Response to reviewer 2

General response:

Dear anonymous reviewer 2,

Thank you for your useful comments on our manuscript "Low contributions of dimethyl sulfide (DMS) chemistry to atmospheric aerosols over the high Arctic Ocean". **We have carefully revised the manuscript as per the comments. Revisions in the text are shown using red highlight. The responses to the reviewer's comments** are marked in blue with the corresponding changes highlighted in red and presented in the following.

Summary/recommendations

The authors present gas-phase MSA and particulate MSA, DMA, SO4, etc. results from ship cruises taken in the northern Atlantic ocean. The results indicate that DMS chemistry may not greatly impact aerosol composition or concentrations in the study region. I think that it is a good look at the problem, but that there is still more research to be done before declaring this finding as fact. The authors did not collect gas-phase DMS, they were only out for a single season, and their study range was small. I think the paper should be published with more caveats and care to not over-state their findings. The paper was well cited and generally well written, but there are several sentence fragments throughout the paper. I have pointed out a few but I do encourage the authors to carefully go through the paper again.

General comments

Introduction: it would be helpful to define the different Arctic regions more rigorously- currently the authors refer to 'low', 'high', and 'coastal'. Please provide latitude ranges or similar.

Response: We revised the introduction as comments. They are all defined in the introduction.

As a general comment, it's helpful to define 'uncommon' acronyms in each major section when they first come up. (For example, AO and PO).

Response: We revised the introduction as comments.

Methods: How did the authors quality control their datasets to remove ship emission influences (that is, emissions from the R/V Xuelong)? This is an important but missing detail.

Response: We did the calibration cruise at the beginning and the end of the cruise. We have already tested it in the lab or on the other cruise in the Southern Ocean (Yan et al., 2019; Yan et al., 2020 a, b) and in the Arctic Ocean (Yu et al., 2020;2021). We deployed a total suspended particulate (TSP) sampler at the top of the mast (20 m height) to minimize the impact of ship emission. Conductive silicon tubing with an inner diameter of 1.0 cm was used to connect to the instrument to avoid the sampling loss of particles. We also removed the data which was impacted by the ship emission.

Methods: there is no discussion of the size range of aerosol collected, which is relevant for climate and human health. Was there any differentiation by size?

Response: Unfortunately, we did not measure particles in a different size. We only focused on the mass concentration of the total suspended particle.

Results: Since there was no direct measurement of DMS in the atmosphere, the findings are somewhat speculatory. The authors should state this limitation and be careful to not definitively state anything. The authors write as if they have solved this particular question for all of the AO (that DMS emissions and chemistry "scarcely impacted the atmospheric aerosols in the high AO" but I don't think this is so definitively solved. This study is a good look but requires more measurements/modeling over more seasons and a much larger area than the ship tracks covered. These caveats need to be discussed.

Response:

It is appreciated that the reviewer pointed out these weaknesses. However, it is very difficult for us to perform a perfect observation during the Arctic Ocean campaign, similar to a typical land station surrounding the Arctic Ocean. We can only obtain a short period of data across the central Arctic during our observation. We presented the details as follows to prove our conclusion of "low contributions of DMS chemistry to atmospheric aerosols over the high Arctic Ocean" was reliable and solid.

Firstly, our previous study reported that deficient DMS levels (<0.5 nmol L^{-1}) and flux (general below 0.5 μmol m^{-2} d^{-1}) were observed in the high latitude (Zhang et al., 2021, in Global Biogeochemical Cycles) due to the nutrient limitation and heavy sea ice cover. Even after sea ice retreat, the DMS levels remained unchanged, and the DMS flux slightly increased to only 1.2 μmol m^{-2} d^{-1}. For the annual changes of DMS levels and flux, we can also conclude that these values were difficult to be changed as the upper layer water mass of Arctic Central was dominated by an increasing fresh water with very low nutrients (Figure 1, Zhang et al., 2021). Although we did not detect the atmospheric DMS and flux in the 2018 cruise, the low DMS flux and air DMS levels could be expected during the 2018 campaign.

