

Response to Referee 1

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.

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*”.
- 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 23–24. Inappropriate use of “condensation”:** We have replaced “*condensation of*” with “*the occurrence of*”, and the text “*the relative lack of coagulation and condensation sinks of pre-existing particles may result in the formation of new particles in the atmosphere*” has been added in lines 23–24.
- 6. P2:** A research report by Leaitch et al. (2016) has been cited in line 26.
- 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 2–3 (page 3), and the revised reference list.
- 8. P3. Provide more information about the use of DMA-CPC:** In the revised manuscript (lines 17–23), 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 24), we have included a measure of uncertainty associated with the $\delta^{34}\text{S}_{\text{ss}}$ value ($21.0 \pm 0.1\%$) 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 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 3–5, 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 15:** We have replaced “*probably*” with “*possibly*”.
- 13. P5, lines 17–19:** We have cited O’Dowd et al. (2002) and Allan et al. (2015), and added a brief explanation.

- 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 1–4, 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 19–21:** 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 18:** We have removed *“(promoted by the values of Henry’s law constants for H₂SO₄ and MSA)”* from the revised text.
- 18. P6, line 24 and P18.** We have moved Figure S3 to the main text.
- 19. P7, line 5. Be consistent with S isotope ratios of anthropogenic SO₄²⁻ (δ³⁴S_{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”*.

Response to Referee 2

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.

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 during Arctic phytoplankton bloom periods provided compelling evidence that there is a connection between DMS emissions and SO_4^{2-} 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 SO_4^{2-} aerosols" (lines 19–20, page 1; lines 28–29, page 8) and "possible association between DMS emissions and the formation of aerosol particles" (lines 4–5, page 3). We have also noted that confirmation of this proposed link requires measurements of inorganic, organic, and halogen species (lines 21–24, 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 SO_4^{2-} 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).
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 17–23, page 3).
- 6. Page 4, line 30:** We have replaced “*3-fold higher*” with “*more than double*”.
- 7. Page 5, lines 8–9. 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 16–20 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 21–24, page 5; line 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 $< 7.0 \text{ m}^{-2}$). 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 23–24, page 2 and lines 12–13, page 6).

12. P6, line 19: We have removed “*hypothesis*” from line 17, 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: Because 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 22, 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 PM_{2.5} 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 26, page 8 – line 2, 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.

Observational evidence for the formation of ~~ocean~~ DMS-derived aerosols during Arctic phytoplankton blooms

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Abstract. The connection between marine biogenic dimethyl sulfide (DMS) and the formation of aerosol particles in the Arctic atmosphere was evaluated by analyzing atmospheric DMS mixing ratio, aerosol particle size distribution and aerosol chemical composition data that were concurrently collected at Ny-Ålesund, Svalbard (78.5° N, 11.8° E) during April and May 2015. Measurements of aerosol sulfur (S) compounds showed distinct patterns during periods of Arctic haze (April) and phytoplankton blooms (May). Specifically, during the phytoplankton bloom period the contribution of DMS-derived SO_4^{2-} to the total aerosol SO_4^{2-} increased by 7-fold compared with that during the preceding Arctic haze period, and accounted for up to 70% of fine SO_4^{2-} particles ($< 2.5 \mu\text{m}$ in diameter). ~~The results also showed that a sharp increase in the atmospheric DMS mixing ratio during Arctic phytoplankton bloom events was directly associated with the formation of sub-micrometer SO_4^{2-} aerosols, and their subsequent growth to climate-relevant particles. The results also showed that the formation of submicron SO_4^{2-} aerosols was significantly associated with an increase in the atmospheric DMS mixing ratio.~~ More importantly, two independent estimates of the formation of DMS-derived SO_4^{2-} aerosols, calculated using the stable S isotope ratio and the non sea salt SO_4^{2-} /methanesulfonic acid ratio, respectively, were in close agreement, providing compelling evidence that the contribution of biogenic DMS to the formation of aerosol particles was substantial during the Arctic phytoplankton bloom period.

메모 포함[박1]: (response to referee #2)
Q1. We have removed 'ocean' from the title.

메모 포함[박2]: (response to referee #2)
Q2. **Overstatement of our results and conclusions:** We agree that our measurements did not provide direct evidence supporting a connection between DMS emissions and the formation and growth of aerosol particles. Therefore, we have revised the sentence.

