Response to Referee 1

We thank Referee 1 for providing insightful suggestions that have considerably improved the readability of the revised manuscript. Our responses to general and specific comments raised by Referee 1 are stated below.

General comments

The link between phytoplankton taxonomic composition and NPF is not convincingly presented and the scientific approach is not clearly outlined: We have added a description and figures to support our key findings in the revised manuscript. In short, the explanation regarding the biological characteristic of the surrounding ocean has been thoroughly complemented in the revised manuscript. Figures for 8-year CN records ($CN_{2.5}$, CN_{10}, and $CN_{2.5-10}$), 8-year transport history at an hourly interval, sea-surface DMSP concentration, and daily MSA concentration have been newly added. We have addressed these issues in detail in our response to specific comments.

Specific Comments

1. **The discussion presented in Chapters 3 is not conclusive, in particular, the description regarding DMSP-to-chlorophyll ratio is confusing (Issue 1). What is the impact of SST, MLD and PIC compared to Chl (Issue 2)? How representative is the PHYSAT database for the period 1997–2010 with regard to your observation period? (Issue 3):**

   (Response to Issue 1) We have thoroughly revised the "Results and Discussion (chapter 3)" part to improve the readability of our manuscript. First, we have added a paragraph explaining the DMSP-relevant processes and their association with marine biota to clarify the meaning of the DMSP-to-chlorophyll ratio (P4, line 34 – P5, line 6; P7, line 16 – P8, line 1; P8, lines 7–10). In short, the conversion of cellular DMSP into DMS is controlled by not only the concentration of DMSP but also, more importantly, the DMSP cleavage enzyme. The phytoplankton species containing high cellular DMSP (i.e., high DMSP-to-chlorophyll ratio) mostly possess an enzyme that can convert cellular DMSP into DMS, whereas phytoplankton species containing low DMSP content (i.e., low DMSP-to-chlorophyll ratio) do not have a DMSP cleavage enzyme. Therefore, the DMSP-to-chlorophyll ratio is commonly used to explain the differences in taxonomic compositions affecting the oceanic DMS-production capacity (e.g., Belviso et al., 2000; Stefels et al., 2007; Tison et al., 2010; Park et al., 2014b and 2018). Explanation regarding the PHYSAT analysis and MSA has also been modified in the revised manuscript (see authors' response 1-3 and 5 for more details).

   (Response to Issue 2) DMSP is produced by marine phytoplankton; however, the dependence of the production of planktonic DMSP on phytoplankton biomass is not straightforward owing to the strong variabilities across taxonomic groups and the interplay with environmental factors. *Gali* et al. (2015) developed a DMSP algorithm based on satellite-derived chlorophyll (to measure phytoplankton biomass) and the light exposure regime (to measure key environmental factors controlling DMSP production). The terms SST and MLD have been used to validate the environmental factors controlling DMSP production. Specifically, euphotic layer depth (Z_{eu}) and mixed layer depth (MLD) dataset were applied to establish a mixing state of the sea surface (stratified vs. mixed water column), and the variability in modeled and measured DMSP was improved by adding sea-surface temperature and $\log_{10}(Z_{eu}/MLD)$ as predictors for the stratified and mixed subsets in the proposed algorithm. Additionally, a sub-model based on particulate inorganic carbon (PIC) was developed to complement DMSP diagnosis in coccolithophore blooms, where satellite chlorophyll concentration may not be reliable. We have added these sentences in the revised supplementary (P2, lines 2–11).

   (Response to Issue 3) The PHYSAT method was developed based on the SeaWiFS dataset, which is available from 1997 to 2010, and is the most widely used algorithm for the estimation of the taxonomic composition of marine phytoplankton. The PHYSAT analysis calculated using the SeaWiFS climatology map was successfully applied to the Southern Ocean and could represent the general seasonal trend of the taxonomic composition of marine phytoplankton in the study area (Alvain and d'Ovidio, 2014). Recently, a regional PHYSAT algorithm for the Mediterranean Sea was developed by applying linear interpolation between SeaWiFS and MODIS wavelengths and reflectance threshold and is available at a global scale (Navarro et al., 2014). However, challenges remain in high-latitude areas such as the Southern Ocean, especially because of the rather sparse matchup available for the calibration and validation of the PHYSAT algorithm (Alvain et al., 2014). We have calculated the taxonomic composition of marine phytoplankton by using the MODIS-based PHYSAT algorithm (see the figure below). In this study area, the MODIS-based PHYSAT algorithm overestimated the dominance of

diatoms compared with the SeaWiFS-based PHYSAT algorithm, whereas the dominance of phaeocystis was underestimated. Nevertheless, the PHYSAT results estimated using both the SeaWiFS archive (from January 1997 to December 2010) and MODIS archive (from January 2002 to December 2016) show more than three times higher dominance of the phaeocystis group in the Bellingshausen Sea than in the Weddell Sea during the summer period. In the revised manuscript, we have changed *"...obtained PHYSAT database, and was estimated between 1997 and 2010"* to *"...obtained from the PHYSAT database estimated using climatology over the SeaWiFS period (1997–2010)"* (P5, lines 18–19). We also have changed *"which was applied from 1997 to 2010"* to *"which was estimated using the SeaWiFS climatology map"* (P6, line 35) for better clarity. An additional paragraph explaining the feasibility of the SeaWiFS-based PHYSAT method has been added in the revised manuscript (P5, lines 12–21; P7, lines 3–11).
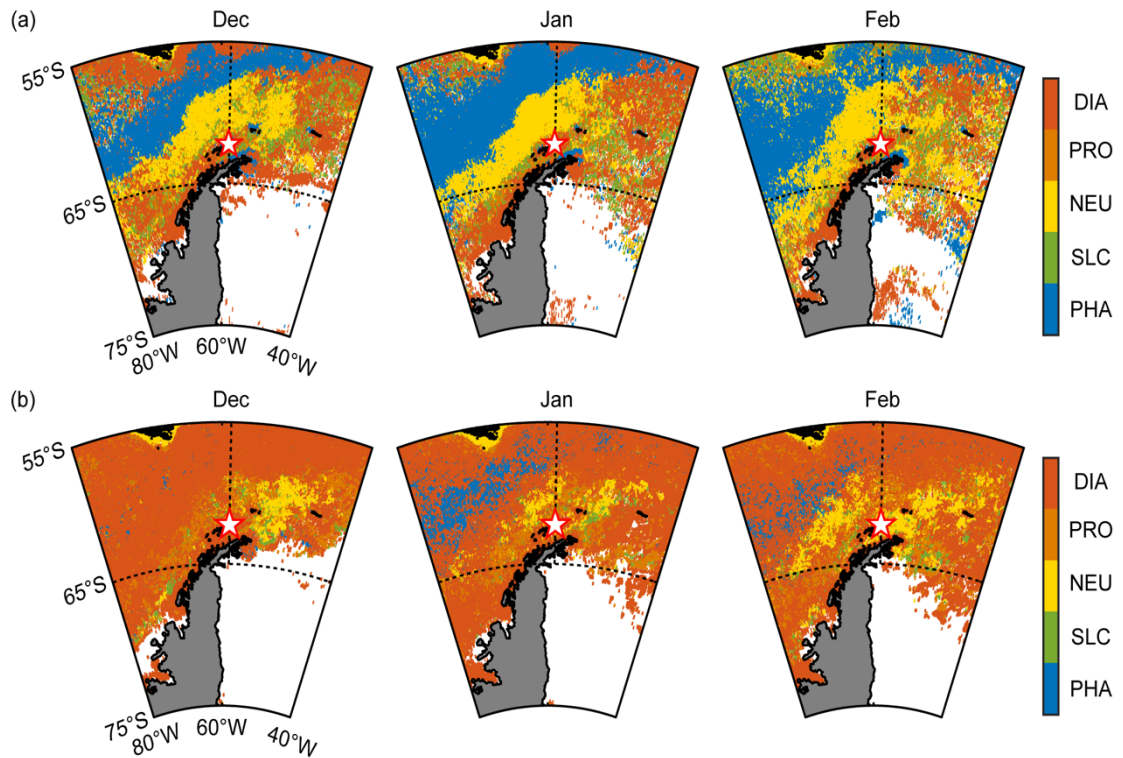
Figure. Phytoplankton taxonomic composition including diatoms (DIA), Prochlorococcus (PRO), nanoeucaryotes (NEU), Synechococcus (SLC). and phaeocystis (PHA) during the months of December, January, and February estimated using the PHYSAT algorithm based on (a) the SeaWiFS climatology map archived from 1997 to 2010, and (b) the MODIS climatology map archived from 2002 to 2016

2. **Need to compare the absolute DMSP concentration, not just the DMSP-to-chlorophyll ratio:** As this referee suggested, we have added figures for sea-surface DMSP concentration near the observation site (Fig. 3b and Fig. S4a). A brief explanation regarding the DMSP concentration has also been added (P7, line 36 – P8, line 1). The DMSP concentration was ~30% higher in the Weddell Sea during the blooming period, possibly owing to intense blooming of DMSP-containing diatoms. This could illustrate that, despite having lower DMSP-to-chlorophyll ratios than phaeocystis, diatoms dominated the overall DMSP production in the Weddell Sea owing to their much larger biomass. However, we cannot fully support this hypothesis owing to the absence of field measurement of taxonomic compositions of phytoplankton in this vast remote ocean.

3. **Provide clear description regarding the results for the PHYSAT analysis:** The PHYSAT method is a bio-optic model specifically developed to identify the dominant phytoplankton groups. Here, *"dominant"* has been defined as situations in which a given phytoplankton group is a major contributor to the total pigment at a given 9 km resolution (Alvain et al., 2005 and 2008). This paragraph has been added to the revised manuscript (P5, lines 19–21). We have replaced *"~35% of the satellite pixels were dominated by diatoms"* to *"the dominance of the diatom was ~35% during the austral summer period*

2

in the Weddell Sea, followed by nanoeucaryotes (20%), phaeocystis (17%), Prochlorococcus (15%), and Synechococcus (14%)" for better clarity (P6, line 37–38).

4. **The statement –"About 38.2% of the hourly mean number concentration of nanoparticles complied with the >90%criterion." Did the remaining 61.8% of the data indicate any significant link to the origin of the air masses?** The formation of new particles in the remote marine atmosphere is complicated owing to multiple sources and complicated processes. The remaining 61.8% of hourly mean number concentration of nanoparticles may have undergone more complicated transport history. Therefore, the hourly mean number concentration of nanoparticles, which do not satisfy the >90% retention over the two ocean domains, were excluded from further analysis. Because it was not easy to find a strong relationship between the formation of nanoparticles and the multiple potential source origin (e.g., marine biota, sea-ice extent, and penguin colony). A total of 101 days were defined as new particle formation events during the eight years and 80 days of new particle formation events occurred when the air mass originated from the ocean domain. The number concentration of the nanoparticle was at its maximum during the productive summer period, and the frequency of new particle formation was the highest when the air mass originated from the ocean domain. Therefore, we focused on the influence of marine biota on the formation of nanoparticles in this study. In the revised manuscript, we have added more description to clarify the scope of the present study (P1, line 16; P1, lines 20–22; P5, line 34 – P6, line 6; P6, lines 11–13). Furthermore, the limitations of the present study and the scope of future studies have also been added (P19, line 33 – P10, line 8).

5. **How many individual filter measurements are represented by each bar shown in Fig. 3? Provide daily MSA concentration data:** 52 individual measurements for MSA (43 measurements for the Bellingshausen Sea and nine measurements for the Weddell Sea) were used for Fig. 3 in the previous version of the manuscript. As this referee suggested, we have added daily MSA concentration data in the revised manuscript (Fig. 4a). An explanation of MSA variation has also been added (P8, lines 16–27; P8, lines 30–33). We have revised the figure for MSA concentration in response to the query raised by referee #2. In the revised figure, we have applied 3-, 4-, and 5-day back trajectories to analyze the potential origin of MSA, because the retention time of PM_{10} is known to be >2 days in the low troposphere. The extension of the air mass back trajectory time points altered the percentage of air mass retention above the major domains. The number of samples that satisfy >90% retention in the Bellingshausen and Weddell Seas was less than 20% of the total MSA samples owing to its longer transport pathway. Inevitably, the air mass origin of MSA was divided into two sectors i.e., the Bellingshausen Sea sector (<58.8ºW) and the Weddell Sea sector (>58.8ºW) by selecting the air mass back trajectories with >50% retention in a given sector. Thus, all the measurements (84 individual samples) were allocated to one of the two sectors. The mean MSA concentration and the number of individual measurements are shown in Fig. 4b.

Technical correction

6. P3, line 34: We have replaced "…*below the detection*" to "*below the detection limit*" (P4, line 3).

Response to Referee 2

We thank Referee 2 for providing valuable suggestions that improved the readability of our revised manuscript. Our responses to this Referee's scientific and technical comments are stated below.

Scientific issues

1. **Need to provide more comprehensive discussion on how this result build upon earlier findings and what exactly is the new scientific message:** The key finding of this study is that the formation of nanoparticles was strongly associated with not only the biomass of phytoplankton but also, more importantly, its taxonomic composition in the Antarctic Ocean; therefore, changes in the taxonomic composition of marine phytoplankton (i.e., DMS(P)-rich species vs. DMS(P)-poor species) may have a significant impact on the aerosol properties in the remote marine environment. In the revised manuscript, we have added a paragraph explaining earlier finding and the key results of this study. Furthermore, the limitations of the present study and the scope of future studies have also been added (P9, line 29 – P. 10, line 8).

2. **Add more recent published work:** In the revised manuscript, we have added recent findings that report the roles of diverse environment factors affecting the formation of new particles (P3, line 10; P3, lines 17–24).

3. **Typical lifetime of PM is generally longer than 2 days that used to identify the origin of measure air mass:** We agree with this referee that the retention time of PM_{10} is known to be >2 days in the lower troposphere. The retention time for aerosol particles with a diameter <10 μm is known to be 3–5 days in the low troposphere (Mishra et al., 2004; Budhavant et al., 2015). Therefore, we have applied 3-, 4- and 5-day air mass back trajectories to identify the potential origin of MSA in the revised manuscript. When applying 3-, 4-, and 5-day air mass back trajectories, the number of samples that satisfy >90% retention in the Bellingshausen and Weddell Seas was less than 20% of the total MSA samples owing to its longer transport pathway. Inevitably, the air mass origin of MSA was divided into two sectors i.e., the Bellingshausen Sea sector (<58.8ºW) and the Weddell Sea sector (>58.8ºW) by selecting the air mass back trajectories with >50% retention in a given sector. We have added these explanations in the revised manuscript (P4, lines 21–24; P8, lines 20–25; P8, lines 30–33).

Minor scientific and technical issues

4. **Need to modify some terminologies:** We have changed "*aerosols*" to "*aerosol particles*" (P1, line 15) and "*secondary organic aerosols*" to "*new particles*" (P1, line 35) in the revised manuscript.

5. The paper written by Kim et al. (2018) is a companion paper submitted to ACP and is under review (*Kim et al., New particle formation events observed at the King Sejong Station, Antarctic Peninsula – Part 1: Physical characteristics and contribution to cloud condensation nuclei, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1180, in review, 2018*).

6. **Why there are only 6 measurements points in the figure, although the measurements cover 8 year?** Unfortunately, we could not obtain the CN data during the summer period in January and February 2014 owing to a malfunction of the analytical system. As we started observation in March 2009, the CN data for January and February 2009 were not included in the analysis. Therefore, six measurement points are represented in Fig. 5.

