Interactive comment on "**Dimethyl sulfide and its role in aerosol formation and growth in the Arctic summer – a modelling study**" *by* **Roya Ghahreman et al.**

Anonymous Referee #3

Received and published: 20 August 2019

Review

This paper uses a NWP model with atmospheric chemistry and aerosol microphysics to compare and evaluate different DMS inventories over the Canadian Arctic. Additionally, the authors investigate the role of DMS in the simulation of Arctic aerosol. There results indicate (in general) a very poor agreement with cruise and aircraft observations of DMS. This is unsurprising given the short temporal timescale of the observations used. It was found that a satellite derived product with 8-day resolution was 'better' than a monthly climatology (Lana).

Overall the paper is very strangely structured and difficult to follow. It also contains typos and labelling mistakes which must be addressed before publication. Additionally, many of the plots need improvement. In general the paper presents some interesting ideas but requires significant editorial and scientific clarification (and potentially revision) before publication. Below are my comments to improve the paper:

Line 31: change 'The atmospheric aerosol' to 'Atmospheric aerosol' (aerosol is plural)

Line 37: Bates et al., 1987 is a extremely outdated reference, use https://journals.ametsoc.org/doi/full/10.1175/BAMS-D-15-00317.1 or https://journals.ametsoc.org/doi/pdf/10.1175/BAMS-D-14-00145.1 for most up to date state of Arctic measurement network.

Line 51: 'On the global scale, the CLAW hypothesis may be flawed,' I think you mean that the impact of the feedback is trivial.

Line 53: 'However, recent atmospheric observation and modeling studies suggest a significant role for DMS(g) in particle formation above oceans, especially in remote areas with low concentrations of pre-existing aerosol such as the Arctic Ocean in summer (Leaitch et al., 2013; Ghahremaninezhad et al., 2016; Quinn et al., 2017).'

This gives the impression that we would potentially see a CLAW feedback in the Arctic. However, greater cloud cover is more likely to warm the Arctic surface rather than cool it so the inference that 'CLAW' could occur is incorrect. Please reword.

Line 67: 'For example, the abstraction pathway (with the ratio of 75% of total OH and DMS oxidation) is the dominant reaction at 300 K (Hynes et al., 1986).'

I'm not sure why a reaction pathway that dominates at 27ïĆřC is relevant to Arctic atmospheric chemistry (even in summer).

Line 93: 'to have' please change to 'had' and 'of comparable level' to 'comparable'

Line 93: Are these seawater concentrations of DMS or atmospheric concentrations?

Line 167: I would suggest changing CLIM1 to LANA. Lana is well known climatology and immediately recognizable to modellers.

Line 189: 'coarse' should be 'coarser'

Figure 1 (and all others): Please do not use the rainbow colour scale in plots it is very difficult to interpret and distorts the results. In particular, your use of the rainbow scale for a difference plot makes interpreting the plot very hard. I suggest using a brewer colour scale or equivalent.

Line 206: 'In the case of simulation using CLIM1, constant (temporally) climatology for the month of July is used, while in the case of simulation using SAT, DMS(aq) is updated approximately every 8 days whenever the satellite-derived DMS(aq) is available'

What do mean by when they are available? Are they sometimes unavailable? Additionally, as I understand this climatology is merged with Lana at high-latitudes so regions in this model run also have static DMS concentration over the month. What percentage of this new DMS product is actually Lana?

Line 208: 'Figure S1 shows the satellite-derived DMS(aq) concentrations for the SAT time intervals, every 8 days, during July and August 2014 (July 1st to 3rd, July 4th to 11th, July 12th to 19th, July 20th to 27th, July 28th to August 4st and August 5th to 12th).'

Does this mean Figure 1 shows the average?

Figure 3: Please see my comments on Figure 1. At the moment it looks like there could be significant differences in DMS concentration over regions where the DMS climatologies are identical?

Figure 4: see comments on figures 1 and 2.

Line 260: "These flux estimates, based on measurements, are comparable with the present simulations."

With both CLIM1 and SAT? So changing the DMS inventory has had no impact on your DMS emissions?

Figure 5: This figure is difficult to understand. What do the grey dots represent? CLIM11+ave-obs is not explained in the caption or the text. Due to the linear y-axis it is very difficult to judge the fit in the model BL (which is arguably the most important region).