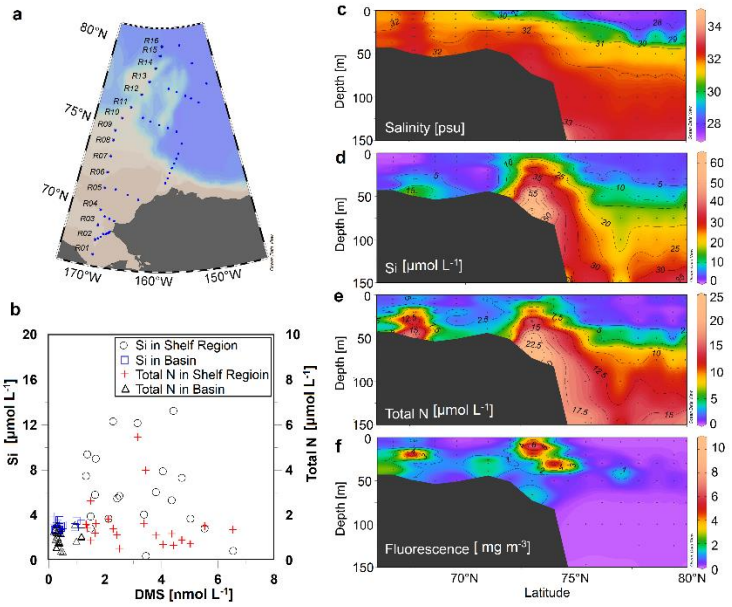

Figure 1 Distribution of nutrients in relation to surface seawater dimethylsufide (DMS) in the western Arctic Ocean. (a) Locations of conductivitytemperature-depth (CTD) stations along T1. The section R, panels (c)–(f), correspond to stations labeled R on the map in (a). (b). Relationships between DMS and Si and total N in surface water of all stations in (a). The panels on the right show depth profiles of (c) salinity, (d) Si, (e) total N, and (f) fluorescence along section R. (Zhang et al., 2021)

Secondly, we found that the MSA mass concentrations decreased from the low latitude Arctic to the high latitude. If the air mass contained high MSA levels could be rapidly transported to high latitude regions, we would observe high MSA in the high Arctic. However, we found deficient gas phase and particle MSA levels over there. In addition, MSA is well known only from DMS oxidation in the atmosphere. This means that the oceanic DMS emission greatly

influences the atmospheric MSA production. Thus, the low emission of DMS in the high Arctic was possibly the main reason for extremely low MSA there.

Thirdly, our observation of aerosol compounds indicated that the contributions of biogenic sulfur decreased significantly (only 1.61%) in high latitude Arctic (Figure 4 in manuscript). This result suggested that the low DMS chemistry contribution to aerosol was found in the high Arctic.

Lines 107-109: provide latitude ranges for LL, ML, SL, and HL. These ranges appear to be in Table S3 but I think this is relevant enough to be in the main text as well.

Response: We agreed that and Table S3 was moved to the main text.

Lines 113-114: "The variations in the MSAg level were not always consistent with those of the MSAp along the cruise tracks, indicating that the formation mechanisms of MSAg and MSAp from oxidation of DMS are different." This is an interesting statement, I think the authors could expand upon it some. Consider providing a supplemental figure or table showing the ratio of MSAg / MSAp. This may be of research interest to some.

Response: Thanks for the reviewer giving this good suggestion. Actually, we are preparing another manuscript to focus on this topic. And we want to compare the difference in this ratio between the Arctic Ocean and the Southern Ocean. Additionally, we have already published one paper about this topic: observation in the Southern Ocean (Yan et al., 2019 c).

Line 119-120: "The variations in MSAp levels in the SL region during leg I and II were associated with the phytoplankton activity in these regions (Fig. S6)." It is not easy to make this association based on looking at Fig S6 and Fig 1. The authors need to do more work here to make this association more clear. It would be nice to have some sort of statistical measure of how well associated MSAp and phytoplankton are - this could be an average for the region or more high-definition. The authors may also consider adding a supplemental figure that shows this association more clearly, like the ratio of phytoplankton to MSAp along the ship tracks, if possible.

Response: Thanks for the reviewer giving this good suggestion. We did not make the

correlations between the Chl a and MSAp in distinct regions. Our description is mainly based on the difference of Chl a, an indicator of phytoplankton activity, values in a large-scale scan. As in figure S6, we could easily find the difference of Chl a in the distinct regions. This difference was consistent with the variations in MSAp along the cruise track. Our purpose here was to note that the high MSA occurred in the regions where phytoplankton activity was high.

Line 209-210: I recommend that the authors state "That is likely the reason why…" as the authors did not do a comprehensive analysis (for example, a principal components analysis) on where emissions were coming from for each species.

Response: Sure, we agreed with this comment, and the sentence was revised in line 230.

Line 212: do the authors have the measurement precision to state that a ~1% difference in amine is significant?

Response: Our measurement precision for amine is 0.12 ng m^{-3}. We did not consider that the 1% difference in amine in SL and LL regions was significant. We just presented the difference but no more description or explanation for this small difference. And, we revised these sentences in lines 230-233 as follows:

Similar to NO_x^-, high fractions of amine aerosols were found in the SL and LL regions. But the amine fraction (5.23%) in the SL region was higher than that (4.2%) in the LL region because of amine emissions from marine surfaces.

Technical comments

Line 22: HL not defined in the abstract

Response: It was revised in line 27. "high latitude (HL) region"

I recommend against using undefined acronyms in your short summary.

