

## ***Interactive comment on “Vertical profile of atmospheric dimethyl sulfide in the Arctic Spring and Summer” by Roghayeh Ghahremaninezhad et al.***

### **Anonymous Referee #2**

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This paper discusses seasonal measurements of DMS in the Arctic atmosphere with special emphasis on vertical distributions and evaluation of sources and impact. I have to say, though, given the title, that I was surprised to see only a handful of the measurements (between 10 – 12 samples total in 2 separate campaigns) were obtained at altitudes greater than 500 m. Given the variability and uncertainty in DMS emission intensity and distribution, this seems to be a marginal data set for evaluation by the modeling tools that were applied here. Furthermore, the measurements are obtained in a very limited geographic range of the Arctic and may not be particularly representative of the Arctic. Still, given the paucity of reported airborne measurements of DMS in the Arctic, there may be interest to see these data presented and discussed, otherwise

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they just disappear into some data repository.

First, I think the description of the measurement technique needs some clarification, even though it is based on methods described 20 years ago. My main questions to be cleared up: 1) Sample times range from 2 – 11 minutes. What is the flow rate, and what are the resulting sample volumes? 2) Reference to Sharma et al. (1999) as a description of the method seems incorrect as listed. The listed Sharma et al. (1999) paper describes a 10 L sample collected on molecular sieve. The reference Sharma et al. (1997) is a typo and should also be 1999. This appears to be the correct reference to the Tenax method. Sharma (1997) is the MSc thesis, which should contain all of the details but is not easily available which is why some extra detail is needed. The Sharma et al. (1999) paper lists the detection limit as  $\pm 6$  pptv in a 2 L sample (defined as 2:1 S/N), and accuracy and precision of  $\pm 12\%$  (not sure what this means, though). This manuscript refers to a  $\pm 12$  pptv uncertainty (not specified if accuracy or precision). Surely this can't be constant for samples of different volumes. Was this determined based on the triplicate standard measurements, or was this somehow referenced back to Sharma et al. (1999)? Please clarify this, esp. since many of the samples reported seem to be in this range. 4) Sample storage tests: how much standard was loaded onto the cartridge? Were different loadings tested? Please also note the axis of the graph in Figure 3. Is this really the DMS mixing ratio in pptv, or some ratio to initial DMS addition? Is the uncertainty in the stability also considered in assessing the overall uncertainty of  $\pm 12$  pptv?

P7, L.22. The GEOS-CHEM model is certainly widely used for many applications, but as I looked at the Mungall et al., 2016 paper, it seemed that there were significant issues with the DMS emissions specific for the region that was studied, and these conclusions suggest that the GEOS CHEM model needs to be applied with caution for specific locations and seasons for dealing with a highly variable compound such as DMS. Just because it was used is not necessarily an endorsement for its applicability for these measurements, at least without some caveats.

P8, L6. Meteorology and profiles. It is unusual to see the vertical profiles of species averaged for an entire campaign. The DMS measurements need to be related to the atmospheric vertical structure and variability during each flight, or there should be some demonstration that a more suitable analysis is found by taking mission averages.

P8, L 16. Please provide the reference(s) to the heterogeneous oxidation of DMS on or within aerosols.

P8, L 24. While sources of DMS and H<sub>2</sub>O are the ocean (though there could be other sources of DMS), different correlations could be observed due to the high spatial and temporal variability of DMS (compared to H<sub>2</sub>O) and effect of temperature on either gas. I assume this discussion of the correlation is related to the argument about the greater presence of clouds in spring vs. summer, and (again) the assertion of a major role of cloud processing of DMS. The presence (note typo on P8, L25) of clouds could potentially have a greater impact on OH production below or above the clouds with subsequent impact on DMS lifetime. Perhaps this was suggested by the statement regarding clouds during transport, but this seems a speculation that could be checked from observations. It is not clear what the authors are saying regarding the effect of clouds. . . .reduction of DMS due to heterogeneous O<sub>3</sub> oxidation, or increase of DMS due to slower OH oxidation? Unfortunately, the data is not available to support either impact so this just remains as speculation.

