

Responses to Co-Editor' comments

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Title: Characteristics of methanesulfonic acid, non-sea-salt sulfate and organic carbon aerosols over the Amundsen Sea, Antarctica

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Note: Co-Editor's comments are high lightened in **blue**, while our responses to reviewers are in **black**. The revisions in the manuscript was highlighted to make easily readable for the Co-Editor.

Prof. Lynn M. Russell

Co-Editor

Atmospheric Chemistry and Physics

USCD, Scripps Institution of Oceanography

United States

Dear Professor Russell,

We have changed the title of our manuscript to "Characteristics of methanesulfonic acid, non-sea-salt sulfate and organic carbon aerosols over the Amundsen Sea, Antarctica" according to Co-Editor's comment. We have attached an electronic copy of manuscript file ready to go to press entitled "Characteristics of methanesulfonic acid, non-sea-salt sulfate and organic carbon 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 Co-Editor's comments. We appreciated that the comments from Co-Editor improved our manuscript a lot. We believe that the comments from Co-Editor 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

1. p.11 line 26 to p.12 line 15: omit this section. Extrapolating from summer to annual is not justified by the measurements or the literature. It detracts from the otherwise solid nature of the paper.

(Response) We thank Co-Editor for Co-Editor's comment. As Co-Editor suggested, we have removed the sentences Co-Editor mentioned in section 3.4 (the sentences were written on page 11, line number 26–page 12, line number 15 in the previous manuscript). We also have removed from the section 3.4 the following sentences: “While nss-SO_x can have multiple sources, including DMS oxidation, volcanic and industrial sulfur emissions, MSA is formed exclusively from DMS (Gondwe et al., 2003). Thus MSA was proposed as a useful tracer to distinguish between marine biogenic and anthropogenic nss-SO_x (Legrand and Pasteur, 1998). Considerable efforts have been devoted to investigating the contribution of biogenically-derived atmospheric sulfur species over the various geographical locations by using the MSA/nss-SO_x ratio observed in aerosols, which shows the spatial (higher ratios at high latitudes) and seasonal variability (summer maxima and winter minima) (e.g., Savoie and Prospero, 1989; Prospero et al., 1991; Bates et al., 1992; Arimoto et al., 2001; Savoie et al., 2002; Yang et al., 2009; Jung et al., 2014; Legrand et al., 2017). Mungall et al. (2018), however, pointed out that the MSA/nss-SO_x 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-SO_x by OH radical in aerosol particles (the order of days to weeks), although it remains useful as a qualitative indicator of marine biological influence.” (these sentences were written on page 11, line number 9–19 in the previous manuscript). Besides, we have changed the title of section 3.4 “Contribution of biogenically-derived nss-SO₄²⁻ to total nss-SO₄²⁻” to “MSA/nss-SO₄²⁻ ratios over the Southern Ocean and the Amundsen Sea” because the contribution of biogenically-derived nss-SO₄²⁻ is not discussed in this section (page 10, line number 24). We also moved the following sentence to the first paragraph of section 3.4 to give information to the readers (page 10, line number 25–28): “There have been several field studies investigating the MSA/nss-SO₄²⁻ ratio in the Southern Ocean during the austral summer (range: 0.32–0.53, the South Pacific (40°S–45°S), Berresheim et al., 1990; range: 0.12–0.24, Cape Grim (40°41'S, 144°41'E), Ayers et al., 1991; range: 0.17–0.32, the South Pacific (30°S–60°S), Bates et al., 1992; range: 0.096–0.49, the South Pacific (40°S–56°S), Jung et al., 2014).” (this sentence was written on page 11, line number 29–32 in the previous manuscript).

2. As reviewers noted, the constant value of MSA/SO4 is not sufficient to show biogenic sulfate fraction for the whole year, so please remove all bio/anthro attribution from article (including title, abstract, conclusions). The title should say what was measured: MSA, sulfate, organic mass.