7. **Clarify the meaning of DMSP-to-chlorophyll ratio:** The conversion of cellular DMSP into DMS is controlled by not only the concentration of DMSP but also, more importantly, the DMSP cleavage enzyme. The phytoplankton species containing high cellular DMSP (i.e., high DMSP-to-chlorophyll ratio) mostly possess an enzyme that can convert cellular DMSP into DMS, whereas phytoplankton species containing low DMSP content (i.e., low DMSP-to-chlorophyll ratio) do not have a DMSP cleavage enzyme. Therefore, the DMSP-to-chlorophyll ratio is commonly used to explain the differences in taxonomic compositions affecting the oceanic DMS-production capacity (e.g., Belviso et al., 2000; Stefels et al., 2007; Tison et al., 2010; Park et al., 2014b and 2018). We have added more description to clarify the meaning of the DMSP-to-chlorophyll ratio in the revised manuscript (P4, line 34 – P5, line 4; P7, lines 15–40).

8. We have changed "DEB" to "DEC" (Figure S3 and S4 in the revised supplement).

Response to Referee 3

We thank Referee 3 for providing valuable comments on our manuscript. Our responses to this Referee's five scientific issues are provided below.

Scientific issues

1. **Only 38% of the data are analyzed. Present a whole analysis, including air masses originating other sectors:** We have a companion paper (*Kim et al., New particle formation events observed at King Sejong Station, Antarctic Peninsula – Part 1: Physical characteristics and contribution to cloud condensation nuclei*) that describes the physical characteristics of the aerosol particles observed at the King Sejong station during the same period. The whole dataset of new particle formation events recorded for eight years was analyzed in detail in this companion paper. In short, Kim et al. (2018) evaluated the numerous physical parameters (e.g., occurrence, formation rate, growth rate, condensation sink, and source rate of condensable vapor) that originated from South America, Antarctic Peninsula, and the Bellingshausen and Weddell Seas, and determined that ~80% of the new particle formation events were observed when the air mass originated from ocean domain (including the Bellingshausen and Weddell Seas) during the productive summer peiod. Therefore, we focused on the influence of marine biota on the formation of nanoparticles in this study. A total of 22,469 hourly mean number concentrations of nanoparticles were measured over the eight years. The linkage between new particle formation and environmental parameters is complicated, owing to the interplay among multiple sources and complicated processes. We have excluded the dataset that did not satisfy >90% retention over the two ocean domains to evaluate the relationship between new particle formation and oceanic biological characteristics. Thus, 8573 hourly mean number concentrations of nanoparticles were used for further analysis. To the best of our knowledge, it is based on the longest observation periods measured in the Antarctic site regarding this subject. We agree with this referee's comment that the previous version of this manuscript lacked explanation regarding the overall trend of nanoparticles, even though it was introduced in the companion paper. Therefore, we have added more description regarding "1) *the general aspect of new particle formation events (e.g., frequency and potential origin)* (P5, line 34 – P6, line 2)' 2) *time-series transport history* (Fig. 1c), 3) *the reason why we focused on the dataset originating from two ocean domains* (P1, lines 20–22; P6, lines 2–6) and 4) *the limitations of the present study and the scope of future studies* (P9, line 33 – P10, line 8)*" in the revised manuscript.

2. **As regards of the filter data, there are only few months of data presented, this should be stated in the abstract:** A total of 84 MSA samples were analyzed daily during the summer periods in 2013 and 2014. As this referee suggested, we have added this information to the Abstract (P1, line 29–30).

3. **MSA concentration is too high (Issue 1) and local penguin emissions near the base should be discussed (Issue 2):**
   (response to issue 1) We have added a new figure representing the daily MSA concentration and a brief explanation of MSA variation in the revised manuscript (Fig. 4a; P8, lines 16–27). The mean MSA concentration during the entire PM_{10} sampling period was 72.6 ± 99.1 ng m^{-3} (ranged from 4.2 to 657.0 ng m^{-3}; 176.0 ± 186.2 ($n=16$), 68.1 ± 36.3 ($n=25$), 40.1 ± 30.9 ($n=27$), and 30.9 ± 26.2 ($n=16$) for Jan. 2013, Feb. 2013, Dec. 2013, and Jan. 2014, respectively), similar to the values observed at six Antarctic sites during the productive summer period. For example, the monthly mean MSA concentrations observed during the summer period were reported to be 59.3, 154.2, 63.0, 180, ~60, and ~100 ng m^{-3} in the Halley, Neumayer, Dumont d'Urville, Palmer, Zhongshan, and Marambio stations, respectively (e.g., Prospero et al., 1991; Minikin et al., 1998; Preunkert et al., 2007; Read et al., 2008; Zhang et al., 2015; Asmi et al., 2018). Note that most of the filter samples for MSA measurement were collected at a duration >3 days in these studies. As shown in Fig. 4a, an extremely high MSA concentration was observed between 14 Jan. 2013 and 17 Jan. 2013 (291.9, 386.6, 657.0, and 489.6 ng m^{-3} at a daily interval). During this period, the air masses originated from the Bellingshausen Sea (>90% air mass retention over the Bellingshausen Sea domain based on 2-, 3-, 4-, and 5-day air mass trajectories analysis). Such a high MSA concentration was also observed at the Antarctic site. For example, ~300 ng m^{-3} of MSA was reported at the Zhongshan Station (located in eastern Antarctic site) for the aerosol particles collected for a sampling duration of 10–15 days. Therefore, the high MSA concentration observed at the King Sejong Station appears to be due to the high biological productivities surrounding the observation site and the relatively shorter sampling duration during the study period.

(response to issue 2) Over the study period of eight years, 16 days of new particle formation events out of 101 events were observed when the air mass originated from the Antarctic Peninsula. Penguin colonies are dispersed throughout the Antarctic Peninsula (Croxall et al., 2002), and the emission of ammonia from these colonies could trigger the formation of nanoparticles (Weber et al., 1998; Croft et al., 2016). However, this issue was beyond the scope of the present study. In the revised manuscript, we have added more description to clarify the scope of the present study (P1, line 16; P1, lines 20–22; P5, line 34 – P6, line 6).

4. **Need to add more recent findings regarding this issue:** We have added more references that report the roles of diverse environment factors affecting the formation of new particles in the revised manuscript (P2, lines 17–24).

5. **Need to carry on a more in-depth analysis with the whole data-set available:** As we mentioned above, we have a companion paper that describes the physical characteristics of the aerosol particles observed at the King Sejong station during the entire sampling period. The number concentration of the nanoparticle was at its maximum during the productive summer period, and the frequency of new particle formation was highest when the air mass originated from the ocean domain. Therefore, we focused on the influence of marine biota on the formation of nanoparticles in this study. In the revised manuscript, we have added more description to clarify the scope of the present study (P1, line 16; P1, lines 20–22; P5, line 34 – P6, line 6). We have added more description and figures to support our key findings in the revised manuscript. In short, the explanation regarding the biological characteristic (e.g., DMSP-relevant process, feasibility of PHYSAT method and oceanic DMS production capacity) of the surrounding ocean was thoroughly complemented in the revised manuscript (P4, line 34 – P5, line 6; P7, lines 3–11; P7, line 16 – P8, line 40). Figures for 8-year CN records ($CN_{2.5}$, CN_{10} and $CN_{2.5-10}$), 8-year transport history at an hourly interval, sea-surface DMSP concentration, and daily MSA concentration have been newly added in the revised manuscript (Fig. 1, Fig. 3b, Fig. 4 and Fig. S4a). Furthermore, the limitations of the present study and the scope of future studies have also been added (P9, line 28 –P10, line 8).

Response to Short Comments

We thank to Dr. Allan for providing insightful suggestions that improved the readability of our revised manuscript. Our responses to Dr. Allan's comments are stated below.

Scientific comment

1. **Need to add explanation on the roles of iodine compounds in new particle formation events:** We agree that iodine-containing compounds which produced by both biotic and abiotic processes are one of the key compounds contributing to new particle formation in the Arctic and Antarctic regions. Unfortunately, we do not have observational evidence (i.e., chemical analysis of halogen compounds) to support the roles of iodine-compounds in formation of new particles in this study. In the revised manuscript, we have added recent findings regarding the potential roles of halogen compounds in the 'Introduction' part (P2, lines 17–22). Molecular-scale measurements of chemical species (e.g., sulfur-, nitrogen-, and halogen-containing compounds) involved in nucleation processes are required to provide direct evidence for the contribution of these compounds to the formation and growth of aerosol particles and to understand their climate feedback roles in the remote marine environment. Therefore, we have added a short paragraph indicating the limitations of the present study and the scope of future studies in this regard (P9, line 33 – P10, line 8).

New particle formation events observed at the King Sejong Station, Antarctic Peninsula – Part 2: Link with the oceanic biological activities

Eunho Jang[1, 2,*], Ki-Tae Park[1,*], Young Jun Yoon[1], Tae-Wook Kim[3], Sang-Bum Hong[1], Silvia Becagli[4], Rita Traversi[4], Jaeseok Kim[5], Yeontae Gim[1]

[1]Korea Polar Research Institute, 26 Songdomirae-ro, Yeonsu-gu, Incheon 21990, South Korea
[2]University of Science and Technology, 217 Gajeong-ro, Yuseong-gu, Daejeon 34113, South Korea
[3]Division of Environmental Science and Ecological Engineering, Korea University, Seoul, South Korea
[4]Department of Chemistry "Ugo Schiff", University of Florence, via della Lastruccia, 3, Sesto F.no (FI), 50019, Italy.
[5]Korea Research Institute of Standards and Science, 267 Gajeong-ro, Yuseong-gu, Daejeon 34113, South Korea
[*]These authors contributed equally to this work

Correspondence to: Ki-Tae Park (ktpark@kopri.re.kr)

Abstract. Marine biota is an important source of atmospheric ~~aerosols~~ aerosol particles in the remote marine atmosphere. However, the relationship between new particle formation and marine biota is poorly quantified. Long-term observations (from 2009 to 2016) of the physical properties of atmospheric aerosol particles measured at the Antarctic Peninsula (King Sejong Station; 62.2°S, 58.8°W) and satellite-derived estimates of the biological characteristics were analyzed to identify the link between new particle formation and marine biota. New particle formation events in the Antarctic atmosphere showed distinct seasonal variations, with the highest values ~~occurring~~ occurred when the air mass originated from the ocean domain during productive austral summer (December, January and February). Interestingly, new particle formation events were more frequent in the air masses that originated from the Bellingshausen Sea than in those that originated from the Weddell Sea. The monthly mean number concentration of nanoparticles (2.5–10 nm in diameter) was >2-fold when the air masses passed over the Bellingshausen Sea than the Weddell Sea, whereas the biomass of phytoplankton in the Weddell Sea was more than ~70% higher than that of the Bellingshausen Sea during the austral summer period. Dimethyl sulfide (DMS) is of marine origin and its oxidative products are known to be one of the major components in the formation of new particles. Both satellite-derived estimates of the biological characteristics (dimethylsulfoniopropionate (DMSP; precursor of DMS) and phytoplankton taxonomic composition) and in situ methanesulfonic acid (~~MSA; oxidation product of DMS~~ 84 daily measurements during the summer period in 2013 and 2014) analysis revealed that DMS(P)-rich phytoplankton were more dominant in the Bellingshausen Sea than in the Weddell Sea. Furthermore, the number concentration of nanoparticles was positively correlated with the biomass of phytoplankton during the period when DMS(P)-rich phytoplankton predominate. These results indicate that oceanic DMS emissions could play a key role in the formation of new particles; moreover, the taxonomic composition of phytoplankton could affect the formation of ~~secondary organic aerosols~~ new particles in the Antarctic Ocean.

메모 포함[KTP1]: (response to referee #2)
Q4. As suggested, we have changed "*aerosols*" to "*aerosol particles*".

메모 포함[WU2]: (response to referee #1 and #3)
We have added this sentence to clarify the scope of this study.

메모 포함[KTP3]: (response to referee #1 and #3)
We have changed "*occurring during austral summer*" to "*occurred when the air mass originated from the ocean domain during the productive austral summer*" to clarify the scope of this study.

메모 포함[WU4]: (response to referee #3)
Q2. As regards of the filter data, there are only few months of data presented, this should be stated in the abstract: As this referee suggested, we have added this information to the Abstract.

메모 포함[KTP5]: (response to referee #2)
Q4. As suggested, we have changed "*secondary organic aerosols*" to "*new particles*".

1 Introduction

Aerosols in the atmosphere significantly influence radiative forcing directly (by scattering incoming radiation) and indirectly (by modifying cloud micro-physical properties) (IPCC, 2013). Considering that the ocean surface accounts for approximately 70% of the total surface of the Earth, the marine aerosols are globally one of the most important natural aerosol systems (O'Dowd et al., 2004; Leck and Bigg, 2005; O'Dowd and De Leeuw, 2007). Marine aerosols consist of primary and secondary aerosols. The primary aerosols are produced via the bubble bursting process and include mostly sea-salts and organic matter (O'Dowd and De Leeuw, 2007). Secondary aerosols (resulting from gas-to-particle conversion) are produced in the marine atmosphere through several different processes (e.g., binary, ternary and ion-induced) (Kulmala et al., 1990; Korhonen et al., 1999; Lee et al., 2003; Lovejoy et al., 2004; Kirkby et al., 2016; Jokinen et al., 2018). Biogenic volatile organic compounds emitted into the marine atmosphere through air-sea gas exchange, and their oxidative products can be involved in new particle formation events via the nucleation of stable clusters of the order of 0.5–1 nm in size (O'Dowd and De Leeuw, 2007).

In the Antarctic, most submicron aerosol particles are derived from natural sources rather than from long-range transport of anthropogenic sources (Ito, 1989; Kyrö et al., 2013). The Southern Ocean has been considered as the most significant source of secondary organic aerosols in the Antarctic atmosphere, especially during the phytoplankton bloom period (Asmi et al., 2010; Yu and Luo, 2010). Recent studies reported that biological (mostly derived by sea-ice algae) and abiotic (photochemical reaction of halogen compounds) processes occurring near sea-ice regions could significantly influence the formation and growth of aerosol particles (Atkinson et al., 2012; Kyrö et al., 2013; Allan et al., 2015; Dall'Osto et al., 2017 and 2018). Molecular level evidence of new particle formation via sequential addition of iodine-containing species, which could be emitted from both open water and sea-ice zone, was reported in the Arctic and Antarctic sites (Sipilä et al., 2016). In addition, the emission of ammonia from seabird colonies in polar regions could act as a key factor contributing to bursts of newly formed particles (Weber et al., 1998; Croft et al., 2016). In addition, high biological activities in melt ponds and at sea-ice edges also have an important influence on the total burden of organic aerosols (Kyrö et al., 2013; Dall'Osto et al., 2017). The Antarctic Peninsula is one of the three areas of the globe facing drastic regional warming, and it undergoes rapid environmental changes (i.e., increased temperature, acidification, shallowing mixed layer depth, sea-ice decline, increased light, increased nutrient supply, reduced salinity and glacial retreat) (Clarke et al., 2007; Deppeler and Davidson, 2017). Such environmental changes in this region could significantly affect the marine ecosystem. Marine phytoplankton influence aerosol properties by releasing various organic compounds back into the atmosphere. The Antarctic Ocean is known to be a significant source of numerous biogenic volatile organic compounds including dimethyl sulfide (DMS), organic nitrogen and halogenated organic compounds (Laturnus et al., 1996; Beyersdorf et al., 2010; Dall'Osto et al., 2017; Giordano et al., 2017). In particular, the Antarctic Ocean is the region with the highest sea-surface DMS concentration (Lana et al., 2011). Observations of both DMS and its oxidative products (i.e., methanesulfonic acid (MSA) and non-sea-salt SO_4^{2-}) in Antarctica show a clear seasonal cycle with a minimum in austral winter and a maximum in austral summer (Prospero et al., 1991; Minikin et al., 1998; Preunkert et al., 2007; Read et al., 2008). DMS is produced by a complex marine biota food-web mechanism (Stefels et al., 2007; Park et al., 2014a). Phytoplankton produce dimethylsulfoniopropionate (DMSP) and then some of the DMSP is converted into gaseous DMS through enzymatic cleavage (Simó, 2001).