1 Introduction

Aerosols are known to have influenced the Earth's radiation budget, by scattering and absorbing incoming solar radiation or forming cloud condensation nuclei (CCN) (Charlson et al., 1992; Haywood and Boucher, 2000). However, large uncertainty remains in assessing the effects of aerosols on radiative forcing (IPCC 2013). Both field and laboratory studies have indicated that sulfate (SO_4^{2-}) is an aerosol component that principally contributes to the formation of CCN (Kulmala, 2003; Boy et al., 2005; Sipilä et al., 2010). Most SO_4^{2-} particles originate from three sources: anthropogenic SO_x , sea salt SO_4^{2-} , and marine biogenic emissions (biogenic SO_4^{2-}). The latter is exclusively produced from the oxidation of dimethyl sulfide (DMS) (Simó, 2011; 2001). Over the past 3 decades, DMS emissions from the upper ocean have been considerably studied because there may be a **direct association close linkage** between marine biota and climate change (Charlson et al., 1987). DMS **is of marine origin**, is produced in the upper ocean via interactions of multiple biological processes (e.g., Stefels et al., 2007; Kettle and Andreae, 2000; Kim et al., 2010; Lee et al., 2012; Park et al., 2014), and some of the DMS **is eventually** emitted into the atmosphere through air-sea gas exchange processes **due to its low solubility in seawater**. Atmospheric DMS is **rapidly** oxidized to methanesulfonic acid (MSA) and H_2SO_4 , both of which influence the sulfurous aerosol budget (Davis et al., 1999; Barnes et al., 2006). **The MSA and H_2SO_4 formed from DMS tend to transform into new particles or condense onto existing aerosol particles because of their high values of Henry's law constant, and eventually become CCN (Charlson et al., 1987). The MSA and H_2SO_4 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, and eventually form CCN (e.g., Korhonen et al., 1999; Vehkamäki et al., 2002; Lee et al., 2003). Several studies have investigated the association of DMS with non sea salt sulfate (nss- SO_4^{2-}), CCN, and aerosol optical depth (AOD) (e.g. Ayers and Gras, 1991; Gabric et al., 2005a; Carslaw et al., 2010), but direct proofs for such associations is limited (Ayers and Cainey, 2007). Despite 30 years of research effort, the contribution of oceanic DMS emission to the formation of aerosol particles and CCN remains poorly quantified.** In the absence of this knowledge, in particular a lack of observational evidence for **a direct an** association between DMS production and the formation and growth of aerosol particles in the marine boundary layer, doubts remain as to the validity of the hypothesized feedback mechanism connecting DMS-derived aerosols to climate (Quinn and Bates, 2011). **Condensation of low-volatility vapors (generally involving H_2SO_4) in the absence of coagulation with larger particles results in the constant formation of new particles in the atmosphere, subsequently grow into Aitken particles (> 50–100 nm in diameter), which probably influence cloud formation, and thereby radiation (Boy et al., 2005; Pierce et al., 2014). The occurrence of low volatility vapors (generally involving H_2SO_4) in conjunction with the relative lack of coagulation and condensation sinks of pre-existing particles may result in the formation of new particles in the atmosphere (Boy et al., 2005). If precursor vapors are sufficient, these particles may grow into large particles (> 50 nm in diameter), which may influence cloud formation and thereby radiation (e.g. Pierce et al., 2014; Leaitch et al., 2016).** However, it is difficult to establish the quantitative relationship between oceanic DMS emission and the formation and growth of aerosol particles in the marine boundary layer. A small number of recent studies have reported that atmospheric DMS mixing ratios are related to the ocean phytoplankton biomass (Preunkert et al., 2008; Park et al., 2013), and that an increase in nss- SO_4^{2-} corresponds to a proportional increase in the MSA concentration in regions of high phytoplankton productivity (Becagli et al., 2012, 2013 and 2016; Zhang et al., 2015), where DMS emissions are also high. However, a mechanistic understanding of the major physical and chemical processes that are involved in the formation of DMS-derived SO_4^{2-} particles and their growth into larger particles remains elusive. Explicitly, DMS emissions may exert greater impacts on aerosol formation in regions where the concentration of background aerosol particles is low, but DMS-producing phytoplankton are abundant. The Arctic atmosphere is an excellent example of an environment that meets these two criteria (e.g., Chang et al., 2011; Browse et al., 2012; Leaitch et al., 2013; Tunved et al., 2013; Willis et al., 2016). The aims of the present study were to investigate **the direct association of DMS emissions with the formation of aerosol particles the possible association between DMS emissions and the formation of aerosol particles**, and to assess the contribution of DMS to total SO_4^{2-} aerosol budget. To this end we analyzed datasets of atmospheric DMS mixing ratio, aerosol particle size distributions, and aerosol chemical composition measured at Ny-Ålesund (Svalbard; 78.5° N, 11.8° E) in April and May 2015. To address the second aim we analyzed the MSA concentration (formed exclusively from the photo-oxidation of DMS) and the stable S isotope composition of aerosol particles.

메모 포함[박3]: (response to referee #1)
Q1. We have added omitted citations to the revised list of references.

메모 포함[박4]: (response to referee #2)
Q3. We have changed "direct association" to "close linkage".

메모 포함[박5]: (response to referee #2)
Q13. **Is there non-biogenic DMS?** More than 90% of DMS is derived from marine ecosystem. Other DMS sources are negligible.

메모 포함[박6]: (response to referee #2)
Q13. We have added this reference.

메모 포함[박7]: (response to referee #1)
Q2. As suggested, we have changed "DMS eventually emitted" to "DMS is emitted"

메모 포함[박8]: (response to referee #1)
Q3. As suggested, we have removed "rapidly".

메모 포함[박9]: (response to referee #1)
Q4. **Modify the description regarding Henry's constant and its association with particle nucleation:** Henry's constants are macroscopic representations of particle formation, while particle nucleation is more a molecular-level process. Therefore, we have revised the sentence.

메모 포함[박10]: (response to referee #2)
Q4. **The DMS-climate feedback may not as significant as other feedback processes:** We believe that the DMS-climate feedback mechanism is yet to be tested in the Arctic environment, where most rapid warming has occurred. In the revised manuscript we have more clearly elaborated the value of our study testing the DMS-climate feedback

메모 포함[박11]: (response to referee #2)
Q3. We have change "a direct association" to "an association".

메모 포함[박12]: (response to referee #1 and Co-editor)
Q5. We have modified the sentence.

메모 포함[박13]: (response to referee #1 and Co-editor)
Q6. We have added this reference.

메모 포함[박14]: (response to referee #1)
Q7. We have added these references.

메모 포함[박15]: (response to referee #2)
General concern: we have changed "direct association" to "possible association".

2 Experimental Methods

The atmospheric DMS mixing ratio was measured at 1–2 h intervals on the Zeppelin observatory, which is located at an elevation of 474 m above sea level (m.a.s.l) and 2 km south and southwest of Ny-Ålesund. The measurement period (April – May) approximately covered the pre- to post-phytoplankton bloom periods. The analytical system includes a component for DMS trapping and elution and a gas chromatography (GC) equipped with a pulsed flame photometric detector (PFPD) enabling DMS quantification. The detection limit of the analytical DMS system was reported to be 1.5 pptv in an air volume of approximately 6 L. Details for the DMS analytical system and the DMS measurement protocol are described in Jang et al (2016).

The distribution of aerosol particle sizes was measured at the Gruebadet observatory, which is approximately 1 km southwest of Ny-Ålesund and approximately 60 m.a.s.l. ~~Both a differential mobility analyzer and a condensation particle counter continuously measured the occurrence and distribution of two discrete small-particle size ranges (3–60 nm and 10–500 nm in differential mobility equivalent diameter).~~ Two discrete systems of scanning mobility particle sizer (SMPS) systems, each of which included a differential mobility analyzer (DMA) and a condensation particle counter (CPC), continuously measured the distribution of small particles in differential mobility equivalent diameter ranges 3–60 nm (combination of TSI 3085 and TSI 3776) and 10–500 nm (TSI 3034), and an aerodynamic particle sizer (APS) analyzed larger particles in the range 0.5–20 μm in diameter (Park et al., 2014; Lupi et al., 2016).

A high volume air sampler equipped with a PM_{2.5} impactor (collecting particles < 2.5 μm in aerodynamic equivalent diameter) was used for collection of aerosol samples. The sampler was mounted on the roof of the Gruebadet observatory, and sampled particles every 3 days between 9 April and 20 May 2015, and later measured concentrations of major ions and the stable S isotope composition on a quartz filter.

