991; Norman et al., 1999). The results are expressed in parts per thousand (‰). The sulfate concentration at Alert consists of the sum of marine biogenic, anthropogenic, and sea salt sulfate with the delta isotopic ratios of +17.5‰, +5‰, +21‰ respectively.

A comparison with the isotopic data indicates that the model underestimates fossil-fuel sulfate and overestimates biogenic sulfate (Fig. 2b and 2c). This difference is particularly pronounced in spring and early summer, when observed biogenic sulfate concentrations are particularly high. An interesting feature is the double peak in biogenic sulfate concentrations during the annual cycle with one peak occurring in May and the other in October (Fig. 2c), which is

essentially captured by the model. The occurrence of October peak at Alert has not been investigated. 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. The correlation coefficient for the mean annual cycle between observed and simulated biogenic sulfate concentrations is 0.73.

Another interesting feature is the relative contribution of biogenic sources to total sulfate concentrations (Fig. 2d). Although absolute sulfate aerosol concentrations in summer are much lower than during other seasons, both observations and model results indicate a much larger contribution of biogenic sources to total sulfate concentration in this season compared to other seasons (Fig. 2d).

4 Sensitivity of sulfate, clouds, and radiation to changes in DMS

Five ensembles of five simulations each were performed, where ensemble members were generated by introducing random perturbations in radiative flux calculations (a total of 25 simulations). The five experiments differ in terms of specified surface seawater DMS concentrations in the Arctic region, defined here as the region from 62.78˚-90˚N. A wide range of different Arctic surface seawater DMS concentration patterns is considered in order to account for substantial uncertainties in surface seawater DMS concentrations. For present day, uncertainty in specified Arctic DMS concentration climatologies arises from a lack of observational data, and concentrations that are highly variable in space and time (Lana et al., 2011; Tesdal et al., 2016a; Galí et al., 2018). Furthermore, very little is known about how DMS concentration may evolve in the future. Outside the Arctic, monthly mean ocean DMS concentrations in the simulations are specified according to the L10 climatology.

For the first set of simulations, DMS concentrations were specified as zero in the Arctic, hereafter referred to as CNTRL. All subsequent simulations are compared to CNTRL in order to estimate the contribution of Arctic DMS emissions to simulated biogenic sulfate concentrations and climate. For the second set of simulations, CLIM, the L10 monthly climatology is used in the Arctic. In 10×CLIM, the L10 climatology was multiplied by a factor of 10 at each model grid point in the Arctic region. In order to analyze the impact of spatial variability in DMS concentrations, a single DMS concentration (i.e. 16.9 nM) was assigned to each grid cell in the Arctic region hereafter referred to as UNFM. The number used for DMS concentration in UNFM is arbitrarily calculated as the grid-point average of 10 times the L10 climatology in the Arctic. It provides an additional scenario for testing the sensitivity of forcings under uniform surface seawater concentrations of DMS and also provides a test of linearity of such responses. Finally, as a further test of spatial variability of DMS, we used a satellite-based estimate of surface seawater DMS concentration (Galí et al., 2018) and multiplied it by a factor of 10 in 10×SAT. A value of 5 nM was applied in 10xSAT over the central Arctic region where satellite observations are not available, upon the observation that available sea-surface DMS measurements in the Arctic winter have an average of ~0.5 nM. Note this earlier version of Galí et al. (2018) satellite based DMS estimation

had a small negative bias in magnitude, however, the spatial distribution remained largely unchanged after correction. Surface seawater DMS concentrations in all simulations are summarized in Fig. 3 and Table 1.

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. The model was integrated over four years for 1998-2001 and annual mean model results during the last three years of the simulations were analyzed (hereafter referred to as 2000). Present-day sea ice amounts and sea surface temperatures (SSTs) are specified according to reanalysis data from Climate Forecast System Version 2 (Saha et al., 2014). In addition, all of the above experiments were repeated for 2048-2051 (referred to as 2050). The projected sea ice amounts and SSTs from a 50-member~~large~~ ensemble of simulations with CanESM2 for the RCP8.5 emission scenario were used in order to represent conditions in 2050 (Sigmond and Fyfe, 2016). ~~-~~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 CanAM4.3, we use mean results from 5 different CanAM4.3 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. In CanESM2, the Arctic is devoid of sea ice in September by 2050, consistent with results from other models for this relatively high emission scenario (Stroeve et al., 2012). The difference of sea ice extent in the simulation time periods is summarized in Fig. S1 (in supplementary data). The Arctic annual mean sea ice fractions are 75.6% (2000) and 50% (2050) for grid cells where the sea ice fraction is 0.15 or larger. Similar to simulations corresponding to year 2000, simulated horizontal winds and temperature were nudged towards specified results from 5 simulations with different meteorological conditions.

The total annual Arctic DMS fluxes for the two simulations time periods are summarized in Table 1. For 2000, DMS emission fluxes are approximately linearly proportional to the mean surface seawater DMS concentrations. For example, the total annual Arctic fluxes for the 10xCLIM are 84% higher than for 10xSAT, corresponding to 76% higher concentrations of Arctic-mean surface seawater DMS. This indicates that the sensitivity of Arctic-mean DMS fluxes to deviations of spatial distributions from the mean surface seawater concentrations is relatively low for 2000. Similar results for 2050 give evidence for relatively low sensitivity of fluxes to spatial distributions of surface seawater DMS concentrations.