Response: It was revised as commented. We did not use acronyms in the short summary.

Line 29: "The Arctic is known for its amplified rate? of global climate change…" there is a missing word, perhaps the authors meant rate? Or Amplification instead of amplified?

Response: We replace the amplified with "amplification".

Line 34: no comma needed between "increase, when"

Response: We deleted the comma.

Line 48 - I recommend adding a citation for "The loss of sea ice in the AO promotes the air-sea

exchanges and subsequently increases dimethyl sulfide (DMS) emissions." - unless the Sharma et al 2012 citation was appropriate for this statement as well? This isn't clear to me.

Response: Sure, we added a citation (Galí et al., 2019) here in line 54.

Galí, M., E. Devred, M. Babin and M. Levasseur (2019). "Decadal increase in Arctic dimethylsulfide emission." Proceedings of the National Academy of Sciences 116(39): 19311-19317.

Line 53: "The observations carried out in the Ny-Ålesund revealed…" Do the authors mean Ny-Alesund region?

Response: Yes, Ny-Ålesund region.

Lines 98-99: the sentence here is a sentence fragment.

Response: We revised the sentence as follows in lines 109-110.

The remote sensing chlorophyll-a data was obtained from MODIS-Aqua (http://oceancolor.gsfc.nasa.gov) with a spatial resolution of 4 km.

Results: the authors are inconsistent in whether or not they capitalize 'leg'.

Response: We checked the whole manuscript. If the leg started the sentence, we used the capitalized "Leg".

Line 116: I think the authors meant "confirming" not "conforming"?

Response: Thanks. We revised the sentence.

Line 135: the sentence here is a sentence fragment, I believe it belonged to the previous sentence?

Response: We revised all these sentences from line 152 to line 156 as follows:

Although obvious sea ice retreat occurred from July to September (Fig. S7), the chlorophyll-a remained at an extremely low level in this region possibly leading to very low DMS emission (Zhang et al., 2021, Figure S6). This would be the main reason for low observed sulfur aerosols. In contrast, our result differed from the observation in the high latitude SO that high atmospheric DMS chemistry contribution was reported when the sea ice retreated (Yan et al., 2020 a).

Lines 167-168: the sentence here is a sentence fragment, I believe it belonged to the previous sentence?

Response: We revised the sentence in lines 187-189 as follows:

Because Na^+ and MSA can be used as a marker for sea salt aerosols and biogenic sulfur aerosols, respectively, the variations in the MSA to Na^+ ratio is useful to understand the contribution of biogenic sulfur species in the marine atmospheric aerosols.

Line 213: sentence fragment.

Response: It was revised in lines 231-233.

But the amine fraction (5.23%) in the SL region was higher than that (4.2%) in the LL region, which could be attributed to the higher biogenic activity in SL than LL (Fig. S6), and the amines mainly originated from marine surfaces.

Line 258: suggest "strong positive…" rather than good. This is a more common way of phrasing it. (E.g. strong, moderate, weak correlations)

Response: It was revised as comments in line 281.

Line 277: Suggest "Sulfuric acid is more effective for new particle formation"

Response: It was revised as comments in line 300.

Line 305: The authors may have meant "concerning" , not "concerned".

Response: It was revised as comments. We used "be highly paid attention" here.

Citation for "Croft, B., Martin, R. V., Leaitch, W. R., Tunved, P., Breider, T. J., D'Andrea, S. D., and Pierce, J. R.: Processes controlling the annual cycle of Arctic aerosol number and size distributions, Atmos. Chem. Phys., 16, 3665–3682, doi:10.5194/acp-16-3665-2016, 2016. " - fix the D'Andrea, and this is erroneously "Crof et al 2016" in the main text, line 59. Fix to Croft.

Response: Sure. It was revised as comments.

Figures/Tables

All figure with spatial maps (e.g. Fig 1): I recommend considering explicitly drawing out the different characterized regions (ML, SL, HL, LL). For example, you could make the land/sea masses a lighter gray, then add dashed lines to separate each region + a label on the figure for

each region. This would make the results more distinct.

Response: It was revised as comments:

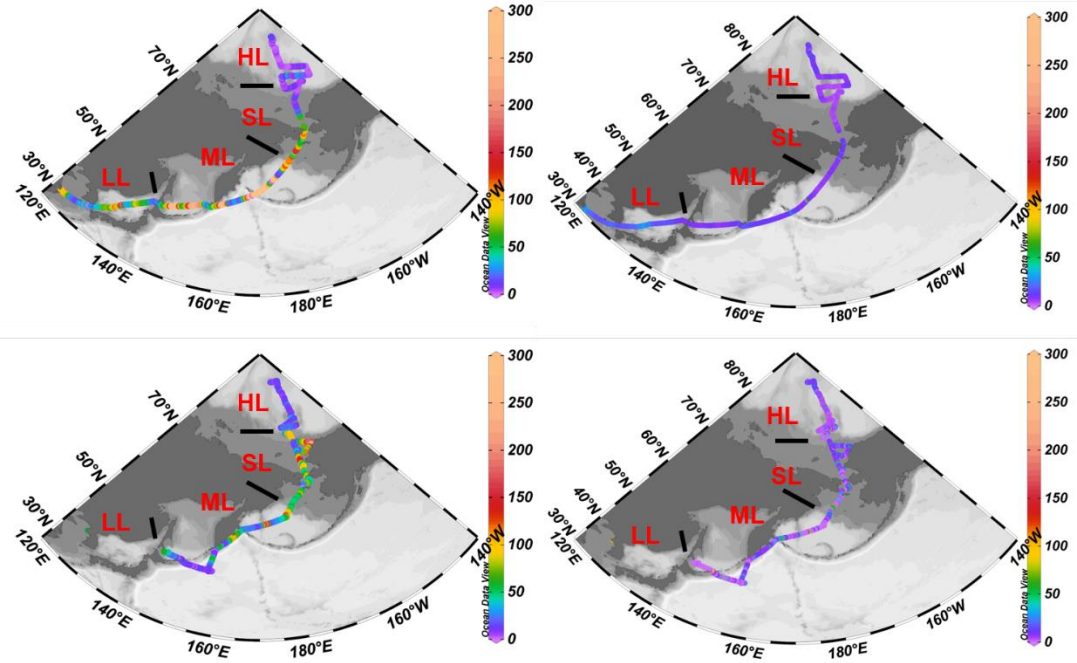

Fig.1 Spatial distribution of the gaseous and particulate MSA concentrations, (a) Particulate MSA concentration during leg I (ng.m^{-3}); (b) Gaseous MSA concentration during leg I; (c) Particulate MSA concentration during leg II (ng.m^{-3}) and (d) Gaseous MSA concentration during leg II. Noted that the distinct regions are marked, and we used the blank line indicating the boundary. LL, ML, SL and HL indicate low latitude (South of 45 ºN), mid-latitude (45 º – 60 ºN), sub-high latitude (60 º – 75 ºN) and high latitude (75 º – 85 ºN).

Figure 2: it's not clear to me why ML and HL have dark cross hatchings over them? This makes the figure harder to read for me. I think the vertical lines are enough to provide distinctions. If the authors want to keep the cross hatchings, please make them a much lighter color, like light gray.

Response: It was revised as comments.

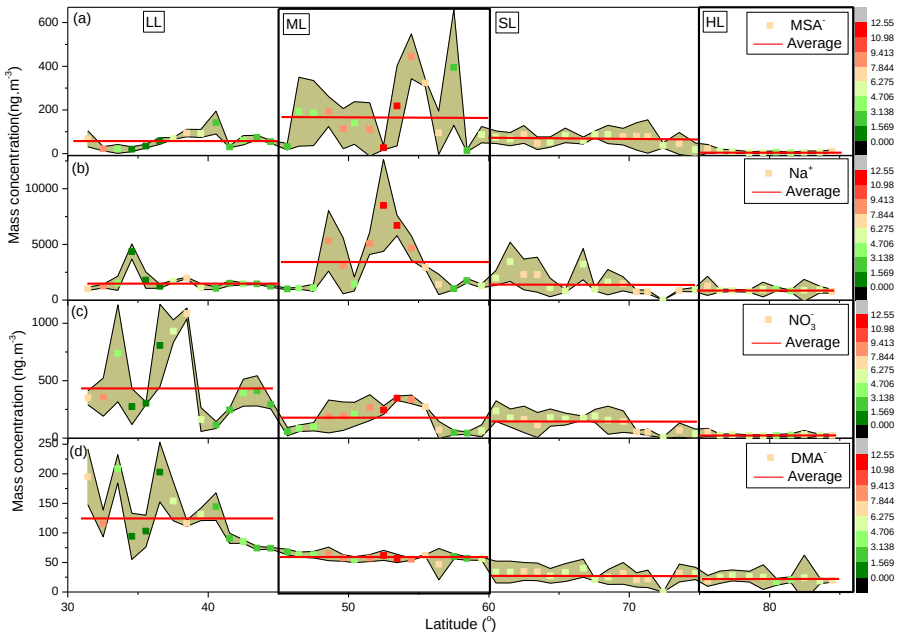

Fig. 2 Latitudinal distributions of different aerosol species from 30-85°N, (a) MSA concentration; (b) Na^+ concentration; (c) NO_3^- concentration; and (d) DMA concentration.

Table S3, Figure S3, Figure S4: define ML, SL, HL, LL in the table or figure caption

Response: Sure. It was revised as comments.