

## Authors' response to Reviewer #5

Reviewer comments in black, Author responses in Red

In this paper, the authors present annual cycles of some important gases that may act as precursors to aerosol formation. They attempt to interpret these either as caused by long range transport, using FLEXPART inverse modeling, or from local sources, using rather hand-waiving arguments but no observations. The motivation is to understand the environment for aerosol formation, but the formation of aerosols itself is not supported by any direct observations. The primary results are sort of unique and should be published, the inverse modeling is what it is and but the results are in my mind a bit unclear and unbalanced since the long-range aspects is supported by rather advanced, but uncertain, methods while the discussion on local sources are rather speculative. Based on this, I believe the manuscript could benefit from a clearer focus on what is the core data provided, what are the central conclusions and what is speculation; hence I recommend major revision.

We thank the reviewer for their comments and criticisms of this manuscript. We have addressed each of the reviewer's comments below. We appreciate that the reviewer pointed out a major need for revision for several aspects that were not well explained. This helped us to streamline and clarify the provided information. We address these aspects point by point in the comments below. Other suggestions by the reviewer extend well beyond the scope of this manuscript, but they are good points that could be addressed in the future. We pointed out these cases in our responses to their comments. The reviewer did highlight several points related to our source region identification methods, using FLEXPART and inverse modelling, that required further description in the text, as we have addressed in the specific comments below. In addition, we took the reviewer's suggestion to weaken the language in parts of discussion to highlight that there are aspects of this work that remain uncertain and could warrant future investigation by the scientific community.

Major comments:

I have no problem with the quality and handling of the observations. A ship is about the worst place one can do measurements but pollution and other effects on the observations are in my opinion handled with care. My concerns are instead in the modeling and the interpretation of the results.

We thank the reviewer for their general agreement with our handling and presentation of pollution in this data, as we did our best to maintain transparency with respect to pollution and its influence in the datasets that accompany this manuscript. Ship based campaigns are indeed challenging in this regard, however, there are currently very limited options to obtain measurement data in the central Arctic Ocean otherwise.

First, I'd like to underscore that while measuring the complete annual cycle is unique for the central Arctic, this is but one annual cycle and how representative this is for the true (average) annual cycle is unclear. I would therefore have liked some of the space devoted to rather speculative discussions on various local sources to have been spent on documenting how representative this year is compared to other years. And while on the topic of representativity; while I understand there may be source regions that does not impact where MOSAiC happened to be, I would like some discussion on

how regional or local is the source contributions from long-range transport are? In short, now we have one annual cycle at one (or series of) location; what does this mean for pan-central-Arctic conditions? Would the very localized Russian smelter emissions, that dominate the SO<sub>2</sub> results, be the same if MOSAiC had been say in the Beaufort gyre? After all, if the motive is Arctic aerosol precursor gases” and how they may change in the future, we need to see the whole Arctic.

We understand the concerns of the reviewer. However, it is challenging to compare this annual cycle with previous years due to the general lack of observations of these precursor gases within the central Arctic Ocean. Understanding these observations over multiples years or from a pan-Arctic perspective are out of the scope of the current work. The other key factor limiting a comparison between our annual cycle and other years is the different locations at different times of year, as also pointed out by the reviewer. We feel that we adequately expressed that localized emissions, such as from the Russian smelter, were observed at *Polarstern’s* location due to evidence and insights obtained from the FLEXPART simulations and inverse modeling. In addition, while our measurements are restricted to specific locations at specific times, our observations generally agree with the general knowledge on prevailing source regions. For example, emissions from Siberia are known as a dominant source region of atmospheric transport in the central Arctic during winter (Moschos et al., 2022; Stohl, 2006; Stohl et al., 2013). It is important to point out that the FLEXPART simulations used emission fields from a known emission inventory (ECLIPSE v6b) and the inverse model made use of the measured timeseries data from the MOSAiC expedition and the FLEXPART footprint emission sensitivity (FES) to assign emissions to grids and subsequently simulate concentrations from those emission grids. Interestingly, the source regions identified are comparable between these different methods. Although the source region analysis is qualitative in nature, which we expressed in the manuscript on Line 288 of the main text, it is less “speculative” that attempting to extrapolate the concentrations of these vapors over a larger area in the Arctic. For this reason, we focused our analysis on the seasonal cycles of these vapors and on various source regions that we could identify, qualitatively, to have an influence on their concentrations.

