

Responses to Reviewers' comments

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Title: Characteristics of biogenically-derived aerosols over the Amundsen Sea, Antarctica

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Note: Reviewers' comments are highlighted in **blue**, while our responses to reviewers are in **black**. The revisions in the manuscript were highlighted to make easily readable for the editors and reviewers.

Dr. Ulrich Pöschl

Chief Executive Editor

Atmospheric Chemistry and Physics

Max Planck Institute for Chemistry,
Mainz, Germany

Dear Doctor Pöschl,

We have attached an electronic copy of manuscript file ready to go to press entitled "Characteristics of biogenically-derived aerosols over the Amundsen Sea, Antarctica" by Jinyoung Jung, Sang-Bum Hong, Meilian Chen, Jin Hur, Liping Jiao, Youngju Lee, Keyhong Park, Doshik Hahm, Jung-Ok Choi, Eun Jin Yang, Jisoo Park, Tae-Wan Kim, and SangHoon Lee for publication in Atmospheric Chemistry and Physics (acp-2019-133). We have modified our text based on the referee's comments. We appreciated that the comments from referees improved our manuscript a lot. We believe that the comments from referees were clearly responded in our modified manuscript.

We are looking forward to hearing about your decision. Thank you for your consideration.

Sincerely yours,

Jinyoung Jung

Anonymous Referee #4:

Major Points:

In section 3.6, the authors attributed the negative correlation of the wind speed and WSOC/Na⁺ and WIOC/Na⁺ to sea surface micro-layer coverage, suggesting that lower wind speed led to higher sea surface micro-layer coverage and higher OC enrichment on sea salt particles. I am not sure if this is clearly supported by the measurements. In order to come to this conclusion, WSOC and WIOC could only be exclusively ocean-generated and internally mixed with sea salt particles. This is not directly supported by the measurements since the particle mixing state was not presented in this study. The correlation of DOC concentration in sea surface micro-layer to wind speed (Figure 7e and Figure 7f) is too weak to account for the strong negative correlation of the wind speed and WSOC/Na⁺ and WIOC/Na⁺ (Figure 7c and Figure 7d). The most intuitive reason of this wind dependence of OC to sea salt ratio is that wind speed is higher in the Southern Ocean and thus more sea salt particles were emitted from the ocean whereas OC is not as strongly affected by wind as sea salt. According to section 3.7, WSOC and WIOC are from secondary and primary sources, respectively, yet their ratio to sodium showed a somewhat similar trend against wind speed in Figure 7c and Figure 7d. This probably means that sodium, instead of OC, was more sensitive to wind speed changes.

(Response) We agree to Referee #4's comments. We realized that our discussion about the negative correlations of the wind speed with WSOC/Na⁺ and WIOC/Na⁺ was not supported by the measurements. To explain these relationships more clearly, we have modified Figures 7c and 7d by adding in situ Chl-a concentrations averaged for each aerosol sampling time (page 35). Although in situ Chl-a was not measured during the cruise from New Zealand to the entrance to the Amundsen Sea due to the limited ship time, our results, shown in Figures 7c and 7d, suggest that the wind speed is a significant factor influencing the WSOC/Na⁺ and WIOC/Na⁺ ratios in our study region. According to Referee #4's comments, we revised our manuscript as follows: "In addition to marine biological activities, the wind speed has been reported to have an effect on the organic mass fraction of sea spray aerosol (Rinaldi et al., 2013; Ceburnis et al., 2016). Gantt et al. (2011) demonstrated that the organic mass fraction of sea spray aerosol depends not only on marine biological activity in oceanic surface waters but also on sea surface wind speed. During the sampling period, both WSOC/Na⁺ and WIOC/Na⁺ ratios showed inverse relationships with mean wind speed ($r = -0.82$, $p < 0.01$ and $r = -0.76$, $p < 0.01$, respectively) (Figs. 7c and 7d). The highest WSOC/Na⁺ and WIOC/Na⁺ ratios were observed in the aerosol sample (i.e., A5) collected in biologically active region (i.e., in situ Chl-a concentration was highest (6.9 mg m^{-3})) under calm conditions (i.e., mean wind speed was lowest (4.4 m s^{-1})), and the WSOC/Na⁺ and WIOC/Na⁺ ratios drastically decreased with increasing mean wind speed. This result suggests that organic matters produced in high biological activity conditions were enriched in the sea surface water under low wind speed conditions, making a favorable condition for transferring organic matters to the atmosphere through the bubble bursting process (Rinaldi et al., 2013). However, as shown in Figures 7c and 7d, the WSOC/Na⁺ and WIOC/Na⁺ ratios observed in the biologically active region (i.e., higher in situ Chl-a concentrations) did not always show higher values when the wind