Sea ice fraction in 2050 in summer and autumn is much lower than in 2000 (Fig. S1). For CLIM and 10xCLIM the total Arctic sulfur flux increases by 33% from the earlier to the later time period due to the reduction in future sea ice fraction. The difference is up to 47% for 10xSAT and 53% for UNFM (Table S1). Regionally, differences in fluxes are strongly

correlated with changing sea ice fractions, with increases in regions with reduced sea ice fraction in 2050 and only minor changes over the open ocean (Fig. 4).

Owing to large DMS emission fluxes in the Atlantic region, the spatial pattern of the biogenic sulfate burden (relative to CNTRL) produces a maximum in this region for all surface seawater DMS data sets (Fig. 5). However, for CLIM diagnosed biogenic sulfate burdens are statistically significant over the Greenland Sea and nearby Baffin Bay for 2050 but not for 2000. Differences in Arctic-mean biogenic sulfate burdens between 2000 and 2050 are relatively small for all of the scenarios, ranging from just -1% for UNFM to +21% for CLIM, despite the relatively large increases in DMS emissions between 2000 and 2050 (Table 1 and Fig. 5). The weak responses in biogenic sulfate burdens to DMS emissions are caused by increased precipitation and aerosol wet removal in the Arctic in 2050 (Tables S1 and S2; Figs. S2 and S3). Thus the wet deposition of biogenic sulfate from Arctic DMS emissions becomes more efficient in the future. Whereas emissions of Arctic DMS increase between 33.3% (CLIM) and 53.2% (UNFM), wet deposition of biogenic sulfate from Arctic DMS emissions increases more strongly, between 42.45% (CLIM) and 72.1% (UNFM) from 2000 to 2050 (Table S2). The fraction of Arctic DMS emissions that is removed by wet deposition increases from between 55.3% (UNFM) and 76.5% (CLIM) in 2000 to between 62.1% (UNFM) and 81.8% (CLIM) in 2050 (Table S3).

On the other hand, projected reductions in anthropogenic sulfur emissions between 2000 and 2050 lead to reductions in total wet deposition of sulfate in the Arctic by -47.7% in CNTRL (Table S1). In the sensitivity experiments with increases in Arctic emissions of DMS between 2000 and 2050 reductions in total sulfate wet deposition in the Arctic between 2000 and 2050 are weaker, i.e. between -7.5% (10×CLIM) and -40.6% (CLIM). Considering the very wide range of surface seawater DMS concentrations applied here, a nearly complete compensation of aerosol production from oceanic DMS by increased wet deposition seems to be a robust feature of the future Arctic, largely independent of DMS emission patterns and amounts.

Changes in CDNC are important for radiative effects of sulfate aerosols. Impacts of climatological DMS emissions on CDNC are not statistically significant in CLIM, i.e. they are within the range of meteorological variability in the ensemble of simulations (Fig. 6). The relatively weak simulated impact of present-day climatological DMS concentrations on CDNC and cloud microphysics is in agreement with previous studies (e.g. Browse et al., 2014; Ridley et al., 2016). Similarly, for 2050, few regions in the Arctic show significant impacts of present-day DMS emissions on CDNC although local increases are up to about 10%. On the other hand, the other sets of simulations (i.e. 10×CLIM, UNFM and 10×SAT) produce significant changes in CDNC, especially for 2050, with increases up to ~10^7 m^{-3} for 10×CLIM. It is interesting to note that although the biogenic sulfate burdens are similar in 2000 and 2050, there are relatively large systematic increases in CDNC due to increased Arctic DMS emissions in 2050 for these simulations.

Increases in CDNC between 2000 and 2050 are related to increases in formation of new particles in the lower Arctic troposphere by between +128 and +269% (Table 2 and Fig. S3) for the range in surface seawater DMS concentration considered. This leads to large-scale increases in CCN concentrations near the surface in the Arctic (Fig. S3), which is in contrast to a more non-uniform response of CCN concentrations to reductions in sea ice fraction according to Browse et

al. (2014), with relatively large simulated increases over the continental Arctic and small decreases over the central Arctic Ocean. Large-scale increases of CCN concentrations and nucleation rates in 2050 in simulations with CanAM4.3 can be attributed to several factors: First, global anthropogenic emissions of sulfur are considerably lower in 2050 compared to 2000, which causes a reduction in the burden of anthropogenic sulfate (-65% in CNTRL) and the associated condensation sink of sulfuric acid in the Arctic atmosphere, which can be expected to facilitate the formation of new particles (Wyslouzil et al., 1991). The condensation sink of sulfuric acid is further reduced by increased wet deposition of aerosols due to increased Arctic precipitation. Finally, increased evaporation of moisture from the ocean leads to increases in relative humidity in the Arctic, which also produces conditions that are more favorable to nucleation in 2050 than 2000. On the other hand, increases in the sulfuric acid condensation sink due to increased emissions of sea salt and organic aerosols from the open ocean are not accounted for in the current version of the model, which may lead to overestimates in nucleation rates in the simulations. According to Browse et al. (2014), the increase in the natural condensation sink due to increased production of sea salt and organic aerosol under ice-free conditions causes a substantial reduction in the near-surface nucleation rate. However, it is likely that increases in production of sea salt under ice-free conditions are accompanied by large increases in wet deposition of sea salt due to increased Arctic precipitation (Struthers et al., 2011), which has not been explicitly accounted for by Browse et al. (2014). In addition, recent observations indicate a dominant role of small particles in activation and formation of cloud condensation nuclei in clean Arctic conditions (Leaitch et al., 2016) implying efficient nucleation of fine mode particles.