(Response) We thank Co-Editor for Co-Editor's comment. As Co-Editor suggested, we have changed the title of our manuscript from “Characteristics of biogenically-derived aerosols over the Amundsen Sea, Antarctica” to “Characteristics of methanesulfonic acid, non-sea-salt sulfate and organic carbon aerosols over the Amundsen Sea, Antarctica” (page 1, line number 1–2). Besides, we have removed all bio/anthropogenic attribution related to nss-SO₄²⁻ according to Co-Editor's comment as follows:

1. We have revised “To investigate the influence of marine biological activity on aerosols,” to “To investigate characteristics of methanesulfonic acid (MSA), non-sea-salt sulfate (nss-SO₄²⁻) and organic carbon (OC) aerosols,” (page 1, line number 12–13). Also, we have revised “atmospheric methanesulfonic acid (MSA)” to “atmospheric MSA” (page 1, line number 15) and “Unlike MSA, mean non-sea-salt sulfate (nss-SO₄²⁻)” to “Unlike MSA, mean nss-SO₄²⁻” (page 1, line number 17).

2. We have revised “Unlike MSA, mean nss-SO₄²⁻ concentration in the Amundsen Sea was comparable to that in the Southern Ocean, suggesting significant influences of marine biological activity on atmospheric sulfur species in the Amundsen Sea.” to “Unlike MSA, mean nss-SO₄²⁻ concentration in the Amundsen Sea was comparable to that in the Southern Ocean.” (page 1, line number 17–18).

3. We have removed “The results from this study provide significant new observational data on biogenically-derived sulfur and organic carbon species in the Amundsen Sea.” from the abstract (this sentence was written on page 2, line number 2–4 in the previous manuscript).

4. We have revised “To understand the influence of marine biological activities on atmospheric biogenically-derived aerosols in the Amundsen Sea,” to “To understand the influence of marine biological activities on atmospheric marine aerosols in the Amundsen Sea,” (page 3, line number 22).

5. We have removed “(3) estimate the contribution of biogenic nss-SO₄²⁻ to total nss-SO₄²⁻” from the objectives of this study (page 3, line number 26–28). We also have removed “The results from this study provide quantitative insight into ambient levels of biogenically-derived sulfur and OC species in the marine boundary layer in the Amundsen Sea.” from the introduction. (this sentence was written on page 4, line number 1–3 in the previous manuscript).

6. We have removed “biogenically-derived” from the following sentence in conclusion: “Characteristics of biogenically-derived atmospheric sulfur (i.e., MSA and nss-SO₄²⁻) and OC (i.e., WSOC and WIOC) species in marine aerosols,” (page 15, line number 5–6).

7. We have removed the following sentence from the conclusion: “Furthermore, biogenically-derived nss-SO₄²⁻ dominated the atmospheric budget of nss-SO₄²⁻ in the Amundsen Sea, contributing ~86% to total nss-SO₄²⁻ (this sentence was written on page 19, line number 6–7 in the previous manuscript). We also revised “, suggesting significant influences of marine biological activities on atmospheric sulfur species” to “, suggesting significant influences of marine biological activities on atmospheric MSA.” (page 15, line number 8) and “There results were attributed to....” to “The higher MSA concentration was attributed to...” (page 15, line number 8–9).

8. We have revised “biogenically-derived aerosols” to “marine aerosols” (page 3, line number 5).

3. Since the assertion that the nssSO4 is biogenic has little supporting evidence, it is weak and should simply be removed. A stronger case could be made with tracers such as BC and back trajectories (or isotopes) but in lieu of that please just remove that term as it is an unsupported assertion (title, p.19 line 2,6, 21, etc.).

(Response) We thank Co-Editor for Co-Editor’s comment. As we have already responded to Co-Editor’s comments #1 and #2, we have changed the title of our manuscript to “Characteristics of methanesulfonic acid, non-sea-salt sulfate and organic carbon aerosols over the Amundsen Sea, Antarctica” (page 1, line number 1–2). We have already changed or removed the unsupported assertions Co-Editor mentioned. Please see our responses to Co-Editor’s comments #1 and #2. Besides, we have revised “biogenically-derived OC species” to “OC species” (page 15, line number 18–19).

