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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Uptake selectivity of Methanesulfonic Acid (MSA) on fine particles over polynya regions of the Ross Sea, Antarctica" *by* Jinpei Yan et al.

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Summary/recommendations: This is an interesting paper that provides a generally clear and thorough analysis of particulate-phase MSA in the Ross Sea region. The authors have done an excellent job presenting several different results that provide a clear picture of MSA-containing particles in this region for particles between ~0.1-2 μm. I particularly appreciated their discussions upon why MSA condenses on some particle types but not others. This study can become a useful resource to the community. However, there were details missing in the methods that made a complete evaluation of this study difficult. My primary concern is that the authors did not make it clear

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Discussion paper

whether the datasets were screened for ship exhaust (pollution) contamination. If this was not done, the results would likely be skewed, in particular those that discuss particle speciation. I have outlined a few other concerns in my review below. If the authors have corrected their data for ship contamination and simply didn't include these details in the manuscript, then I recommend that this study be published after the revisions discussed below. If the authors have not corrected their data for ship contamination, then I request either evidence that ship contamination was not a problem during the entire study, or that the data be corrected and reanalyzed. Thanks very much for the comments. The shipboard observations are a challenge and specifically in the marine environment. It is the case that ship emissions may impact the observation data. In this study, to minimize the impact of self-contaminations of the vessel on the observation results, the air inlet connecting to the monitoring instruments is fixed to a mast at 20 meters above the sea surface located at the bow of the R/V. Note that the major pollution source is from the chimney, which is located at the stern of the R/V and about 25 meters above the sea level. Hence, the pollution emissions from the vessel mainly located at the downwind of the sampling inlet, especially when the vessel is running. As high-time-resolution observations are used in this study, the self-contaminations from the vessel have been eliminated from the measurement results. The wind speed and wind directions were also monitoring during the observation period, which were used to determine if the observations were affected by the self-contaminations or not in this study. The data have been corrected to eliminate the impact of ship contamination in this study.

General comments: The paper should be edited throughout for grammar. The authors use tense in confusing ways–they often use past tense when present tense is more appropriate and clear. For example, Lines 43-44: "The chemical components and sources of aerosol particles in the marine atmosphere were rather complicated" (italics mine). The chemical components and sources are still complicated, and the present tense should be used here. Please check tense use throughout. As well, verb endings are often incorrect. For example, line 25: "... deriving from the oxidation. . .".

This should be “derived” here. Thanks for the suggestion, we have revised the tense throughout the manuscript.

Define the size ranges meant by ‘fine particles’ and ‘coarse particles’ in this paper. Different studies use different definitions. In this study the size range of ‘fine particles’ is from 0.1 to 2.0μm, size range of ‘submicron particles’ is from 0.1 to 1.0μm, and the size range of ‘coarse particles’ is from 1.0 to 2.0μm. We have added the definition in the manuscript.

The authors must define each acronym when it is first used. For instance, MP1 and MP2 are brought up in lines 139-144 but are not explained or defined. Same for ‘MA’ (line 148). MP1 and MP2 represent the high MSA population regions, MA represents the high MSA mass region. We have added the description in the manuscript.

There needs to be discussion in the methods of: –Particle size range of the IGAC and SPAMS. In the results, there is mention that particles between .1-2.5 μm were considered from the SPAMS. Is this the size range used throughout the study? Not being able to capture particles <100 nm is a limitation of this study and should be acknowledged and discussed. The measurement particle size range of $\sim$10μm for IGAC, and 0.1$\sim$2.5μm for SPAMS. It is true that particles smaller than 100nm cannot be detected by the SPAMS in this study. Note that most of the MSA particles were in the range of 0.1 to 1.0μm in the marine atmosphere (Ayers et al., 1997), indicating that the MSA particles measured in this study represent most of the MSA particles in the marine atmosphere. We have added the discussion in the manuscript.

–Very important: how the authors corrected for potential contamination of pollution from the ship. Although pollution from the ship likely wouldn’t impact the MSA measurements, it would impact the total aerosol population and mass concentrations. As well, contamination from ship pollution could alter the speciation of the particles, skewing the authors’ results. Did the authors exclude time periods in which the ship pollution would have impacted the measurements? As mentioned above, to minimize the impact

of self-contaminations of the vessel on the observation results in this study. The following methods were used: 1) The air inlet connecting to the monitoring instruments was fixed to a mast at 20 meters above the sea surface located at the bow of the R/V. Note that the major pollution source is from the chimney, which is located at the stern of the R/V and about 25 meters above the sea level. Hence, the pollution emissions from the vessel mainly located at the downwind of the sampling inlet, especially when the vessel was running. 2) The wind speed and wind directions were also monitoring during the observation period, which were used to determine if the observations were affected by the self-contaminations or not in this study. 3) As high-time-resolution observations were used in this study, the self-contaminations from the vessel were eliminated from the measurement results. In this study, the NOx concentration was also monitored simultaneously during the cruise (Fig. SS1). The NOx concentrations were extremely low and remained stable in this study, indicating that the sampling gases were rarely affected by the ship emissions. We have checked the data when high NOx concentrations were present. The data have been excluded, when the observations were impacted by the ship pollution in this study.

