

Atmos. Chem. Phys. Discuss., referee comment RC1
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Comment on acp-2022-454

Anonymous Referee #1

Referee comment on "Low contributions of dimethyl sulfide (DMS) chemistry to atmospheric aerosols over the high Arctic Ocean" by Miming Zhang et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-454-RC1>, 2022

Zhang et al. measured ion concentrations in total suspended particulates (TSP) on a ship cruise from mid-latitudes to the Arctic during July-September, and focused on the analysis of MSA data. They found low MSA concentrations over the Arctic Ocean and concluded low contributions of dimethyl sulfide (DMS) chemistry to atmospheric aerosols over the high Arctic Ocean. The gas-phase and particle MSA dataset is useful to the community. However, the scientific justification throughout the manuscript is weak. The manuscript lacks novelty. The research method used is not supportive enough towards their conclusions. It has not reached the standard of publication at ACP, unless major revision is done.

General comments

1. Atmospheric DMS was not measured in this study. The only seawater DMS data was from a previous study in 2014. Therefore, a correlation between DMS and MSA cannot be reached. Both the emissions of DMS, the oxidation, and transport can affect the abundance of MSA observed during the cruise. The authors should not make strong claim of the contribution of DMS chemistry to atmospheric aerosols in the abstract and conclusion. They would not able to quantify the contributions of marine DMS chemistry to the atmospheric aerosols (Line 66).

2. The aerosols collected in this study is TSP. While MSA and sulfate are mostly in fine-mode aerosols, coarse-mode sea salt mass can make a large contribution to TSP. It is not surprising that they found a large contribution (88.78%) of sea salt aerosols to the total aerosols in the high Arctic Ocean on the ship 20m above the sea surface because coarse-mode sea salt could be important. The authors should clarify the potential contribution of

coarse-mode sea salt to TSP and what impacts coarse-mode sea salt can have in the high Arctic Ocean. Please give more information on why we care about TSP over the Arctic, instead of fine aerosols? We don't expect the coarse aerosols to go higher up and contribute to CCN etc.

3. The fraction of MSA in TSP would be significantly affected by the coarse-mode sea salt mass, making it a less useful indicator of biogenic contribution to aerosol mass. Please clarify what we can learn from 1.61% of TSP (comprising fine and coarse aerosols) as MSA, as in the abstract.

4. More information is needed on how MSA is formed in the atmosphere.

Other comments

1. Line 17: It is not clear whether the numbers for MSA concentration are for gaseous MSA or particle MSA. It would be good to list both of them, make comparison, and explain the difference, since the gaseous and particle MSA data is the key in this manuscript.

2. Lines 19 and 20: Is it 88.78% of total suspended particle mass? 1.61% of total suspended particle mass?

3. Line 22: HL is not defined.

4. Line 41: Grammar issue with "such as".

5. Introduction section: Since MSA is a focus of this study, there should be some introduction on how MSA is produced and lost in the atmosphere.

6. Section 2.2: Please provide more information about the sampling inlet setup (20 m above the sea surface) such as the flow and estimate the sampling loss of particles of different size during sampling.

7. Section 2.3: It would be helpful to provide time resolution of the AIM-IC data. Is there any interference for the gas-phase MSA measurements, e.g. from MSIA?

8. Line 113-114: Please provide more information on how different are the formation mechanisms of MSAg and MSAP. Please also discuss the influence of gas-particle partitioning of MSA.

9. Line 115-116: It is not that obvious from Fig. S3 that MSAg concentration decreased with temperature during LL-leg I. Please provide numbers or a scatter plot. In addition, does this negative correlation occur during LL-leg I or during the whole study? Why and why not? Does MSAP depend on temperature? Why and why not? What is the role of temperature-dependent gas-particle partitioning?

10. Line 115: Does SO refers to Southern Ocean? Please define.

11. Line 128: It is not clear how to come to this conclusion "...indicating that the high value of MSAP during ML-leg I may have also been affected by long-term transport...". Please clarify.

12. Line 131: It is not clear how to come to this conclusion "...It should be noted that MSAP and nss-SO₄²⁻ concentrations changed scarcely during this period, suggesting that the DMS chemistry has little effect on the atmospheric aerosols in the high latitude of AO...". Please clarify.

13. Line 134-135: Grammar issue. Should be one sentence.

14. Lines 146 and 150: Na should be Na⁺. Please check throughout the manuscript.

15. Line 149-151: It is really difficult to see by eye that MSA and Na⁺ concentrations increased with wind speed. Please provide more information, such as numbers, scatter plots, etc.

16. Line 168: Grammar issue starting with "Since...".

17. Line 168-169: The authors state "...The variations in the MSA to Na⁺ ratio is useful to understand the contribution of biogenic sulfur species in the marine atmospheric aerosols..." Note that MSA is produced in fine aerosols while a large fraction of Na⁺ is present in coarse aerosols. The MSA/Na⁺ ratio in TSP may not be that useful to understand the contribution of biogenic sulfur species in the marine atmospheric aerosols. The information obtained from MSA/Na⁺ ratio in TSP could be significantly affected by freshly emitted coarse sea salt. Please discuss what that ratio stands for under this

scenario. The aerosols in upper marine atmosphere would be different from the aerosols 20 m above the ocean surface.

18. Line 169-170: MSA/Na⁺ ratio doesn't lead to the conclusion of low MSA contribution from DMS chemistry in the HL. What is the role of Na⁺ here?

19. Line 189: "...SSAs are strongly associated with the wind speed (Fig. S3)..." It is not clear how this conclusion is drawn from Fig. S3.

20. Line 195: It would be helpful to show MSA/nssSO₄ ratio and discuss the biogenic versus anthropogenic sulfur emissions and chemical formation.

21. Line 200: Grammar issue with "such as."

22. Line 213: Grammar issue with "because".

23. Line 227: "...Nss-SO₄²⁻ correlated well with MSA in the HL region, indicating that nss-SO₄²⁻ is mainly derived from the DMS..." Please show the scatter plot to get more information of the correlation. It is not clear to see by eye.

24. Line 230-231: "...MSA is generated through alternative routes, including gas phase reaction and reactive uptake on the existing particles..." Please provide more discussion on the chemical formation of MSA in the atmosphere. MSA formation also takes place in clouds in the marine boundary layer. Would it be fewer clouds in the HL for MSA formation?

25. Line 232-234: Please explain why low temperature was in favor of MSA formation in the atmosphere. Also, please show the scatter plot and correlation coefficient of MSA versus temperature.

26. Line 235-236: Again, please show the scatter plot and correlation coefficient of MSA versus temperature. It is difficult to tell by eye.

27. Line 242-244: "...However, we did not find an obvious relationship between the MSA concentration and RH in the HL region. The MSA levels changed little with the RH in this region (Fig. 4b), indicating that RH had little effect on the MSA formation in the HL

region..." Please explain why.

28. Line 249-250: Isn't DMS flux a function of DMS levels and transfer velocity? This does not logically make sense. Do you mean "transfer velocity increased with wind speed"?

29. Line 265: What type of radicals? How do they affect MSA formation? More discussion is needed.

30. Line 266-268: "...The observation results confirmed that the low MSA concentration was determined by the low DMS concentration in the high AO and further demonstrated that low contribution of DMS chemistry was determined by the low DMS emissions in this region..." Why not determined by the radicals?