According to the first indirect effect of aerosols on climate, increases in CDNC may lead to smaller cloud droplets which are associated with more efficient scattering of incoming solar radiation and therefore stronger cloud radiative forcings, determined here as the difference in total-sky minus clear-sky shortwave radiative fluxes at top of the atmosphere (Soden et al., 2004). Subsequently, the cloud radiative forcing associated with biogenic DMS from Arctic DMS emissions is determined as difference in cloud radiative forcing between sensitivity experiments and CNTRL. As shown in Fig. 7 the cloud radiative forcing due to Arctic DMS emissions is small and not statistically significant for CLIM for both time periods. However, for 10×CLIM and 10×SAT the cloud radiative forcing in the Arctic due to Arctic DMS emissions is significant with maximum of up to -4 Wm^{-2} for the Atlantic side of the Arctic for 10xCLIM in 2050, qualitatively in agreement with differences in CDNC. Overall, the mean cloud radiative forcing in the Arctic due to Arctic DMS emissions increases by between 108% (CLIM) and 145% (UNFM) from 2000 to 2050 (Table 2). All DMS data sets produce similar patterns of changes, with systematically ~~increased~~enhanced cloud radiative forcings for the Atlantic region of the Arctic where loss of sea ice leads to particularly large increases in DMS emissions in all cases.

On regional scales, differences in cloud radiative forcing due to Arctic DMS emissions in Fig. 7 are generally smaller than changes in cloud radiative forcing associated with changes in meteorological conditions and anthropogenic aerosol precursor emissions between 2000 and 2050 (Fig. S4). However, averaged over the Arctic, differences are similar. For instance, the mean cloud radiative forcing in the Arctic in CLIM is -0.13 and -0.27 Wm^{-2} for 2000 and 2050 respectively (difference of -0.14 Wm^{-2}). Similarly, CNTRL produces a difference in cloud radiative forcings of -0.65 Wm^{-2} in total cloud radiative forcing between 2000 and 2050. It is evident that the cloud radiative forcing from Arctic DMS (Fig. 7) acts to enhance negative cloud radiative forcings in the central Arctic and counteracts positive forcings in the Atlantic

Arctic and north of Siberia (~~cf. Figs. 7 and~~ Fig. S4), especially for 10xCLIM (the Arctic mean difference for 10×CLIM is -0.84 Wm^{-2} between 2000 and 2050).

Mean results in the Arctic are summarized in Fig. 8 which provides an indication of robust aerosol and cloud responses at ~~relationships between~~ pan-Arctic scale~~mean results~~, despite large differences in amount and spatial distribution of surface seawater DMS concentration in the different cases. For instance, Arctic-mean sulfate burdens due to Arctic DMS emissions are similar for present-day and future conditions (Fig. 8b) despite strongly increased DMS emissions in 2050 resulting from~~form~~ sea ice retreat. On the other hand, biogenic DMS emissions lead to more efficient formation of cloud droplets in the future in the Arctic. Therefore, cloud droplet number concentrations and cloud radiative forcing increase systematically as sea ice extent declines from 2000 to 2050 for each Arctic DMS, despite the low sensitivity of biogenic sulfate burdens to changes in sea ice. This provides evidence for a negative feedback of Arctic DMS emissions on Arctic radiative forcing, assuming that DMS concentrations in the ocean and atmospheric oxidant concentrations do not change between 2000 and 2050. To a good first approximation, the strength of the feedback is proportional to the mean surface seawater DMS concentration in the Arctic despite low sensitivity of sulfate burdens. The simulated responses of clouds and radiative forcing to changes in sea ice extent are found to be robust for a wide range of surface seawater DMS concentration scenarios.

5 Conclusions

Simulated sulfate concentrations from the Canadian Center for Climate Modeling and Analysis Atmospheric Model (CanAM4.3) were compared to observations from various shipboard campaigns and in-situ observations at Alert in Canada with a particular emphasis on the role of biogenic emissions. We found that the model reproduced seasonal variations in observed biogenic sulfur concentrations at Alert, although the model overestimates the biogenic contribution to total sulfate somewhat. Observed biogenic sulfur concentration maxima in May and in October are well reproduced by the model. Furthermore, comparisons with ship-based measurements from different field campaigns yield good agreement with simulated sulfate concentrations, especially in regions with large contributions of biogenic sulfate. However, it is plausible that the current Arctic surface seawater DMS concentrations are underestimated because models and climatological data sets do not yet account for substantially enhanced concentrations in melt ponds, and near the ice edge (e.g. Mungall et al., 2016; Ghahremaninezhad et al., 2016; Hayashida et al., 2017; Gourdal et al. 2018). In addition, large uncertainties exist for nucleation parameterizations (e.g. Zhang et al., 2010).