For measurement of major ions (Na⁺, K⁺, Mg²⁺, SO₄²⁻, Cl⁻, and MSA), a 47-mm (diameter) disk filter was punched out from a PM_{2.5} aerosol quartz filter. All major ions collected on the disk filter were extracted in 50 mL Milli-Q water and analyzed by a Dionex ion chromatography system (Thermo Fisher Scientific Inc., USA). The concentrations of major anions were determined using a Dionex model ICS-2000 with an IonPac AS 15 column and the concentrations of major cations were determined using a Dionex model ICS-2100 with an IonPac CS 12A column. Three times the standard deviations of blank measurements were used as detection limits (0.01 to 0.26 ng mL⁻¹) (Kang et al., 2015).

For measurement of the stable S isotope ratio (³⁴S/³²S), all S compounds on half of a PM_{2.5} quartz filter were extracted in 50 mL Milli-Q water. The filtrate was treated with 50–100 μL of 1M HCl to adjust the solution to pH = 3–4. Then, 100 μL of 1M BaCl₂ was added to cause all S-as SO₄²⁻ to precipitate as BaSO₄. After a 24-h precipitation period at room temperature, the BaSO₄ precipitate was recovered by filtration on a membrane filter and finally dried for 24 h. Each membrane filter containing BaSO₄ was packed into a tin cup and analyzed by isotope ratio mass spectrometer (IsoPrime100, IsoPrime Ltd, UK) coupled to an elemental analyzer (Vario EL, Elementar Co, German). The resulting S isotope ratio of a sample ($\delta^{34}\text{S}$) was expressed as parts per thousand (‰) relative to the ³⁴S/³²S ratio in a standard (Vienna Cañon Diablo Troilite) (Krouse and Grinenko, 1991).

$$\delta^{34}\text{S} (\text{‰}) = \left\{ \left(\frac{{}^{34}\text{S}/{}^{32}\text{S}}{\text{sample}} / \left(\frac{{}^{34}\text{S}/{}^{32}\text{S}}{\text{standard}} - 1 \right) \right) \times 1000 \right. \\ (1)$$

Information about the S isotope ratio of aerosol particles and the concentrations of major ions enabled the estimation of the relative contributions of biogenic DMS (f_{bio}), anthropogenic SO_x (f_{anth}), and sea-salt SO₄²⁻ (f_{ss}) to the total aerosol SO₄²⁻ concentration. The concentration of ss-SO₄²⁻ was first calculated by multiplying the Na⁺ concentration (as a sea spray marker) by 0.252 (the seawater ratio of SO₄²⁻/Na⁺) (Keene et al., 1986). The nss-SO₄²⁻ fraction of the total SO₄²⁻ was then calculated by subtracting the fraction of ss-SO₄²⁻ from the total SO₄²⁻. Finally, the fraction of biogenic SO₄²⁻ was estimated by solving the following equations:

$$\delta^{34}\text{S}_{\text{measured}} = \delta^{34}\text{S}_{\text{bio}} f_{\text{bio}} + \delta^{34}\text{S}_{\text{anth}} f_{\text{anth}} + \delta^{34}\text{S}_{\text{ss}} f_{\text{ss}} \\ (2)$$

$$f_{\text{bio}} + f_{\text{anth}} + f_{\text{ss}} = 1 \\ (3)$$

$$f_{\text{ss}} = 0.252 \text{ Na}^+ / \text{total SO}_4^{2-} \\ (4)$$

In solving Eq. (2)–(4), the published S isotope end-member values of DMS-derived SO₄²⁻ ($\delta^{34}\text{S}_{\text{bio}} = 18 \pm 2\text{‰}$), anthropogenic SO₄²⁻ ($\delta^{34}\text{S}_{\text{anth}} = 5 \pm 1\text{‰}$), and sea-salt SO₄²⁻ ($\delta^{34}\text{S}_{\text{ss}} = 21.0 \pm 0.1\text{‰}$) were used (McArdle and Liss, 1995; Norman et al., 1999; Böttcher et al., 2007; Lin et al., 2012).

메모 포함[박16]: (response to referee #1 and #2)
Q8. Provide more information about the use of DMA-CPC: Two discrete SMPS systems were used to measure the distribution of aerosol particle sizes. We have added more detailed information and references.

메모 포함[박17]: (response to referee #1)
Q9. we have included a measure of uncertainty associated with the $\delta^{34}\text{S}_{\text{ss}}$ value ($21.0 \pm 0.1\text{‰}$) for sea salts.

메모 포함[박18]: (response to referee #1)
Q9. We have added this reference.

3 Results and Discussion

3.1 Atmospheric DMS and aerosol particles

The atmospheric DMS mixing ratio measured at Zeppelin observatory changed abruptly (by several orders of magnitude) within a few days of measurement and occasionally reached a level of 400 pptv, particularly during phytoplankton bloom events (Fig. 1a). The monthly mean DMS mixing ratio for May (47 ± 91 pptv) was ~~3-fold higher more than double~~ than the April mean (18 ± 18 pptv). The 3-day integrated concentrations of MSA were broadly consistent with the concentrations of DMS; the lowest concentration (< 50 ng m⁻³) occurred in April and the highest value (approximately 200 ng m⁻³) occurred in May (Fig. 1a). The strong positive correlation between the MSA concentrations and the corresponding DMS mixing ratios ($r = 0.84$, $n = 14$, $P < 0.05$; Fig. 1c) supports the assumption that the photochemical oxidation of biogenic DMS was the ~~only~~ major source of MSA in our study area. ~~Variations in DMS explained approximately 70% of the observed variance in the MSA concentration; the remaining variance was probably associated with variations in MSA that formed elsewhere, and was subsequently advected to the measurement site, and also with variations in the efficiency of photochemical oxidation of DMS.~~