To clarify this in the text, we have toned down various phrases which imply that our observations could be extrapolated to the entire central Arctic Ocean region. These changes are as follows:

- Line 128: “The results of this study are valuable for improving our understanding of these key aerosol precursor vapors and their potential sources from direct observations within the central Arctic, a region which is currently undergoing rapid changes.”
- Line 289: “These datasets offer unique insights on the seasonal variability of these vapors during the MOSAiC expedition in the high Arctic.”
- Line 436: “Our observations highlight the combined influence of both natural and anthropogenic sources of atmospheric SA during the year, where the highest concentrations occurred in winter/spring.”
- Line 462: “This is particularly clear during March and April when the MSA concentration in our measurements starts to increase despite the observation that chl-a concentrations are still low at the northernmost latitudes.”
- Line 597: “Our observations suggest that IA concentrations are also strongly linked to seasonal changes in sea ice conditions.”
- Line 650: “Our results show the influence of both natural and anthropogenic sources on SA concentrations.”
- Line 654: “Localized anthropogenic emissions in Siberia/Northern Russia, especially from the region of Norilsk in Northern Russia, contribute substantially to our observed SA concentrations during winter.”

- Line 658: “Our analyses additionally show that biological activity in the open ocean areas south of the marginal ice zone within the Arctic region contributes to enhanced MSA concentrations, an important component of aerosol formation and growth, during late spring through summer.”
- Line 663: “Transport from regions south of the marginal ice zone appear to be the primary driver of MSA concentrations in our observations over the sea ice.”
- Line 675: “Our observations provide circumstantial evidence that the current seasonal cycles of SA, MSA, IA, and SO<sub>2</sub> in the central Arctic Ocean are linked to sea ice conditions and solar radiation due to their role in biological activity and air mass transport from southern regions.”

Other examples of where the language of our conclusions was weakened are given in our responses to the detailed comments below.

This could have been addressed using for example CAMS; come to think of it, a comparison to – or maybe even an evaluation of – CAMS would have been really interesting and I’m a bit surprised to see that CAMS is not even mentioned. The inverse modeling is what it is. There is a saying about models: “shit in, shit out”. I’m not saying these results are shit, I’m just observing that 30-day back-trajectories are awfully long and uncertain, and that the results are very much dependent in the emissions inventories used. It also seems to be a systematic problem with limiting the domain at 60 °N.

The reviewer raises an important point here. Future analyses could implement and use our dataset to extrapolate over the larger central Arctic region using further modelling such as CAMS, but we conclude that such an analysis is beyond the scope of the work presented here. Our work intends to highlight the novel, year-long seasonal cycle of these condensable vapors and suggest the prevailing source regions that contributed to their concentrations in our observations. This is apparent from the title of the manuscript. To address the concerns of the reviewer about the scope of our work, and to clarify the extent to which we can extrapolate our observations, we have updated the title to indicate that our observations are reported “during the MOSAiC expedition.” The title now reads:

“The annual cycle and sources of relevant aerosol precursor vapors in the central Arctic during the MOSAiC expedition.”

We thank the reviewer for pointing out the discussion of the 30-day backward particle dispersion simulations, as this is a point that should be clarified further in the text. First, we would like to specify that FLEXPART is an air tracer dispersion model, which is different than the back trajectory analyses to which the reviewer refers to in their comment. Dispersion simulations are systematically better than backward trajectories in the sense that they account for the stochastic nature of atmospheric transport and that the source receptor results are in essence probabilistic (given the use of thousands of particles rather than few single-particle back trajectories) (Stohl et al., 2002).

Please note that we stated that the FLEXPART simulations were carried out for periods “up to 30 days backward in time” on Line 234. This statement was meant to generally describe the FLEXPART simulations that were conducted during the MOSAiC expedition, which are openly available for scientific community to use. These FLEXPART simulations are available for 1, 7, 10, and 30 days backward in time. We did not intend to imply that we used 30-day simulations for each of our

analyses in this work, but we understand the confusion, as we did not explicitly state the length of the FLEXPART simulations that we used in every instance. For example, refer to Fig. S12 which utilized 10-day backward simulations. For clarity we have added the description of the FLEXPART air tracer ages used for various aspects of our analysis in the text. Refer to our response to the first detailed comment below for the locations where the text was updated.