메모 포함[WU6]: (response to referee #2)
Q2. We have added these references.

메모 포함[KTP7]: (response to referee #2, 3 and Short Comment)
Q2 and Q4. Need to add more recent findings: We have added recent findings that report the roles of diverse environment factors affecting the formation of new particles.

2

The biogenic emission of DMS from the ocean is the largest natural sulfur source to the atmosphere (Andreae, 1990) and, the oxidation of DMS in the marine atmosphere is a key process contributing to the formation of new particles (Park et al., 2017). Once the DMS in the sea-surface is emitted into the atmosphere, it is rapidly converted into SO_2 and MSA through the photochemical oxidation process. The SO_2 and MSA formed from DMS tend to contribute to the formation of new particles via nucleation processes and eventually serve as nuclei for cloud formation (Charlson et al., 1987). Secondary organic aerosols contribute to a large fraction of the submicron aerosol mass in the atmosphere, affecting clouds and climate (Jimenez et al., 2009). However, secondary organic aerosols are a source of considerable uncertainty in understanding current climate change (Shiraiwa et al., 2017). In particular, the formation of new particles in the remote marine atmosphere and their association with marine biota remains poorly quantified.

In this study, we aimed to identify the link between new particle formation and marine biota at a remote Antarctic site, where biological productivity is the highest in the global ocean. To this end, we analyzed the physical and chemical properties of aerosol particles at King Sejong Station (62.2°S, 58.8°W) on the Antarctic Peninsula from 2009 to 2016. To study the oceanic biological characteristics surrounding the observation site, we analyzed satellite-derived estimates including the chlorophyll concentration, total DMSP concentration and taxonomic composition of marine phytoplankton.

2 Experimental Methods

2.1. Aerosol measurements

King Sejong Station (62.2°S, 58.8°W) is located on the Antarctic Peninsula where severe climate change is occurring (Fig. 1a). The aerosol observatory is approximately 400 m southwest of the main facilities of King Sejong Station and approximately 10 m above sea level (m.a.s.l). Continuous observations of the physical properties of aerosol particles in the Antarctic atmosphere were conducted between March 2009 and December 2016 at the observatory. The number concentration of aerosol particles (CN) was measured using two condensation particle counters (CPCs) that have different measurement range limits for particle diameter: particles larger than 2.5 nm ($CN_{2.5}$; TSI model 3776) and particles larger than 10 nm (CN_{10}; TSI model 3772) (Fig. 1a). The difference in the particle number concentration between the two CPCs (i.e., particles between 2.5 and 10 nm in diameter) was interpreted as an indication of the existence of newly formed nanoparticles ($CN_{2.5-10}$) (Fig. 1b). An Aethalometer (Magee Scientific model AE16) was used to analyze the concentration of black carbon by measuring light-absorbing particles at the 880 nm wavelength. To avoid local influence on the aerosol properties, data with a black carbon concentration of >100 ng were excluded. Data were also excluded when the wind direction was between 355° and 55°. This is because the north-eastern direction is designated the local air pollution sector due to emissions from the power generators and crematory. More detailed information regarding the aerosol particle analysis at King Sejong Station is provided by Kim et al. (2017 and 2018).

For analysis of chemical properties of aerosol particle, an air sampler equipped with a PM_{10} impactor (collecting particles <10 μm in aerodynamic equivalent diameter) was used to collect aerosol particles. The sampler was mounted on the roof of the aerosol observatory and sampled particles every 24 hours during the summer period in 2013 and 2014 (explicitly, from 14 January to 28 February in 2013, and from 2 December 2013 to 18 January

3

메모 포함[KTP8]: (response to referee #3)
Q1. Only 38% of the data are analyzed. Present a whole analysis, including air masses originating other sectors: We have added new figure that represent the 8-year nanoparticle measurement and corresponding air mass transport history.

메모 포함[KTP9]: (response to referee #2)
Q5. This paper is a companion paper submitted to ACP and is under review (https://doi.org/10.5194/acp-2018-1180).

2014). Half of a 47-mm Teflon filter was used to measure major ions including MSA. The MSA (and the other ions) collected on the filter was extracted into about 10 mL (18 MΩ Milli-Q) in ultrasonic bath for 20 min. MSA was determined by an ion chromatography system (Dionex, Thermo Fisher Scientific Inc.) following the procedure described by Becagli et al. (2012). For MSA, reproducibility on real samples was better than 5%. Filter

5   blank concentrations for methanesulphonate were always below the detection limit.

2.2. Air-mass back trajectories

The air mass back trajectories and meteorological parameters were obtained using the Hybrid Single-Particle Lagrangian Integrated Trajectories model (Draxler and Hess, 1998). In general, the growth rate of sub-micron

10 particles in the remote marine environment ranges from 0.2 to 5.0 nm h^{-1} (Järvinen et al., 2013; Weller et al., 2015; Kerminen et al., 2018), and the mean growth rate of the aerosol particles measured at the King Sejong Station was 0.68 ± 0.27 nm h^{-1} during the eight years (Kim et al., 2018). Therefore, the 2-day air mass back trajectories and hourly positions were determined and combined with satellite-derived geographical information to identify the travel history of the air mass arriving at the observation site. Daily geographical information on sea-ice, land, and

15 ocean area was obtained from the Sea Ice Index at a 25-km resolution provided by the National Snow and Ice Data Center (NSIDC). The oceanic region adjacent to the observation site was surrounded by two different ocean basins, namely, the Bellingshausen and Weddell seas. To evaluate the influence of the oceanic biological characteristics on the occurrence of new particle formation, we limited our analysis to the air masses that had exposure predominantly to the ocean area. Specifically, the origin of the hourly air mass arriving at the observation

20 site was divided into two ocean domains (i.e., the Bellingshausen and Weddell seas). Then, all air mass back trajectories were grouped into one of the two ocean domains by only selecting the 2-day air mass back trajectories that had >90% retention in a given ocean domain.
A total of 84 PM$_{10}$ samples for MSA analysis were collected daily during the summer periods in 2013 and 2014. The retention time of the aerosol particles with a diameter <10 μm is known to be approximately 3–5 days in the

25 atmosphere (Mishra et al., 2004; Budhavant et al., 2015). Therefore, 3-, 4-, and 5-day air mass back trajectories were applied to identify the potential origin of MSA during the sampling period.

2.3. Phytoplankton biomass, DMSP and taxonomic composition analysis

Satellite-derived ocean color provide a good measure of analyzing the phytoplankton characteristics of the Southern Ocean (Siegel et al., 2013; Haentjens et al., 2017). The phytoplankton biomass of the two ocean domains

30 was estimated by calculating the chlorophyll concentration from the Moderate Resolution Imaging Spectroradiometer on the Aqua (MODIS-Aqua) satellites at 4 km resolution during the study period (2009–2016). The trajectory concentration of the air masses originating from the two ocean domains was calculated from the ratio of the number of hourly trajectory points passing over each grid cell (1° × 1°) to the total number of hourly trajectory points (Kim et al., 2011), as shown in Fig. 1a 2a. We limited our analysis of satellite-derived chlorophyll

35 concentration to the ocean area for which the trajectory concentration was approximately over 0.1% (55–65°S, 40–60ºW for the Weddell Sea and 55–65°S, 60–80ºW for the Bellingshausen Sea). DMSP is produced by marine phytoplankton and is the most important precursor of oceanic DMS production. However, the dependence of the

4

메모 포함[KTP10]: (response to referee #1)
Q6. As suggested, we have replaced "*below detection*" to "*below detection limit*".

메모 포함[WU11]: (response to referee #2)
Q3. Typical lifetime of PM is generally longer than 2 days that used to identify the origin of measure air mass: We agree with referee #2 that retention time of PM$_{10}$ is known to be >2 days in the lower troposphere. Therefore, we have applied 3-, 4- and 5-day air mass back trajectories to identify the potential origin of MSA in the revised manuscript.

oceanic DMS emission on phytoplankton biomass and DMSP concentration is not straightforward owing to the strong variabilities across taxonomic groups and its interplay with environmental factors. Nevertheless, temporal and spatial distribution of sea-surface DMSP could be an indicator of contemporary DMS emission. In particular, the DMSP-to-chlorophyll ratio could represent the potential DMS production capacity of the ocean because the phytoplankton species with higher cellular DMSP content (i.e., higher DMSP-to-chlorophyll ratio) mostly possess an enzyme that can convert cellular DMSP into DMS, whereas phytoplankton species containing lower DMSP content (i.e., lower DMSP-to-chlorophyll ratio) do not have a DMSP cleavage enzyme (Stefels et al., 2007, Park et al., 2014b and 2018). The total DMSP concentration in the sea-surface was estimated using the algorithm developed by Galí et al. (2015). The algorithm for the total DMSP concentration was based on the satellite-derived chlorophyll concentration and the light exposure regime (see Supplement for more information). We estimated the taxonomic phytoplankton composition of the two ocean domains using the PHYSAT method. This method is a bio-optic model that was specifically developed to identify the dominant phytoplankton groups from ocean color measurements. Phytoplankton groups are generally characterized by specific pigment, shape and size that have different light scattering and absorption properties (Alvain et al., 2005). ~~Therefore, the PHYSAT method can be used to classify sea-surface phytoplankton into five groups including diatoms, Prochlorococcus, nanoeucaryotes, Synechococcus and phaeocystis simultaneously with the chlorophyll concentration (Alvain et al., 2008; Mustapha et al., 2014). The monthly data set of phytoplankton groups at a resolution of 9 km was obtained from the PHYSAT database (http://log.univ-littoral.fr/Physat), and was estimated between 1997 and 2010. The period of the PHYSAT analysis did not coincide with the period of aerosol analysis. This is because the PHYSAT method was developed and calibrated based on daily global data derived from Sea-viewing Wide Field-of-view Sensor (SeaWiFS) that had been operated from September 1997 to December 2010.~~ The PHYSAT method was first developed in 2005 and was used to classify sea-surface phytoplankton into four groups: diatoms, Prochlorococcus, nanoeucaryotes, and Synechococcus (Alvain et al., 2005). Subsequently, the modified PHYSAT method, which can estimate the contribution of the phaeocystis group, was reported in 2008 (Alvain et al., 2008). The PHYSAT method was developed and calibrated based on global data obtained from the sea-viewing wide field-of-view sensor (SeaWiFS) operated from September 1997 to December 2010. In this study, the monthly dataset of five phytoplankton groups at a resolution of 9 km was obtained from the PHYSAT database (http://log.univ-littoral.fr/Physat) estimated using climatology over the SeaWiFS period (1997–2010). Note that "dominant" has been defined as situations in which a given phytoplankton group is a major contributor to the total pigment in a given 9 km resolution (Alvain et al., 2005 and 2008).

3 Results and Discussion

3.1. Seasonal variabilities of nanoparticles at King Sejong Station

The number concentration of aerosol particles increased gradually from early spring, peaked in the austral summer period (December, January and February) and then began to decrease (Fig. ~~S1~~ 1a). The number concentration of nanoparticles (2.5–10 nm in diameter, $CN_{2.5-10}$), which is an indication of newly formed particles, also shows distinct seasonal variation (Fig. 1b). The physical aspects of new particle formation ~~event~~ events (e.g., number of new particle formation event days, formation, and growth rate) observed at the same site are explained in detail

5

in Kim et al. (2018) ~~that submitted in this issue~~. The observation site is surrounded by ocean, sea-ice, and land domains, which may influence new particle formation in different ways. The 2-day air mass back trajectory combined with geographical information showed that approximately ~~78.5% of the hourly trajectory points were assigned to the ocean, followed by sea-ice (16.3%) and land (5.2%) during the austral summer period (Fig. S2)~~

5 66% of the hourly trajectory points were assigned to the ocean, followed by sea-ice (29%) and land (6%) during the entire study period (Fig. 1c and Fig. S1a). The percentage of hourly trajectory points that passed over the ocean domain were at their maximum during the summer period (79%) when the extent of sea-ice was at its minimum (Fig. S1b). Kim et al. (2018) reported that a total of 101 days were defined as new particle formation events during the eight years and 80 days of new particle formation events occurred when the air mass originated

10 from the ocean domain. Furthermore, 16 days of new particle formation events were observed for the air masses originating from the Antarctic Peninsula. The remaining five days of events were considered as those of South American origin (three events) and undefined (two events) (see Kim et al. 2018 for the detailed definition and categorization of new particle formation events). The relationship between new particle formation and environmental parameters is complicated, owing to the interplay among multiple sources and complicated

15 processes. The number concentration of the nanoparticles was at its maximum during the productive summer period, and the frequency of new particle formation was the highest when the air mass originated from the ocean domain. Therefore, we focused on the influence of marine biota on the formation of nanoparticles. The hourly mean concentration of nanoparticles matched with the hourly air mass back trajectory in this study. A total of 22,469 hourly mean number concentrations of nanoparticles were measured above the Antarctic atmosphere over

20 the eight years. Approximately 38.2% of the hourly mean number concentration of nanoparticles, which satisfy the >90% retention of hourly trajectory points over the two ocean domains, were used to estimate the link between new particle formation and the oceanic biological characteristics around the observation site. The remaining 61.8% of the hourly mean number concentration of nanoparticles, which do not satisfy the >90% retention over the two ocean domains, were excluded from further analysis. Interestingly, the monthly mean number concentration of

25 nanoparticles that originated from the Bellingshausen Sea was highest in January (835.9 ± 2673.2 cm^{-3}) and ~2.5-times greater than that which originated from the Weddell Sea (332.0 ± 920.7 cm^{-3}; Fig. ~~1b~~ 2b and Table S1). The differences in the number concentration of nanoparticles that originated from the two ocean domains were particularly noticeable during the austral summer period (567.5 ± 249.3 cm^{-3} for the Bellingshausen Sea and 262.3 ± 66.1 cm^{-3} for the Weddell Sea). However, the differences were not evident between March and November (97.2 ± 51.1 cm^{-3} for the Bellingshausen Sea and 73.2 ± 56.9 cm^{-3} for the Weddell Sea; Fig. ~~1b~~ 2b).

