

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 1 for providing numerous specific suggestions, which have considerably improved the readability of our revised manuscript. Our responses to this Referee's comments are presented below. The revised manuscript was uploaded in the form of a supplement.

Technical Comments

1. P2, lines 5 and 8. Check the references: We have added omitted citations to the revised list of references.
2. P2, line 12: We have changed “DMS eventually emitted” to “DMS is emitted”.

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3. P2, line 13: We have removed “rapidly” from line 13.
4. P2, line 14–16. Modify the description regarding Henry's constant and its association with particle nucleation: We agree with this referee that Henry's constants are macroscopic representations of particle formation, while particle nucleation is more a molecular-level process. Therefore, we made the following changes: “The MSA and H₂SO₄ formed from DMS tend to transform into new particles via multiple nucleation processes (i.e., binary, ternary, and ion-induced) or condense onto existing particles because of their low volatility nature”.
5. P2, lines 23–25. Inappropriate use of “condensation”: We have replaced “condensation of” with “the occurrence of large source of”, and the text “because of the high level of production of condensable vapors and the relative lack of a condensation sink of pre-existing particles” has been added in lines 24–25.
6. P2: A research report by Leaitch et al. (2016) has been cited in line 27.
7. P3: Reports by Chang et al. (2011), Browse et al. (2012), Leaitch et al. (2013), Turnved et al. (2013), and Willis et al. (2016) have been cited in lines 3–4 (page 3), and the revised reference list.
8. P3. Provide more information about the use of DMA-CPC: In the revised manuscript (lines 19–23), we have stated that two discrete SMPS systems were used to measure the distribution of aerosol particle sizes in our study.
9. P4. Provide uncertainty for the $\delta^{34}\text{S}$ value for sea salt: In the revised manuscript (P4, line 25), we have included a measure of uncertainty associated with the $\delta^{34}\text{S}_{\text{ss}}$ value (21.0 ± 0.1‰ for sea salts. This uncertainty was estimated from direct measurements reported by Böttcher et al. (2007); this paper has been cited and added to the reference list as supporting evidence.
10. P5. Provide more description about the relation between atmospheric DMS mixing ratios and MSA concentration: We agree that some of the MSA, particularly that

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measured in April, was probably formed elsewhere and subsequently transported to the measurement site. Local DMS production explained much of the variation in MSA in May, but did not account for all MSA variations observed in that month (see Fig. 1a and c). Part of the remaining variance in MSA in May can be explained by MSA that was introduced to the measurement site as a result of long-range transport, and the efficiency of photochemical oxidation of DMS. Therefore, we have added a short paragraph (lines 4–6, page 5) noting that transport of MSA to the site was a possibility.

11. P15: We have added the time series for solar irradiance to Figure 1b.

12. P5, line 16: We have replaced “probably” with “possibly”.

13. P5, lines 18–20: We have cited O’Dowd et al. (2002) and Allan et al. (2015), and added a brief explanation to lines 18–19.

14. P6. Need an explicit explanation about changes in aerosol properties during the months of April and May: As this referee noted, our observations are not sufficient to prove that neutralization processes enhanced the growth of aerosol particles in the accumulation mode during the Arctic haze period. Therefore, to clarify the interpretation of our measurements we have revised the paper as follows (see lines 3–6, page 6): “The transition of aerosol microphysical properties from a distribution dominated by an accumulation mode (Arctic haze period) to a distribution dominated by nucleation and Aitken mode atmospheric particles (phytoplankton bloom period) was probably driven by the combination of three factors, including changes in air mass transport, incoming solar radiation and condensation sink processes (Tunved et al., 2004 and 2013)”.

15. P6, line 8. Inappropriate reference: we have removed the Sharma et al. (2012) citation from the text and the reference list.

16. P6, lines 20–22: We have added a brief overview of recent field observations reported in Willis et al. 2016, which is conceptually similar to our work.

17. P6, line 20: We have removed “(promoted by the values of Henry’s law constants

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for H₂SO₄ and MSA)” from the revised text.

18. P6, line 25 and P18. We have moved Figure S3 to the main text.

19. P7, line 6. Be consistent with S isotope ratios of anthropogenic SO₄²⁻ ($\delta^{34}\text{S}_{\text{anth}}$): We have replaced “(0–8‰)” with “(5 ± 1‰)”.

20. P8. Need to acknowledge potential contribution of anthropogenic SO₄²⁻ to NPF events during the phytoplankton bloom period: The contribution of anthropogenic SO₄²⁻ to fine SO₄²⁻ particles (< 2.5 μm in diameter) was still significant in May (30–60%). Unfortunately, we could not accurately estimate the relative contribution of biogenic versus anthropogenic SO₄²⁻ to the formation and growth of aerosol particles. However, recent field observations indicate that a considerable amount of SO₄²⁻ in aerosol particles having a diameter < 0.49 μm is biogenic (> 63%), based on size-segregated sulfur isotope analysis in the Arctic atmosphere. Therefore, we have added a short paragraph indicating the limitations of our study (lines 3–10, page 8), and cited Ghahremaninezhad et al. (2016) in support of our argument.

21. P8, line 28: We have removed the sentence including “confirming”.

Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/acp-2016-1161/acp-2016-1161-AC1-supplement.pdf>

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