We would also like to point out that the FLEXPART simulations were not performed specifically for our analysis; we simply used the results of the FLEXPART simulations as a tool to gain further insights into possible source regions in our analysis and used inverse modelling to fill in gaps for species not in the emission inventories. It is for this reason that the domain was limited to 60 °N, as the simulations were performed independently from the analysis presented in this manuscript.

It would have been useful to show the emissions that goes into these calculations in the paper. For example, the inverse modeling of SO<sub>2</sub> does not fit all that well with the observations; most of the modeled annual cycle have values well above the instrument threshold, while there is almost no useful SO<sub>2</sub> observations at all (above the instrument threshold). While the few spikes in SO<sub>2</sub> coincide with the inverse modeling maximum, the modeling indicates a sharp increase in winter starting already in December, the measured SO<sub>2</sub> have as low values as there is later, in summer, and still the modelled concentrations for summer is as high as early in the year. I guess I wonder how the inverse modeling works when there isn't any data? I have no problem believing the peak in the inferred concentrations is connected to the peaks in the observations; I just think the linkage is weak.

We do not understand the reviewer's concerns presented in this comment. The SO<sub>2</sub> emissions included in the emission inventory, as well as their geographic extent, are presented in Fig. S11. We also specified in the caption of Fig. S11 that the emission inventory uses anthropogenic SO<sub>2</sub> emissions and that FLEXPART converts this SO<sub>2</sub> to particulate sulfate, or SO<sub>4</sub>-S. This was also specified on Line 366, where we stated that "FLEXPART treated anthropogenic SO<sub>2</sub> emissions as SO<sub>4</sub>-S, which yields the SO<sub>4</sub>-S weighted influence from anthropogenic sources." Please also note that we explicitly referred to Fig. S11 on Line 371: "Refer to Fig. S11 for a more detailed description of anthropogenic SO<sub>4</sub>-S emissions from each source region in the emission inventory."

The concentrations simulated by the inverse model are also shown in their respective figures (Figs. 4, 5, & 6), both as the source region footprint maps (panel a) and then their simulated influence during the year (panel b). These footprint maps were determined using the inverse model itself, as described in detail in Section 2.6 of the methods. These figures clearly show the emissions and their geographic locations, determined from the inverse model, that were combined with the FLEXPART air tracer data to calculate the simulated concentrations presented in panel b of these figures.

We disagree with the reviewer's statements about the inverse model results of SO<sub>2</sub> in Fig. 4. There are two separate y-axes in Fig. 4: the left axis for the SO<sub>2</sub> mixing ratios simulated by the inverse model and the right axis for the measured SO<sub>2</sub> mixing ratios. None of the simulated SO<sub>2</sub> mixing ratios exceed the instrumental detection limit of 1 ppb. The detection limit associated with the measurements, indicated by the grey shaded area, may have caused confusion here. To clarify that the shaded region showing the detection limit is associated with the measurement data on the right axis, we added a statement to the figure caption to state this on Line 207:

"The measured SO<sub>2</sub> mixing ratio, presented as a rolling 10-minute median during the year, is included on the right axis for context. Note that the shaded area shows the regions where the SO<sub>2</sub> measurement data are below the detection limit."

We do not understand why the reviewer suggested that the inverse model shows a sharp increase in SO<sub>2</sub> in December, as this is not shown in the data presented in Fig. 4b. To calculate the simulated concentrations, we used the complete SO<sub>2</sub> timeseries for the year, including the data below the detection limit. While the values below the SO<sub>2</sub> detection limit are not relevant for quantitative analysis, these low values could still be used to determine the footprint map in the inverse model (i.e., identifying regions with very low emissions). In general, the inverse model and source region identification work are qualitative, as we specified on Line 288 of the main text. The key point here, with which the reviewer agrees, is that the simulated SO<sub>2</sub> mixing ratios spiked while we observed measured spikes in SO<sub>2</sub> mixing ratios that correspond to air masses from the Russian smelter region, which is also consistent with geographic extent of known sulfur sources in the ECLIPSE v6b emission inventory data as shown in Fig. S11.