30

3.2. Biological characteristics surrounding the observation site

In general, the abundance and composition of phytoplankton show distinct spatial and seasonal variation in the Antarctic Ocean (Sullivan et al., 1993). Primary production in the Antarctic Ocean is strongly controlled by various factors such as iron limitation, light availability and mixed layer depth (Arrigo et al., 1999; Sedwick et al.,

35 2007; Park et al., 2013a). The composition of the phytoplankton community is poorly studied in the Antarctic Ocean except for the marginal zone at the Antarctic Peninsula. Nevertheless, both phaeocystis and diatoms (mainly *Phaeocystis antarctica* and *Fragilariopsis cylindrus*) are well known as dominant phytoplankton groups in the Antarctic Ocean during the phytoplankton bloom period (Kropuenske et al., 2009; Arrigo et al., 2010). Both

6

diatom and phaeocystis, considered the most ecologically important phytoplanktonic groups, contribute >20% of the global primary productivity, and are particularly abundant at high latitudes (Schoemann et al., 2005; Malviya et al., 2016). The monthly mean chlorophyll concentration around the observation site (55ºS–65ºS, 40ºW–80ºW) began to increase in October and reached its maximum in November and December during the study period (Fig.

5 ~~S3a~~ S2a). The biological characteristics of the two ocean domains showed notable differences. The monthly mean chlorophyll concentration in the Weddell Sea (0.49 ± 0.07 mg m^{-3}) was ~70% higher than that of the Bellingshausen Sea (0.29 ± 0.05 mg m^{-3}) during the phytoplankton growth period (October–February; Fig. ~~2a~~ 3a and Fig. ~~S3b~~ S2b). The PHYSAT analysis, ~~which was applied from 1997 to 2010,~~ which was estimated using the SeaWiFS climatology map, revealed that the distribution of the dominant phytoplankton groups showed distinct

10 patterns (Fig. ~~2b~~ 3c). Diatoms were the most abundant, and ~~~35% of the satellite pixels were dominated by diatoms during the austral summer period in the Weddell Sea~~ the dominance of the diatom was ~35% during the austral summer period in the Weddell Sea, and followed by nanoeucaryotes (20%), phaeocystis (17%), Prochlorococcus (15%), and Synechococcus (14%) (Fig. ~~2b~~ 3c and Fig. ~~S4b~~ S3b). Conversely, the dominance of phaeocystis increased significantly and accounted for more than 50% in the Bellingshausen Sea during the austral summer

15 period, while the contribution of the diatom decreased below 10% (Fig. ~~2b~~ 3c and Fig. ~~S4a~~ S3a). Although, the period considered for the SeaWiFS archive (from September 1997 to December 2010) did not coincide with the period of aerosol particle observation at the King Sejong Station (from March 2009 to December 2016), the PHYSAT analysis performed using the SeaWiFS climatology map was successfully applied to the Southern Ocean and could represent the general seasonal trend of the taxonomic composition of marine phytoplankton in the study

20 area (Alvain and d'Ovidio, 2014, Mustapha et al., 2014). Recently, a regional PHYSAT algorithm for the Mediterranean Sea was developed by applying linear interpolation between SeaWiFS and MODIS wavelengths and reflectance threshold (Navarro et al., 2014). However, challenges remain in high-latitude areas such as the Southern Ocean, especially because of the rather sparse matchup available for the calibration and validation of the PHYSAT algorithm (Alvain et al., 2014).

25 ~~The production of DMS and DMSP is species-specific.~~ Enzymatic cleavage of planktonic DMSP into DMS is the major source of DMS and, the production of DMS and DMSP is species-specific. For example, diatoms, picoplankton (i.e., Synechococcus and Prochlorococcus) and most nanoeucaryotes are known to be DMS(P)-poor species. Conversely, phaeocystis and dinoflagellates have a high cellular DMSP content and many of them possess a DMSP cleavage enzyme that can convert DMSP into gaseous DMS (Keller, 1989; Stefels et al., 2007; Park et

30 al., 2014b). ~~In particular, Phaeocystis antarctica was reported to be a dominant species in terms of DMS production in the Antarctic Ocean during the bloom period (Gibson et al., 1990; Schoemann et al., 2005), exhibiting a cellular DMSP concentration in phaeocystis 10–100 times that of diatoms (Hatton and Wilson, 2007; Stefels et al., 2007). Therefore, the DMSP to chlorophyll ratio can reflect the differences in the taxonomic composition of the two ocean domains (Park et al., 2018).~~ The conversion of cellular DMSP into DMS is controlled by not only the

35 concentration of DMSP but also, more importantly, the DMSP cleavage enzyme. DMS is often produced following the local chlorophyll maxima, leading to a lag period (several weeks to months) (Polimene et al., 2012). This phenomenon is evident when the concentration of DMSP is largely contributed by DMSP-poor species such as diatoms. Most DMSP-poor species do not possess a DMSP cleavage enzyme, and therefore, the conversion of DMSP into DMS occurs when the cellular DMSP is released into the ocean as a form of dissolved matter.

40 Subsequently, some dissolved DMSP degrades into gaseous DMS through the bacterial DMSP enzymatic

7

메모 포함[KTP17]: (response to referee #1)
Q1. The discussion presented in Chapters 3 is not conclusive: We have added this paragraph to improve the readability.

메모 포함[KTP18]: (response to referee #1)
Q1. How representative is the PHYSAT database for the period 1997-2010 with regard to your observation period? We have replaced "*which was applied from 1997 to 2010*" to "*which was estimated by SeaWiFS climatology map*"to clarify the meaning.

메모 포함[KTP19]: (response to referee #1)
Q3. Provide clear description regarding the results for the PHYSAT analysis: We have replaced "*~35% of the satellite pixels were dominated by diatoms*" to "*the dominance of the diatom was ~35%.... and Synechococcus (14%)*".

메모 포함[KTP20]: (response to referee #1)
Q1. How representative is the PHYSAT database for the period 1997-2010 with regard to your observation period? An additional paragraph explaining the feasibility of the SeaWiFS-based PHYSAT method has been added.

메모 포함[WU21]: (response to referee #1)
Q1. The description regarding DMSP-to-chlorophyll ratio is confusing: We have added a paragraph explaining the DMSP-relevant processes and their association with marine biota to clarify the meaning of the DMSP-to-chlorophyll ratio in this chapter.

cleavage (Simó, 2001; Stefels et al., 2007). However, a larger proportion of dissolved DMSP is assimilated into bacterial tissues through demethylation processes, which do not produce gaseous DMS (Todd et al., 2007; Reisch et al., 2011). A direct correlation between the local chlorophyll concentration and atmospheric DMS mixing ratio in the absence of lag periods was observed in the Arctic Ocean where *Phaeocystis pouchetii* (containing both high

5 cellular DMSP and DMSP cleavage enzyme) dominates (Park et al., 2013b). Moreover, the DMS production capacity in the Arctic Ocean was more significantly controlled by the abundance of DMSP-rich phytoplankton than the total biomass of phytoplankton (Park et al., 2018). These results indicate that the blooming of phytoplankton species containing higher cellular DMSP content results in a much higher DMS production capacity than the blooming of DMSP-poor phytoplankton species. Therefore, the DMSP-to-chlorophyll ratio is

10 commonly used to explain the differences in taxonomic compositions affecting the oceanic DMS-production capacity (e.g., Belviso et al., 2000; Stefels et al., 2007; Tison et al., 2010; Park et al., 2014b and 2018). In particular, *Phaeocystis antarctica* was reported to be a dominant species in terms of DMS production in the Antarctic Ocean during the bloom period (Gibson et al., 1990; Schoemann et al., 2005), exhibiting a cellular DMSP concentration in phaeocystis several times that of diatoms (Hatton and Wilson, 2007; Stefels et al., 2007). The sea-surface DMSP

15 concentration surrounding the observation site was estimated using a newly developed algorithm and was 30% higher in the Weddell Sea than in the Bellingshausen Sea during the summer period, possibly owing to intense blooming of DMSP-containing diatoms in the Weddell Sea (Fig. 3b and Fig S4a). This could illustrate that, despite having lower cellular DMSP content than phaeocystis, diatoms dominated the overall DMSP production in the Weddell Sea owing to their much larger biomass. However, the DMSP to chlorophyll ratio in the Bellingshausen

20 Sea (110.2 ± 27.8 mmol g^{-1}) was ~2-fold higher than that of the Weddell Sea (72.2 ± 8.3 mmol g^{-1}) between December and February in 2009–2016 (Fig. 2c 3d and Fig. S5 S4b), possibly owing to the relatively higher contribution of the DMSP-rich phaeocystis group in the Bellingshausen Sea.

3.3. Influence of phytoplankton on aerosol formation

25 Biogenic trace gases produced by marine phytoplankton (i.e., DMS, isoprene, and halogenated gases) are known to be the key compounds contributing to the formation of new particles in the remote marine environment; however, quantifying the relationship between new particle formation events and marine biology is a major challenge (Brooks and Thornton, 2018). MSA in the marine atmosphere forms exclusively from the photooxidation of DMS, and shows strong seasonal variation (Ayers and Gras, 1991; Savoie et al., 1993; Preunkert

30 et al., 2008). A previous study has reported that the highest values for both MSA and the scattering Ångström exponent (SAE; qualitative examination of the aerosol optical mean size) were observed at the Marambio Station (64.3°S, 56.6°W) on the Antarctic Peninsula during austral summer in 2013–2015 (Asmi et al., 2018). The MSA concentration of the fine aerosol particles measured at the King Sejong Station during the summer period in 2013 and 2014 was broadly consistent with the number concentration of nanoparticles. The MSA concentration shows

35 distinct daily variations. The mean MSA concentration was 72.6 ± 99.1 ng m^{-3} (ranged from 4.2 to 657.0 ng m^{-3}) (Fig. 4a), similar to the values observed at six Antarctic sites during the productive summer period (e.g., Prospero et al., 1991; Minikin et al., 1998; Preunkert et al., 2007; Read et al., 2008; Zhang et al., 2015; Asmi et al., 2018). To identify the potential origin of MSA, air mass back trajectories were determined and the retention time above

8

each domain was averaged for the corresponding 24 h sampling time. When applying 3-, 4-, and 5-day air mass back trajectories, the number of samples that satisfy >90% retention in the Bellingshausen and Weddell Seas was less than 20% of the total MSA samples owing to its longer transport pathway. Inevitably, the air mass origin of MSA was divided into two sectors i.e., the Bellingshausen Sea sector (<58.8ºW) and the Weddell Sea sector

5 (>58.8ºW) by selecting the air mass back trajectories with >50% retention in a given sector. Although the limited number of samples of MSA (84 samples at daily intervals) collected during the summer periods in 2013 and 2014 may not be sufficient to identify its source origin exactly, the MSA concentration also showed distinct differences depending on the air mass origin. The inflow of the air masses from the Bellingshausen Sea increased the concentration of MSA in the aerosol particles. Notably, the MSA concentration that originated from the

10 Bellingshausen Sea sector (87.6 ± 110.0, 86.6 ±110.0, and 83.9 ± 109.0 ng m^{-3} for 3-, 4-, and 5-day air mass back trajectories based estimates, respectively) was ~7~ 3-times higher than that which originated from the Weddell Sea sector (27.4 ± 19.3, 30.6 ± 27.5, and 33.9 ± 30.4 ng m^{-3} for 3-, 4-, and 5-day air mass back trajectories based estimates, respectively) during the austral summer period in 2013–2014 (Fig. ~3~ 4b). Although the period of satellite observations and in situ chemistry analysis is not exactly the same, both ~multi-year~ satellite-derived

15 biological characteristics and aerosol chemistry data support the interpretation that there was higher abundance of DMS(P)-rich phytoplankton in the Bellingshausen Sea than in the Weddell Sea during the austral summer period (Fig. ~2~ 3, Fig. ~3~ 4, Figs. ~S3~ S2, ~S4~ S3 and ~S5~ S4).

In the 8-year record, the monthly mean chlorophyll concentration was positively correlated with the monthly mean number concentration of nanoparticles for the air masses that originated from the Bellingshausen Sea in January

20 and February (r^2 = 0.69, n = 12, P < 0.05; Fig. ~4~ 5a). During this period, the contribution of the DMS(P)-rich phaeocystis to the chlorophyll concentration was highest in the Bellingshausen Sea (i.e., DMSP to chlorophyll ratio >100 mmol g^{-1}; dominance of phaeocystis >50%; dominance of diatoms <10%; Fig. ~2~ 3, Fig. ~S4a~ S3a and Fig. ~S5~ S4). Conversely, the increase in the chlorophyll concentration was not correlated with the increase in the number concentration of nanoparticles in the Weddell Sea (Fig. ~4b~ 5b). As a consequence, the higher occurrence

25 of nanoparticles from the Bellingshausen Sea inferred from our analysis was likely to be associated with a higher abundance of DMS(P)-rich phytoplankton, whereas the lower occurrence of nanoparticles from the Weddell Sea appeared to be associated with a higher abundance of DMS(P)-poor phytoplankton.

4 Conclusions

30 The physical properties of aerosol particles measured above the remote Antarctic Peninsula over 8 years were analyzed in conjunction with the satellite-derived biological characteristics around the observation site. These results show that the formation of nanoparticles was strongly associated not only with the biomass of phytoplankton but, more importantly, also its taxonomic composition in the Antarctic Ocean. Previous studies have reported that diatoms have a competitive advantage under conditions in which the mixed layers are shallow

35 and the light levels are relatively high. Conversely, phaeocystis is well adapted to conditions in which mixed layers are deep and light levels are variable (e.g., Weber and El-Sayed, 1987; Arrigo et al., 1999; Goffart et al., 2000; Alvain et al., 2008; Arrigo et al., 2010). These results are consistent with the distribution of phytoplankton

메모 포함[KTP26]: (response to referee #2)
Q3. Typical lifetime of PM is generally longer than 2 days: We agree with referee #2 that the retention time of PM$_{10}$ is known to be >2 days in the lower troposphere. Therefore, we have applied 3-, 4- and 5-day air mass back trajectories to identify the potential origin of MSA.

메모 포함[KTP27]: (response to referee #2)
Q3. Typical lifetime of PM is generally longer than 2 days: We have estimated the MSA concentration originated from Bellingshausen Sea sector by applying 3-, 4- and 5-day air mass back trajectories.

메모 포함[KTP28]: (response to referee #2)
Q3. Typical lifetime of PM is generally longer than 2 days: We have estimated the MSA concentration originated from Weddell Sea sector by applying 3-, 4- and 5-day air mass back trajectories.

groups in the Bellingshausen and Weddell seas. Given that the mixed layer depth in the Bellingshausen Sea (45.6 ± 4.1 m) was relatively deeper than that of the Weddell Sea (36.2 ± 3.8 m; Fig. S6) during the austral summer period, the growth of DMS(P)-rich phaeocystis may therefore be more favorable in the Bellingshausen Sea. Sea-surface warming and freshening is commonly associated with a shallowing of the mixed layer depth (Capotondi et al., 2012). The warming trend has shown the spatial complexity across the Antarctic Ocean in recent decades (Turner et al., 2005). Therefore, all regions of the Antarctic Ocean will experience different changes in phytoplankton productivity and taxonomic composition in response to the climate change (Deppeler and Davidson, 2017). Changes in the marine ecosystem could significantly influence the formation of aerosols and clouds in the remote marine atmosphere (Wang et al., 2018). Continuous measurements of the physiochemical properties of aerosols and key nucleation compounds are needed to improve our knowledge regarding the association between marine biota and the formation of aerosol particles and to understand their climate feedback roles in the remote marine environment.

