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Interactive comment on "Observational evidence for the formation of ocean DMS-derived aerosols during Arctic phytoplankton blooms" *by* Ki-Tae Park et al.

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We thank Referee 2 for providing insightful suggestions, which have considerably improved the readability of the revised manuscript. Our responses to this referee's one general and several comments are stated below. The revised manuscript was uploaded in the form of supplement.

A general (major) concern

Overstatement of our results and conclusions: A set of unique concurrent measurements (including atmospheric DMS and MSA concentrations, S isotope composition of aerosols, aerosol particle concentrations, and satellite-based biomass) that we made

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during Arctic phytoplankton bloom periods provided compelling evidence that there is a connection between DMS emissions and SO42– aerosol formation in the Arctic atmosphere. However, our measurements did not provide direct evidence supporting the connection between DMS emissions and the formation and growth of aerosol particles. We agree with Referee 2 that to directly confirm that DMS emissions lead to new particle formation, measurements of inorganic, organic, and halogen species are also needed. Therefore, we have amended the revised manuscript (Abstract, Results, and Conclusion) by changing the text reading "the direct association of DMS emissions with the formation of aerosol particles" to "the significant association between DMS emissions and the formation of submicron SO42– aerosols" (lines 19–20, page 1; lines 28–29, page 8) and "possible association between DMS emissions and the formation of aerosol particles" (lines 5–6, page 3). We have also noted that confirmation of this proposed link requires measurements of inorganic, organic, and halogen species (lines 22–25, page 5; lines 1–4, page 9).

Specific Comments

1. We have removed 'ocean' from the title

2. Abstract. Tone down the conclusion in our results reading "the direct association of DMS emissions with the formation and growth of aerosol particles": As stated in our response to the general concern above, we have changed the above statement to "the formation of submicron SO42– aerosols was significantly associated with an increase in the atmospheric DMS mixing ratio" (lines 19–20, page 1).

3. Introduction. A direct link between marine biota and climate change may not be accurate: The connection between marine biota and climate change is far from simple, because multiple and complicating processes are interwoven, and consequently our assertion of a "direct link" may not appropriate. Therefore, we have changed "direct association" to "close linkage" (lines 9–10, page 2) and ". . ..a direct association. . .." to ". . ..an association. . .." (lines 19–20, page 2).

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4. Introduction. The DMS-climate feedback may not as significant as other feedback processes, for example the iodine-feedback: We believe that the DMS-climate feedback mechanism is yet to be tested in the Arctic environment, where most rapid warming has occurred. The critical testing has not been possible in the Arctic, mainly because of the absence of atmospheric DMS data. The absence of data necessary to perform this critical testing greatly enhances the utility of our present study. In the revised manuscript we have more clearly elaborated the value of our study testing the DMS-climate feedback (lines 17–19, page 2). We agreed with Referee 2 that chemical species other than DMS may be equally or even more important than DMS in climate feedback. We have addressed this issue in response 8 below.

5. Page 3. Provide more information about the online aerosol size distribution measurements: We have addressed this issue in our response (8) to comments of Referee 1 (see lines 19–23, page 3).

6. Page 4, line 31: We have replaced “3-fold higher” with “more than double”.

7. Page 5, lines 9–10. The greater formation of nucleation mode particles in May than in April is not a direct cause of the strong correlation between DMS/MSA and nucleation mode particles: Our description of Figure 1 was misleading. In the revised manuscript we have changed “the observed nucleation events also concurrently occurred with high atmospheric DMS mixing ratios” to “The observed increase in nucleation mode particles coincided with high atmospheric DMS mixing ratio and MSA concentration”.

8. Page 5. Is DMS only responsible for the formation of small particles?: Figure 1a shows that approximately 45% of the variance in small particle formation was explained by DMS. This does not mean that DMS was the only contributor to small particle formation. Iodine may have contributed to explaining the remaining variance. We explicitly stated in lines 18–22 that iodine could be an important contributor, as demonstrated in an iodine-rich coastal environment.

9. Page 5. This study did not provide direct evidence that DMS-derived sulfate was

responsible for the formation of nucleation mode particles: We agree that molecular-scale measurements of chemical species actually involved in nucleation processes are required to provide direct evidence for the DMS-derived aerosol formation events. Therefore, we have added a short paragraph indicating the limitation of our study in this regard (lines 22–25, page 5; lines 1–4, page 9).

10. Page 6. Need more evidence (such as box model) to support strong statements regarding DMS-derived aerosol formation: As supporting evidence that DMS can substantially contribute to the fine-mode particle formation we observed in the Arctic atmosphere, we have cited the modeling work of Chang et al. (2011), who showed that atmospheric DMS mixing ratios > 100 pptv are sufficient to account for the formation of ultrafine particles, particularly when background particle concentrations are low (condensation sink < 7.0 m-2). To support the confidence of our statement, we have added a short paragraph (lines 10–13, page 8) describing the results of Chang et al. (2011) and have added this paper to the revised list of references.

11. Page 6. What role had super-micron particles in surface area or as a condensation sink?: Because super-micron particles have the greater surface area than sub-micron particles, and the condensation sink is proportional to the surface area of aerosol particles, the increase in super-micron particles would be expected to increase the condensation sink and depress the nucleation rate. We have added a short paragraph explaining the link between increase in super-micron particles and decrease in the nucleation rate (line 25, page 2 and lines 14–15, page 6).

12. P6, line 19: We have removed “hypothesis” from line 19, page 6.

13. Is there non-biogenic DMS?: More than 90% of DMS is derived from marine ecosystem. Other DMS sources are negligible. Reference to the publications of Kettle and Andreae (2000) and Stefels et al. (2007) have been added to support the assertion that almost all DMS was of marine origin (lines 10–11, page 2).

14. The title of section 3.3 “Aerosol formation from biogenic DMS” is inadequate: Be-

cause the data presented in the present study does not provide evidence supporting a direct link between aerosol particle formation and DMS-derived sulfate aerosol, we have changed “Aerosol particles formed from biogenic DMS” to “SO42– aerosol particles formed from biogenic DMS” (line 23, page 6).

15. Is there closure between the SMPS/DMPS/APS derived mass distributions, or integrated mass distributions up to 2.5 microns and sulphate/seasalt?: This issue was beyond the scope of the present study, and our data did not enable it to be addressed because the total mass of the aerosol samples and the concentration of organic carbon collected on the PM2.5 filters were not analyzed.

16. How much of the DMS is available for nucleation/condensation of sulphuric acid to the aerosol phase and over what timescale?: This question was also beyond the scope of the present study. The oxidation rate of atmospheric DMS in our study area could not be resolved because we did not concurrently measure key parameters (e.g., halogen species) affecting the conversion efficiency of DMS to SO2. Therefore, we can only say that an atmospheric DMS mixing ratio >100 ppt might be sufficient for the formation of ultrafine aerosol particles when background particle concentrations are low in the remote Arctic marine boundary layer. We have addressed this issue in our response 10.

17. Our results were overly stated in Conclusion: We have already addressed this issue in our response to a Referee 2’s general concern. We have clearly stated in the revised Conclusion (line 28, page 8 – line 4, page 9) that our measurements (atmospheric DMS and MSA concentrations, S isotope composition of aerosols, aerosol particle concentrations, and satellite-based biomass) were not direct evidence supporting a connection between DMS emissions and SO42– aerosol formation in the Arctic atmosphere.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/acp-2016-1161/acp-2016-1161-AC2-

supplement.pdf

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