The SA source is very diverse compared to that of SO<sub>2</sub> and almost all of it is over land surfaces (except for the Labrador Sea and a little in the Bering Sea). Prudhoe Bay is discussed and while it does fall within the green box, it seems like the largest sources in that box are further south, over land. In the discussion the ocean and DMS is discussed as a source, however, the modeling has no chemistry and land surfaces seems to dominate as source regions. There are no measurements of DMS; instead MSA is presented. But MSA lifetime is order of a few hours (line 519), so it is in fact incorrect to even talk about long-range transport of MSA (such as e.g. line 507). It has to be DMS that is emitted, transported and then oxidized, to MSA and also SO<sub>2</sub>. To deal with this, chlorophyll-a is used as a proxy - for what, DMS or MSA emissions? It seems to me there are an awful number of hopeful guesses in this chain that I think is not very clearly described. That most of the DMS is probably long-range transported is no news; that has been published decades ago. Much of the source regions for MSA (or is it DMS?) are discussed to be tied to biological activity in the MIZ, but it appears that the authors have no clear picture of where the MIZ typically is located during the biologically most active part of the season. In the North-American sector, Labrador Sea qualify, but else most of the sources are – again – over land. In the North-Atlantic, the source regions are way south of where the MIZ would be; there's for example nothing in the Greenland Sea/Fram Strait, nothing north of Svalbard and nothing over the Siberian shelf area. Where does the DMS emitted here end up if not in the Arctic? This goes back to my previous question about locality; how local are the impact from different source regions?

It is unclear why the reviewer expresses such confusion between DMS and MSA in this manuscript. It is true that DMS is emitted, which is then subsequently oxidized during transport to form MSA. We stated this repeatedly throughout the text, for example in the following locations:

- The paragraph in the introduction, starting on Line 64.
- Line 82: “The concentration of gaseous DMS and its oxidation products (SA and MSA)...”
- Line 293: “...during which we expect DMS emissions that lead to the observed maximum MSA concentrations.”
- Line 469: “...was used to evaluate the connection between DMS emissions (and subsequent formation of MSA) with biological activity...”
- Line 479: “...increase in chl-a and MSA concentrations is unsurprising, as gaseous DMS, and subsequently MSA, is a product of biological activity...”
- Line 490: “...potential air mass exposure to oceanic regions with biological influence, and hence potential DMS (MSA) emissions.”
- Line 509: “...or important sources of DMS, the precursor of MSA, ...”

- Line 525: "...we can infer that MSA production from DMS..."
- Line 527: "...we conclude that transport of DMS from the regions > 60°N in Fig. 6a, followed by subsequent chemical processing during transport, could explain our MSA measurements..."
- Line 608: "...organisms that produce DMS (MSA)..."

Based on this, and the other statements that the reviewer pointed out about lack of chemistry in the models used in our analysis, it should be clear that any reference to emissions during the discussion of MSA refer to emissions of DMS, followed by the subsequent oxidation and production of MSA. We made it a point to mention this in the discussion of each aspect of the MSA discussion, in addition to the points already mentioned (emissions: Line 496; transport: Line 517, Line 520-524; and the chl-a proxy, Line 470).

We also do not understand why the reviewer suggests that most of the sources of MSA were determined to be above land. From Fig. 6b, many of the inverse model polygons are located over the ocean or near coastal areas. The two exceptions to this are polygon "a" and "f", or the blue and brown polygons, respectively. We would like to point out that for these polygons, we wrote the following in the figure caption on Line 537 as follows:

"Due to the limited domain of the FLEXPART simulations (> 60°N), source regions polygons "a" and "f" may represent the contribution of MSA transport from regions further south than the polygons depicted on the map, such as the oceanic regions on the western coast of North America and the Bering Sea, respectively."

This is also explicitly stated on Line 505:

"Note that due to the limited domain of the FLEXPART simulations (> 60°N), source regions polygons a and f in Fig. 6a may represent the contribution of MSA transport from regions further south than the polygons depicted on the map, which could be associated with oceanic regions on the western coast of North America and Bering Sea, respectively."

Given the knowledge that DMS can be transported over distance, as pointed out by the reviewer, and subsequently oxidized during transport, this is a plausible hypothesis.

We also made it a point to discuss the influence of transport and air mass history for MSA. Emissions alone are not enough to describe measurements over the Arctic. There must be emissions combined with transport. We stated this on Line 487, where we introduced the chl-a proxy:

"The source regions of the observed air masses in the central Arctic would need to correspond with the regions of enhanced biological activity to explain the MSA measured at the ship. Therefore, we coupled the FLEXPART air tracer simulations with the oceanic chl-a concentrations to calculate an index that quantifies potential air mass exposure to oceanic regions with biological influence, and hence potential DMS (MSA) emissions."

