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.

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.

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.

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*.”

References:

Ardyna, M., M. Babin, M. Gosselin, E. Devred, L. Rainville, and J.-É. Tremblay (2014), Recent Arctic Ocean sea ice loss triggers novel fall phytoplankton blooms, Geophys. Res. Lett., 41, doi:10.1002/2014GL061047.

Arrigo, K.R., van Dijken, G.L., 2015. Continued increases in Arctic Ocean primary production. Prog. Oceanogr. 136, 60–70.

Bates, N. R., Moran, S. B., Hansell, D. A., & Mathis, J. T. (2006). An increasing CO2 sink in the Arctic Ocean due to sea-ice loss. Geophysical Research Letters, 33(23).

Fiddes, S. L., Woodhouse, M. T., Nicholls, Z., Lane, T. P., and Schofield, R.: Cloud, precipitation and radiation responses to large perturbations in global dimethyl sulfide, Atmos. Chem. Phys., 18, 10177-10198, https://doi.org/10.5194/acp-18-10177-2018, 2018.

Grandey, B. S., and Wang, C.: Enhanced marine sulphur emissions offset global warming and impact rainfall, Sci. Rep., 5, 13055, doi:10.1038/srep13055, 2015.

Jahne, B., Munnich, K.O., Bosinger, R., Dutzi, A., Huber, W., and Libner, P.: On the parameters influencing air-water gas exchange, J . Geophys. Res., 92, 1937-1949, 1987.

Kettle, A. J., and Andreae, M. O.: Flux of dimethylsulfide from the oceans: A comparison of updated data sets and flux models, J. Geophys. Res.-Atmos., 105, 26793–26808, doi:10.1029/2000JD900252, 2000.

Lana, A., Bell, T. G., Simó, R., Vallina, S. M., Ballabrera-Poy, J., Kettle, A. J., Dachs, J., Bopp, L., Saltzman, E. S., Stefels, J., Johnson, J. E., and Liss, P. S.: An updated climatology of surface dimethlysulfide concentrations and emission fluxes in the global ocean, Global Biogeochem. Cycles, 25, GB1004, doi:10.1029/2010GB003850, 2011.

Leaitch, W. R., Sharma, S., Huang, L., Macdonald, A. M., Toom-Sauntry, D., Chivulescu, A., von Salzen, K., Pierce, J. R., Shantz, N. C., Bertram, A., Schroder, J., Norman, A.-L., and Chang R. Y.-W.: Dimethyl sulphide control of the clean summertime Arctic aerosol and cloud, Elementa: Science of the Anthropocene, 1, 000017, doi:10.12952/journal.elementa.000017, 2013.

Loose, B., McGillis, W. R., Perovich, D., Zappa, C. J., & Schlosser, P. (2014). A parameter model of gas exchange for the seasonal sea ice zone. Ocean Science, 10(1), 17-28.

Memery, L., and Merlivat L.: Modeling of gas flux through bubbles at the air-water interface, Tellus Ser. B, 37, 272-285, 1985.

Merlivat, L., and Memery L.: Gas exchange across an air-water interface: Experimental results and modeling of bubble contribution to transfer, J. Geophys. Res., 88, 707-724, 1983.

Monahan, E. C., and Spillane M. C.: The role of whitecaps in air-sea gas exchange, in Gas Transfer at Water Surfaces, edited by W. Brutsaert, and G.H. Jirka, pp. 495-504, D. Reidel, Norwell, Mass., 1984.

Nicolaus,M., Katlein, C.,Maslanik, J., Hendricks, S., 2012. Changes in Arctic sea ice result in increasing light transmittance and absorption. Geophys. Res. Lett. 39, L24501. http://dx.doi.org/10.1029/2012GL053738.

Nightingale, P. D., Malin, G., Law, C. S., Watson, A. J., Liss, P. S., Liddicoat, M. I., Boutin, J., and Upstill-Goddard, R. C.: In situ evaluation of air-sea gas exchange parameterizations using novel conservative and volatile tracers, Global Biogeochem. Cy., 14, 373y., 14doi:10.1029/1999GB900091, 2000.

Quinn, P. K., and Bates, T. S.: The case against climate regulation via oceanic phytoplankton sulphur emissions, Nature, 480, 51–56, doi:10.1038/nature10580, 2011.

Rainville, L., Lee, C. M., and Woodgate, R. A.: Impact of wind-driven mixing in the Arctic Ocean, Oceanography, 24(3), 136-145, 2011.

van der Loeff, M. M. R., Cassar, N., Nicolaus, M., Rabe, B., & Stimac, I. (2014). The influence of sea ice cover on air-sea gas exchange estimated with radon-222 profiles. Journal of Geophysical Research: Oceans, 119(5), 2735-2751.

Wanninkhof, R., Asher, W. E., Ho, D. T., Sweeney, C., & McGillis, W. R. Advances in quantifying air-sea gas exchange and environmental forcing, Annu. Rev. Mar. Sci., 1:213–44, 2009.