speeds exceeded 7 m s^{-1} . According to previous studies (Gantt et al., 2011; Rinaldi et al., 2013), the wave breaking caused by high wind speeds ($> 8 \text{ m s}^{-1}$) thoroughly mixes the organic-enriched sea surface layer with the underlying relatively organic-poor waters resulting in the homogeneous water column and considerably reduces the organic mass fraction of sea spray aerosol. Therefore, the inverse relationships between WSOC/Na⁺, WIOC/Na⁺ ratios and the mean wind speed suggest that the wind speed is a significant factor influencing the organic mass fractions of sea spray aerosols in our study region.” (page 14, line number 3–19). Besides, we added the references (i.e., Rinaldi et al., 2013; Ceburnis et al., 2016) to the manuscript (page 26, line number 4–6; page 20, line number 29–30). We also revised “the SML coverage” to “the wind speed” (page 14, line number 20; page 14, line number 27–28; page 18, line number 23). We also revised “In addition, these ratios showed strong correlations with DOC concentration in the Amundsen Sea when the wave breaking thoroughly mixes the SML with the underlying water.” to “In addition, these ratios showed strong correlations with DOC concentration in the Amundsen Sea when the wave breaking thoroughly mixes the organic-enriched sea surface layer with the underlying relatively organic-poor water.” (page 18, line number 25). We believe that we revised our manuscript properly according to Referee #4’s comments.

Minor Comments:

1. P2 Line 11- P3 Line 5: These two paragraphs are about marine and biological CCN. Not sure such a detailed introduction is needed since CCN is mentioned in only section 1 and not discussed in later sections.

(Response) This study focuses on the characteristics of biogenically-derived aerosols, especially atmospheric sulfur and organic carbon species, over the Amundsen Sea. In the two paragraphs Referee #4 mentioned about, we described the formation processes and the importance of biogenically-derived atmospheric sulfur-containing and organic aerosols in the marine environment. Although Referee #4 questioned whether the detailed introduction is needed, we believe that the introduction of biogenically-derived sulfur-containing and organic aerosols is necessary.

2. P9 Line10-16: Since there are only 14 samples (or in this case 9 samples), removal of outliers needs better discussion. Please specify how "the highest mean wind speed" affects DMS flux and possibly provide either reference or calculation.

(Response) As Referee #4 suggested, we discussed the influence of wind speed on DMS flux. DMS fluxes typically rely on gas transfer velocity, which is frequently parameterized as a function of wind speed (Wanninkhof, 2014). Measurement and parameterization of the gas transfer velocity are more challenging and subject to greater uncertainty, particularly at high wind speeds (Smith et al., 2018). As the gas transfer velocity increases with increasing wind speed, DMS flux can be overestimated especially, in higher latitudes where DMS is commonly found at high concentrations in surface water, and where both low temperatures and high winds are typical (McGillis et al., 2000). Despite the uncertainty in DMS flux (Fig. 4), given the lifetime of DMS is approximately 1–2 days (Kloster et al., 2006; Read et al., 2008), our results

revealed that the local sea–air DMS flux affected directly atmospheric MSA concentration in the Amundsen Sea, and that the higher atmospheric MSA concentrations observed over the Amundsen Sea compared to those over the Southern Ocean and in coastal Antarctic regions were attributed to the higher DMS concentrations produced by *P. antarctica* and to the higher DMS fluxes in the Amundsen Sea. (page 9, line number 17–26). We also added the references (i.e., Wanninkhof, 2014; Smith et al., 2018, McGillis et al., 2000) to References (page 27, line number 28–29; page 27, line number 4–6; page 24, line number 4–5).

3. P9 Line 28: "the variation trend of " Can the authors provide correlation coefficient?

(Response) We already described about the correlation coefficients between nss-SO₄²⁻ and MSA in the Amundsen Sea and Southern Ocean (page 10, line number 2–5).

4. P11 Line 12: "somewhat overestimated... showing negative values" Can the authors discuss possibilities causing this overestimation?

(Response) We already described the limitation in quantifying biogenically-derived nss-SO₄²⁻ using the MSA/nss-SO₄²⁻ ratio (page 10, line number 25–29). Nevertheless, according to Referee #4's comment, we added the following sentences to the manuscript. "However, if we apply for the highest MSA/nss-SO₄²⁻ ratio (0.7) obtained from this study, the contributions of biogenically-derived nss-SO₄²⁻ over the Southern Ocean and the Amundsen Sea decrease to $38 \pm 20\%$ (range: 17–73%) and $62 \pm 23\%$ (range: 28–100%), respectively. This result suggests that the estimate of biogenically-derived nss-SO₄²⁻ contribution to total nss-SO₄²⁻ using the MSA/nss-SO₄²⁻ ratio could have limitation (Mungall et al., 2018) as mentioned above." (page 11, line number 18–22). We believe that we revised our manuscript properly according to Referee #4's comments.

5. P11 Line 14: "variation trend" Again consider show correlation coefficient.

(Response) According to Referee #4's comment, we added the correlation coefficient between NO₃⁻ and anthropogenic nss-SO₄²⁻ in the sentence (page 11, line number 23).