To again address the reviewer's comments about locality of emissions in the Arctic—we are reporting observations from a given point in time and space across a large region. It's reasonable to assume that emissions of DMS from other known source regions, as mentioned by the reviewer, could end up in the Arctic, but these regions were not identified to be influential in our measurements at *Polarstern's* location at those times of year. We are not arguing against other sources—we are simply identifying sources associated with our dataset, which still adds value given the limited number of

observations in the central Arctic region—particularly for the precursor vapors highlighted in this study. Again, to make this clearer, we have added “during the MOSAiC expedition” to the title of the manuscript.

Finally, the results for IA are very interesting but the hand-waiving on the reasons for the two peaks, in spring and autumn, is not very impressive. Almost no references are given and no observations are offered to support the arguments of melting and refreezing blocking up the brine channels(?). This is nothing but a hypothesis; an interesting one but it feels the authors dwell on this for quite a while in the hope to convince the reader, while the fact is that we have clue. I stagger at the words “provides evidence” (Line 624); I can’t find a shred of “evidence” here other than what could be called “circumstantial evidence”!

The mechanism of IA formation and its sources in the atmosphere are still not well understood and require further investigation, which is the key takeaway from Section 3.3. We do not intend to convince the reader of anything in this section, but rather, we present a discussion of what has been hypothesized about the seasonality of iodine compounds from previous studies and compare them with our study. We do not understand the reviewer’s comment that there are no references given, as our discussion includes a number of citations to previous studies. In fact, we cited various sources that identify seasonal peaks in iodine compounds and others that propose mechanisms that could support why these peaks are observed, including the study that the proposed mechanism of iodine compound transport through brine channels before the melt season. We again want to point out that the brine channel transport is a hypothesis presented from previous decades by Saiz-Lopez et al. in 2007 and 2015, which was cited appropriately in the text; we are not proposing a new mechanism here. We have changed the wording on Line 624 from “provides evidence” to “suggests”, according to the reviewer’s concern with our statement about the studies that discusses iodine transport through brine channels in the sea ice.

Our analysis of the seasonal cycle in this work is not sufficient to resolve these processes, but a more detailed analysis of IA is planned to address some of these ongoing hypotheses. We specified this in the main text on Line 648:

“However, our analysis focuses on the seasonal cycle, which is not sufficient to resolve the relative contributions from these processes on IA concentrations. As such, atmospheric iodine processes, especially in the Arctic, require further investigation. A more detailed analysis of atmospheric IA formation mechanisms during the MOSAiC expedition will be given in a separate study.”

We have also removed the statement about brine channels in the paragraph discussing IA in the conclusions on line 670:

“In addition, the thinning sea ice could facilitate the exchange of iodine into the atmosphere and further reaction with O<sub>3</sub> to form IA.”

Finally, for a paper that confesses to not deal with aerosols in general or especially not with NPF, there’s quite a bit of text on it.

This comment appears to be a follow up to one of our responses to reviewer #4, where we stated:

“Please note that this paper is not focused on the role of precursor gases in aerosol formation and growth, and any mention of these processes in the introduction is used to give context for our measurements and analysis. The scope of this work focuses on understanding the seasonal cycle of vapor that are known to be potential aerosol precursors and the observed source regions that contributed to their concentrations in our measurements.”

To highlight why these aerosol precursor vapors are relevant, it is necessary to include discussions on their potential implications for aerosol process or new particle formation (NPF). Otherwise, there is no context for our measurements. The observations and the science presented in this manuscript do not focus on aerosol processes or NPF, but the paper would be incomplete without discussion connecting these condensable vapors to aerosol processes.

Detailed comments:

Line 234: Awfully long trajectories(!); the added uncertainty from this should be addressed. There is a literature on this; use it.