The concentration of aerosol particles in the 3–10 nm diameter range (a nucleation mode), which is an indicator of recent nucleation, occasionally exceeded 3000 cm⁻³. These small particles formed more frequently in May than in April (blue line in Fig. 1a). ~~As a result, the observed increase in nucleation mode particles coincided with high atmospheric DMS mixing ratio and MSA concentration. Therefore, the 3-day mean DMS mixing ratios and the MSA concentrations were both significantly correlated with the 3-day mean concentrations of nucleation mode particles ($r = 0.66$, $n = 14$, $P < 0.05$, Fig. 1e 1d; and $r = 0.71$, $n = 14$, $P < 0.05$, Fig. 1d 1e, respectively); the observed nucleation events also concurrently occurred with high atmospheric DMS mixing ratios (Fig. 1a).~~ Approximately 45% of the variability in the 3-day mean concentrations of nucleation mode particles can be explained by overall variations in the concentrations of DMS and MSA; some of the remaining variability will be associated with variations in the intensity of solar radiation, which influences the efficiency of photochemical oxidation of DMS. In fact, high atmospheric DMS mixing ratios found in mid-April (77.1 ± 51.5 pptv; 14–17 April) was not followed by the formation of nucleation mode particles (42.6 ± 49.5 cm⁻³) and MSA (21.4 ng m⁻³), ~~probably possibly~~ due to the low intensity of solar irradiation (80.4 ± 81.9 W m⁻²) (Fig. 1a and e, c and d). We cannot completely rule out the possibility that sources other than DMS (e.g., iodine) contributed to the formation of nucleation mode particles (Fig. 1a, e and d and e). ~~The emission of iodine is an alternative explanation for the particle nucleation event in the Arctic atmosphere during our study period (O'Dowd et al., 2002; Allan et al., 2015). Recent field observations in an iodine-rich coastal environment have shown that species containing iodine contribute to the formation of new aerosol particles via direct molecular-scale observations of nucleation in an iodine-rich coastal environment (Sipilä et al., 2016). As all chemical species (including H₂SO₄, iodine species, and organic vapors) that are directly involved in the nucleation process were not measured during the observational periods of the present study, we are unable to pinpoint the major contributor; however, these strong correlations indicate that the small aerosol particles that were formed newly were probably derived from recently released biogenic DMS.~~

3.2 Aerosol particles formed during periods of Arctic haze (April) and phytoplankton blooms (May)

Arctic haze, formed originally from emissions of pollution in North Europe, Siberia and North America, and its transport to the Arctic environment, has been reported to influence the aerosol characteristics of the Arctic atmosphere during early spring (April and earlier periods). The Arctic haze is a mixture of SO₄²⁻ and particulate organic matter, plus minor contributions of ammonium, nitrate, dust, black carbon, and heavy metals (Quinn et al., 2007). The concentrations of nss-SO₄²⁻ during this period reached 2000 ng m⁻³, and the mean level in April was 2-fold greater than that in May (Fig. 2a). However, information about the concentrations of nss-SO₄²⁻ only did not enable differentiation of the strengths of two major sources (anthropogenic SO_x vs. biogenic DMS). Additional measurements of particle concentrations enabled quantification of the contributions of anthropogenic SO_x and biogenic DMS to the total nss-SO₄²⁻.

~~Anthropogenic SO_x from the continents that reaches Ny Ålesund will have undergone alterations during long-range transport. For example, hydrophilic H₂SO₄ particles derived from anthropogenic SO_x tend to grow into particles larger than 100 nm (accumulation mode), especially following partial or total neutralization by NH₃, which in turn yields hygroscopic compounds such as NH₄HSO₄ and (NH₄)₂SO₄. In contrast, the photochemical oxidation of DMS (yielding biogenic SO₄²⁻) occurs locally and the resulting biogenic particles more tend to develop into small particles (nucleation mode, 3–10 nm; and Aitken mode, 10–100 nm). These established explanations are generally consistent with our observations. The transition of aerosol microphysical properties from a distribution dominated by an accumulation mode (Arctic haze period) to a distribution dominated by~~

메모 포함[박19]: (response to referee #2)
Q6. As suggested, we have changed “3-fold higher” to “more than double”.

메모 포함[박20]: (response to referee #1)
Q10. Provide more description about the relation between atmospheric DMS mixing ratios and MSA concentration: Local DMS production explained much of the variation in MSA, but did not account for all MSA variations. Therefore, we have added a short paragraph noting that long range transport of MSA to the site was a possibility.

메모 포함[박21]: (response to referee #2)
Q7. Our description of Figure 1 was misleading. In the revised manuscript we have newly added this sentence.

메모 포함[박22]: (response to referee #2)
Q7. Our description of Figure 1 was misleading. In the revised manuscript we have removed this sentence.

메모 포함[박23]: (response to referee #1)
Q10. We have added “and MSA (21.4 ng m⁻³)”.

메모 포함[박24]: (response to referee #1)
Q12. We have replaced “probably” to “possibly”.

메모 포함[박25]: (response to referee #1)
Q13. We have cited O'Dowd et al. (2002) and Allan et al. (2015), and added a brief explanation.

메모 포함[박26]: (response to referee #2)
Q8. 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. Iodine may have contributed to explaining the remaining variance. We have added statement that iodine could be an important contributor.

메모 포함[박27]: (response to referee #2)
Q9. This study did not provide direct evidence: 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.

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). Specifically, the large accumulation mode particles outnumbered the small nucleation and Aitken mode particles during early spring (April) but the concentration of those large particles decreased rapidly from April to May, with particles smaller 100 nm becoming dominant in May (Fig. 3 and S1). ~~The high concentration of small particles (<100 nm) during phytoplankton bloom period (May) constitutes compelling evidence for new particle formation derived from local DMS emission (Sharma et al., 2012; Tunved et al., 2013).~~ A similar sharp transition (large-to-small particles) in the dominant particle type was also identified in previous observations at the same site (Engvall et al., 2008). Our data on the particle size distributions showed that particles > 100 nm were more abundant in April, whereas small particles (< 100 nm) were more abundant in May (Fig. 3 and S1). As a result, the total surface area of aerosol particles in April was 2-fold greater than that observed in May, whereas the concentration of particles in April was 3-fold less than that in May (Fig. 3). ~~As the condensation sink is proportional to the surface area of aerosol particles, it will decrease with decreasing intensity of Arctic haze.~~ The concentrations of nss-SO₄²⁻ measured in April did not correlate with the levels of biogenic MSA (P > 0.05; blue circles in Fig. 2b). On the contrary, a strong correlation (r = 0.75, n = 7, P < 0.05; red circles in Fig. 2b) between these two parameters was found in May. The greater DMS contribution to the formation of nss-SO₄²⁻ in May than in April is broadly consistent with the 2-fold greater chlorophyll concentration observed in May compared to April (Fig. S2). These observations support ~~our hypothesis~~ that the formation of new particles resulting from the photo-oxidation of biogenic DMS, followed by a gas-to-particle conversion ~~(promoted by the values of Henry's law constants for H₂SO₄ and MSA)~~ is an important source of Aitken mode (10–100 nm) particles in the Arctic atmosphere. ~~Recent field observation also provided the evidence that the growth of nucleation mode particles in the summertime Arctic atmosphere can be mediated by the presence of secondary marine organic aerosol including MSA (Willis et al., 2016).~~