6. P 17 Line 7: "negatively correlated with the relative biomass of *P. antarctica* ($r=0.79$, $p < 0.05$)" Should be "-0.79" if using the Pearson correlation coefficient. Otherwise, the authors should specify the sign. This also applies to some other place where r is less than 0 including some of the figures.

(Response) According to Referee #4's comment, we revised " $r = 0.79$ " to " $r = -0.79$ " (page 17, line number 26). We also revised the correlation coefficient in Figure 10 (page 38).

7. P18 Line 18: Consider citing this article (Bromwich et al., 2013).

(Response) As Referee #4 suggested, we added Bromwich et al. (2013) to the manuscript with the following sentence: "West Antarctica is one of the fastest-warming regions globally." (page 19, line number 3). We also added Bromwich et al. (2013) to reference (page 20, line number 23–25).

Editorial Comments:

1. P4 Line 20: "Procedural blanks (n = 4)..." Change to "Four procedural blanks..."

(Response) According to Referee #4's comment, we revised "Procedural blanks (n = 4)..." to "Four procedural blanks..." (page 4, line number 20).

2. P13 Line 15:" reflecting that Na⁺ was formed from bubble by local wind speed. " Delete "speed".

(Response) According to Referee #4's comment, we deleted "speed" from the sentence (page 13, line number 24).

3. P16 Line 15-16:" our results strongly suggested that the submicron WSOC observed in the Amundsen Sea might be formed..." Remove "might"

(Response) According to Referee #4's comment, we revised "might be formed" to "was formed" (page 17, line number 2).

4. Figure 8c: Check unit

(Response) According to Referee #4's comment, we revised the unit of WSOC in Figure 8c " $\mu\text{g m}^{-3}$ " to " $\mu\text{gC m}^{-3}$ " (page 36).

References

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- Rinaldi, M., Fuzzi, S., Decesari, S., Marullo, S., Santolero, R., Provenzale, A., Hardenberg, Von, J., Ceburnis, D., Vaishya, A., O'Dowd, C. D. and Facchini, M. C.: Is chlorophyll-a the best surrogate for organic matter enrichment in submicron primary marine aerosol? *J. Geophys. Res. Atmos.*, 118(10), 4964–4973, doi:10.1002/jgrd.50417, 2013.
- Smith, M. J., Walker, C. F., Bell, T. G., Harvey, M. J., Saltzman, E. S. and Law, C. S.: Gradient flux measurements of sea-air DMS transfer during the Surface Ocean Aerosol Production (SOAP) experiment, *Atmos. Chem. Phys.*, 18(8), 5861–5877, doi:10.5194/acp-18-5861-2018, 2018.
- Wanninkhof, R.: Relationship between wind speed and gas exchange over the ocean revisited, *Limnol. Oceanogr. Methods*, 12, 351–362, doi:10.4319/lom.2014.12.351, 2014.

Anonymous Referee #3:

Major comments:

1. The first major comment reiterates the comment already raised by anonymous reviewer #4. The authors suggest that the inverse relationship between WSOC/Na⁺ and WIOC/Na⁺ with wind speed implies that organics in the atmosphere are controlled by winds due to the breakage of the surface microlayer for periods of times associated with high winds. However, I believe that there are several inconsistencies in the text that will need to be clearly addressed prior to publication, mainly:

a. Both WSOC/Na⁺ and WIOC/Na⁺ are inversely related to wind speeds but the authors claim that only WIOC have a primary source whereas WSOC is mostly formed by oxidation of biogenic precursors. How do the authors then explain that both ratios correlated well with wind speed?