As mentioned above, the statement about FLEXPART simulations “up to 30 days backward in time” is general to the FLEXPART data produced during the MOSAiC campaign, but it is not specific to the length of the dispersion simulations used in each aspect of our analysis. For example, we specified for the chl-a influence index in the caption of Fig. 1 on Line 303, as well as in the caption of Fig. S12 in the supplemental information that we used 10-day backward simulations. Backward dispersion simulations up to 30 days were used in the inverse model, but this should not be a major concern as more weight was placed on the shorter simulations, and many of the simulated particles were outside of the domain after 30 days. We have updated the main text to specify the length of FLEXPART simulations used for each aspect of the analysis as follows:

- Line 226: “Specifically, we used the ECLIPSE v6b (Evaluating the Climate and Air Quality Impacts of Short-Lived Pollutants) emission inventory and 10-day backward air tracer simulations from FLEXPART to estimate source regions of anthropogenic sulfur, from SO<sub>2</sub> emissions, as described in section 3.1.”
- Line 283: “The calculation of these emission fields in the inverse model used the FLEXPART FES air tracer from 1, 7, 10, and 30 day simulations backward in time, where the shorter simulations were given more weight in the estimated emissions.”
- Line 495: “The index, called the sea surface chl-a influence index, was obtained by multiplying the residence time (in seconds) of the FLEXPART air tracer (based on 10-day backward simulations) below 100 m altitude with the corresponding chl-a concentration maps (in mg·m<sup>-3</sup>).”

Without directly providing the citation the reviewer is referring to here, we can only assume that they are referring to one of the studies from ASCOS (Tjernström et al., 2012, 2014). We feel that our updated descriptions specifying the lengths of the backward particle dispersion simulations used in our analyses are sufficient, as well as our claims that the source region analyses are qualitative in nature.

Lines 335-341: Here the peak SA concentrations in early spring is discussed as if it and the so-called Arctic haze are two things that here happen to coincide. Aren't they two sides of the same coin?

We thank the reviewer for their comment. In this section we are stating that the high winter/spring concentrations of SA are resulting from transport of air masses further south associated with Arctic haze—so indeed, two sides of the same coins, as the reviewer stated. This is stated clearly on Line 339:

“... which suggests that Arctic haze, or anthropogenic pollution, is a key source of the high SA concentrations in winter and the dominant source of SA during the annual cycle.”

Line 368-369: I think this is an overstatement, or maybe wishful thinking. There is some agreement, but I’m not very much impressed.

We thank the reviewer for their comment. In this statement, we were highlighting the general consistency in the timing of temporal spikes for each sulfur species in Fig. 2, which occurs predominately during January and February. We have weakened the sentence on Line 368 – 369 according to the reviewer’s concern, as follows:

“Overall, the SO<sub>4</sub>-S simulations peak during the same time of year as the measured gas phase sulfur species, especially in January and February when we observed temporal spikes of each species.”

Line 439-440: I wonder what the source is here; I think the authors are wrong. Most of the long-range meridional transport occurs in association with weather systems, and with those there is usually plenty of precipitation. In the Arctic, there is very little convective precipitation at all, except over land in summer.

It is not clear on the point with which the reviewer’s disagrees here. The reviewer agrees that there is precipitation during poleward transport in summer and that there is convective precipitation over land. It is worth noting that the prevailing anthropogenic sources are located on land, and therefore, we feel that our statement about convective precipitation increasing in summer, thereby decreasing northward transport of anthropogenic pollutants, is not inaccurate. We would also like to point out that we did not specify that the convective precipitation was occurring in the Arctic itself but during transport from regions further south. The reviewer’s main concern here seems to be with the use of “convective” to describe the precipitation. To address their concern, we have rephrased the sentence on Line 439 – 440 follows:

“In contrast, there is more precipitation during summer, which limits northward transport of anthropogenic pollution...”

Lines 441-442: Maybe I misunderstand, but here the authors seem to suggest that Arctic haze, which occurs in spring, is the source of SA in summer. That would mean SA has a life-time of months. At the very least drop the “demonstrates”

The point here was to compare the general SA concentrations during summer to the higher concentrations observed during the Arctic haze period. We did not intend to suggest that Arctic haze remained a source of SA in summer. We rephrased this sentence to be clearer and avoid such confusion. Line 441 now reads as follows:

“Based on this, our results show that SA concentrations from DMS emissions in summer are smaller in magnitude than anthropogenic sulfur sources from Arctic haze in spring.”

Lines 500-516: The link between DMS and MIZ biology is certainly not new, but the results here indicate regions that are either over land or way south of any MIZ, so this doesn't work.