In this study, we have focused on the relationship between the formation of nanoparticles and marine biota. The formation of secondary aerosols contributes significantly to the atmospheric aerosol number and accounts for half of the global cloud condensation nuclei (Merikanto et al., 2009; Sullivan et al., 2018). Our results indicate that changes in the taxonomic composition of marine phytoplankton (i.e., DMS(P)-rich species vs. DMS(P)-poor species) could have a significant impact on the aerosol properties in the remote marine environment. Precursors other than biogenic DMS could play a key role in the formation of new particles in the Antarctic atmosphere. In fact, 16 days of new particle formation events out of 101 events were observed when the air mass originated from the Antarctic Peninsula during the study period. Penguin colonies are dispersed throughout the Antarctic Peninsula (Croxall et al., 2002), and the emission of ammonia from these colonies could trigger the formation of nanoparticles (Weber et al., 1998; Croft et al., 2016). Moreover, iodine molecules produced by biotic and abiotic processes near sea-ice region are known to influence the formation of aerosol particles (Allan et al., 2015; Sipilä et al., 2016). Future studies are required to minimize the knowledge gaps related to multiple precursors and their source origins. Specifically, continuous measurements of the physiochemical properties of aerosol particles and molecular-scale measurements of chemical species (e.g., sulfur-, nitrogen-, and halogen-containing compounds) involved in nucleation processes are required to provide direct evidence for the contribution of these compounds to the formation and growth of aerosol particles and to understand their climate feedback roles in the remote marine environment.

Author contributions

KTP, JEH, JSK, TWK and YYJ designed the study. YYJ and YTG analyzed the physical properties of the aerosol particles. HSB, SB and RT operated the air sampler and analyzed the MSA. KTP and JEH wrote the manuscript.

Acknowledgements

10

메모 포함[KTP29]: (response to Short comment)
Q1. **Need to add explanation on the roles of iodine compounds in new particle formation events:** we have added a short paragraph indicating the roles of iodine-molecules on the formation of new particles and the limitation of our study.

메모 포함[KTP30]: (response to referee #2)
Q1. Need to provide more comprehensive discussion on how this result build upon earlier findings and what exactly is the new scientific message: In the revised manuscript, we have revised and added a paragraph explaining the key results of this study. Furthermore, the limitations of the present study and the scope of future studies have also been added.

(response to referee #3)
Q1, Q3, and Q5. Present a whole analysis, including air masses originating other sectors: we have a companion paper that describes the physical characteristics of the aerosol particles observed at the King Sejong station during the entire sampling period. The relationship between new particle formation and environmental parameters is complicated, owing to interplay among multiple sources and complicated processes. The number concentration of the nanoparticle was at its maximum during the productive summer period, and the frequency of new particle formation was highest when the air mass originated from the ocean domain. Therefore, we focused on the influence of marine biota on the formation of nanoparticles in this study. In the revised manuscript, we have added more description to clarify the key finding of this study. Furthermore, the limitations of the present study and the scope of future studies have also been added.

References

Alvain, S., Moulin, C., Dandonneau, Y., and Breon, F. M.: Remote sensing of phytoplankton groups in case 1 waters from global SeaWiFS imagery, Deep-Sea Res. Pt. I, 52, 1989–2004, https://doi.org/10.1016/j.dsr.2005.06.015, 2005.

5 Alvain, S., Moulin, C., Dandonneau, Y., and Loisel, H.: Seasonal distribution and succession of dominant phytoplankton groups in the global ocean: A satellite view, Global Biogeochem. Cy., 22, https://doi.org/10.1029/2007gb003154, 2008.

Alvain, S., and d'Ovidio, F.: Phytoplankton diversity in the Southern Ocean: a satellite view, in: Biogeographic Atlas of the Southern Ocean, edited by: Broyer, C. D., Koubbi, P., Griffiths, H. J., Raymond, B., d'Udekem
10 d'Acoz, C., Scientific Committee on Antarctic Research, Cambridge, UK, 260–265, 2014.

Allan, J. D., Williams, P. I., Najera, J., Whitehead, J. D., Flynn, M. J., Taylor, J. W., Liu, D., Darbyshire, E., Carpenter, L. J., Chance, R., Andrews, S. J., Hackenberg, S. C., and McFiggans, G.: Iodine observed in new particle formation events in the Arctic atmosphere during ACCACIA, Atmos. Chem. Phys., 15, 5599–5609, https://doi.org/10.5194/acp-15-5599-2015, 2015.

15 Andreae, M. O.: Ocean-Atmosphere Interactions in the Global Biogeochemical Sulfur Cycle, Mar. Chem., 30, 1–29, https://doi.org/10.1016/0304-4203(90)90059-L, 1990.

Arrigo, K. R., Robinson, D. H., Worthen, D. L., Dunbar, R. B., DiTullio, G. R., VanWoert, M., and Lizotte, M. P.: Phytoplankton community structure and the drawdown of nutrients and CO2 in the southern ocean, Science, 283, 365–367, https://doi.org/10.1126/science.283.5400.365, 1999.

20 Arrigo, K. R., Mills, M. M., Kropuenske, L. R., van Dijken, G. L., Alderkamp, A.-C., and Robinson, D. H.: Photophysiology in two major southern ocean phytoplankton taxa: photosynthesis and growth of Phaeocystis antarctica and Fragilariopsis cylindrus under different irradiance levels, Integr. Comp. Biol., 50, 950–966, https://doi.org/10.1093/icb/icq021, 2010.

Asmi, E., Frey, A., Virkkula, A., Ehn, M., Manninen, H., Timonen, H., Tolonen-Kivimäki, O., Aurela, M., Hillamo,
25 R., and Kulmala, M.: Hygroscopicity and chemical composition of Antarctic sub-micrometre aerosol particles and observations of new particle formation, Atmos. Chem. Phys., 10, 4253–4271, https://doi.org/10.5194/acp-10-4253-2010, 2010.

Asmi, E., Neitola, K., Teinilä, K., Rodriguez, E., Virkkula, A., Backman, J., Bloss, M., Jokela, J., Lihavainen, H., De Leeuw, G., Paatero, J., Aaltonen, V., Mei, M., Gambarte, G., Copes, G., Albertini, M., Fogwill, G. P.,
30 Ferrara, J., Barlasina, M. E., and Sánchez, R.: Primary sources control the variability of aerosol optical properties in the Antarctic Peninsula, Tellus B, 70, 1414571, https://doi.org/10.1080/16000889.2017.1414571, 2018.

Atkinson, H. M., Huang, R.-J., Chance, R., Roscoe, H. K., Hughes, C., Davison, B., Schönhardt, A., Mahajan, A. S., Saiz-Lopez, A., Hoffmann, T., and Liss, P. S.: Iodine emissions from the sea ice of the Weddell Sea,
35 Atmos. Chem. Phys., 12, 11229-11244, https://doi.org/10.5194/acp-12-11229-2012, 2012.

Ayers, G. P., and Gras, J. L.: Seasonal relationship between cloud condensation nuclei and aerosol methanesulphonate in marine air, Nature, 353, 834–835, https://doi.org/10.1038/353834a0, 1991.

11

메모 포함[WU31]: (response to referee #1)
We have added this reference.

메모 포함[WU32]: (response to referee #2, 3 and Short comment)
We have added this reference.

메모 포함[WU33]: (response to referee #2, 3 and Short comment)
We have added this reference.

Becagli, S., Scarchilli, C., Traversi, R., Dayan, U., Severi, M., Frosini, D., Vitale, V., Mazzola, M., Lupi, A., Nava, S., and Udisti, R.: Study of present-day sources and transport processes affecting oxidised sulphur compounds in atmospheric aerosols at Dome C (Antarctica) from year-round sampling campaigns, Atmos. Environ., 52, 98–108, https://doi.org/10.1016/j.atmosenv.2011.07.053, 2012.

5   Belviso, S., Christaki, U., Vidussi, F., Marty, J.-C., Vila, M., and Delgado, M.: Diel variations of the DMSP-to-chlorophyll a ratio in Northwestern Mediterranean surface waters, J. Marine Syst., 25, 119-128, https://doi.org/10.1016/S0924-7963(00)00011-7, 2000.

Beyersdorf, A. J., Blake, D. R., Swanson, A., Meinardi, S., Rowland, F. S., and Davis, D.: Abundances and variability of tropospheric volatile organic compounds at the South Pole and other Antarctic locations, Atmos.
10   Environ., 44, 4565–4574, https://doi.org/10.1016/j.atmosenv.2010.08.025, 2010.

Brooks, S. D., and Thornton, D. C. O.: Marine Aerosols and Clouds, Annu. Rev. Mar. Sci., 10, 289-313, https://doi.org/10.1146/annurev-marine-121916-063148, 2018.

Budhavant, K., Safai, P. D., and Rao, P. S. P.: Sources and elemental composition of summer aerosols in the Larsemann Hills (Antarctica), Environ. Sci. Pollut. R., 22, 2041–2050, https://doi.org/10.1007/s11356-014-
15   3452-0, 2015.

Capotondi, A., Alexander, M. A., Bond, N. A., Curchitser, E. N., and Scott, J. D.: Enhanced upper ocean stratification with climate change in the CMIP3 models, J. Geophys. Res., 117, C04031, https://doi.org/10.1029/2011JC007409, 2012.

Charlson, R. J., Lovelock, J. E., Andreae, M. O., and Warren, S. G.: Oceanic Phytoplankton, Atmospheric Sulfur,
20   Cloud Albedo and Climate, Nature, 326, 655–661, https://doi.org/10.1038/326655a0, 1987.

Clarke, A., Murphy, E. J., Meredith, M. P., King, J. C., Peck, L. S., Barnes, D. K., and Smith, R. C.: Climate change and the marine ecosystem of the western Antarctic Peninsula, Philos. T. Roy. Soc. B, 362, 149–166, https://doi.org/10.1098/rstb.2006.1958, 2007.

Croft, B., Wentworth, G. R., Martin, R. V., Leaitch, W. R., Murphy, J. G., Murphy, B. N., Kodros, J. K., Abbatt, J.
25   P., and Pierce, J. R.: Contribution of Arctic seabird-colony ammonia to atmospheric particles and cloud-albedo radiative effect, Nat. Commun., 7, 13444, https://doi.org/10.1038/ncomms13444, 2016.

Croxall, J. P., Trathan, P. N., and Murphy, E. J.: Environmental change and Antarctic seabird populations, Science, 297, 1510-1514, https://doi.org/10.1126/science.1071987, 2002.

Dall'Osto, M., Ovadnevaite, J., Paglione, M., Beddows, D. C. S., Ceburnis, D., Cree, C., Cortés, P., Zamanillo,
30   M., Nunes, S. O., Pérez, G. L., Ortega-Retuerta, E., Emelianov, M., Vaqué, D., Marrasé, C., Estrada, M., Sala, M. M., Vidal, M., Fitzsimons, M. F., Beale, R., Airs, R., Rinaldi, M., Decesari, S., Facchini, M. C., Harrison, R. M., O'dowd, C., and Simó, R.: Antarctic sea ice region as a source of biogenic organic nitrogen in aerosols, Sci. Rep.-UK, 7, 6047, https://doi.org/10.1038/s41598-017-06188-x, 2017.

Dall'Osto, M., Simó, R., Harrison, R. M., Beddows, D. C. S., Saiz-Lopez, A., Lange, R., Skov, H., Nøjgaard, J.
35   K., Nielsen, I. E., and Massling, A.: Abiotic and biotic sources influencing spring new particle formation in North East Greenland, Atmos. Environ., 190, 126–134, https://doi.org/10.1016/j.atmosenv.2018.07.019, 2018.

12

메모 포함[WU34]: (response to referee #1)
We have added this reference.

메모 포함[WU35]: (response to referee #1)
We have added this reference.

메모 포함[WU36]: (response to referee #2)
We have added this reference.

메모 포함[WU37]: (response to referee #2, 3 and Short comment)
We have added this reference.

메모 포함[WU38]: (response to referee #3)
We have added this reference.

메모 포함[WU39]: (response to referee #2, 3 and Short comment)
We have added this reference.

Deppeler, S. L., and Davidson, A. T.: Southern Ocean phytoplankton in a changing climate, Front. Mar. Sci., 4, 40, https://doi.org/10.3389/fmars.2017.00040, 2017.

Draxler, R. R., and Hess, G. D.: An overview of the HYSPLIT_4 modelling system for trajectories, Aust. Meteorol. Mag., 47, 295–308, 1998.

5 Galí, M., Devred, E., Levasseur, M., Royer, S.-J., and Babin, M.: A remote sensing algorithm for planktonic dimethylsulfoniopropionate (DMSP) and an analysis of global patterns, Remote Sens. Environ., 171, 171–184, https://doi.org/10.1016/j.rse.2015.10.012, 2015.

Gibson, J. A. E., Garrick, R. C., Burton, H. R., and McTaggart, A. R.: Dimethylsulfide and the algaPhaeocystis pouchetii in antarctic coastal waters, Mar. Biol., 104, 339–346, https://doi.org/10.1007/BF01313276, 1990.

10 Giordano, M. R., Kalnajs, L. E., Avery, A., Goetz, J. D., Davis, S. M., and DeCarlo, P. F.: A missing source of aerosols in Antarctica–beyond long-range transport, phytoplankton, and photochemistry, Atmos. Chem. Phys., 17, 1–20, https://doi.org/10.5194/acp-17-1-2017, 2017.

Goffart, A., Catalano, G., and Hecq, J. H.: Factors controlling the distribution of diatoms and *Phaeocystis* in the Ross Sea, J. Marine Syst., 27, 161–175, https://doi.org/10.1016/S0924-7963(00)00065-8, 2000.

15 Haëntjens, N., Boss, E., and Talley, L. D.: Revisiting Ocean Color algorithms for chlorophyll a and particulate organic carbon in the Southern Ocean using biogeochemical floats, J. Geophys. Res.-Oceans, 122, 6583–6593, https://doi.org/10.1002/2017jc012844, 2017.

Hatton, A. D., and Wilson, S. T.: Particulate dimethylsulphoxide and dimethylsulphoniopropionate in phytoplankton cultures and Scottish coastal waters, Aquat. Sci., 69, 330–340, 20 https://doi.org/10.1007/s00027-007-0891-4, 2007.

IPCC: Climate change 2013: The physical science basis, Intergovernmental panel on Climate Change, Cambridge University Press, New York, USA, 571–740, 2013.

Ito, T.: Antarctic submicron aerosols and long-range transport of pollutants, Ambio, 34–41, 1989.

Järvinen, E., Virkkula, A., Nieminen, T., Aalto, P. P., Asmi, E., Lanconelli, C., Busetto, M., Lupi, A., Schioppo, 25 R., Vitale, V., Mazzola, M., Petäjä, T., Kerminen, V.-M., and Kulmala, M.: Seasonal cycle and modal structure of particle number size distribution at Dome C, Antarctica, Atmos. Chem. Phys., 13, 7473–7487, https://doi.org/10.5194/acp-13-7473-2013, 2013.