(Response) We agree to Referee #3's comments. To explain these relationships more clearly, we have modified Figures 7c and 7d by adding in situ Chl-a concentrations averaged for each aerosol sampling time (page 35). Although in situ Chl-a was not measured during the cruise from New Zealand to the entrance to the Amundsen Sea due to the limited ship time, our results, shown in Figures 7c and 7d, suggest that the wind speed is a significant factor influencing the WSOC/Na⁺ and WIOC/Na⁺ ratios in our study region. According to Referee #3's comments, we revised our manuscript as follows: "In addition to marine biological activities, the wind speed has been reported to have an effect on the organic mass fraction of sea spray aerosol (Rinaldi et al., 2013; Ceburnis et al., 2016). Gantt et al. (2011) demonstrated that the organic mass fraction of sea spray aerosol depends not only on marine biological activity in oceanic surface waters but also on sea surface wind speed. During the sampling period, both WSOC/Na⁺ and WIOC/Na⁺ ratios showed inverse relationships with mean wind speed ($r = -0.82$, $p < 0.01$ and $r = -0.76$, $p < 0.01$, respectively) (Figs. 7c and 7d). The highest WSOC/Na⁺ and WIOC/Na⁺ ratios were observed in the aerosol sample (i.e., A5) collected in biologically active region (i.e., in situ Chl-a concentration was highest (6.9 mg m^{-3})) under calm conditions (i.e., mean wind speed was lowest (4.4 m s^{-1})), and the WSOC/Na⁺ and WIOC/Na⁺ ratios drastically decreased with increasing mean wind speed. This result suggests that organic matters produced in high biological activity conditions were enriched in the sea surface water under low wind speed conditions, making a favorable condition for transferring organic matters to the atmosphere through the bubble bursting process (Rinaldi et al., 2013). However, as shown in Figures 7c and 7d, the WSOC/Na⁺ and WIOC/Na⁺ ratios observed in the biologically active region (i.e., higher in situ Chl-a concentrations) did not always show higher values when the wind speeds exceeded 7 m s^{-1} . According to previous studies (Gantt et al., 2011; Rinaldi et al., 2013), the wave breaking caused by high wind speeds ($> 8 \text{ m s}^{-1}$) thoroughly mixes the organic-enriched sea surface layer with the underlying relatively organic-poor waters resulting in the homogeneous water column and considerably reduces the organic mass fraction of sea spray aerosol. Therefore, the inverse relationships between WSOC/Na⁺, WIOC/Na⁺ ratios and the mean wind speed suggest that the wind speed is a significant factor influencing the organic mass fractions of sea spray aerosols in our study region." (page 14, line number 3–19). Besides, we added the references (i.e., Rinaldi et al., 2013; Ceburnis et al., 2016) to the manuscript (page 26, line number 4–6; page 20, line number 29–

30). We also revised “the SML coverage” to “the wind speed” (page 14, line number 20; page 14, line number 27–28; page 18, line number 23). We also revised “In addition, these ratios showed strong correlations with DOC concentration in the Amundsen Sea when the wave breaking thoroughly mixes the SML with the underlying water.” to “In addition, these ratios showed strong correlations with DOC concentration in the Amundsen Sea when the wave breaking thoroughly mixes the organic-enriched sea surface layer with the underlying relatively organic-poor water.” (page 18, line number 25). We believe that we revised our manuscript properly according to Referee #3’s comments.

b. The authors hypothesize that WIOC is of primary origin, but their measurements indicate no correlation between WIOC and Na⁺. The authors attribute this to transport but their measurements clearly indicate a correlation between Na⁺ and wind speed, suggesting transport cannot completely rule out a correlation between WIOC and Na⁺. Also, why does WIOC correlate with the relative abundance of *P. Antarctica*? Presumably, this implies that a part of WIOC is formed from oxidation of BVOC?

(Response) In section 3.7, we discussed the insignificant relationships between WIOC and Na⁺ in the fine modes over the Southern Ocean and the Amundsen Sea. As mentioned in section 3.6, the wave breaking caused by high wind speed thoroughly mixes the organic-rich sea surface layer with the underlying relatively organic-poor waters resulting in the homogeneous water column and considerably reduces the organic mass fraction of sea spray aerosol. Thus, these insignificant relationships between WIOC and Na⁺ in the fine modes could result from the differences in local wind speeds and local biological activities, such as sea surface DOC concentration, because wind speed, a key factor determining sea spray aerosols, controls the local flux rather than local concentration of marine particles (page 15, line number 33–page 16, line number 6). Besides, as Referee #3 pointed out, the WIOC production by secondary processes cannot be excluded. Thus, we added the following sentence to the manuscript: “However, the WIOC production by secondary processes cannot be completely excluded either (Ceburnis et al., 2016), but we have no evidence of that (page 16, line number 11–12).

Referee #3 asked why WIOC concentration showed a good correlation with the relative biomass of *P. antarctica* in the Amundsen Sea. As we already described in section 3.6, phytoplankton exudates include exopolymer gels consisting of polysaccharides, which are insoluble, thermally stable, highly surface active, highly hydrated and readily sequester dissolved organic matter (Quinn and Bates, 2011 and references therein). Besides, *P. antarctica*, which was a dominant phytoplankton species in the Amundsen Sea (see section 3.2), generates a substantial amount of extracellular polysaccharide mucus in its colonial matrix, and it has been suggested that large amounts of this material ultimately enter the DOC pool (Smith et al., 1998). Thus, the significant correlation ($r = 0.87$, $p < 0.05$) between WIOC and the relative biomass of *P. antarctica* in the Amundsen Sea suggests that water-insoluble organic matter released by *P. antarctica* was emitted in the Amundsen Sea via bubble bursting and breaking waves (page 15, line number 7–10). We believe that we revised our manuscript properly according to Referee #3’s comments.

c. Finally, why do WSOC concentrations (presumably of secondary origin, i.e., from the oxidation of BVOC) correlate strongly with Na^+ and with DOC? The authors hypothesize that the WSOC relation on Na^+ is due to higher surface area (from salt particles) and therefore higher WSOC concentrations are a result of a larger condensation sink. However, the authors present no compelling evidence to substantiate that hypothesis. Can the author examine the relation between WSOC and average short wavelength radiation? This could provide the evidence needed to argue that WSOC is formed from secondary sources. The linkage between WSOC and WIOC with biology needs to be better articulated.