We performed model simulations to understand the sensitivity of sulfate aerosols and cloud radiative forcing to projected changes in sea ice and climate conditions between 2000 and 2050. Several model experiments were performed using a wide range of different surface seawater DMS concentrations in order to account for uncertainties in present-day and future DMS and to explore the sensitivity of aerosol/climate interactions to differences in spatial patterns of DMS. Results of the simulations indicate that the enhanced wet removal efficiency from increased precipitation in 2050 largely counteracts the impact of the increase in DMS emissions on sulfate burden in the Arctic. Annual mean Arctic sulfate burden differences between 2000 and 2050 are small for any given scenario (differences ranging between -1 and 21%)

despite large increases in DMS emission between 2000 and 2050 due to sea ice retreat (between +33 and +53%). The sensitivity of modeled DMS fluxes into the atmosphere and sulfate burdens to spatial variations in surface seawater DMS is relatively weak.

Similar to previous studies (e.g. Browse et al., 2014; Ridely et al., 2016) we found weak impacts of climatological DMS emissions on cloud radiative forcings for present day conditions (simulation CLIM). However, ~~in the future~~for 2050 simulations, biogenic DMS emissions lead to considerable impacts on simulated Arctic aerosol and cloud processes owing to conditions that are conducive to the formation of fine particles in the Arctic. In 2050, increased emissions of DMS from large ice-free regions of the Arctic ocean are associated with increased biogenic sulfate aerosol nucleation rates (between +128 and +269%) and cloud droplet number concentrations (between +35 and +133%) and thus enhanced cloud albedos, resulting in negative cloud radiative forcing of biogenic sulfate in the Arctic. The difference in cloud radiative forcing between years 2050 and 2000 based on simulations for four different Arctic surface seawater DMS data sets ranges from between -0.14 Wm^{-2} (CLIM) to -0.84 Wm^{-2} (10×CLIM). Thus our model results provide evidence for a negative Arctic climate feedback. The essential ingredient of the feedback is a response of DMS emissions and cloud droplet number concentrations to sea ice retreat due to changes in radiative forcings in the climate system. This differs from CLAW which is rooted in the assumption of a change in biological production of DMS in the ocean in response to a change in radiative forcings. Furthermore, the strength of the Arctic climate feedback is proportional to the mean surface seawater DMS concentration in the Arctic. Consequently, potential future changes in primary productivity (Yool et al., 2005; Vancoppenolle et al., 2013), mixing and phytoplankton habitat (Harada, 2016) in the Arctic Ocean (Levasseur, 2013) may act to enhance the strength of the Arctic feedback. ~~Additional uncertainty in the strength of the feedback arises from the fact that atmospheric oxidant concentrations are assumed to be steady in our study.~~

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. 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. 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.

Acknowledgement

Shipping non-sea-salt sulfate data was obtained from NOAA PMEL Atmospheric Data Server (available at: *https://saga.pmel.noaa.gov/data/*). We thank Jim Christian and two anonymous referees for providing helpful ~~an internal~~ reviews of the manuscript. We also thank Martine Lizotte and Hakase Hayashida for providing very helpful discussions of surface ocean DMS concentrations. Funding for this work was provided by the Natural Sciences and Engineering Research Council of Canada through the NETCARE project of the Climate Change and Atmospheric Research Program.

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Table 1. Arctic mean surface seawater DMS concentration, total sulfur emission flux, and associated mean biogenic sulfate burden for 2000 (2050)

	DMS (nM)	Emissions (TgS/yr)	SO_4^{2-} Burden (kilotonnes)
CLIM	1.96	0.24 (0.32)	2.13 (2.58)
10×CLIM	19.58	2.41 (3.22)	20.05 (20.55)
UNFM	16.88	1.88 (2.87)	16.82 (16.64)
10×SAT	11.19	1.31 (1.92)	10.31 (11.63)

Table 2. Arctic mean aerosol nucleation rate, CDNC, and cloud radiative forcing in 2000 (2050) associated with emissions of DMS in the Arctic.

	Nucleation rate ($\times 10^6$ m^{-2}s^{-1})	CDNC ($\times 10^6$ m^{-3})	Cloud radiative forcing (Wm^{-2})
CLIM	0.02517 (0.09294)	0.34716 (0.81023)	-0.13 (-0.27)
10×CLIM	0.41751 (0.98998)	2.9886 (4.1781)	-0.75 (-1.59)
UNFM	0.27358 (0.62366)	2.3019 (3.4893)	-0.40 (-0.98)
10×SAT	0.20618 (0.54223)	1.7853 (2.4045)	-0.55 (-1.18)