Jimenez, J. L., Canagaratna, M. R., Donahue, N. M., Prevot, A. S., Zhang, Q., Kroll, J. H., DeCarlo, P. F., Allan, J. D., Coe, H., Ng, N. L., Aiken, A. C., Docherty, K. S., Ulbrich, I. M., Grieshop, A. P., Robinson, A. L., 30 Duplissy, J., Smith, J. D., Wilson, K. R., Lanz, V. A., Hueglin, C., Sun, Y. L., Tian, J., Laaksonen, A., Raatikainen, T., Rautiainen, J., Vaattovaara, P., Ehn, M., Kulmala, M., Tomlinson, J. M., Collins, D. R., Cubison, M. J., Dunlea, E. J., Huffman, J. A., Onasch, T. B., Alfarra, M. R., Williams, P. I., Bower, K., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Salcedo, D., Cottrell, L., Griffin, R., Takami, A., Miyoshi, T., Hatakeyama, S., Shimono, A., Sun, J. Y., Zhang, Y. M., Dzepina, K., Kimmel, 35 J. R., Sueper, D., Jayne, J. T., Herndon, S. C., Trimborn, A. M., Williams, L. R., Wood, E. C., Middlebrook, A. M., Kolb, C. E., Baltensperger, U., and Worsnop, D. R.: Evolution of organic aerosols in the atmosphere, Science, 326, 1525–1529, https://doi.org/10.1126/science.1180353, 2009.

Jokinen, T., Sipilä, M., Kontkanen, J., Vakkari, V., Tisler, P., Duplissy, E.-M., Junninen, H., Kangasluoma, J., Manninen, H. E., Petäjä, T., Kulmala, M., Worsnop, D. R., Kirkby, J., Virkkula, A., and Kerminen, V.-M.:

13

Ion-induced sulfuric acid–ammonia nucleation drives particle formation in coastal Antarctica, Sci. Adv., 4, eaat9744, https://doi.org/10.1126/sciadv.aat9744, 2018.

Keller, M. D., Bellows, W. K., and Guillard, R. R. L.: Dimethyl sulfide production in marine phytoplankton, in: Biogenic sulfur in the environment, ACS Symposium Series, 393, edited by: Saltzman E. S., and Cooper W. J., American Chemical Society, Washington, DC, USA, 167–182, 1989.

Kerminen, V.-M., Chen, X., Vakkari, V., Petäjä, T., Kulmala, M., and Bianchi, F.: Atmospheric new particle formation and growth: review of field observations, Environ. Res. Lett., 13, 103003, https://doi.org/10.1088/1748-9326/aadf3c, 2018.

Kim, J., Yoon, Y. J., Gim, Y., Kang, H. J., Choi, J. H., Park, K.-T., and Lee, B. Y.: Seasonal variations in physical characteristics of aerosol particles at the King Sejong Station, Antarctic Peninsula, Atmos. Chem. Phys., 17, 12985–12999, https://doi.org/10.5194/acp-17-12985-2017, 2017.

Kim, J., Yoon, Y. J., Gim, Y., Choi, J. H., Kang, H. J., Park, K.-T., Park, J., and Lee, B. Y.: New particle formation events observed at the King Sejong Station, Antarctic Peninsula – Part 1: Physical characteristics and contribution to cloud condensation nuclei, ~~Manuscript submitted for publication,~~ Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1180, in review, 2018.

Kim, T.-W., Lee, K., Najjar, R. G., Jeong, H.-D., and Jeong, H. J.: Increasing N abundance in the northwestern Pacific Ocean due to atmospheric nitrogen deposition, Science, 334, 505–509, https://doi.org/10.1126/science.1206583, 2011.

Kirkby, J., Duplissy, J., Sengupta, K., Frege, C., Gordon, H., Williamson, C., Heinritzi, M., Simon, M., Yan, C., Almeida, J., Tröstl, J., Nieminen, T., Ortega, I. K., Wagner, R., Adamov, A., Amorim, A., Bernhammer, A. – K., Bianchi, F., Breitenlechner, M., Brilke, S., Chen, X., Craven, J., Dias, A., Ehrhart, S., Flagan, R. C., Franchin, A., Fuchs, C., Guida, R., Hakala, J., Hoyle, C. R., Jokinen, T., Junninen, H., Kangasluoma, J., Kim, J., Krapf, M., Kürten, A., Laaksonen, A., Lehtipalo, K., Makhmutov, V., Mathot, S., Molteni, U., Onnela, A., Peräkylä, O., Piel, F., Petäjä, T., Praplan, A. P., Pringle, K., Rap, A., Richards, N. A. D., Riipinen, I., Rissanen, M. P., Rondo, L., Sarnela, L., Schobesberger, S., Scott, C. E., Seinfeld, J. H., Sipilä, M., Steiner, G., Stozhkov, Y., Stratmann, F., Tomé, A., Virtanen, A., Vogel, A. L., Wagner, A. C., Wagner, P. E., Weingartner, E., Wimmer, D., Winkler, P. M., Ye, P., Zhang, X., Hansel, A., Dommen, J., Donahue, N. M., Worsnop, D. R., Baltensperger, U., Kulmala, M., Carslaw, K. S., and Curtius, J.: Ion-induced nucleation of pure biogenic particles, Nature, 533, 521, https://doi.org/10.1038/nature17953, 2016.

Korhonen, P., Kulmala, M., Laaksonen, A., Viisanen, Y., McGraw, R., and Seinfeld, J. H.: Ternary nucleation of H_2SO_4, NH_3, and H_2O in the atmosphere, J. Geophys. Res.-Atmos., 104, 26349–26353, https://doi.org/10.1029/1999JD900784, 1999.

Kropuenske, L. R., Mills, M. M., van Dijken, G. L., Bailey, S., Robinson, D. H., Welschmeyer, N. A., and Arrigo, K. R.: Photophysiology in two major Southern Ocean phytoplankton taxa: Photoprotection in Phaeocystis antarctica and Fragilariopsis cylindrus, Limnol. Oceanogr., 54, 1176–1196, https://doi.org/10.4319/lo.2009.54.4.1176, 2009.

Kulmala, M., and Laaksonen, A.: Binary nucleation of waer-sulfuric acid system: Comparison of classical theories with different H_2SO_4 saturation vapor pressures, J. Chem. Phys., 93, 696–701,

https://doi.org/10.1063/1.459519, 1990.

Kyrö, E.-M., Kerminen, V.-M., Virkkula, A., Dal Maso, M., Parshintsev, J., Ruíz-Jimenez, J., Forsström, L., Manninen, H. E., Riekkola, M.-L., Heinonen, P., and Kulmala, M.: Antarctic new particle formation from continental biogenic precursors, Atmos. Chem. Phys., 13, 3527–3546, https://doi.org/10.5194/acp-13-3527-2013, 2013.

Lana, A., Bell, T. G., Simó, R., Vallina, S. M., Ballabrera-Poy, J., Kettle, A. J., Dachs, J., Bopp, L., Saltzman, E. S., Stefels, J., Johnson, J. E., and Liss, P. S.: An updated climatology of surface dimethlysulfide concentrations and emission fluxes in the global ocean, Global Biogeochem. Cy., 25, https://doi.org/10.1029/2010gb003850, 2011.

Laturnus, F., Wiencke, C., and Klöser, H.: Antarctic macroalgae—sources of volatile halogenated organic compounds, Mar. Environ. Res., 41, 169–181, https://doi.org/10.1016/0141-1136(95)00017-8, 1996.

Leck, C., and Bigg, E. K.: Source and evolution of the marine aerosol—A new perspective, Geophys. Res. Lett., 32, https://doi.org/10.1029/2005GL023651, 2005.

Lee, S.-H., Reeves, J. M., Wilson, J. C., Hunton, D. E., Viggiano, A. A., Miller, T. M., Ballenthin, J. O., and Lait, L. R.: Particle formation by ion nucleation in the upper troposphere and lower stratosphere, Science, 301, 1886–1889, https://doi.org/10.1126/science.1087236, 2003.

Lovejoy, E. R., Curtius, J., and Froyd, K. D.: Atmospheric ion-induced nucleation of sulfuric acid and water, J. Geophys. Res.-Atmos., 109, https://doi.org/10.1029/2003JD004460, 2004.

Malviya, S., Scalco, E., Audic, S., Vincent, F., Veluchamy, A., Poulain, J., Wincker, P., Iudicone, D., de Vargas, C., Bittner, L., Zingone, A., and Bowler, C.: Insights into global diatom distribution and diversity in the world's ocean, P. Natl. Acad. Sci. USA, 113, E1516–E1525, https://doi.org/10.1073/pnas.1509523113, 2016.

Merikanto, J., Spracklen, D. V., Mann, G. W., Pickering, S. J., and Carslaw, K. S.: Impact of nucleation on global CCN, Atmos. Chem. Phys., 9, 8601–8616, https://doi.org/10.5194/acp-9-8601-2009, 2009.

Minikin, A., Legrand, M., Hall, J., Wagenbach, D., Kleefeld, C., Wolff, E., Pasteur, E. C., and Ducroz, F.: Sulfur-containing species (sulfate and methanesulfonate) in coastal Antarctic aerosol and precipitation, J. Geophys. Res.-Atmos., 103, 10975–10990, https://doi.org/10.1029/98JD00249, 1998.

Mishra, V. K., Kim, K.-H., Hong, S., and Lee, K.: Aerosol composition and its sources at the King Sejong Station, Antarctic peninsula, Atmos. Environ., 38, 4069–4084, https://doi.org/10.1016/j.atmosenv.2004.03.052, 2004.

Mustapha, Z. B., Alvain, S., Jamet, C., Loisel, H., and Dessailly, D.: Automatic classification of water-leaving radiance anomalies from global SeaWiFS imagery: application to the detection of phytoplankton groups in open ocean waters, Remote Sens. Environ., 146, 97–112, https://doi.org/10.1016/j.rse.2013.08.046, 2014.

O'Dowd, C. D., Facchini, M. C., Cavalli, F., Ceburnis, D., Mircea, M., Decesari, S., Fuzzi, S., Yoon, Y. J., and Putaud, J.-P.: Biogenically driven organic contribution to marine aerosol, Nature, 431, 676–680, https://doi.org/10.1038/nature02959, 2004.

O'Dowd, C., and De Leeuw, G.: Marine aerosol production: a review of the current knowledge, Philos. T. Roy. Soc. A, 365, 1753–1774, https://doi.org/10.1098/rsta.2007.2043, 2007.

Park, J., Park, T., Yang, E. J., Kim, D., Gorbunov, M. Y., Kim, H.-C., Kang, S. H., Shin, H. C., Lee, S., and Yoo, S.: Early summer iron limitation of phytoplankton photosynthesis in the Scotia Sea as inferred from fast repetition rate fluorometry, J. Geophys. Res.-Oceans, 118, 3795–3806, https://doi.org/10.1002/jgrc.20281,

2013a.

Park, K.-T., Lee, K., Yoon, Y.-J., Lee, H.-W., Kim, H.-C., Lee, B.-Y., Hermansen, O., Kim, T.-W., and Holmén, K.: Linking atmospheric dimethyl sulfide and the Arctic Ocean spring bloom, Geophys. Res. Lett., 40, 155–160, https://doi.org/10.1029/2012GL054560, 2013b.

5   Park, K.-T., Lee, K., Shin, K., Yang, E. J., Hyun, B., Kim, J. M., Noh, J. H., Kim, M., Kong, B., Choi, D. H., Choi, S.-J., Jang, P.-G., and Jeong, H. J.: Direct Linkage between Dimethyl Sulfide Production and Microzooplankton Grazing, Resulting from Prey Composition Change under High Partial Pressure of Carbon Dioxide Conditions, Environ. Sci. Technol., 48, 4750–4756, https://doi.org/10.1021/es403351h, 2014a.

Park, K.-T., Lee, K., Shin, K., Jeong, H. J., and Kim, K. Y.: Improved method for minimizing sulfur loss in analysis
10   of particulate organic sulfur, Anal. Chem., 86, 1352–1356, https://doi.org/10.1021/ac403649m, 2014b.

Park, K.-T., Jang, S., Lee, K., Yoon, Y. J., Kim, M.-S., Park, K., Cho, H.-J., Kang, J.-H., Udisti, R., Lee, B.-Y., and Shin, K.-H.: Observational evidence for the formation of DMS-derived aerosols during Arctic phytoplankton blooms, Atmos. Chem. Phys., 17, 9665–9675, https://doi.org/10.5194/acp-17-9665-2017, 2017.

15   Park, K.-T., Lee, K., Kim, T.-W., Yoon, Y. J., Jang, E.-H., Jang, S., Lee, B.-Y., and Hermansen, O.: Atmospheric DMS in the Arctic Ocean and its relation to phytoplankton biomass. Global Biogeochem. Cy., 32, 351–359. https://doi.org/10.1002/2017GB005805, 2018.

Polimene, L., Archer, S. D., Butenschön, M., and Allen, J. I.: A mechanistic explanation of the Sargasso Sea DMS "summer paradox", Biogeochemistry, 110, 243–255, https://doi.org/10.1007/s10533-011-9674-z, 2012.

20   Preunkert, S., Legrand, M., Jourdain, B., Moulin, C., Belviso, S., Kasamatsu, N., Fukuchi, M., and Hirawake, T.: Interannual variability of dimethylsulfide in air and seawater and its atmospheric oxidation by-products (methanesulfonate and sulfate) at Dumont d'Urville, coastal Antarctica (1999–2003), J. Geophys. Res.-Atmos., 112, https://doi.org/10.1029/2006JD007585, 2007.

Preunkert, S., Jourdain, B., Legrand, M., Udisti, R., Becagli, S., and Cerri, O.: Seasonality of sulfur species
25   (dimethyl sulfide, sulfate, and methanesulfonate) in Antarctica: Inland versus coastal regions, J. Geophys. Res.- Atmos., 113, https://doi.org/10.1029/2008JD009937, 2008.

Prospero, J. M., Savoie, D. L., Saltzman, E. S., and Larsen, R.: Impact of Oceanic Sources of Biogenic Sulfur on Sulfate Aerosol Concentrations at Mawson, Antarctica, Nature, 350, 221–223, https://doi.org/10.1038/350221a0, 1991.

30   Read, K. A., Lewis, A. C., Bauguitte, S., Rankin, A. M., Salmon, R. A., Wolff, E. W., Saiz-Lopez, A., Bloss, W. J., Heard, D. E., Lee, J. D., and Plane, J. M. C.: DMS and MSA measurements in the Antarctic Boundary Layer: Impact of BrO on MSA production, Atmos. Chem. Phys., 8, 2985–2997, https://doi.org/10.5194/acp-8-2985-2008, 2008.

Reisch, C. R., Moran, M. A., and Whitman, W. B.: Bacterial catabolism of dimethylsulfoniopropionate (DMSP),
35   Front. Microbiol., 2, 172, https://doi.org/10.3389/fmicb.2011.00172, 2011.

Savoie, D. L., Prospero, J. M., Larsen, R. J., Huang, F., Izaguirre, M. A., Huang, T., Snowdon, T. H., Custals, L., and Sanderson, C. G.: Nitrogen and Sulfur Species in Antarctic Aerosols at Mawson, Palmer Station, and Marsh (King George Island), J. Atmos. Chem., 17, 95–122, https://doi.org/10.1007/BF00702821, 1993.

16

메모 포함[WU45]: (response to referee #1)
We have added this reference.

메모 포함[WU46]: (response to referee #1)
We have added this reference.

메모 포함[WU47]: (response to referee #1)
We have added this reference.

Schoemann, V., Becquevort, S., Stefels, J., Rousseau, W., and Lancelot, C.: Phaeocystis blooms in the global ocean and their controlling mechanisms: a review, J. Sea Res., 53, 43–66, https://doi.org/10.1016/j.seares.2004.01.008, 2005.