(Response) As we already described in section 3.7, submicron WSOC showed a strong correlation ($r = 0.94$, $p < 0.01$) with submicron Na^+ in the Amundsen Sea (Fig. 8b). In addition, we also found a significant correlation ($r = 0.93$, $p < 0.01$) between WSOC and MSA concentrations in the Amundsen Sea (Fig. 8c). However, in the Southern Ocean, WSOC showed no significant relationship with submicron Na^+ or MSA. MSA is produced by atmospheric oxidation of DMS, which is released as a gas phase from marine biological activities and thus can be used as an indicator of secondary aerosols of marine biogenic origin (Miyazaki et al., 2011). As described in section 3.2, Kim et al. (2017) observed extremely high DMS concentrations ($> 150 \text{ nM}$) in surface water during our cruise, and MSA concentration showed a strong correlation with DMS flux in the Amundsen Sea. Consequently, the strong correlations between WSOC, Na^+ and MSA in the Amundsen Sea implies that the Amundsen Sea that has the most productive polynya in the Antarctic is a strong source region of BVOCs, and that WSOC was formed by the condensation of BVOCs released from sea surface onto preexisting submicron sea spray aerosols through gas-to-particle conversion due to a higher surface-to-volume ratio of submicron aerosols (Romakkaniemi et al., 2011). On the other hand, the poor correlations between WSOC, Na^+ , and MSA in the Southern Ocean implies the differences in local source strength of BVOCs and that the presence of DMS in seawater and its subsequent oxidation to MSA were not necessarily linked to the formation of submicron WSOC over the Southern Ocean (Miyazaki et al., 2016). Therefore, our results provide the evidence that WSOC was formed by secondary processes in the Amundsen Sea (page 16, line number 13–26).

Referee #3 questioned that why WSOC concentration showed a strong correlation with Na^+ . However, WSOC concentration did not show a good correlation with DOC, but WSOC/ Na^+ ratio did, as shown in Figure 7e. We already discussed the reason why WSOC/ Na^+ was correlated with DOC in section 3.6 (page 14, line number 20–page 15, line number 17).

Unfortunately, we do not have any short wavelength radiation data. However, it would be interesting to investigate the relationship between WSOC and short wavelength radiation in the future study. Although we cannot discuss the relationship between WSOC and short wavelength radiation, the significant relationship between WSOC, MSA and Na^+ in the Amundsen Sea (Figs. 8b and 8c) suggest that WSOC was formed by the condensation of BVOCs released from sea surface onto preexisting submicron sea spray aerosols through gas-to-particle conversion as we mentioned above.

Referee #3 pointed out that the linkage between WSOC and WIOC with biology needs to be better articulated. However, in section 3.6, we already discussed the significant relationship between WIOC concentration and the relative biomass of *P. antarctica* in the Amundsen Sea, suggesting that water-insoluble organic matter released by *P. antarctica* was emitted in the Amundsen Sea via bubble bursting and breaking waves (page 15, line number 9–10). Besides, in section 3.8, we investigated the fluorescence properties of WSOC over the Southern Ocean and the Amundsen Sea. Interestingly, fluorescence intensity of C1 (i.e., protein-like component) showed a significant positive relationship with the relative biomass of diatoms ($r = 0.89$, $p < 0.01$); however, it was negatively correlated with the relative biomass of *P. antarctica* ($r = -0.79$, $p < 0.05$) (Figs. 10c and 10d). These results suggest that protein-like component is most likely produced as a result of biological processes of diatoms, which play a key role in forming the submicron WSOC observed over the Southern Ocean and the Amundsen Sea, and that phytoplankton community structure is a significant factor affecting atmospheric OC species since the submicron WIOC was quite related to the relative biomass of *P. antarctica* (page 17, line number 24–page 18, line number 2). Thus, we believe that we discussed properly the linkage between WSOC and WIOC with biology in our manuscript.

2. What happens to the correlations in Figures 7 if the authors looked at WSOC and WIOC instead of WSOC/Na⁺ and WIOC/Na⁺? This could remove the cross-correlation with wind speed (I encourage the authors to add these graphs as part of the SI).

(Response) As Referee #3 suggested, we added the plots of WIOC concentration in fine mode versus mean wind speed and WSOC concentration in fine mode versus mean wind speed to Figure S6 in Supplementary material. We also added the following sentence to our manuscript: “In addition, no relationship was found between the submicron WIOC and mean wind speed (Fig. S6).” (page 15, line number 32).

3. The authors often mention correlation strength even though the p-value for the regression is higher than the specified threshold (in this case 0.05). A p-value larger than the threshold implies there is no confidence in rejecting the null hypothesis (which is: the variables are not correlated). I suggest the authors reframe their results accordingly.

