Dear Reviewer,

Thanks for the great comments on the paper. Please find the answers we provided for your comments and questions, below.

Your Sincerely,

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Referee #1

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).

Thank you- We refereed to Shupe et al. (2013).

(Page 2, line 23) Shupe et al., (2013) provided the evidence for the formation of clouds and transport of moisture and aerosol particles, likely accompanied warm air masses, from lower latitudes into the central Arctic during summer.

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.

Please note that we tried to compare DMS(g) mixing ratio (and source) during summer and spring. The linkage between DMS and aerosol/clouds/CCNs is important, however, this topic is beyond the scope of our study. We used CTM simulations to compare with our observations.

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.

The sampling altitudes are mentioned in table 1. Some samples were collected whithin the ABL. To show the boundary layer height, we referred to Aliabadi et al., (2016). Also, we added ice fraction (Figs S1, S3) to show the ice coverage during field campaigns.

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.

Agreed, we removed section 3.1 and Figs 5, S1, S2 and S4, and instead we added:

(Page 10, Line 11) The relatively larger observed DMS(g) away from open water sources springtime relative to summer suggests longer DMS(g) lifetimes in April than July, possibly due to lower OH mixing ratios enabling more long-range range transport of DMS(g) (Li et al., 1993). Lower water vapour and higher DMS mixing ratios during the spring compared with the summer (Fig. S2) suggests that more of the April DMS(g) originated from open water sources further away from the observations point than in summertime. The greater ice cover and increased presence of DMS(g) at higher altitudes during April suggests an origin from further south than in summertime. More water vapour will initially accompany that DMS(g), but the Arctic is cold in April, especially aloft, and the low water vapour indicates significant loss via cloud processes during transport. Some of the water vapour loss will occur via the ice phase, and DMS oxidation in the aqueous-phase was likely relatively insignificant during this time (Henry's Law constant for DMS is relatively small: 0.14 mol/L-atm) or the DMS(g) values at their origin were much higher than the present observations.

Ozone depletion during spring was observed within the boundary layer (Fig S2) and is well documented in the literature (e.g. Barrie et al., 1989). Ozone depletion may further decrease OH near the surface and enhance DMS(g) lifetimes in the boundary layer due to reduced oxidation rates, contributing to the relatively larger springtime DMS(g) in our measurements. However, if DMS is present in the ozone-depleted boundary layer, halogen oxides, such as BrO radical, can be more important during winter/spring than summer and could oxidize DMS(g) (von Glasow et al., 2004, Chen et al., 2016).

DMS (g) vertical profiles are sensitive to the boundary layer height. For the summertime, Arctic the boundary layer height on various days (275±164 m), for the July 2014 campaign, is reported in Aliabadi et al. (2016). They showed that the profiles of the potential temperature exhibited a positive vertical gradient throughout the aircraft campaign (their Fig. 4). In addition, using vertical profiles of wind speed, they derived a positive gradient Richardson number (Ri) with a median of 2.5 (Their Fig. 7) throughout the aircraft campaign. The magnitude of the positive gradient Richardson number is an indicator of the strength of thermal stability in the atmospheric boundary layer. Due to the strong thermally stable conditions during the field campaign, mixing was weaker compared to well-mixed boundary layers at mid latitudes. As a result the summertime measurements show a strong decrease in DMS(g) above the boundary layer. Although there is no reference for the April 2015 campaign boundary layer, we expect similar boundary layer characteristics in the stable Arctic boundary layer at high latitudes due to the even more reduced thermal forcing with large sun angles in the month of April compared to the month of July. The springtime measurements show a more uniform vertical profile suggesting transport in the free troposphere from open water sources that were relatively farther distance from the observation point in springtime than in summer.

Aerosol number concentrations and size distributions during the July, 2014 study are discussed by Willis et al. (2016) and Burkart et al. (2017), who show that increases in the number concentrations of smaller particles (5-20 nm), believed to reflect new particle formation (NPF), occurred principally near the surface during July 12th 2014. The highest levels of DMS(g) during the July study also occurred near the surface (Fig. 4a), and both Willis et al (2016) and Burkart et al. (2017) noted increased MSA near the surface associated with two case studies of NPF. In the clean conditions of the Arctic summer (e.g. CO in Fig. S2), the low-level DMS may contribute to

NPF. The springtime Arctic differs in that the aerosol mass near the surface is much higher, resulting in a higher condensation sink that, in addition to other potential factors, inhibits NPF. During the springtime flights, there was no evidence for NPF near the surface, and only a few instances aloft. Unfortunately, no sampling for DMS coincided with those few events, and we cannot say if they were connected with the DMS(g) aloft.

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.

Figure S2 shows vertical distribution for measurement and modeling simulations.

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.