Sedwick, P. N., Garcia, N. S., Riseman, S. F., Marsay, C. M., and DiTullio, G. R.: Evidence for high iron requirements of colonial Phaeocystis antarctica at low irradiance, Biogeochemistry, 83, 83–97, https://doi.org/10.1007/s10533-007-9081-7, 2007.

Shiraiwa, M., Li, Y., Tsimpidi, A. P., Karydis, V. A., Berkemeier, T., Pandis, S. N., Lelieveld, J., Koop, T., and Pöschl, U.: Global distribution of particle phase state in atmospheric secondary organic aerosols, Nat. Commun., 8, 15002, https://doi.org/10.1038/ncomms15002, 2017.

Siegel, D. A., Behrenfeld, M. J., Maritorena, S., McClain, C. R., Antoine, D., Bailey, S. W., Bontempi, P. S., Boss, E. S., Dierssen, H. M., Doney, S. C., Eplee Jr., R. E., Evans, R. H., Feldman, G. C., Fields, E., Franz, B. A., Kuring, N. A., Mengelt, C., Nelson, N. B., Patt, F. S., Robinson, W. D., Sarmiento, J. L., Swan, C. M., Werdell, P. J., Westberry, T. K., Wilding, J. G., and Yoder, J. A.: Regional to global assessments of phytoplankton dynamics from the SeaWiFS mission, Remote Sens. Environ., 135, 77–91, https://doi.org/10.1016/j.rse.2013.03.025, 2013.

Simó, R.: Production of atmospheric sulfur by oceanic plankton: biogeochemical, ecological and evolutionary links, Trends. Ecol. Evol., 16, 287–294, https://doi.org/10.1016/S0169-5347(01)02152-8, 2001.

Sipilä, M., Sarnela, N., Jokinen, T., Henschel, H., Junninen, H., Kontkanen, J., Richters, S., Kangasluoma, J., Franchin, A., Peräkylä, O., Rissanen, M. P., Ehn, M., Vehkamäki, H., Kurten, T., Berndt, T., Petäjä, T., Worsnop, D., Ceburnis, D., Kerminen, V.-M., Kulmala, M., and O'Dowd, C.: Molecular-scale evidence of aerosol particle formation via sequential addition of HIO 3, Nature, 537, 532, https://doi.org/10.1038/nature19314, 2016.

Stefels, J., Steinke, M., Turner, S., Malin, G., and Belviso, S.: Environmental constraints on the production and removal of the climatically active gas dimethylsulphide (DMS) and implications for ecosystem modelling, Biogeochemistry, 83, 245-275, https://doi.org/10.1007/s10533-007-9091-5, 2007.

Sullivan, C. W., Arrigo, K. R., McClain, C. R., Comiso, J. C., and Firestone, J.: Distributions of phytoplankton blooms in the southern ocean, Science, 262, 1832–1837, https://doi.org/10.1126/science.262.5141.1832, 1993.

Sullivan, R. C., Crippa, P., Matsui, H., Leung, L. R., Zhao, C., Thota, A., and Pryor, S. C.: New particle formation leads to cloud dimming, npj Climate and Atmospheric Science, 1, 9, https://doi.org/10.1038/s41612-018-0019-7, 2018.

Tison, J.-L., Brabant, F., Dumont, I., and Stefels, J.: High-resolution dimethyl sulfide and dimethylsulfoniopropionate time series profiles in decaying summer first-year sea ice at Ice Station Polarstern, western Weddell Sea, Antarctica, J. Geophys. Res.-Biogeo., 115, https://doi.org/10.1029/2010JG001427, 2010.

Todd, J. D., Rogers, R., Li, Y. G., Wexler, M., Bond, P. L., Sun, L., Curson, A. R. J., Malin, G., Steinke, M., and Johnston, A. W. B.: Structural and regulatory genes required to make the gas dimethyl sulfide in bacteria, Science, 315, 666–669, https://doi.org/10.1126/science.1135370, 2007.

17

Turner, J., Colwell, S. R., Marshall, G. J., Lachlan-Cope, T. A., Carleton, A. M., Jones, P. D., Lagun, V., Reid, P. A., and Iagovkina, S.: Antarctic climate change during the last 50 years, Int. J. Climatol., 25, 279–294, https://doi.org/10.1002/joc.1130, 2005.

Wang, S., Maltrud, M. E., Burrows, S. M., Elliott, S. M., and Cameron-Smith, P.: Impacts of shifts in phytoplankton community on clouds and climate via the sulfur cycle, Global Biogeochem. Cy., 32, 1005–1026, https://doi.org/10.1029/2017GB005862, 2018.

Weber, L. H., and El-Sayed, S. Z.: Contributions of the net, nano- and picoplankton to the phytoplankton standing crop and primary productivity in the Southern Ocean, J. Plankton Res., 9, 973–994, https://doi.org/10.1093/plankt/9.5.973, 1987.

Weber, R. J., McMurry, P. H., Mauldin, L., Tanner, D. J., Eisele, F. L., Brechtel, F. J., Kreidenweis, S. M., Kok, G. L., Schillawski, R. D., and Baumgardner, D.: A study of new particle formation and growth involving biogenic and trace gas species measured during ACE 1, J. Geophys. Res.-Atmos., 103, 16385–16396, https://doi.org/10.1029/97JD02465, 1998.

Weller, R., Schmidt, K., Teinilä, K., and Hillamo, R.: Natural new particle formation at the coastal Antarctic site Neumayer, Atmos. Chem. Phys., 15, 11399–11410, https://doi.org/10.5194/acp-15-11399-2015, 2015.

Yu, F. Q., and Luo, G.: Oceanic Dimethyl Sulfide Emission and New Particle Formation around the Coast of Antarctica: A Modeling Study of Seasonal Variations and Comparison with Measurements, Atmosphere-Basel, 1, 34–50, https://doi.org/10.3390/atmos1010034, 2010.

Zhang, M., Chen, L., Xu, G., Lin, Q., and Liang, M.: Linking phytoplankton activity in polynyas and sulfur aerosols over Zhongshan Station, East Antarctica, J. Atmos. Sci., 72, 4629–4642, https://doi.org/10.1175/JAS-D-15-0094.1, 2015.

메모 포함[WU52]: (response to referee #2, 3 and Short comment) We have added this reference.

메모 포함[WU53]: (response to referee #1) We have added this reference.

18
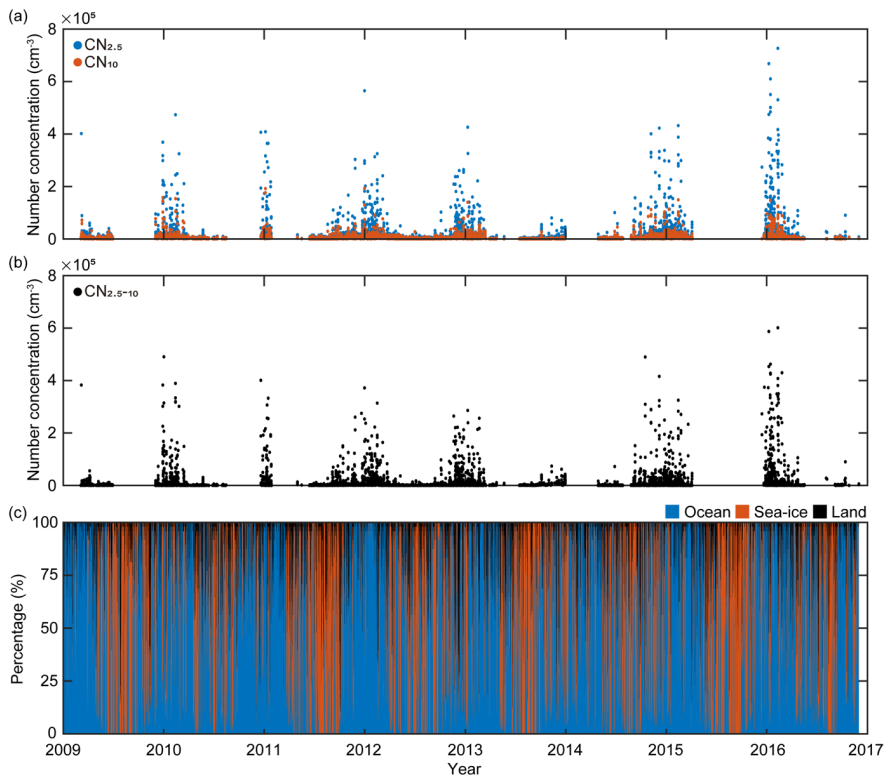
Figure 1: (a) Hourly variations in the number concentration of particles >2.5 nm in diameter ($CN_{2.5}$, blue symbols) and the number concentration of particles >10 nm in diameter (CN_{10}, red symbols), (b) hourly variations in the number concentration of nanoparticles (ranging from 2.5 to 10 nm in diameter) calculated using the differences between $CN_{2.5}$ and CN_{10}, and (c) hourly variations in the retention time of 2-day air mass back trajectories over the three domains including ocean (blue), sea-ice (red), and land (black) domains from March 2009 to November 2016.

메모 포함[WU54]: (response to referee #3)
We have added new figure that represent the 8-year nanoparticle measurement and corresponding air mass transport history.
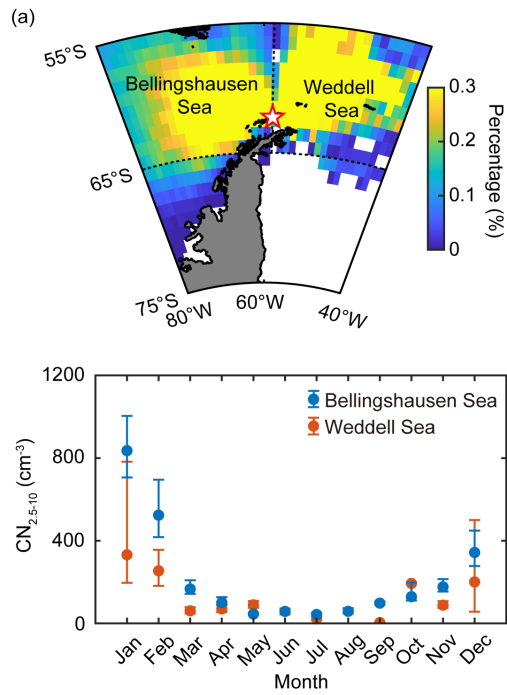
Figure 1 2: (a) Back-trajectories of the air masses arriving at King Sejong Station (62.2ºS, 58.8ºW; star symbol), Antarctic Peninsula. The colors indicate the percentage (%) of the air mass located at that spot during the 2 days prior to arriving at the observation site. Note that the air mass back-trajectories that did not have >90% retention in the two selected ocean domains (i.e., Bellingshausen and Weddell Seas) were excluded. (b) Seasonal variation of nanoparticles (2.5–10 nm in diameter, $CN_{2.5-10}$) observed at King Sejong Station between March 2009 and December 2016. Blue and red symbols indicate the number concentration of nanoparticles that originated from the Bellingshausen and Weddell seas, respectively. The error bars indicate the 95% confidence interval estimated by bootstrap method from the monthly $CN_{2.5-10}$ data.
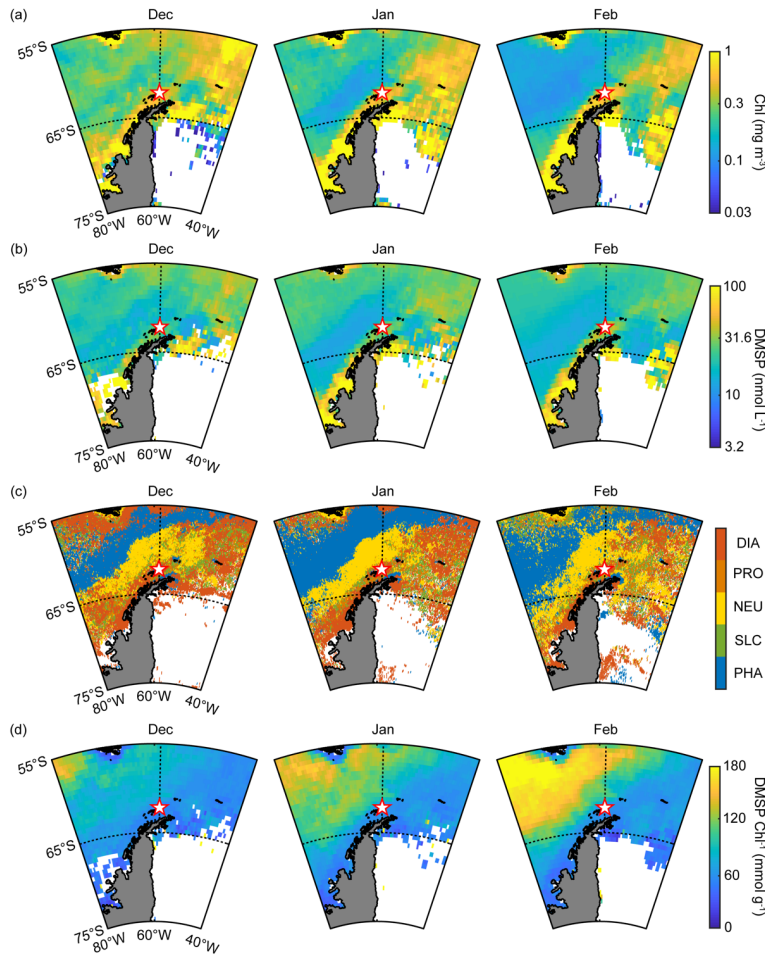
Figure 2 3: (a) Monthly mean chlorophyll concentration during the months of December, January, and February in 2009–2016, (b) monthly mean DMSP concentration during the months of December, January, and February in 2009–2016, (b c) phytoplankton taxonomic composition including diatoms (DIA), Prochlorococcus (PRO), nanoeucaryotes (NEU), Synechococcus (SLC) and phaeocystis (PHA) during the months of December, January and February in 1997 2010 estimated using the PHYSAT method with the climatology map obtained from SeaWiFS archive, and (e d) monthly mean DMSP to chlorophyll ratio during the months of December, January, and February in 2009–2016. Note that the phytoplankton taxonomic composition was estimated using the SeaWiFS (Sea-viewing Wide Field-of-View Sensor) ocean color sensor which is available from September 1997 to December 2010.

Figure 4: (a) Daily concentration of MSA collected at the sampling site during the summer periods in 2013 and 2014 (explicitly, from 14 January to 28 February in 2013, and from 2 December 2013 to 18 January 2014). (b) The mean MSA concentration that potentially originated from the Bellingshausen Sea sector (<58.8ºW) and the Weddell Sea sector (>58.8ºW) estimated by applying 3-, 4-, and 5-day air mass back trajectories during the austral summer periods in 2013 and 2014. The error bars indicate 1 standard deviation (1σ) from the mean values.
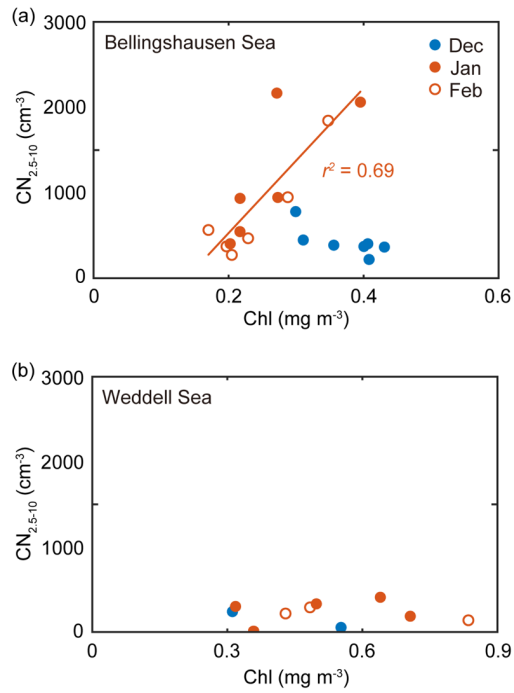
Figure 4 5: (a) Relationship between the monthly mean chlorophyll concentration for the Bellingshausen Sea (55°S–65°S, 60°W–80°W) and the monthly mean number concentration of nanoparticles that originated from the Bellingshausen Sea in 2009–2016. (b) Relationship between the monthly mean chlorophyll concentration for the Weddell Sea (55°S–65°S, 40°W–60°W) and the monthly mean number concentration of nanoparticles that originated from the Weddell Sea in 2009–2016. The filled blue, filled red and open red symbols indicate the data obtained in December, January and February, respectively. The solid lines represent the best fit.