3.3 Aerosol particles formed from biogenic DMS SO₄²⁻ aerosol particles formed from biogenic DMS

The use of an asymptotic value in a plot of nss-SO₄²⁻/MSA ratio versus MSA concentration is a convenient method for estimating the fraction of biogenic SO₄²⁻ aerosols. As the MSA concentration in an aerosol sample increases, the contribution of biogenic SO₄²⁻ to the total nss-SO₄²⁻ will also increase while the contributions of other sources will decrease. In this case, the nss-SO₄²⁻/MSA ratio tends to approach an asymptotic value as the MSA concentration increases. Therefore, this asymptotic value adequately represents the nss-SO₄²⁻/MSA ratio derived exclusively from DMS (Udisti et al., 2012, 2016). The biogenic SO₄²⁻/MSA ratio has been reported to vary considerably in space and time (Gondwe et al., 2004), because the ratio is sensitive to temperature, and to a lesser extent photochemical species or reactions with halogen radicals (Bates et al., 1992). When this method was applied to data for aerosol samples (PM₁₀) collected in 2014 at the Gruebadet observatory, in a vicinity of our DMS measurement site, the biogenic SO₄²⁻/MSA ratio was estimated to be 3.0 (Udisti et al., 2016) (Fig. S3 Fig. 4). In other polar locations, a ratio of 2.6 was reported, including for Alert station (82.5°N, 62.3°W; 210 m.a.s.l.) (Norman et al., 1999) and Concordia station (75.1°S, 123.3°E; 3233 m.a.s.l.) (Udisti et al., 2012). We estimated the amount of biogenic SO₄²⁻ by multiplying the biogenic SO₄²⁻/MSA ratio (3.0) by the MSA concentration in each aerosol sample. The fraction of anthropogenic SO₄²⁻ was estimated by subtracting the combined ss-SO₄²⁻ plus biogenic SO₄²⁻ concentration from the total SO₄²⁻ concentration. Another method for estimating biogenic SO₄²⁻ is to use S isotope ratios (δ³⁴S) of SO₄²⁻ aerosols, because the δ³⁴S values of biogenic DMS (18 ± 2‰) are greater than those of anthropogenic SO₄²⁻ ~~(0–8‰)~~ (5 ± 1‰) but less than that of sea salt (21 ± 0.1‰) (e.g., Wadleigh, 2004; Lin et al., 2011; Oduro et al., 2012). A wide range in δ³⁴S (0–8‰) has been reported for anthropogenic SO₂ compared with values reported for other sources (Krouse and Grinenko, 1991). Surprisingly, Patris et al (2000) reported consistent regional-scale δ³⁴S values for anthropogenic SO₂. For example, in remote Arctic regions (including Ny-Ålesund and Alert) the S isotope ratios measured for SO₄²⁻ aerosols during the Arctic haze period in a single year mostly fell within the narrow range of 5–6‰ (McArdle and Liss, 1995; Norman et al., 1999), probably because regional-scale mixing processes averaged the signals (Partis et al., 2000). During the study period the δ³⁴S values measured at Ny-Ålesund ranged from 4.6 to 10.3. The δ³⁴S values were higher in May (8.8–10.3‰) than in April (4.6–8.2‰), reflecting changes in S sources.

The contributions of anthropogenic and biogenic SO₄²⁻ to the total SO₄²⁻ aerosols, estimated using two independent methods, are shown in Fig. 4 Fig 5 (and Fig. S4 S3). In April and May the contribution of ss-SO₄²⁻ to total SO₄²⁻ was small (< 3% of total aerosol particles < 2.5 μm in diameter). It was estimated that approximately 90% of the total SO₄²⁻ was of anthropogenic origin in April, when the Arctic haze was most intense. This estimation is consistent with measurements of anthropogenic SO₄²⁻ in PM₁₀ aerosols collected in April 2014 at the same site (Udisti et al., 2016). In May, following the period of Arctic haze, the contribution of

메모 포함[박28]: (response to referee #1)

Q14. 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.

메모 포함[박29]: (response to referee #1)

Q14 and 15. We have removed this sentence and references.

메모 포함[박30]: (response to referee #2)

Q11. 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

메모 포함[박31]: (response to referee #2)

Q12. We have removed "hypothesis"

메모 포함[박32]: (response to referee #1)

Q17. We have removed "(promoted by the values of Henry's law constants for H₂SO₄ and MSA)" from the revised text.

메모 포함[박33]: (response to referee #1)

Q16. We have added a brief overview of recent field observations reported in Willis et al. 2016, which is conceptually similar to our work.

메모 포함[박34]: The title of section 3.3 is

inadequate: We have changed "Aerosol particles formed from biogenic DMS" to "SO₄²⁻ aerosol particles formed from biogenic DMS"

메모 포함[박35]: (response to referee #1)

Q18. We have moved Figure S3 to the main text.

메모 포함[박36]: (response to referee #1)

Q19. Be consistent with S isotope ratios of anthropogenic SO₄²⁻ (δ³⁴S_{anth}): We have replaced "(0–8‰) to (5 ± 1‰)".

DMS-derived SO_4^{2-} sharply increased up to 70% of fine SO_4^{2-} particles ($< 2.5 \mu\text{m}$ in diameter), and this corresponded to the Arctic phytoplankton bloom (Fig. 4a and S4 5a and S3). The concentrations of DMS-derived SO_4^{2-} estimated using the MSA-based approach were approximately 50% higher than the values based on the S-isotope method. The differences may be the result of uncertainties associated with uncertainties in assigned S isotope end-member values, and the ratio of biogenic SO_4^{2-} to MSA. Nonetheless, a robust correlation between these two estimates was found during April and May ($r = 0.74$, $n = 13$, $P < 0.05$; Fig. 4b 5b).