(Response) We agree to Referee #3’s comment. According to Referee #3’s comments, we revised our manuscript as follows: “Atmospheric MSA concentration showed no relationship with either in situ sea surface Chl-a concentration ($r = 0.029$, $p > 0.05$) (Fig. S2) or the relative biomass of *P. antarctica* ($r = 0.30$, $p > 0.05$).” (page 9, line number 4–6), “but no correlation was found between atmospheric MSA and DMS flux ($r = 0.18$, $p > 0.05$, Fig. 4b)” (page 9, line number 9–10), “atmospheric MSA concentration showed a significant relationship with DMS flux ($r = 0.78$, $p < 0.05$).” (page 9, line number 12), “A similar result was found between atmospheric MSA and DMS concentrations in surface water, showing no correlation ($r = 0.20$, $p > 0.05$).” (page 9, line number 14–15), “whereas no relationship was found between them in the Southern Ocean ($r = 0.51$, $p > 0.05$) (Fig. S3)” (page 10, line number 3). We also removed “Similar results were found between fluorescence intensity of C2 and the relative biomass of diatoms and *P. antarctica*.” because the fluorescence intensity of C2 showed no relationship with the relative biomass of diatoms or

and *P. antarctica* (this sentence was written on page 17, line number 7-8 in the unrevised manuscript). Besides, we modified Figure 10 (page 38), and revised "...by showing significant positive relationships between the relative biomass of diatoms and protein-like components in marine aerosols in the Amundsen Sea." to "...by showing the significant positive relationship between the relative biomass of diatoms and protein-like component in marine aerosols in the Amundsen Sea." (page 19, line number 1–2). We also revised "Protein-like components also showed positive relationships with the relative biomass of diatoms; however, they were negatively correlated with the relative biomass of *P. antarctica*. These results suggest that protein-like components are most likely produced as a result of biological processes of diatoms, which play a crucial role in forming the submicron WSOC observed over the Southern Ocean and the Amundsen Sea, and that phytoplankton community structure is a significant factor affecting atmospheric organic carbon species." to "Protein-like component also showed a significant positive relationship with the relative biomass of diatoms; however, it was negatively correlated with the relative biomass of *P. antarctica*. These results suggest that protein-like component is most likely produced as a result of biological processes of diatoms, which play a crucial role in forming the submicron WSOC observed over the Southern Ocean and the Amundsen Sea, and that phytoplankton community structure is a significant factor affecting atmospheric organic carbon species." (page 1, line number 30–page 2, line number 2).

Minor comments:

P7 L29: specify at least once what +/- refers to, one standard error or the one standard deviation?

(Response) As Referee #3 suggested, we added (mean \pm one standard deviation) to the sentence (page 7, line number 29).

P9 L9-L10: The authors describe a weak but not significant relation between MSA and DMS. If the p-value is not significant than the authors cannot justify that a positive or negative correlation (see major comment 2). Please adjust elsewhere in the text where applicable.

(Response) As Referee #3 already pointed out in major comment 3, we revised our manuscript according to Referee #3's comment. Please see our response to major comment 3.

P9 L13: The authors should provide more thorough reasoning for removing the point to the right. Indeed, removing that one point changes the correlation between DMS and MSA from non-existent to significant, due to the small sample number.

(Response) As Referee #3 suggested, we discussed the influence of wind speed on DMS flux. DMS fluxes typically rely on gas transfer velocity, which is frequently parameterized as a function of wind speed (Wanninkhof, 2014). Measurement and parameterization of the gas transfer velocity are more challenging and subject to greater uncertainty, particularly at high wind speeds (Smith et al., 2018). As the gas transfer velocity increases with increasing wind speed, DMS flux can be overestimated especially, in higher latitudes where DMS is commonly found at high concentrations in surface water, and where both low temperatures and high winds are typical (McGillis et al., 2000). Despite the uncertainty in DMS flux (Fig.

4), given the lifetime of DMS is approximately 1–2 days (Kloster et al., 2006; Read et al., 2008), our results revealed that the local sea–air DMS flux affected directly atmospheric MSA concentration in the Amundsen Sea, and that the higher atmospheric MSA concentrations observed over the Amundsen Sea compared to those over the Southern Ocean and in coastal Antarctic regions were attributed to the higher DMS concentrations produced by *P. antarctica* and to the higher DMS fluxes in the Amundsen Sea. (page 9, line number 17–26). We also added the references (i.e., Wanninkhof, 2014; Smith et al., 2018, McGillis et al., 2000) to References (page 27, line number 28–29; page 27, line number 4–6; page 24, line number 4–5).

P9 L27-30: “Unlike MSA, the mean nss-SO₄²⁻ concentration in the Amundsen Sea. . .” please provide justification to how the trend for nss-sulfate was comparable to that of MSA. Visual inspection of the data is not sufficient. Also, I am not sure how the authors arrived at the conclusion that nss-sulfate is influenced by marine and anthropogenic sources given the information in that sentence.

