

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

Anonymous Referee #1

Received and published: 22 February 2017

General – The authors present a unique combination of observations at Ny-Ålesund, Svalbard made during April and May of 2015 that are used to connect biogenic sulphate (from DMS oxidation) to new particle formation (NPF). I find the connection reasonably compelling. The paper is well structured and mostly well written, but I have technical questions about some of the discussions that need to be addressed. As well, there are recent relevant references that should be considered. The subject is appropriate for ACP, and it should be published if the authors attend to some details of the discussions in various areas of the paper.

Comments:

1. Check the references. Some referred to in the text are not in the list.

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2. Page 2, line 12 - "DMS is emitted. . ."
3. Page 2, line 13 – remove "rapidly" or quantify it.
4. Page 2, line 15 - The attraction between H₂SO₄ and water is an important factor in particle nucleation, and so in a broad sense there is a connection between Henry's Law constant and the ability of H₂SO₄ to contribute to particle nucleation, but Henry's law constants are macroscopic representations while particle nucleation is a molecular-level process. Are you saying that the Henry's Law constants control particle nucleation, or are you mostly referring to what factors might control the condensation of H₂SO₄ and MSA onto existing particles? Please elaborate, and also note that condensation may occur in the absence of water on a particle.
5. Page 2, lines 21-22 – The use of condensation here in the context of new particle formation contradicts your sentence above that states that MSA and H₂SO₄ are either transformed into new particles or condense on existing particles. Please correct.
6. Page 2, line 23 – For a reference to these small particles influencing Arctic cloud, see Leaitch et al. (ACP, 2016).
7. Page 2, lines 29-33 – Many references exist that are relevant to the discussion here. For example: Engvall et al. (ACP, 2008); Chang et al. (JGR, doi:10.1029/2011JD015926, 2011); Browse et al (ACP, 2012); Leaitch et al., Elementa (2013); Tunved et al. (ACP, 2013); Willis et al. (ACP, 2016); Croft et al. (Nat. Comm., 2017).
8. Page 3, Lines 15-19 – Was one DMA and CPC used to measure both indicated size ranges, or were there two DMA-CPC systems? Please provide either a reference for these measurements or the details of sampling and calibration.
9. Page 4, line 21 – The delta 34S value for sea salt is expressed without uncertainty, whereas the deltas for anthropogenic and biogenic S have uncertainties (or ranges). Please elaborate.

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10. Page 4, lines 30-32 – Are you trying to say that local oxidation of DMS was responsible for most of the MSA as opposed to more distant production of MSA? Figure 1b suggests that about 70% of the variance in MSA was connected directly with your measured DMS, but some MSA may still have been advected into the area. E.g. MSA levels below about 30-40 ng m⁻³ do not appear to be strongly associated with DMS. Please provide a more detailed and clearer discussion.

11. Figure 1 – This figure is impressive. It would be helpful to have the time series of solar irradiance in 1a or as a separate panel, even with SR represented in 1b, 1c and 1d. It would also help with your discussion on lines 9-11 of page 5.

12. Page 5, line 10 – “possibly” rather than “probably”.

13. Page 5, line 11 – add a reference for iodine (e.g. O’Dowd, Nature or Allan, ACP).

14. Page 5, lines 24-28 – I have a few little concerns about these sentences. It is unclear why you suggest that neutralization especially enhances growth into the accumulation mode. Acidic sulphate is hygroscopic, just as the salts; slightly more so. Are you trying to say that very acidic solutions will inhibit further transfer of acids from the gas phase? Sulphate accumulates in particles >100 nm through condensation of H₂SO₄ from the gas-phase, coagulation and through aqueous-phase production of H₂SO₄ (e.g. in cloud droplets). Neutralization of acidic sulphate may play a significant role in these processes, but NH₃ may also be an important factor in the production of new particles in the Arctic (e.g. Croft et al., Nat. Comm., 2017). You indicate that the sulphate from DMS oxidation gives rise to smaller particles, but particle production and evolution doesn’t care whether the H₂SO₄ is anthropogenic or biogenic. In a situation of relatively low gaseous precursor concentrations (as can be the case in the Arctic and from DMS), low existing particle surface area will help nucleation. Available sunlight will be a factor in the production of gas-phase H₂SO₄, but H₂SO₄ from biogenic sources is the same as H₂SO₄ from anthropogenic sources. The discussion here needs improvement. Also, remove “established explanations”.

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15. Page 5, line 32 – Sharma et al did not present particle number concentrations.
16. Page 6, lines 3-10 – This is particularly interesting because the decreases in the number concentrations and surface areas of the 100-1000 nm particles that are associated with the increase in sub-30 nm particles from April to May appear to be much lower than the decrease needed for NPF at Alert (Leitch et al., Elementa, 2013). Possibly one reason is the proximity to the sources: open water is close by Svalbard year round with potential for higher concentrations of SO₂ from DMS, whereas Alert is more distant from large areas of open water. Willis et al (ACP, 2016) document a case of NPF and growth associated with open water in the summertime Arctic that may be conceptually similar to your work.
17. Page 6, lines 10-12 - Reference is made to the Henry's Law constants being responsible for gas-to-particle nucleation. Please elaborate, considering comment 4 above.
18. Page 6, Fig. S3 – This figure seems to be a fundamental part of the discussion, and I believe it belongs in the main text rather than a supplement. This figure also demonstrates that some lower concentrations of MSA may result from distant transport to the site.
19. Page 6, line 29 – Why here do you give a range (0-8) for the deltaS of anthropogenic sulphate, when above (for equation 2) you give a value of 5(+/-1)? Please be consistent.
20. Page 7, Figures 4 and 5 – These are compelling results, but you need to acknowledge that there is still a significant fraction of the sulphate that is anthropogenic in origin, and that you cannot identify the origin of the H₂SO₄ contributing to the new particles. It is possible that the NPF is due to anthropogenic SO₂ (or a combination of anthropogenic and biogenic SO₂), and that the correlations result mostly from the coincidence of a lower condensation sink with the relative increase in biogenic S.

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21. Page 8, line 9 – Please change “confirming” to “indicating” or “showing”. There is no hypothesized relationship to confirm.

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