Correct, we added more information:

(Page 11, Line 4) DMS (g) vertical profiles are sensitive to the boundary layer height. For the summertime, Arctic the boundary layer height on various days (275±164 m), for the July 2014 campaign, is reported in Aliabadi et al. (2016). They showed that the profiles of the potential temperature exhibited a positive vertical gradient throughout the aircraft campaign (their Fig. 4). In addition, using vertical profiles of wind speed, they derived a positive gradient Richardson number (Ri) with a median of 2.5 (Their Fig. 7) throughout the aircraft campaign. The

magnitude of the positive gradient Richardson number is an indicator of the strength of thermal stability in the atmospheric boundary layer. Due to the strong thermally stable conditions during the field campaign, mixing was weaker compared to well-mixed boundary layers at mid latitudes. As a result the summertime measurements show a strong decrease in DMS(g) above the boundary layer. Although there is no reference for the April 2015 campaign boundary layer, we expect similar boundary layer characteristics in the stable Arctic boundary layer at high latitudes due to the even more reduced thermal forcing with large sun angles in the month of April compared to the month of July. The springtime measurements show a more uniform vertical profile suggesting transport in the free troposphere from open water sources that were relatively farther distance from the observation point in springtime than in summer.

Also, please see: (Page 12, line 17)

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.

The detection limit is ~ 7 pptv (Page 6, line 10). The measurement at 3000 m was below detection limit during July (mentioned in Table 1). For July 17, again the measurement was below 7 pptv at ~1000 m. However, for July 12th, simulation suggested a local (Lancaster Sound) influence:

(Page 9, line 21) However, relatively high DMS mixing ratios (> 15 pptv) were observed for July 12th at high altitudes (> 800 m), and FLEXPART results shows influence of local source, Lancaster Sound for that day (mentioned in Section 4.2). On this day, NETCARE results do not follow the usual DMS vertical pattern of high DMS at the surface declining with altitude to near zero above the MBL. Instead, high concentrations aloft on July 12 imply convective transport into the free troposphere and potentially an extended photochemical lifetime due to reduced water vapor or limited sunlight.

During April, DMS(g) samples were collected above ice and snow surfaces, and heat fluxes were negligible. Figure S3 shows the ice fraction during the April 2015 campaign.

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.

We changed Figs 7 and 8. New figures show back-trajectories for 1000 m.

Please note that we are interested to know the potential source of DMS, and as a result, we plot back-trajectories. More information is added to the manuscript to address your comments regarding the FLEXPART simulation:

(Page 14, line 2) FLEXPART-ECMWF modeling was used to explore the origin of air samples measured along the Polar 6 flight tracks. Figures 6 and 7 show the potential source regions of these air samples four days before the releases along the flight path. More specifically, the response function is shown to all releases of a passive tracer, which in this case has properties of dry air. If this response function would be folded with an emission flux of the tracer the concentration of this tracer at the release location along, the flight paths could be calculated. We chose to show the potential emission sensitivity after four days. Sharma et al., (1999) showed that atmospheric DMS(g) lifetime was 2.5 to 8 days in the high Arctic. More details about FLEXPART and the potential emissions sensitivity (PES) could be found in Stohl et al. (2005) and references therein.

Figure 6 shows two examples of FLEXPART-ECMWF PES for 4-day back trajectories in July 2014: an influence from a broad area and especially Lancaster Sound (local region) and north on July 12th (Figure 6, left panel), and Hudson Bay, and Baffin Bay (south) on July 19th (Figure 6, right panel). A more detailed analysis of PES reveals that the measured air mass descended from >1500 m on July 19th, which may explain the low DMS(g) mixing ratios.

Figure 7 shows some examples of FLEXPART-ECMWF PES simulations for 4-day back trajectories during April 2015. For the flights near Alert and Eureka on April 9 and 11, some DMS may have originated from ice-free areas of the Nares Strait and Baffin Bay (Figure 7, upper left and right panels, respectively). For the April 13 flight, the Norwegian Sea, North Atlantic Ocean and Hudson Bay are additional potential source regions (Figure 7, lower left panel). The highest

DMS, measured on April 20 near Inuvik is associated with the North Pacific Ocean (Figure 7, lower right panel).

Assuming a DMS atmospheric lifetime of 1 to 4 days, these results suggest that the DMS(g) measured during July 2014 originated primarily from the local region over Baffin Bay and the Canadian Arctic Archipelago. For spring 2015, the DMS(g) sampled was from a range of sources, including Baffin Bay, possibly the Norwegian Sea, the North Atlantic Ocean and the North Pacific Ocean.

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.

The ice fraction is shown on Fig S3 for April 2015. Please see:

(Page 9, line 27) During April, DMS(g) samples were collected above ice and snow surfaces, and heat fluxes were negligible. Figure S3 shows the ice fraction during the April 2015 campaign.