3.4 Relationship between concentrations of DMS-derived SO_4^{2-} and the concentrations of aerosol particles

In both April and May, the concentrations of biogenic SO_4^{2-} particles estimated using the MSA-based and S-isotope-based methods were significantly correlated with the concentration of small aerosol particles in nucleation (from 3–10 nm, $r = 0.71$, $n = 14$, $P < 0.05$; Fig. 5a 6a) and Aitken modes (from 10–100 nm, $r = 0.89$, $n = 14$, $P < 0.05$; Fig. 5b 6b). However, when the Arctic haze prevailed in April, the concentration of large particles in accumulation mode (from 100 nm–1 μm , $P > 0.05$; Fig. 5e 6c) was not significantly correlated with the concentration of biogenic SO_4^{2-} , but was strongly correlated with the concentration of anthropogenic SO_4^{2-} ($r = 0.92$, $n = 7$, $P < 0.05$; the inset in Fig. 5e 6c). In contrast, when Arctic haze moderated in May and the abundance of phytoplankton began to increase, the concentration of biogenic SO_4^{2-} was strongly correlated with the concentration of accumulation mode particles ($r = 0.91$, $n = 7$, $P < 0.05$; Fig. 5e 6c). A strong correlation between biogenic SO_4^{2-} particles and the surface areas of particles provided additional evidence that biogenic SO_4^{2-} significantly contributed to small particle formation (Fig. S5 S4). Moreover, the formation and growth of sub-micrometer particles with diameters between 3 and 100 nm coincided with high concentrations of biogenic SO_4^{2-} (Fig. 6 7 and S6 S5). It is noteworthy that the contribution of anthropogenic SO_4^{2-} to fine SO_4^{2-} particles ($< 2.5 \mu\text{m}$ in diameter) was still considerable (30–60%) during the bloom period (May) (Fig. 5a and S3). Therefore, in May anthropogenic SO_4^{2-} may also have partly contributed to the formation and growth of small aerosol particles ($< 100 \text{ nm}$). However, we could not accurately estimate the exact contributions of biogenic versus anthropogenic SO_4^{2-} to the formation and growth of aerosol particles in the absence of information on the chemical composition of size-segregated aerosol particles. The chemical composition of size-segregated aerosol particles was recently measured in the Arctic atmosphere during summer months. More than 60% of the aerosol particles having a diameter $< 0.49 \mu\text{m}$ was found to be derived from biogenic SO_4^{2-} (Ghahremaninezhad et al., 2016). According to a study based on an aerosol microphysics box model (Chang et al., 2011), the atmospheric DMS mixing ratios observed during phytoplankton bloom periods in our study were sufficiently high for the formation of ultrafine aerosol particles, when background particle concentrations are low (i.e. DMS mixing ratio $> 100 \text{ pptv}$; condensation sink $< 7.0 \text{ m}^{-2}$) (Fig. 1a and b).

These direct observations provide compelling evidence that biogenic DMS released from the Arctic Ocean contributed significantly to the formation of new aerosol particles, and their subsequent growth to larger climate-relevant particles ($> 50\text{--}100 \text{ nm}$ in diameter). In particular, new particles appeared to be formed more rapidly in May, and grew more efficiently to large climate-relevant particles, possibly because of the presence of a large source of condensable vapor (generally involving H_2SO_4) formed from the oxidation product of DMS and the low concentration of background aerosol particles ($> 100 \text{ nm}$). During phytoplankton bloom events, the Arctic environment surrounding the DMS observation site was found to have three key characteristics (small surface area of particles, high concentrations of low volatility condensable vapors, and high solar radiance) that can greatly facilitate the formation of new particles from biogenic DMS.

4 Conclusion

The Arctic Ocean is warming faster than any other ocean region (IPCC 2013). Consequently, a corresponding decrease in the sea ice extent may impact on primary production and the sea-to-air flux of climate-relevant gases including CO_2 and DMS (Gabric et al., 2005b). The loss of Arctic sea ice may also result in a new source of aerosol particles and CCN from DMS, possibly counterbalancing the decrease in surface albedo by an increase in cloud albedo (Levasseur 2013; Woodhouse et al., 2013).

This study provided the observational evidence confirming indicating direct relationships between an increase in atmospheric DMS and the formation and growth of aerosol particles, and also an increase in the total mass concentration of nss-SO_4^{2-} during Arctic phytoplankton blooms. Concurrent measurements of a suite of parameters (DMS, satellite-derived phytoplankton biomass, concentration and chemical composition of particles) supported the assertion that oceanic emission of DMS significantly affects the properties of sub-micrometer particles in the Arctic atmosphere. This study demonstrated the close association between an increase in DMS and increases in the total mass concentration of nss-SO_4^{2-} and MSA during the period of Arctic

메모 포함[박37]: (response to referee #1)

Q20. Need to acknowledge potential contribution of anthropogenic SO_4^{2-} to NPF events during the phytoplankton bloom period: Unfortunately, we could not accurately estimate the relative contribution of biogenic versus anthropogenic SO_4^{2-} to the formation and growth of aerosol particles. However, recent field observations indicate that a considerable amount of SO_4^{2-} in aerosol particles having a diameter $< 0.49 \mu\text{m}$ 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, and cited Ghahremaninezhad et al. (2016) in support of our argument.

메모 포함[박38]: (response to referee #2)

Q10. 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 $> 100 \text{ pptv}$ are sufficient to account for the formation of ultrafine particles, particularly when background particle concentrations are low (condensation sink $< 7.0 \text{ m}^{-2}$). To support the confidence of our statement, we have added a short paragraph describing the results of Chang et al. (2011) and have added this paper to the revised list of references.

메모 포함[박39]: (response to referee #2)

We have removed “compelling”.

메모 포함[박40]: (response to referee #2)

We have removed “significantly”.

phytoplankton blooms. It also confirmed that the increase in DMS-derived SO_4^{2-} occurred concurrently with the formation and growth of aerosol particles. Measurements of the atmospheric DMS mixing ratio, MSA, S isotope ratio, aerosol particle size distribution, and satellite-based biomass indicated that there was a connection between oceanic DMS emissions and the formation of aerosol particles in the Arctic atmosphere during the phytoplankton bloom period. Further measurements of the chemical composition of marine aerosols less than 100 nm in size are needed to provide more direct evidence for the contribution of key nucleating compounds (i.e., DMS, iodine-containing species, and organic vapors) to the formation of aerosol particles, and to investigate how aerosol particles in the Arctic are responding to climate change.

Author contribution

K.-T.P., K.L., Y.J.Y and B.-Y.L designed the study. K.-T.P and S.J carried out the field campaign for DMS and aerosol measurements. M.-S.K, K.-H.S analyzed S-isotope. K.P, H.-J.C and R.U analyzed aerosol particle size distribution. J.-H.K analyzed major ions including MSA. K.-T.P., K.L and Y.J.Y wrote the manuscript.