23

Supplement for

New particle formation events observed at the King Sejong Station, Antarctic Peninsula – Part 2: Link with the oceanic biological activities

Eunho Jang et al.

Correspondence to: Ki-Tae Park (ktpark@kopri.re.kr)

Calculation of the sea surface DMSP concentration

~~We estimated the total dimethylsulfoniopropionate (DMSPt) concentration in the sea-surface using the algorithm developed by Galí et al. (2015). The DMSPt algorithm was implemented using the link between DMSPt and the chlorophyll concentration depending on the light exposure regime of the marine phytoplankton.~~ Several studies reported that DMSP and DMS were strongly linked to with several environmental parameters such as solar radiation, sea-surface temperature, and mixing state of the sea-surface (Vallina and Simo, 2007). *Gali et al.* (2015) developed a DMSP algorithm based on satellite-derived chlorophyll (to measure phytoplankton biomass) and the light exposure regime (to measure key environmental factors controlling DMSP production; i.e., solar radiation and mixing state). In this algorithm, euphotic layer depth (Z_{eu}) and mixed layer depth (MLD) dataset were applied to establish a mixing state of the sea surface (stratified vs. mixed water column), and the variability in modeled and measured DMSP was improved by adding sea-surface temperature and $\log_{10}(Z_{eu}/MLD)$ as predictors for the stratified and mixed subsets in the proposed algorithm. Additionally, a sub-model based on particulate inorganic carbon (PIC) was developed to complement DMSPt diagnosis in coccolithophore blooms, where satellite chlorophyll concentration may not be reliable. The database was divided into three subsets including 'stratified water ($Z_{eu}/MLD > 1$)', 'mixed water ($Z_{eu}/MLD < 1$)' and 'undefined water (Z_{eu} or MLD is unavailable)' based on the ratio between the euphotic layer depth (Z_{eu}) and the mixed layer depth (MLD). The $DMSP_t$ concentrations in stratified, mixed and undefined water were calculated using Equations (S1), (S2) and (S3), respectively:

$$\text{Log}_{10}DMSP_t = 1.70 + 1.14\log_{10}Chl_t + 0.44\log_{10}Chl_t^2 + 0.063SST - 0.0024SST^2 \tag{S1}$$
$$\text{Log}_{10}DMSP_t = 1.74 + 0.81\log_{10}Chl_t + 0.60\log_{10}(Z_{eu}/MLD) \tag{S2}$$
$$\log_{10}DMSP_t = -1.052 - 3.185\log_{10}PIC - 0.783(\log_{10}PIC)^2 \tag{S3}$$

The level-3 product of the Moderate Resolution Imaging Spectroradiometer on the Aqua (MODIS-Aqua) satellites was used for the chlorophyll concentration (Chl_t), sea surface temperature at nighttime (SST) and the calcite concentration (PIC). The monthly mixed layer depth (MLD) was retrieved by Monthly Isopycnal and Mixed-layer Ocean Climatology (MIMOC) at a resolution of 0.5°. All of the MODIS-Aqua products at a resolution of 4 km were averaged onto a 0.5° interval grid of MIMOC climatology to run the DMSPt algorithm. The euphotic layer depth (Z_{eu}) was calculated using satellite-derived chlorophyll data as shown in Equation (S4) (Morel et al., 2007).

$$\log_{10}Z_{eu} = 1.524 - 0.436\log_{10}Chl_t - 0.0145(\log_{10}Chl_t)^2 + 0.0186(\log_{10}Chl_t)^3 \tag{S4}$$

References

Galí, M., Devred, E., Levasseur, M., Royer, S.-J., and Babin, M.: A remote sensing algorithm for planktonic dimethylsulfoniopropionate (DMSP) and an analysis of global patterns, Remote Sens. Environ., 171, 171–184, https://doi.org/10.1016/j.rse.2015.10.012, 2015.

Morel, A., Huot, Y., Gentili, B., Werdell, P. J., Hooker, S. B., and Franz, B. A.: Examining the consistency of products derive d from various ocean color sensors in open ocean (Case 1) waters in the perspective of a multi-sensor approach, Remote Sens. Environ., 111, 69–88, https://doi.org/10.1016/j.rse.2007.03.012, 2007.

Vallina, S. M. and Simó, R.: Strong relationship between DMS and the solar radiation dose over the global surface ocean, Science, 315, 506–508, 2007.

메모 포함[KTP60]: (response to referee #1)
Q1. What is the impact of SST, MLD and PIC compared to Chl?
We have added detailed explanation regarding the DMSP algorithm in the revised supplementary.

Table S1. Monthly average, 1 standard deviation and 95% confidence interval of nanoparticles (2.5–10 nm in diameter, $CN_{2.5-10}$) that originated from the Bellingshausen and Weddell Seas during the study period. A t-test was used to determine if there is a statistically significant difference between the means number concentration nanoparticles originated from the two selected ocean domains.

		Avg.	Std.	95% confidence interval*		p-value (t-test)
				Upper bound	Lower bound	
Jan.	Weddell sea	332.0	920.7	196.3	782.4	0.0003
	Bellingshausen sea	835.9	2673.2	705.9	1004	
Feb.	Weddell sea	254.3	284.0	181.4	355.8	0.0010
	Bellingshausen sea	523.7	2130.7	417.2	695.4	
Mar.	Weddell sea	60.7	60.3	47.0	78.2	< 0.0001
	Bellingshausen sea	166.6	550.3	142.6	208.5	
Apr.	Weddell sea	70.0	103.6	53.9	96.4	0.0245
	Bellingshausen sea	100.9	272.4	87.7	126.6	
May	Weddell sea	89.6	74.5	75.2	108.4	< 0.0001
	Bellingshausen sea	45.2	56.2	41.0	50.9	
Jun.	Weddell sea	58.0	22.0	NaN**	NaN	NaN
	Bellingshausen sea	57.7	62.1	52.7	64.1	
Jul.	Weddell sea	22.9	17.9	15.1	35.1	0.0031
	Bellingshausen sea	42.8	56.8	36.8	51.6	
Aug.	Weddell sea	-	-	NaN	NaN	NaN
	Bellingshausen sea	58.3	78.6	47.5	73.6	
Sep.	Weddell sea	3.7	-	NaN	NaN	NaN
	Bellingshausen sea	97.9	85.9	91.0	105.5	
Oct.	Weddell sea	193.1	160.5	NaN	NaN	NaN
	Bellingshausen sea	129.0	405.1	110.3	197.7	
Nov.	Weddell sea	88.0	61.0	74.3	107.4	< 0.0001
	Bellingshausen sea	176.7	331.9	154.0	214.3	
Dec.	Weddell sea	200.5	380.5	56.5	499.9	0.2111
	Bellingshausen sea	343.0	1138.8	277.6	449.0	

*confidence interval was estimated by bootstrap method that was calculated from 10,000 subsamples generated by random sampling with replacement from monthly $CN_{2.5-10}$ data.
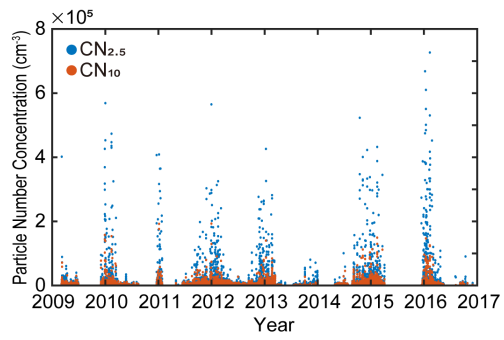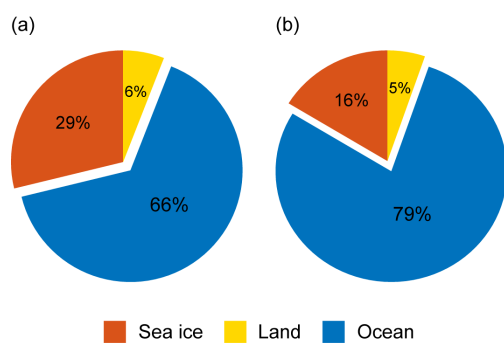**number of monthly $CN_{2.5-10}$ data <10 was excluded from the bootstrap and t-test.

Figure S1: Hourly variations in the number concentration of particles > 2.5 nm in diameter (CN$_{2.5}$, blue symbols) and the number concentration of particles > 10 nm in diameter (CN$_{10}$, red symbols) from March 2009 to November 2016.

(a)   (b)

6%
29%
66%

16%   5%
79%

Sea ice   Land   Ocean

Figure S2 S1: Percentage of the hourly trajectory points that passed over the three major areas surrounding the observation site including sea-ice (red), land (yellow) and ocean (blue) to the total number of hourly trajectory points in the 2-day air-mass trajectory during (a) the overall period (from January to December) and (b) the austral summer period (December, January and February) between March 2009 and November 2016.
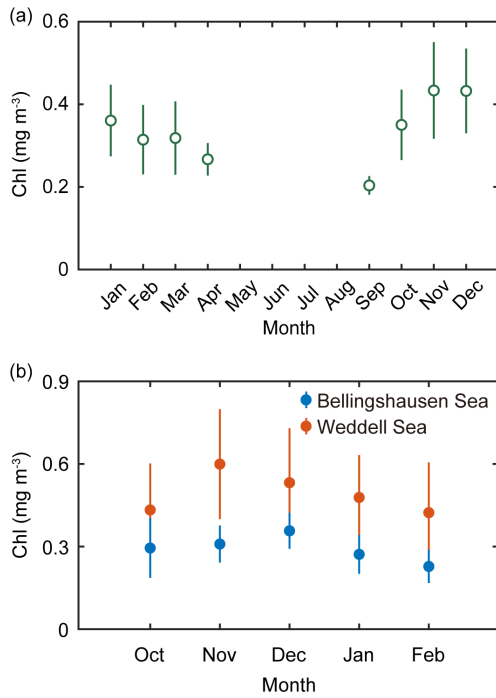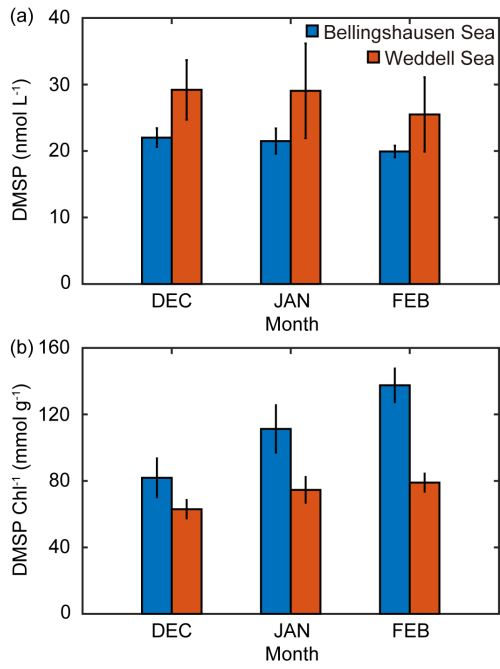
Figure ~~S3~~ S2: (a) Monthly mean chlorophyll concentration around the observation site between 2009 and 2016 (55ºS–65ºS, 40ºW–80ºW). (b) Monthly mean chlorophyll concentration for the two selected ocean domains including the Weddell (red symbols; 55ºS–65ºS, 40ºW–60ºW) and Bellingshausen (blue symbols; 55ºS–65ºS, 60ºW–80ºW) seas during the phytoplankton bloom period (October–February). Note that the monthly mean chlorophyll concentration was not available from May to August due to insufficient satellite-derived values (less than 10%) during the austral winter period.
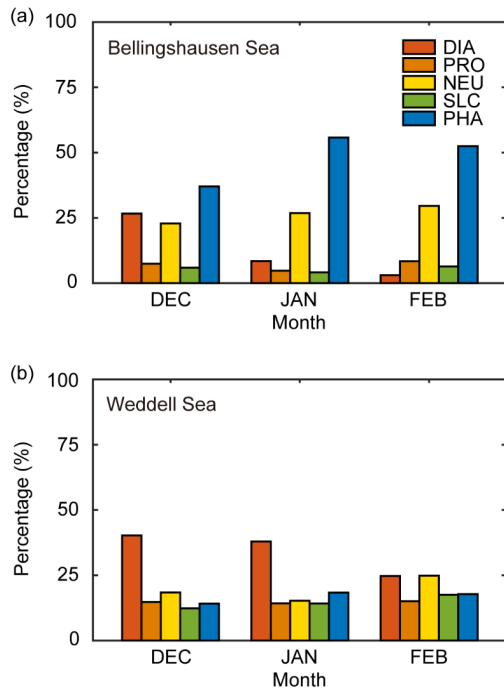
Figure ~~S4~~ S3: The percentage of the dominant phytoplankton groups in the two ocean domains including (a) the Bellingshausen and (b) Weddell seas estimated using the PHYSAT method with the climatology map obtained from SeaWiFS archive during the austral summer period ~~using the SeaWiFS archive (from September 1997 to December 2010) between 1997 and 2010.~~

(a)

(b)

Figure ~~S5~~ **S4:** (a) Monthly mean DMSP concentration and (b) ~~M~~ monthly mean DMSP to chlorophyll ratio in the Bellingshausen (blue bars) and Weddell (red bars) ~~s~~ Seas during the austral summer period between March 2009 and November 2016.

5
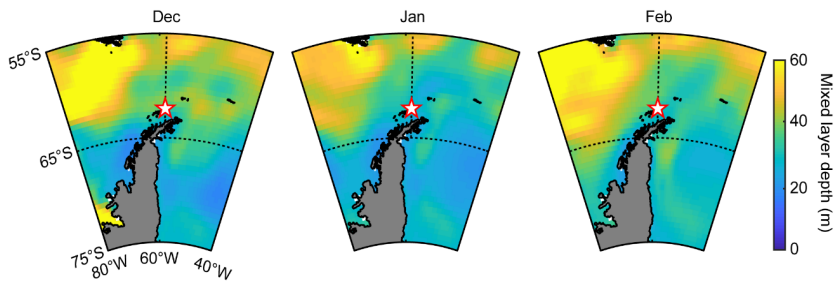
Figure S6 S5: Mixed layer depth retrieved using the Monthly Isopycnal and Mixed-layer Ocean Climatology (MIMOC) during the austral summer period surrounding King Sejong Station (red star symbol).