Line 275: 'The scatter plot in Figure 6 shows the statistical comparison of the model simulations (SAT and CLIM11) with the observation results. Overall, observation and model results are of similar magnitude, but not correlated. The simulation using SAT is in slightly better agreement with the measurement based on root mean squared error and mean bias values of 27.6 and -4.7 compared to 29.5 and -6.6 for the simulation using CLIM11, also better correlation coefficient (as shown in Fig. 6).'

I'm unsure how model and observations can be of similar magnitude but not correlated? This figure shows a terrible agreement between the model and observations-there is really no other interpretation. Additionally, you have included a regression line on what is clearly not a linear relationship and which is not statistically relevant to a model evaluation (typically you would add a one to one line to highlight agreement). Overall, it is unsurprising that the model is unable to capture aircraft point measurements even with a relatively higher (8-day) DMS resolution – which is likely the reason SAT is a slightly better fit. This looks like a clear example of sample bias and I question the usefulness of this comparison.

Line 302: 'These are the physical parameters affecting the sea-air flux. Overall the model is in good agreement with observations, given the model resolution,'

Really? That is not how I would interpret this plot. The most significant differences, particularly towards the end of the month, seem to coincide with the models failure to simulate sea surface temperatures. I'm also unsure what resolution you are referring to here, spatial or temporal?

Line 329: You refer to a figure 8c but figure 8 is not labelled as such.

Figure 9: Again a rainbow colour bar is a bad choice in general but for a difference plot doubly so. Additionally, the plots are also labelled in the caption (a,b,c) but not in the plot.

Line 344 (section 4.2.1):" indicating that the melt pond sources did not contribute to the two high DMS(g) events observed onboard the Amundsen."

This simulation is interesting however it is unclear which DMS climatology that you have used in this simulation, as static monthly climatology (which is very unlikely to capture specific plume events) or the 8-day resolution SAT inventory. The use of either is unlikely to reproduce specific DMS events observed on 2-3 day timescale (particularly Lana) – which I would argue is the reason for your poor model evaluations. Therefore, it seems a stretch to rule out melt ponds as an important DMS source. Overall, this section seems entirely divorced from any other part of the paper.

Section 4.2.3: In the simulation CLIM1-ave+Obs you have (as I understand it) used observations from the NETCARE campaign to update the Lana climatologies. Why update Lana, when SAT has a higher temporal resolution and you have previously shown that SAT is better (i.e. Fig 10)?

Line 380 (Fig 10): 'The statistical evaluations in this figure indicates a significant improvement in CLIM 11 model-observation comparison with this update (Fig. 10).'

Really? I don't see a significant improvement. Correlation between observations and the model is low for all three climatologies, although SAT is the best (again I would argue because it has a higher temporal resolution), which begs the question why it wasn't used for the update. Additionally, as in the previous figure, you have included a regression line rather than a one-to-one line which would make it easier to judge the comparison.

Section 4.3: Throughout this section is unclear exactly what simulations you are com-

paring. It seems you have switched from comparing DMS climatologies to comparing models with and without DMS. To make the paper flow better I would suggest beginning the results section with the impact of adding DMS to the model and then discussing the impact of different DMS inventories.

Line 458: 'In general, GEM-MACH suggests the enhancement of particles between 50 to 100 nm to be higher than particles between 10 to 50 nm for the high Arctic. This difference between Abbatt et al., (2019) and GEM-MACH results could be partly due to missing other natural sources (e.g., organics, see Burkart et al., 2017; Willis et al, 2016) in the model. Possible inadequacy in model representation of particle nucleation process may also contribute to the size discrepancy between model and observation.'

The enhancement of larger particles is more likely the result of too larger a condensation sink leading to condensation of SO2 rather than new particle formation. This could result from an underestimation of sink processes or an overestimation of other aerosol sources.

Line 471: 'The model simulation in this study compares well with the observations'

I'd say reasonable well, given the number of observations. However, at least 20% of the time the model is almost a factor of 10 lower than the observations. Why is the model so wrong on July 14th?

Line 529: 'By adding DMS(g) in the GEM-MACH model, the atmospheric SO2 concentration increased (up to ∼100% for some regions). This increase in may play a significant role in the growth and nucleation of aerosols.'

Doe this improve the models representation of SO2?

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-470, 2019.