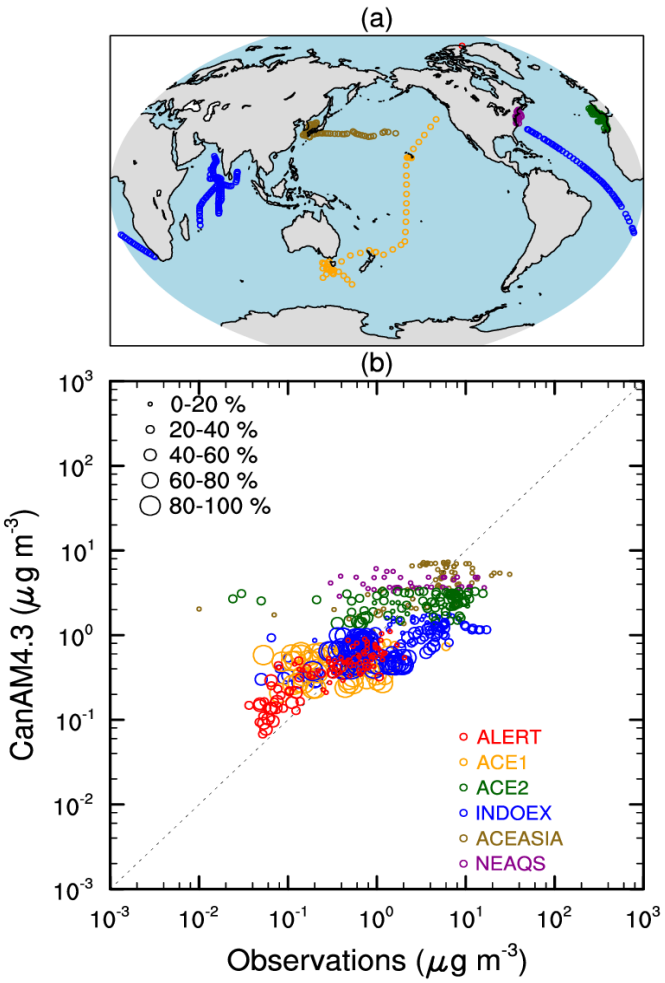

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Figure 1: (a) Locations for Alert (red circle) and ship-based nss-SO_4^{2-} observations used for comparisons with CanAM4.3? model results. (b) Comparison of model and observed nss-SO_4^{2-} . For ship based observations, the size of the markers represent percentage of contribution of DMS to total nss-SO_4^{2-} derived from model results. For Alert, the percentage contribution to total nss-SO_4^{2-} is based on isotopic composition.

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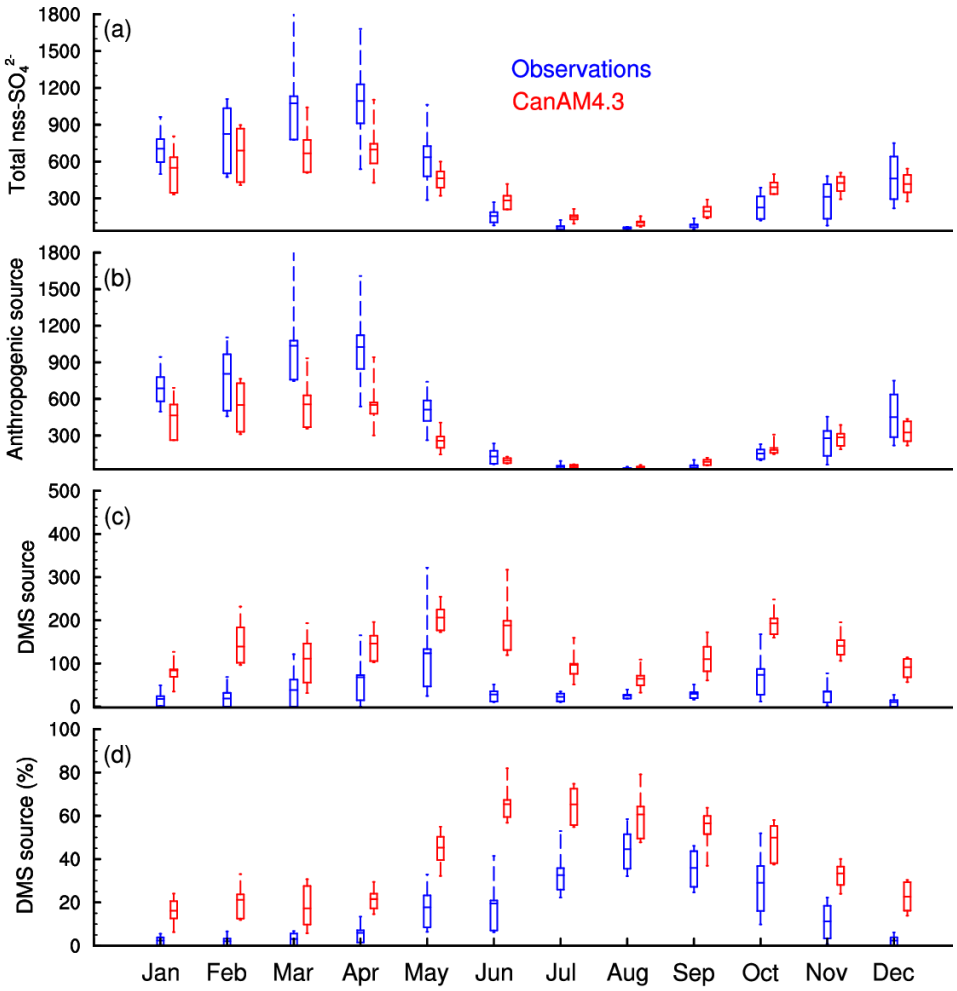

780 **Figure 2: Multi-year mean monthly concentrations of total (a), fossil-fuel (b), biogenic (c) sulfate concentrations from simulations and observed nss-SO_4^{2-} during 1994 to 2002 at Alert, (d) relative contribution of DMS source to total sulfate concentration at Alert. The whiskers represent minimum and maximum and the horizontal line inside the box represents the mean for the whole period. The box height represents the interquartile range of 25th and 75th percentiles. Unit: ng m^{-3}**