As discussed in our response to the general comment above, the polygons associated with DMS (MSA) are not predominately over land but are south of the marginal ice zone or are near coastal areas. In the same discussion, we also specified that regions south of the marginal ice zone were influential in our data. We refer to lines 501 – 505 where we discussed these topics:

“The key insight obtained from the inverse model results, shown in Figs. 6a and 6b, is that regions south of the marginal ice zone appear to be the most influential on MSA concentrations over the central Arctic. More specifically, the inverse model identifies several oceanic regions as potential sources of MSA in our observations, where the Kara, Barents, Norwegian, and Labrador Seas are the most prevalent source regions during spring and summer (polygons b, c, and d in Fig. 6a).”

We stated this again on Lines 509 – 512:

“Previous research has shown that the regions identified in Fig. 6a are biologically active or important sources of DMS, the precursor of MSA, from May to August (Hulswar et al., 2022; Lana et al., 2011; Leck and Persson, 1996a; Terhaar et al., 2021), which is consistent with the chl-a satellite data and again highlights the importance of air mass transport from biologically active source regions further south on our MSA measurements.”

In these statements, we intended to emphasize regions south of the marginal ice zone and did not mean to imply that the DMS was sourced directly at the marginal ice zone, according to our qualitative source region analyses.

The statement on Line 513 - 515 is a comparison of our results to previous studies that have also identified DMS or MSA in the aerosol phase south of the marginal ice zone, which is consistent with our results. To minimize confusion, we have changed “near” on Line 514 to “south of”.

Line 569: During the ASCOS expedition, CCN-limited conditions happened during less than one day, out of an ice camp that lasted ~3 weeks. It has been seen also in other datasets, so I wouldn't call it rare, but it is still “unusual” rather than “common” or as here “often” occurring.

We thank the reviewer for highlighting this information. This is a good point. Our intent in this sentence was to highlight the generally low background aerosol concentrations in the summertime Arctic atmosphere, which can indeed lead to periods of CCN-limited conditions. We have rephrased the sentence on Line 569 to be more accurate, according to the reviewer's suggestion:

“Once in the aerosol phase, secondary particles containing MSA are sufficiently hygroscopic such that they may enhance CCN concentrations in the summertime Arctic atmosphere, which can experience periods of CCN-limited conditions (Mauritsen et al., 2011).”

Lines 580-583: Drop this; when you get to it, let us know but for now there is no information here.

We thank the reviewer for this suggestion. Instead of completely dropping this text, which does not provide much new information as the reviewer noted, we have rephrased this sentence to point out to the reader that future studies are necessary to resolve such processes. Line 580 now reads as follows:

“Note that these aerosol processes involving MSA are beyond the scope of the seasonal analysis presented here, however, future work should aim to investigate the role of MSA in event-level analyses of the mechanism of NPF and aerosol chemistry in the central Arctic region.”

Line 624: I stagger at the words “provides evidence”! I can’t find a shred of evidence here and, moreover, the whole sentence sound like a contradiction: “... provides evidence ... could be ...”.

We have changed the wording on Line 624 from “provides evidence” to “suggests” to account for the reviewer’s concern.

Line 663-664: This is a really myopic perspective. There is certainly clouds around the whole year, that must have formed on some sort of particle, so if IA is not present all the year, it has to be something else – duh!

It is unclear what the reviewer intends to criticize with this comment. Please note that the statement on lines 663 – 664 is not discussing CCN or the composition of all aerosols in the Arctic during the year. Instead, this statement is related to previous observations of the chemical mechanism of NPF in the central Arctic, which due to a very limited number of measurements over the central Arctic Ocean with sufficient instrumentation to resolve this process, is not well characterized. We mentioned IA in the context of two studies that did characterize the mechanism of NPF in the Arctic: one in the central Arctic during autumn, and another from land-based sites in Villum and Svalbard. There has been a lot of emphasis placed on the role of IA as a primary driver for secondary aerosol formation in the Arctic, but it’s worth presenting here that the mechanism of NPF during the whole year is likely more complicated. We have decided to keep this discussion in the text.

Lines 687-688: It actually doesn’t; look where the MIZ is in summer!

We thank the reviewer for pointing this out. In a similar fashion to the comment on Lines 500 – 516, we have changed the word “near” to “south of” on Line 687, which is consistent with our results.

## References

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