4. No figure in this paper shows a correlation between sulfate, msa, or organic aerosol mass with chl or biomass (no relationship is evident in 7c,d), so remove all discussion of biological coupling: p.19 line 16-18; p.15 line 6 to p.16 line 8.

(Response) We thank Co-Editor for Co-Editor’s comment. As Co-Editor suggested, we have removed all discussion of biological coupling, which was written on page 19, line number 16–18, and on page 15, line number 6–page 16, line number 8 in the previous manuscript (please see the second paragraph in the conclusions on page 15 and the first paragraph of section 3.6 on page 12). We also removed “In addition, the submicron WIOC concentration was quite related to the relative biomass of *P. antarctica*, suggesting that extracellular polysaccharide mucus produced by *P. antarctica* was a significant factor affecting atmospheric WIOC concentration in the Amundsen Sea.” from the abstract. (this sentence was written on page 1, line number 25–28 in the previous manuscript). Besides, we have removed Figures 7e, 7f from the manuscript (page 31), and S5 from the Supplement.

5. This statement “A good correlation was found between the relative biomass of *P. antarctica* and the submicron WIOC concentration.” In the conclusions does not seem supported by a figure; remove. Fig.11 shows relationships to fluorescence intensity not mass concentration, and the latter two things are not equivalent.

(Response) We thank Co-Editor for Co-Editor’s comment. As Co-Editor suggested, we have removed “A good correlation was found between the relative biomass of *P. antarctica* and the submicron WIOC concentration, suggesting that extracellular polysaccharide mucus generated by *P. antarctica* is a significant source of atmospheric WIOC in the Amundsen Sea.” from the conclusions (page 15, the third paragraph). Besides, we have revised “Moreover, the fluorescence properties of WSOC revealed that the majority of WSOC (i.e., protein-like components) was most likely derived from BVOCs as a result of biological

processes of diatoms, by showing the significant positive relationship between the relative biomass of diatoms and protein-like component in marine aerosols in the Amundsen Sea.” to “Moreover, the fluorescence properties of WSOC revealed that protein-like components are most likely produced as a result of biological processes of diatoms.” (page 15, line number 19–20). We have also revised “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.” to “These results suggest that protein-like component is most likely produced as a result of biological processes of diatoms in the Amundsen Sea (page 1, line number 28–30).

6. Section 3.8 does discuss the measured biology and provides interesting context, but a relationship beyond that to the aerosol should not be discussed unless it is explicitly shown.

(Response) We thank Co-Editor for Co-Editor’s comment. As Co-Editor suggested, we have revised “Consequently, our results suggest that protein-like components are 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* (see section 3.6).” to “Consequently, our results suggest that protein-like components are most likely produced as a result of biological processes of diatoms.” (page 14, line number 31–32). We also have revised “The high BIX values also supported that the majority of WSOC was derived from biological processes.” to “The high BIX values also supported that the fluorescence properties of WSOC were influenced by marine biological activities.” (page 15, line number 3).

7. The term enrichment is used problematically in many places, e.g. p.14 lines 1,4, where it is not clear what is enriched with respect to what. Compared to seawater? Other sizes? Clarify or remove this word.

(Response) We thank Co-Editor for Co-Editor’s comment. As Co-Editor suggested, we have clarified the meaning of “enriched or enrichment” by revising it to other expressions as follows:

1. We have revised “WSOC and WIOC were highly enriched in the submicron sea spray particles, ” to “WSOC/Na⁺ and WIOC/Na⁺ ratios in the fine mode aerosol particles were higher, ” (page 1, line number 23–24).
2. We have revised “About ~80% (median values for all data) of MSA was enriched in the fine mode aerosols.” to “About ~80% (median values for all data) of MSA existed in the fine mode aerosols.” (page 7, line number 17–18).
3. We have revised “Both WSOC and WIOC mainly existed in fine mode particles, and the enrichment (i.e., the percentage of WSOC or WIOC present in fine aerosol particles) of WSOC and WIOC in fine mode particles were ~93% and ~74%, respectively (median value for all data).” to “Both WSOC and WIOC mainly existed in fine mode particles, and the percentages of WSOC and WIOC present in fine aerosol particles were ~93% and ~74%, respectively (median value for all data).” (page 11, line number 13–14).
4. We have revised “During the cruise, ~76% of Na⁺, a tracer of sea spray, was enriched in the coarse mode particle (Fig. 7a).” To “During the cruise, ~76% of Na⁺, a tracer of sea spray, was associated with the coarse mode particle (Fig. 7a).” (page 12, line number 17).
5. We have revised “WSOC and WIOC were highly enriched in the fine mode sea spray particles,” to “WSOC/Na⁺ and WIOC/Na⁺ ratios in the fine mode aerosol particles were higher than those in the coarse mode aerosol particles,” (page 12, line number 20–21).
6. We have revised “however, WIOC was much more enriched in the fine mode sea spray particles than WSOC,” to “however, WIOC/Na⁺ ratio in the fine mode aerosol particles was much higher than WSOC/Na⁺,” (page 12, line number 27).

7. We have revised “the higher enrichment of OC in sea spray aerosols,” to “the higher OC/Na⁺ ratios” (page 12, line number 29–30).

8. We have revised “although WIOC was highly enriched in the fine mode sea spray particles (Fig. 7b).” to “although WIOC/Na⁺ ratio in the fine mode aerosol particles was much higher (Fig. 7b).” (page 13, line number 3).

9. We have revised “the high enrichment of WIOC in the fine mode sea spray particles” to “the high WIOC/Na⁺ ratio in the fine mode aerosol particles” (page 13, line number 12).

10. We have revised “However, WSOC and WIOC were highly enriched in the submicron sea spray particles,” to “However, the higher WSOC/Na⁺ and WIOC/Na⁺ ratios were observed in the submicron aerosol particles,” (page 15, line number 14–15).

8. Sections 3.6 and 3.7 are confusing and repetitive, and they should be combined and shortened. Fig. 8 shows neither OC depends on wind speed, so please remove all discussion of that as it is not relevant. Fig. 7c,d shows a negative correlation of OC/Na to wind speed, but I expect that is entirely because Na is positively correlated to wind speed and OC has no dependence. If so, remove this discussion and delete p. 19 line 15-18.

(Response) We thank Co-Editor for Co-Editor’s comment. As Co-Editor suggested, we have combined sections 3.6 and 3.7. First of all, we have changed the title of section 3.6 to “WIOC/Na⁺ and WSOC/Na⁺ ratios and relationships of WIOC and WSOC with Na⁺ over the Southern Ocean and the Amundsen Sea” (page 12, line number 9–10). We also have removed all discussion related to relationships of WIOC/Na⁺ (Fig. 7c), WSOC/Na⁺ (Fig. 7d), WIOC (Fig. 8a) and WSOC concentrations (Fig. 8b) with wind speed (please see section 3.6). Besides, we have removed “We found significant inverse relationships between WSOC/Na, WIOC/Na ratios and the mean wind speed, suggesting that the wind speed affected the organic mass fractions of sea spray aerosols in our study region.” from the conclusions (this sentence was written on page 19, line number 14–15 in the previous manuscript). We also have removed Figures 7c, 7d, 8a, and 8b from the manuscript. In addition, we have changed section number (i.e., section 3.8 to section 3.7) and Figure numbers (e.g., Figure 9 to Figure 8).

Other minor comments: p.2 Line 23: suggest Sanchez et al. 2018 PNAS is more relevant.

(Response) We thank Co-Editor for Co-Editor’s comment. As Co-Editor suggested, we have revised “Lana et al., 2012” to “Sanchez et al., 2018” (page 2, line number 18–19).