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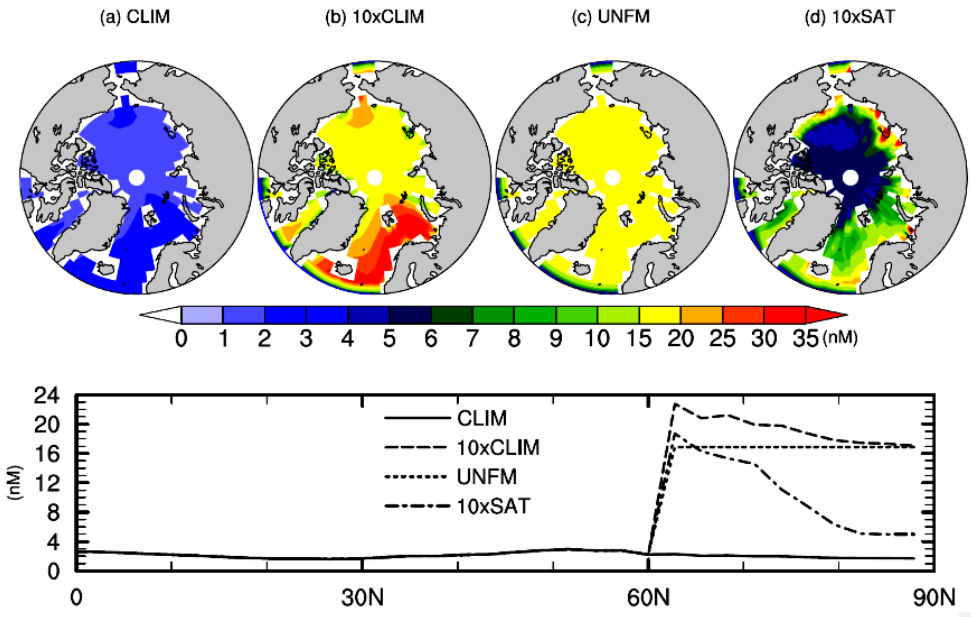

Figure 3: Annual mean distribution of surface seawater DMS concentration used in sensitivity simulations; (a) L10 DMS climatology, (b) L10 DMS climatology multiplied by 10 in the Arctic region, (c) uniform distribution of DMS, (d) satellite based DMS climatology multiplied by 10. The bottom panel shows zonal mean results.

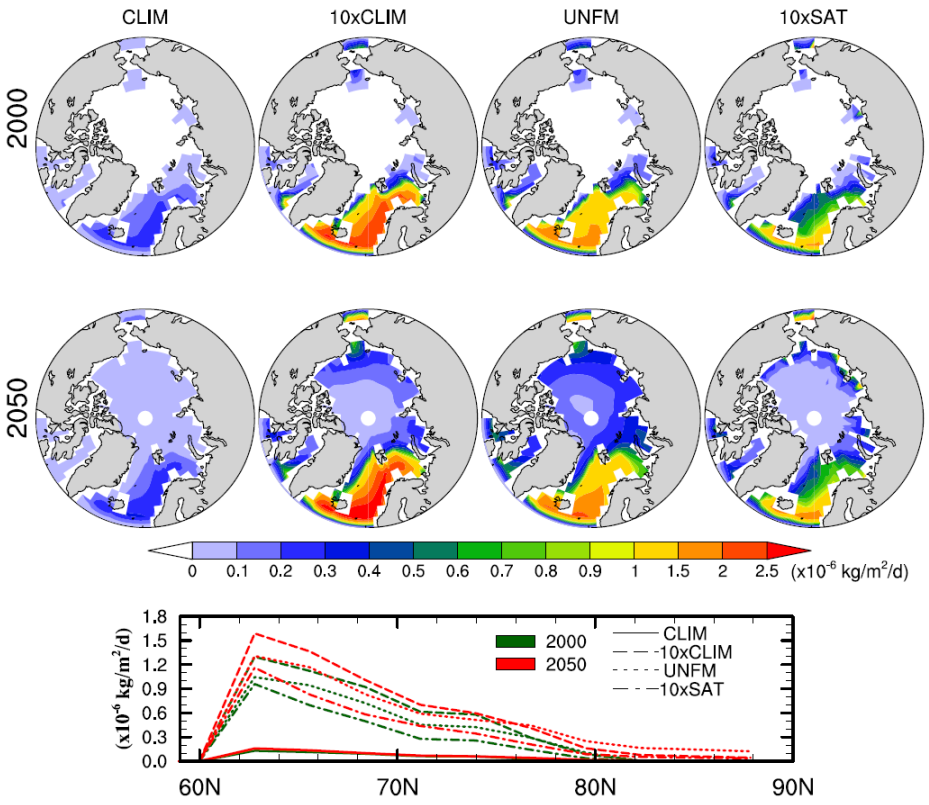

Figure 4: DMS emission fluxes for the two different periods, similar to Fig. 3. Zonal mean results 2000 and 2050 are shown in the bottom panel.