(Response) We already described about the correlations between nss-SO₄²⁻ and MSA in the Amundsen Sea and Southern Ocean (page 10, line number 2–7). Besides, the reason why we believe that nss-SO₄²⁻ was affected by both marine and anthropogenic sources is already described in section 3.4 (page 10, line number 19–page 12, line number 3).

P10 L17: “. . . showing a similar variation trend to that of MSA”. Quantify the agreement.

(Response) As Referee #3 suggested, we added “(r = 0.92, p < 0.01)” to the sentence (page 10, line number 32).

P11 L14-19: “We expected much higher WSOC and WIOC concentrations in the Amundsen Sea than the Southern Ocean because of extremely high Chl-a concentrations. . .”. This result could be consistent with findings of Quinn et al. (2014) who argued that organic enrichment in the aerosol phase is likely controlled by the dissolved organic carbon (DOC) pool rather than chlorophyll concentrations.

(Response) According to Referee #3’s comment, we added “(Quinn et al., 2014, see section 3.6) to the sentence (page 12, line number 14).

P10 L20-21: Specify how the enrichment of WSOC and WIOC in the aerosol phase was calculated.

(Response) According to Referee #3’s comment, we added “(i.e., the percentage of WSOC or WIOC present in fine aerosol particles)” to the sentence (page 12, line number 16–17).

P10 L23: “For example, O’Dowd et al. (2004) observed that the contribution of OC fraction to the submicrometer aerosol mass increased from 15% to 63% between low and high biological activity periods in the North Atlantic.” I am confused by this sentence. The measurements presented in this article show no relation of WSOC and WIOC on chl-a concentrations (as explicitly written in the previous paragraph). Please clarify.

(Response) We referred to O’Dowd et al. (2004) to described that WIOC in fine mode aerosol particles is

dominant OC species during bloom periods, as our result showed it. We, therefore, revised our manuscript as follows: “For example, O’Dowd et al. (2004) observed a dominant water-insoluble OC fraction (~45%) in fine marine aerosol collected during periods of phytoplankton bloom in the North Atlantic.” (page 12, line number 20–21).

P11 L31: “the dominance of WIOC suggests that the bubble bursting process by local wind speeds is a significant formation mechanism of atmospheric WIOC in our area.” If this is true, then why don’t the author observe lower WIOC concentration over the Amundsen Sea compared to the Southern Ocean given that wind speed in the Southern Ocean were larger compared to over the Amundsen Sea?

(Response) We agree to Referee #3’s comment. Therefore, we revised our manuscript as follows: “the dominance of WIOC suggests that water-insoluble organic matter exuded by phytoplankton is more accumulated in sea surface water and emitted into the marine atmosphere via bubble bursting and breaking waves by local wind.” (page 12, line number 26–28).

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We already have revised our manuscript according to Referee #1's comments and uploaded the revised manuscript file on 19 March 2019 before open discussion started. Just in case, we show our responses to Referee #1 again.

Referee #1:

1. The authors present measurements of aerosol and ocean carbon from the southern ocean and Amundsen Sea linking biological processes to marine particle concentrations and composition. This is an important processes, though not well understood, partly due to the interdisciplinary aspect of the subject. Better understanding this process possibly has significant implication on understanding future climate change. The authors do well to discuss and provide evidence for their claims. I am suggesting the article be accepted after major revisions. The main concerns I have involve the claims made in section 3.3-3.4, specifically that a significant amount of nssSO₄ is from anthropogenic sources. While possible, I do not think they have provided strong evidence. Previous studies in the SO have shown that almost no continental/anthropogenic pollution is transported over the southern ocean particularly south of ~50S (See Hudson et al. 1998). Hudson et al. 1998 measurements are in a different part of the Southern Ocean, I expect the result to be similar. A back trajectory analysis would strengthen your case if there really is anthropogenic sources.

(Response) We thank for the comments from Referee #1. Referee #1 pointed out a significant amount of anthropogenic nss-SO₄²⁻ in our study regions. We have already described that concentration of anthropogenic nss-SO₄²⁻ estimated using the MSA/nss-SO₄²⁻ ratio of 0.508 observed at Palmer Station, varied from 0.10–0.63 μg m⁻³ (mean: 0.34 ± 0.18 μg m⁻³) in the Southern Ocean, and 0.13–0.30 μg m⁻³ (mean: 0.19 ± 0.079 μg m⁻³) in the Amundsen Sea (Figs. 5c and 5d) (page 11, line number 2–16). To clarify the context, we have added a short explanation for the contributions of anthropogenic nss-SO₄²⁻ in the Southern Ocean and the Amundsen Sea to section 3.4 (page 11, line number 16–17).