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메모 포함[박41]: (response to referee #2)

Q17. Our results were overly stated in Conclusion:

We have clearly stated in the revised Conclusion 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 SO_4^{2-} aerosol formation in the Arctic atmosphere connection.

메모 포함[박42]: (response to referee #1)

We have added this reference.

메모 포함[박43]: (response to referee #2)

We have added this reference.

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We have added this reference.

메모 포함[박45]: (response to referee #1)
We have added this reference.

메모 포함[박46]: (response to referee #2)
We have added this reference.

메모 포함[박47]: (response to referee #1 and #2)
We have added this reference.

메모 포함[박48]: (response to referee #2)
We have added this reference.

메모 포함[박49]: (response to referee #1)
We have added this reference.

메모 포함[박50]: (response to referee #2)
We have added this reference.

메모 포함[박51]: (response to referee #2)
We have added this reference.

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We have added this reference.

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We have added this reference.

메모 포함[박55]: (response to referee #1)
We have added this reference.

메모 포함[박56]: (response to referee #1)
We have added this reference.

메모 포함[박57]: (response to referee #1)
We have added this reference.

메모 포함[박58]: (response to referee #1)
We have added this reference.

메모 포함[박59]: (response to referee #1)
We have added this reference.

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메모 포함[박60]: (response to referee #1)
We have removed this reference.

메모 포함[박61]: (response to referee #2)
We have added this reference.

메모 포함[박62]: (response to referee #1)
We have added this reference.

메모 포함[박63]: (response to referee #1)
We have added this reference.

메모 포함[박64]: (response to referee #1)
We have added this reference.

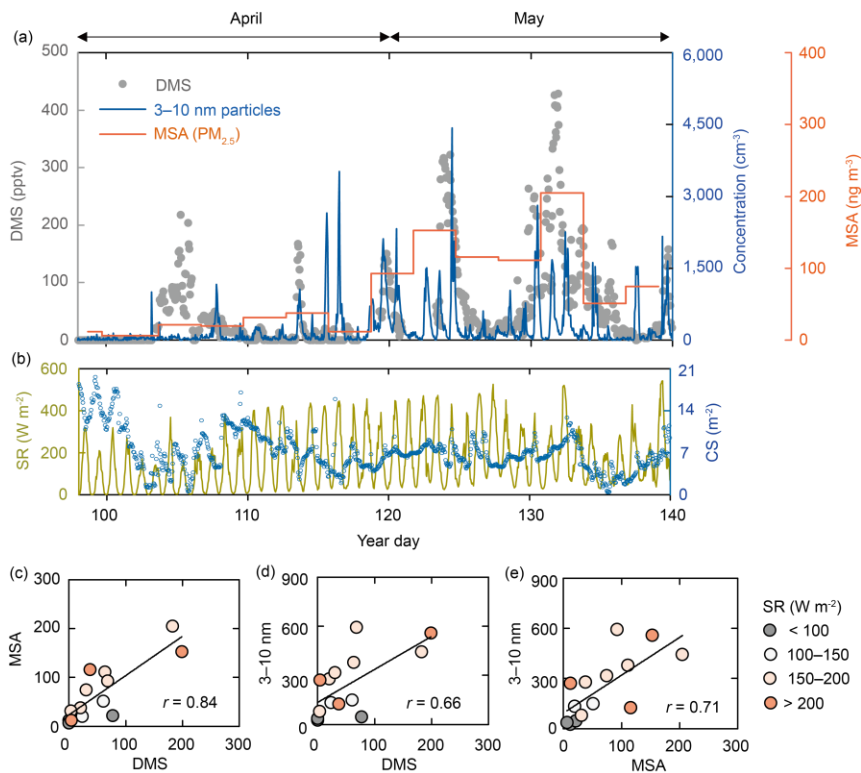


Figure 1: (a) The mixing ratios of atmospheric DMS measured at the Zeppelin observatory (gray circles), the concentration of nucleation mode particles (3–10 nm in diameter) measured at the Gruevbadet observatory (1 km distant from Zeppelin station) (blue line) and the concentration of MSA collected at the observatory (red line) during April and May 2015. (b) The solar irradiance (SR) measured in Ny-Ålesund (yellow line), and the condensation sink (CS) calculated by Kerminen et al. (2004) (blue circles). (c) The relationship between the atmospheric DMS mixing ratio, averaged for 3 days, and the corresponding MSA concentration. (d) The relationship between the atmospheric DMS mixing ratio, averaged for 3 days, and the corresponding concentration of nucleation mode particles. (e) The relationship between the MSA concentration and the corresponding concentration of nucleation mode particles. The colors of the circles in Fig. 1b–d 1c–1e indicate the intensity of solar irradiance (SR; integrated over a wavelength range of 200–3600 nm) measured in Ny-Ålesund.

메모 포함[박65]: (response to referee #1)
 Q11. As suggested, we have added the time series for solar irradiance to Figure 1b.

메모 포함[박66]: (response to referee #2)
 Q10. We have added the calculated condensation sink.

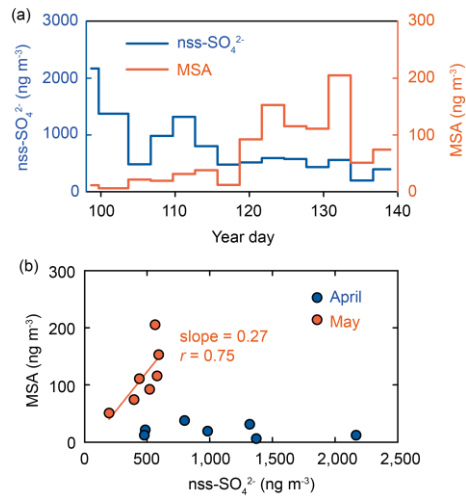


Figure 2: (a) The concentrations of nss-SO_4^{2-} (blue line), and MSA (red line) at the Gruvebadet observatory in April and May 2015. (b) The relationship between nss-SO_4^{2-} and MSA measured in April (blue circles; Arctic haze period) and in May (red circles; phytoplankton bloom period). The red solid line shown in Fig. 2b indicates the best fit.