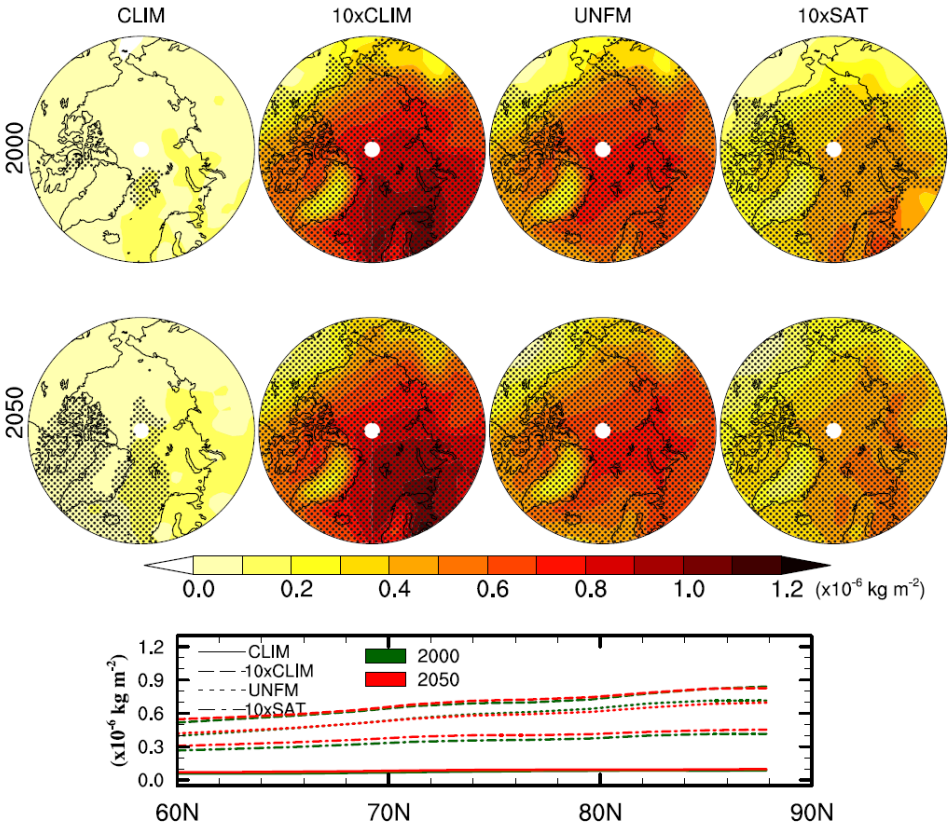

Figure 5: Biogenic sulfate burden due to Arctic DMS emissions for each of the 4 scenarios. The stippling represents regions where the burden difference relative to CNTRL is significant at 95% confidence level. Zonal mean results for 2000 and 2050 are shown in the bottom panel.

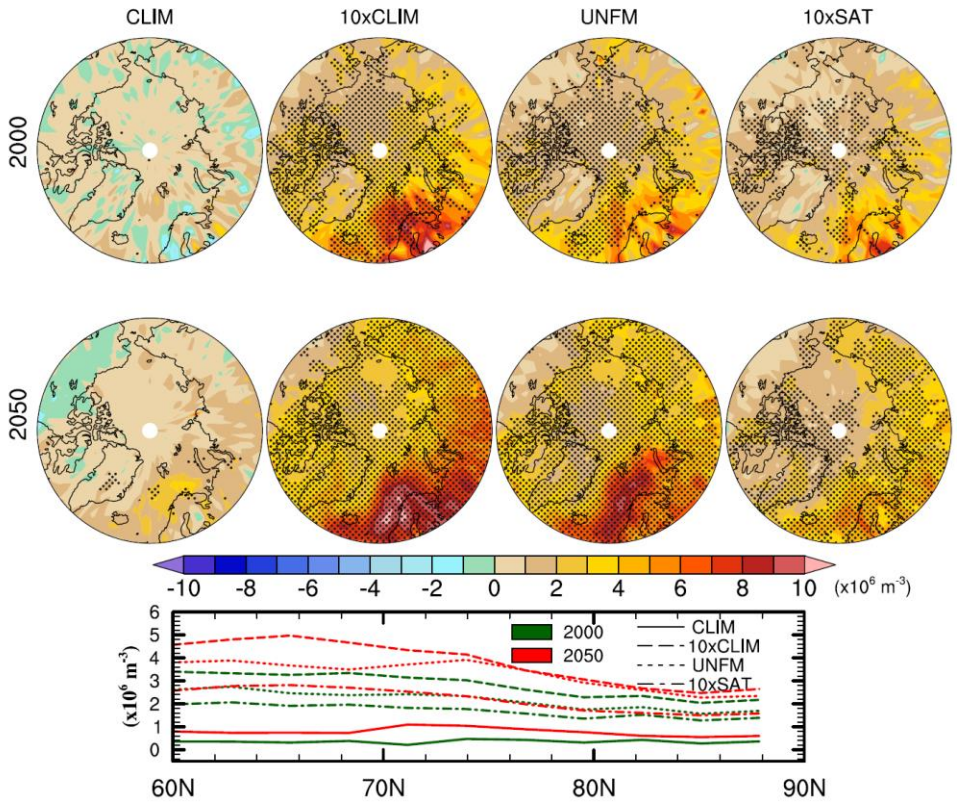

Figure 6: Change in cloud droplet number concentration at first model level above the surface due to Arctic DMS emissions (relative to CNTRL). Stippling represents change significant at 95% confidence level. Zonal mean results are shown in the bottom panel.