–Length of the tubing used for sampling and whether there were corrections for particle and vapor losses within the tubing and associated uncertainties. The length of the tubing is about 20 meter. It is true that the particle and vapor would lose in the tubing. In this study, conductive silicon tubing was used to minimize the particle and vapor lost in the tubing. A high velocity sampling system was also used in this study with a gas velocity of about 4.25m/s in the tubing. The residence time of the gases in the tubing is about 4.7 seconds. The measurements of aerosol particles with and without the tubing have been carried out. There are few differences between the measuring results with and without the tubing. Hence, the particle and vapor losses in the tubing can be neglected in this case.

Line 125: ‘and their populations’ is confusing here. I see later in the text that particle populations are determined by the SPAMS. Please make that clear here. We have

added the description in the manuscript.

Lines 145-147: The authors provide the MSA particle population. It would be of interest to provide the total particle population as well, for comparison. I see this comparison is made in section 3.3; perhaps the authors can refer the reader to that section for comparisons of MSA-containing particles to the total particle concentration. Thanks for the suggestion, we have added the notice to refer the reader to the section for comparisons of MSA-containing particles to the total particle concentration.

Sections 3.3.1-3.3.7: For consistency and to provide a complete picture for each speciation, I suggest briefly including results from Figs 4 and 5 for each species subtype. Figs 4 and 5 are currently inconsistently discussed between sections. (For example, provide the mean fraction that each subtype contributes to the total population [Fig 4] within each section and so forth.) The authors are not limited by space for this journal, and this discussion is currently unsatisfying. Thanks for the suggestion. In this study, we focused on the characteristics of MSA particles, hence, we did not discuss other species in this manuscript to make the manuscript clear and concise. Here, we provided the complete picture for each speciation in the SI (seen in Fig. S6). The mean fraction of each subtype contributes to the total population during leg I and leg II are also illustrated in the SI (seen in Fig. S8). We have added the discussion in this section to smooth the discussion between Fig 4 and Fig. 5.

Section 3.3.7: discuss sources of NOx, HNO3 in the marine atmosphere here We have added the discussion of NOx, HNO3 sources in the marine atmosphere in section 3.3.7.

Figures/tables:

Supplemental figures are not currently referenced in order in the main text. For instance, lines 68-72 jump from Figure S1 to Figure S4. Please update the SI figures to reflect the order they are referenced in the text. We have updated the Figure order in SI.

Figure 3: it is very hard to read the speciation (Na+, C 4 H, etc) on each mass spectra plot. I recommend increasing the font size, if possible. We have increased the font size in Figure 3.

Figure 4: It is very hard to read the legends on this plot. I highly recommend increasing the font size; there is likely enough space to make each legend into 2 columns. As well, the percents in the pie chart are difficult to read. We have enlarged the legend and the percents in the pie chart in Figure 4.

Figure S4: There need to be units on the colorbars for sea ice coverage (presumably percent) and Chl-a. As well, the units can be included in the figure caption (that should be updated to Fig. S4 instead of Fig. 4). We have revised in the Figure S4 in the SI.

Figure 6. I quite like this schematic; all of the text could be larger for clarity. Thanks very much, we have enlarged the text size in the Figure 6.

Technical comments:

I suggest defining 'polynya' the first time it's mentioned in the abstract and main text, as it is not a common term. 'polynya'is an area of open sea water surrounded by ice. We have added the definition of 'polynya' in the abstract and main text.

Line 11: change to "lacking in knowledge" We have changed in Line 11.

Lines 17-18: do the authors mean that MSA uptake favored sea salt particles? Suggest rewording. We have revised in the manuscript.

Line 104: W should be capitalized and there is a missing negative in the denominator of'W/cm -2 ' We have revised in the manuscript.

Line 116: 'cloud effect'is confusing, I suggest rewording. I assume the authors are referring to the loss of data due to clouds? It is the case that the satellite data of Chl-a is affected by the clouds, resulting in the loss of data. We have revised in the manuscript.

Line 119: please define ‘centration data’ We have checked the information. This is a typo here. It is ‘ We used the sea ice concentration data from the daily. . . ’.

Line 179: Do the authors mean “was consistent” instead of “consisted”? We have changed in the manuscript.

Line 278: do the authors mean ‘species’ rather than ‘particles’? For SPAMS detection, particle is determined individually to provide single particle chemical compositions and size. Hence, different types of particles can be identified (such as Na, Mg, SO42- etc.). Here, we obtained the particle count of different types of particle but did not the particle mass concentration with SPAMS. Hence, ‘particles’ was used here.

Line 284: the word ‘except’ here is confusing. It is unclear to me what the authors intend by this statement. We have revised in the manuscript.

References

Ayers, G.P., Cainey, J.M., Gillett, R.W., Ivey, J.P. Atmospheric sulphur and cloud condensation nuclei in marine air in the Southern Hemisphere, Phil. Trans. R. Soc. Lond. B, 1997, 352, 203-211

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2019-811/acp-2019-811-AC1-supplement.pdf

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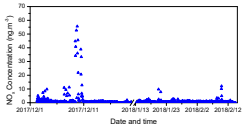

Fig. SS1 Time series of NO_x concentration during the observation

Fig. 1.