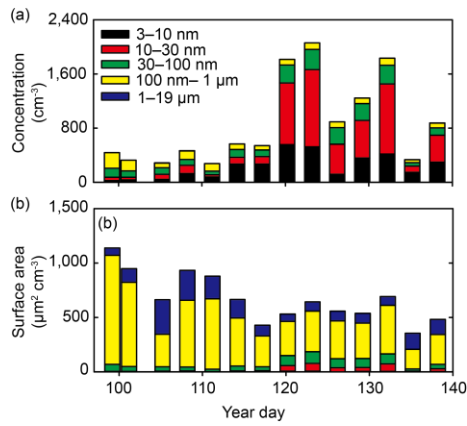


Figure 3: (a) Concentration, and (b) surface area of aerosol particles, including nucleation mode (3–10 nm), Aitken mode (10–100 nm), accumulation mode (100 nm–1 μm) and coarse mode (1–19 μm) particles in April and May 2015.

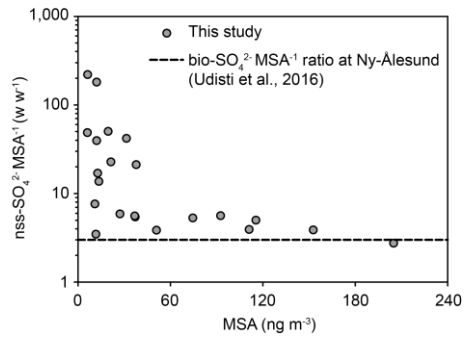


Figure 4: Relation between the nss-SO₄²⁻/MSA¹ ratio and the MSA concentration (PM_{2.5}; April and May 2015; grey circles). The dashed line indicates the bio-SO₄²⁻/MSA¹ ratio measured at Ny-Ålesund in 2014 (Udisti et al., 2016).

메모 포함[박67]: (response to referee #1)
Q18. We have moved Figure S3 to the main text.

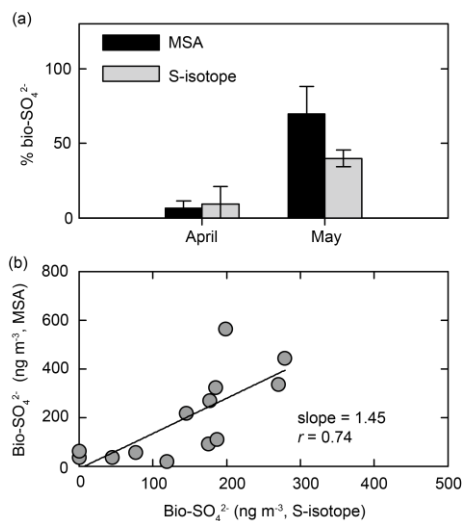


Figure 4.5: Biogenic (bio) SO_4^{2-} estimated using MSA and S isotope. (a) Biogenic SO_4^{2-} as a percentage of the total aerosol SO_4^{2-} burden. Black bars: biogenic SO_4^{2-} estimated using MSA; grey bars: biogenic SO_4^{2-} estimated using stable S isotope. (b) The relationship between the concentrations of biogenic SO_4^{2-} estimated using MSA, and the concentrations of biogenic SO_4^{2-} estimated using stable S isotope. The black solid line represents the best fit.

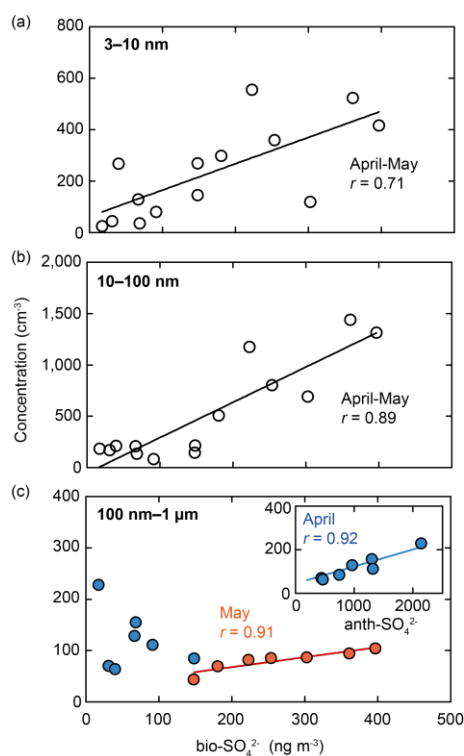


Figure 5.6: Relationships between the concentration of biogenic (bio) SO_4^{2-} and the concentrations of particles of various sizes in April and May. (a) Particles 3–10 nm, (b) particles 10–100 nm, and (c) particles 100 nm–1 μm . The black solid lines in Fig. 5a and 5b 6a and 6b indicate the best fit between biogenic SO_4^{2-} and the particle concentration during April and May 2015. Blue and red circles in Fig. 5e 6c indicate data obtained in April (Arctic haze period) and May (phytoplankton bloom period), respectively. The red solid line in Fig. 5e 6c indicates the best fit between biogenic SO_4^{2-} and particle concentration in May 2015, and the blue solid line in the inset of Fig. 5e 6c indicates the best fit between anthropogenic (anth) SO_4^{2-} and particle concentration in April 2015.

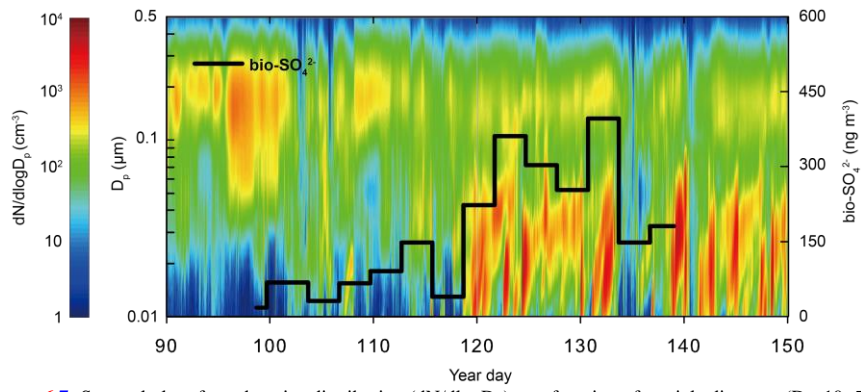


Figure 6 7: Spectral plot of number size distribution ($dN/d\log D_p$) as a function of particle diameter (D_p , 10–500 nm) and year day during April and May 2015. The black line represents the concentration of biogenic SO_4^{2-} .