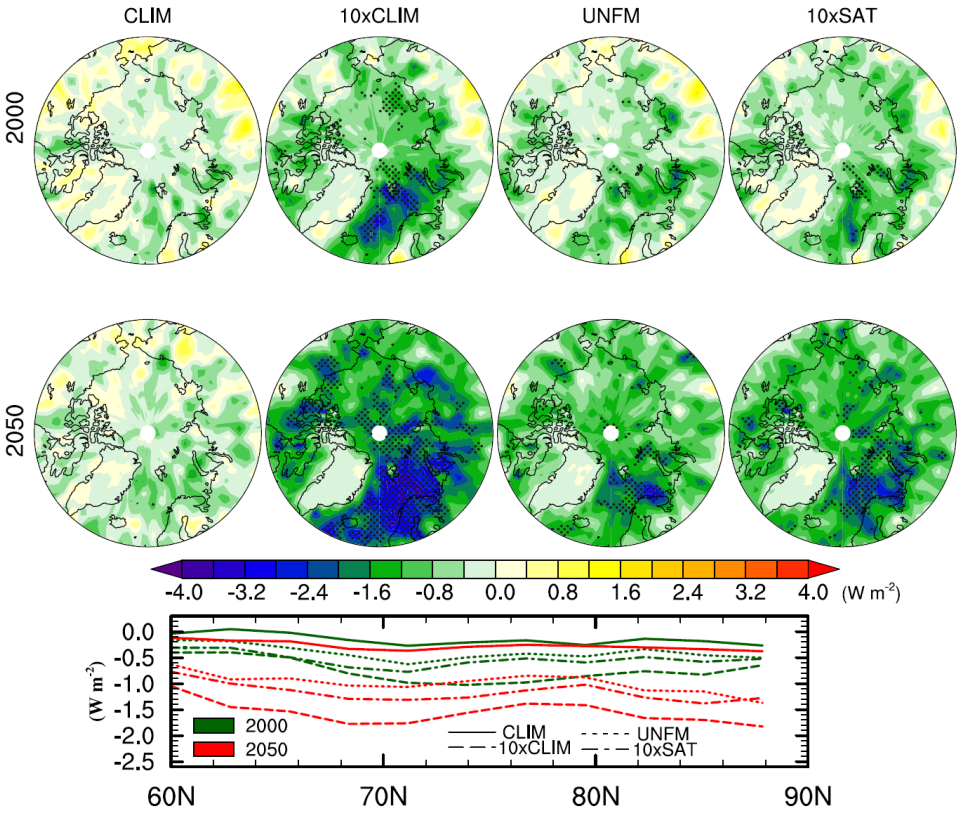

Figure 7: Cloud radiative forcing in the Arctic due to ocean DMS emissions. Stippling represents radiative forcing significant at 95% confidence level. Zonal mean results are shown in the bottom panel.

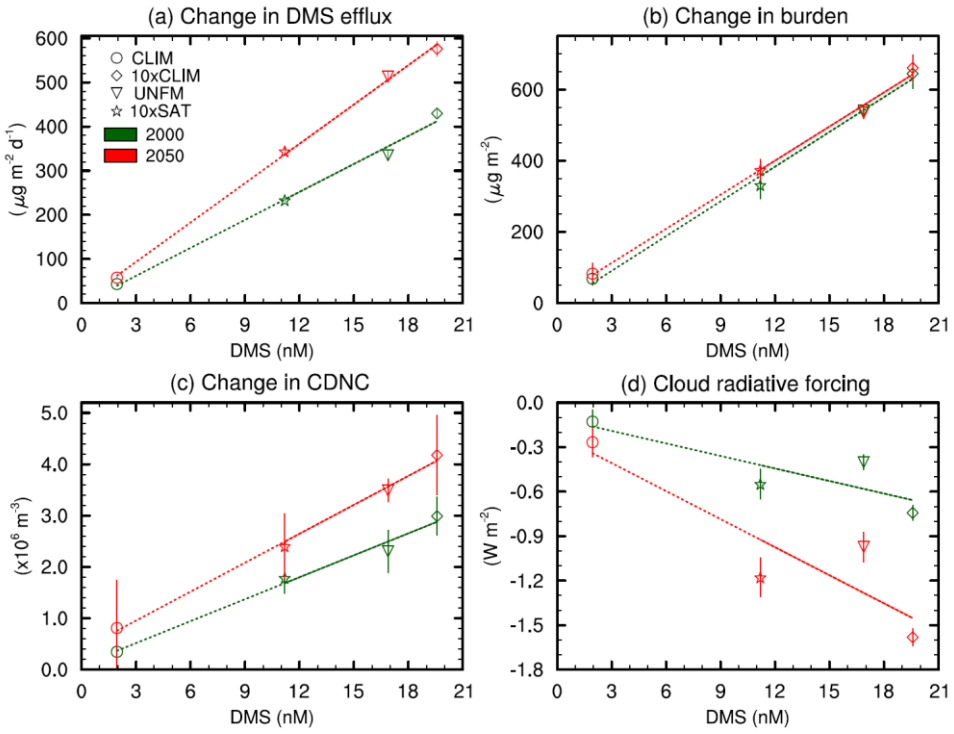

Figure 8: Relationship between annual Arctic mean DMS emission fluxes, sulfate burden, CDNC, and cloud forcing and the mean DMS concentration in the Arctic. The vertical lines represent 95% confidence interval based on two-tailed t-test. Dotted lines represent regression between four scenarios.