As referee #1 suggested, we have read the paper (Hudson et al., 1998). However, Hudson et al. (1998) reported only the number concentrations of condensation nuclei observed over the Southern Ocean, so we could not refer to it. To compare the anthropogenic nss-SO₄²⁻ concentrations estimated in this study to previously published results, we have compared our anthropogenic nss-SO₄²⁻ concentration estimated in this study to the results by Jung et al. (2014) who reported that mean concentration of nss-SO₄²⁻ observed over the South Pacific (10°S–55°S, January–March 2009) was 0.25 ± 0.17 μg m⁻³. Our mean concentration of anthropogenic nss-SO₄²⁻ in the Southern Ocean was a factor of 1.4 higher than the result by Jung et al. (2014). In comparison, our mean concentration of anthropogenic nss-SO₄²⁻ in the Amundsen Sea was a factor of 1.3 lower than the result by Jung et al. (2014) (page 11, line number 17–21).

As referee #1 suggested, we have calculated 7-day air mass backward trajectories and added it to the Supplement (Fig. S4). We also have added short explanations for the backward trajectory results to section 3.4 (page 11, line number 21–26).

2. The nssSO₄ likely correlates strongly to MSA in the Amundsen Sea simply because the nssSO₄ source (MSA) is local. It is common for nssSO₄ to be transported large distance, often in the free troposphere, resulting in little to no correlation with local sources (MSA). (see Sanchez et al. 2018). While MSA is often used as a tracer, a correlation may not always be found. Gas phase and aqueous phase oxidation of MSA to sulfate is known to take place, potentially leading to MSA having a lifetime of only a few days (Mungall et al 2018).

(Response) As referee #1 suggested, we have investigated the relationship between nss-SO₄²⁻ and MSA in the Southern Ocean and the Amundsen Sea and added the result to the Supplement (Fig. S3). We also have added the following sentences to section 3.3. “Indeed, nss-SO₄²⁻ showed a strong correlation ($r = 0.98$, $p < 0.01$) with MSA in the Amundsen Sea, whereas the relationship in the Southern Ocean was statistically insignificant ($r = 0.51$, $p > 0.05$) (Fig. S3), suggesting that the local emission of DMS is a significant source of nss-SO₄²⁻ in the Amundsen Sea and that nss-SO₄²⁻ in the Amundsen Sea was formed by the condensation of DMS products onto existing particles (Sanchez et al., 2018). On the other hand, the statistically insignificant relationship between nss-SO₄²⁻ and MSA could result from strong influence of anthropogenic sources and low biological activity in the Southern Ocean” (page 9, line number 30–page 10, line number 2). We also have added Sanchez et al. (2018) as a reference in the manuscript.

As Referee #1 mentioned, we have read the paper (Mungall et al., 2018) and added the following sentences to section 3.4. “Mungall et al. (2018), however, pointed out that the MSA/nss-SO₄²⁻ ratio could have limitation that may preclude its use in quantitatively unravelling the chemical and biological processes at play in the marine boundary layer due to the conversion of MSA to nss-SO₄²⁻ by OH radical in aerosol particles (the order of days to weeks), although it remains useful as a qualitative indicator of marine biological influence” (page 10, line number 20–24). We also have added Mungall et al. (2018) as a reference in the manuscript

3. While nitrate can be attributed to anthropogenic emission, particle nitrate concentration (and nssSO₄ concentration) concentration can also be enhanced by aqueous phase processing (in clouds). On Figure 5d, with the exception of samples A1 and A13 (both of which are between 45S and 50S on figure 1a) the nitrate concentration seems fairly consistent.

(Response) We have agreed to Referee #1’s opinion. However, we have used NO₃⁻ solely as an indicator of anthropogenic contribution in this study. Moreover, we have focused on the characteristics of biogenically-derived sulfur and organic carbon species in our study regions. Thus, we believe that further discussions on NO₃⁻ are beyond the scope of our manuscript.

4. I would be surprised to find that, at times, anthropogenic sources account for more than half the nssSO₄ even in the Amundsen Sea (your range is 39%-138%). There are no relatively nearby anthropogenic sources to account for this. More evidence would be needed to make such a claim.

(Response) Referee #1 pointed out that the contribution of anthropogenic nss-SO₄²⁻ to total nss-SO₄²⁻ in the Amundsen Sea was unreasonably high (range: 39–138%), even though the influence of anthropogenic sources is weak in the Amundsen Sea. However, what Referee #1 pointed out was not the contribution of anthropogenic nss-SO₄²⁻, but that of biogenically-derived one in the Amundsen Sea. In our manuscript, we have described in section 3.4 that mean concentrations of biogenically-derived nss-SO₄²⁻ over the Southern Ocean and the Amundsen Sea were estimated to be $0.31 \pm 0.19 \mu\text{g m}^{-3}$ (range: 0.074–0.57 $\mu\text{g m}^{-3}$) and $0.56 \pm 0.30 \mu\text{g m}^{-3}$ (range: 0.19–1.1 $\mu\text{g m}^{-3}$), accounting for $52 \pm 28\%$ (range: 24–101%) and $86 \pm 32\%$ (range: 39–138%) of total nss-SO₄²⁻, respectively (Fig. 5b) (page 11, line number 5–8). We therefore believe that Referee #1 might have misunderstood our manuscript.

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