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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Vertical profile of atmospheric dimethyl sulfide in the Arctic Spring and Summer" *by* Roya Ghahreman et al.

Anonymous Referee #1

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This paper addresses aspects of the linkage between surface DMS emissions and Arctic CCN/cloud dynamics. Arctic clouds play a critical role in the surface radiation balance and thus sea ice dynamics. The prevalence of stable atmospheric conditions over sea ice can inhibit a direct connection between local surface emissions and clouds, thus long range transport and downward-mixing of CCN and aerosol precursors may be more important, as in Lunden et al. (2010). See Shupe et al. (2013) for further evidence that long range transport of moisture, CCN, etc. are important for arctic cloud dynamics (doi:10.5194/acp-13-9379-2013).

Studies referenced by the authors reveal that seasonal DMS emissions in the marginal ice zone (MIZ) and open water can be quite large, with significant variability in space

and time. Diagnosing a link between these emissions and aerosol/cloud dynamics ultimately requires CTM simulations (and physically realistic, validated cloud/aerosol models!). This appears to be the authors intention here, but to me their approach seems backwards. They use modeling to explain the source of DMS measured on their flights. Most of the measurements are close to the surface and presumably within the atmospheric boundary layer (ABL), over ice covered seas, the MIZ and open water. Except for flights over ice covered seas in spring, the source of DMS near the surface is less interesting that the fate of that DMS, since the source is most certainly local but the potential effects on aerosol and cloud dynamics may be far afield.

I'm not sure section 3.1 provides much insight. It's not clear what 'cloud processing of DMS' means, and correlations of DMS with water vapor, CO and ozone are subject to such a wide variety of dynamic and chemical influences that they are difficult to interpret. Conclusions here are vague. Fig. S4 merely reveals that DMS emission peaks in spring and water vapor peaks in summer, which is expected based on existing information. The vertical distribution of all these species is much more interesting. The authors are using an advanced CTM to simulate vertical distribution and transport. I'd like to see model results for H2O, CO and O3 plotted with the observations on Fig. S3, in a manner similar to that shown for DMS in Fig.6. It's important to know how well the model reproduces the vertical distribution of these tracers. I think the authors should include a discussion of the vertical structure of the atmosphere in this section. Is there a well-defined atmospheric boundary layer on each flight? At what height? Does the profile of potential temperature, water vapor or vertical wind velocity indicate a well-mixed ABL and strong inversion? Is there evidence of atmospheric stability or of convection and mixing into the free troposphere? What does the CTM assume or simulate for vertical mixing on the flight dates and how does that compare with the observed profiles? How do profiles differ between open water, MIZ and ice covered regions? On p.10 the authors talk about limited vertical mixing and atmospheric stability during the April flights, but present no data or analysis to support this statement.

In all studies of atmospheric DMS over the ocean in the absence of deep convection, the concentration of DMS is elevated and well-mixed below the ABL inversion, dropping to near zero above the inversion. This is due to limited vertical transport and a relatively short photochemical lifetime. If NETCARE results differ from the usual DMS vertical distribution over the ocean, that's an interesting finding, implying convective transport into the free troposphere and perhaps an extended photochemical lifetime due to reduced water vapor or limited sunlight, all of which can be diagnosed with the CTM. In this regard, the vertical profile results for DMS in Section 3.2 are quite interesting. From Fig.4a it looks like atmospheric DMS is often elevated and highly variable near the surface in mid-summer. At 3000m the concentrations are near zero and I presume this represents the background free troposphere (are these measurements at the detection limit?). But, the few measurements on Jul12/17 at 1000m are significantly elevated. Are these within a deep ABL or are they evidence of long range transport in the FT? The latter would be very interesting.

Fig. 7 shows CTM back trajectories for the 0-200m level, so they presumably apply to aircraft measurements at the lowest altitude only. Are the colors in Figs. 7/8 meant to signify surface source regions for low-level air sampled by the aircraft? I don't understand what is meant my 'air mass residence time in seconds before arriving at the aircraft location'. The authors should define 'potential emission sensitivity'. Also, the blue line indicating the flight path is not visible and probably too small properly plot on a map of this scale. Maybe it's best to just put a dot on the map indicating the flight location. Are the authors suggesting DMS measured near the surface over the MIZ and open water in mid-summer near Devon Island originated as far away as Hudson Bay? That doesn't make sense to me. So I'm not sure about the significance of Figs. 7 and 8. It would be more interesting if these were forward trajectories rather than back trajectories.

For July flights over the ice edge and open water in mid-summer (a time of extended daylight hours and perhaps greater photochemical oxidation) the author's conclusion

that DMS sampled near the surface represents local emissions is quite reasonable. DMS within the ABL at 0-200m is likely to have originated from surface emissions quite near the sampling site under these conditions. I would be much more interested in back trajectories for measurements at the 1000m level, especially if this is above the ABL. If not, then the statement on p.9 that 'The decline in DMS mixing ratios with height may be due to a combination of weak vertical mixing and photochemical reactions' is entirely comparable to conditions that exist over the ocean in most other locations in the world. This seems to be supported by the CTM results in Fig. 6. I would think they could provide evidence of vertical structure in the atmosphere to further support this conclusion.

For April flights, the measurements indicate high DMS concentrations near the surface, which is understandable given algal blooms associated with the spring melt, but also elevated concentrations aloft, which is very interesting. It would be good to know the fractional ice coverage for the April flights and if the surface exhibited extensive melting with open leads. We also need to see an analysis of the vertical structure of the atmosphere for these flights. Fig. 8 should show back trajectories for the upper flight levels to help diagnose the significance of long range transport and potential source regions. This seems more relevant than the source of surface atmospheric DMS, which is likely to be local and may be inhibited by atmospheric stability from contributing CCN and cloud dynamics aloft. Though it would be interesting to see a forward model trajectory for the surface air and an analysis of the potential to influence to aerosol aloft over remote, ice covered regions. Heat fluxes in the MIZ, where open water is present, might drive sufficient convection to transport surface emissions from the MIZ and open water to ice-covered regions, where cloud formation is driven by mixing from above. Hopefully, the CTM can reproduce these processes.

In summary, although the amount of data is limited, I think the authors have interesting measurements from an important region of the world, and they have a very powerful modeling system at their disposal. But I don't think they have done sufficient analysis

of the specific meteorological and atmospheric conditions on each flight for me to fully understand the observations, and I don't think they've made best use of the model's capabilities to diagnose the fate of DMS in the Arctic ABL. I'm very interested in this topic and look forward to a revised version of this contribution.

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