P9, L 25. The authors reference again the work reported by Mungall et al., 2016 who measured DMS at the same general location and time period (as far as I can tell) as the airborne data for July, 2014. It would be helpful to discuss the airborne measurements and the impact of the very large variability in DMS emissions that were found in Mungall et al. Perhaps there were times of reasonable overlap between the ship based and airborne measurements that could also be compared. It seems that the large DMS mixing ratios observed during parts of the Mungall et al. cruise were not sampled by the aircraft. Furthermore, the reported range of a factor of 60 in the calculated DMS flux should further raise a cautionary flag about how well climatological

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models and emission estimates might be applied to the data.

P10, L 19. Here and elsewhere in the manuscript the possibility of long-range transport of DMS is mentioned. In this context, it would be helpful if the authors could provide some reasonable estimate of the lifetime of DMS and the possible transport times that are considered “long-range” vs. local.

P10, L 25. I am not a fan of averaged vertical profiles for variable species (such as aerosols) without some indication of the variability of the measurements over the different profiles. I assume that the higher particle number is associated with the specific profile in July where the measured DMS is about 100 ppt. Or is the higher level of aerosol more widespread? Also, the springtime levels of DMS are even higher than those found in summer, but no significant aerosol production here? This level of discussion and interpretation is incomplete and not particularly informative.

P11, L 3. Thin aerosol layers are common in the Arctic spring. It seems very unlikely that the high altitude peak in aerosol is in any way related to DMS. If the authors really consider the DMS source likely, perhaps some analysis of what sort of conditions might be necessary to produce the observed aerosol abundance.

P11, L 13. Do I understand correctly that the GEOS-CHEM results shown in Figure 6 are compared only for the 2 – 10 minute sample integration time of the measurements? It would be interesting to see some examples of the variability of GEOS CHEM results to perhaps provide a more detailed context for the very limited number of actual samples that were collected. Often models track measurement features, but may be slightly off in time or altitude.

P11, L 23. I am not sure what the authors want to convey when they claim that the GEOS CHEM data and the measurements are “within their respective uncertainties”. Perhaps “variability” is a better term than uncertainties in this case. There are clearly more uncertainties in the model results beyond just the calculated mixing ratios.

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P11, L25. From Figure 6, the variability with altitude actually seems less in the summer compared to spring, though the gradient is less in the spring, as noted. Summer range about 20 – 40 ppt at low altitudes, <10 ppt at max altitude; Spring 30 – >50 ppt at all altitudes. Could the authors please quantify the difference in seasonal variability at different altitudes to demonstrate their assertion?

P12, L16. 30% difference might also be due to incorrect distribution and intensity of model DMS emissions.

P13, L5. I am concerned about the conclusions from the Flexpart analysis where the authors claim a large influence for specific regions. As noted, these analyses suggest where the sampled air spent time in the boundary layer. This analysis must be combined with an emission distribution to provide the source attribution. For example, without additional information, it is misleading to suggest that North Pacific was a significant source of DMS measured in the Arctic. It is a potential source. If the authors have some other data (ocean color, for example) to indicate DMS emission in the North Pacific at that time, then that could be a basis for the claim. Without this emission information, the Flexpart analysis can only indicate “potential” regions.

P13, L9. Could the authors make some estimate, based on typical DMS lifetimes, what the source region DMS mixing ratios would need to be to support the observed mixing ratios in spring?

P13, L16. The link established in this data set between aerosol formation and DMS is very weak and essentially presented as a given with no detailed analysis. While we all seek global significance of atmospheric chemistry and a potential link to climate, I think that it is an overreach to state that, without much greater data coverage and analysis, the observed vertical variation will significantly impact the Earth’s radiation budget.

Figures 1 and 2. These are not particularly helpful figures to identify the sample locations. I would suggest combining each season’s flights into several composites that showed 1) a map of flight tracks, 2) an altitude-longitude (and/or latitude) cross-section

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that identified the location of the samples and the flight tracks. Given only a few flights, a color code could make things relatively easy to follow.

Figure 3. Already noted..Vertical axis mislabeled?

Figure 5. Possible to add shading to each mean profile to show range or std deviation in each bin?

Figure 7. I could not distinguish the blue lines for the flight tracks. Are these calculated for a single sample or some altitude? Please specify. I certainly do not understand the units for potential emission sensitivities. I thought that these indicated the time spent in the boundary layer (in this case 0 – 200 m) during the period of the back trajectories (4 days here). Is there a factor of 1000 missing here?

Table 1. It would be helpful to add the flight number and altitude to this table.

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