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. The revised manuscript was uploaded in the form of a supplement.

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 (CN2.5, CN10, and CN2.5-10), 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 (Zeu) 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 log10(Zeu/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).

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

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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 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 PM10 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).

Please also note the supplement to this comment: https://www.atmos-chem-phys-discuss.net/acp-2018-1181/acp-2018-1181-AC1-supplement.pdf

Fig. 1. PHYSAT analysis based on (a) SeaWiFS climatology map (1997-2010), and (b) MODIS climatology map (2002-2016)