

### **General impressions:**

This work uses satellite-based information on sea ice leads in the Arctic combined with GEOS-Chem model information to estimate the contribution of leads to the sea salt aerosol budget and bromine concentrations during November to April. It is a fairly straightforward study, and was relatively easy to follow. The contribution will be useful. However, I think it would be work further emphasizing the limitations of their approach, and under-emphasizing the links to climate, which I felt were a bit too bold. My impression is also that the writing and figures in the manuscript are not yet of sufficient quality for ACP. I recommend that the authors do several more rounds of editing before resubmission, since I only had time to point out some of the issues below. After addressing these issues, I would re-consider recommending it for ACP.

We add a section of “Uncertainties Discussion” to emphasize the limitations of our approach and underemphasize the links to climate change by largely removing that discussion (see more in below comments and revised manuscript).

Throughout this document, text in red is the response to the Reviewer's comment and we refer to lines within the updated manuscript.

### **Specific comments:**

I recommend that the authors further clarify, and when appropriate, emphasize the limitations of their approach in detecting leads. For example,

1. L146: "The lead area fraction includes open water leads and thin ice-covered leads 3 km and wider." Please discuss with references the portion of leads that are smaller than 3 km. This information was briefly touched on in the methods and conclusions, but should be further clarified and expanded upon. Depending on how many leads are being missed, it may merit that the authors clearly state in the title, abstract, introduction and conclusions that they are only focusing on large leads, to avoid misleading readers (no pun intended) about the meaning of their findings.

We add throughout the paper that this study quantifies emissions from large leads, in the abstract (lines 35-37), Sect 2.1 (lines 165-167) and to the newly added uncertainties discussion (lines 539-541).

2. L152: "more than 50% of the total lead area visible in 500 m MODIS images was detected" Please address what fraction of leads will be missed with a 500 m resolution (or at least what is known about that question).

There is only one paper the authors are aware of that addresses this question (Qu et al., 2019) that analyzed 1 day of MODIS data in April 2015 for part of the Beaufort sea that suggested 13-34.5% of leads were  $\leq 1$  km, but they did not specify how much of that was  $\leq 500$  m.

We revise the text to clarify as follows: "We use the AMSR-E lead area product for this study as it avoids cloud interference when detecting leads and provides nearly consistent daily resolution. A limited quantitative validation by Röhrs and Kaleschke (2012) of one day (March 21, 2006) of the AMSR-E product against Moderate Resolution Image Spectroradiometer (MODIS) showed 50% of the total lead area visible in 500 m MODIS images was detected in the AMSR-E product. However, leads greater than 3 km in size ("large leads") are detected with certainty by the AMSR-E product (Röhrs and Kaleschke, 2012), so our results effectively estimate emissions from large leads only." Lines 161-167

3. I was unconvinced about the links to climate, and felt they were over-emphasized as written.

a. For example: Abstract: "Thus, lead SSA emissions could have significant impacts on Arctic climate." There is a missing step in the logic here. Just because leads are increasing and they emit SSA doesn't mean that there will be significant impacts on Arctic climate. What is the evidence for a link here?

To reduce emphasis on linking lead emissions to climate, we remove this sentence.

b. Relatedly, L. 484: "...could also affect aerosol-cloud interactions, which largely have a warming effect in the Arctic from trapping of longwave radiation during the cold season (Cox et al., 2015; Stramler et al., 2011)." As written, this statement is not correct. Clouds have a warming effect in the Arctic from trapping of longwave radiation during the cold/dark season, but the warming vs. cooling effect from aerosol-cloud interactions in the Arctic is not well understood (e.g., Morrison et al., 2012; Schmale et al., 2021; Tan et al., 2023; Zamora and Kahn, 2024). Some studies suggest aerosol-cloud interactions can actually cool the surface during winter (e.g., Villanueva et al., 2022), although others disagree.

This section of the conclusion is reduced and revised: "Future trends in Arctic sea ice predicted by climate models suggest a possible future increasing trend in lead area (Intergovernmental Panel On Climate Change, 2023), which would increase lead emissions. The additional SSA from leads in regions where the background aerosol concentrations are low could also affect local aerosol-cloud interactions, but the overall warming or cooling effect of these additional aerosols remains uncertain (Cox et al., 2015; Schmale et al., 2021; Stramler et al., 2011; Tan et al., 2023; Villanueva et al., 2022)." Lines 574-580

4. I recommend that the authors either include in the analysis or cite other relevant datasets.

a. For example: L421: "This further highlights the need for observations in other regions to better understand the impacts of lead emissions." I believe there are other observations. For example, Villum Research Station has historic Na data. The 2008 NASA ARCTAS campaign has Br concentrations. There may also be data from ship campaigns and other aircraft campaigns as well.

- Th model data is monthly-mean output that is not at a sufficient temporal resolution for meaningful comparison against aircraft data (like NASAARCTAS). We therefore prioritize long-term stations instead.
- There is a gap in data from 2003-2007 at Villum, which is a majority of our study period. Additionally, Villum data is not publicly available.

We include Pallas (see Figures 7 and 8 in the main text) as an additional station with observations available from 2003-2008. We add a description of the Pallas observations within Sect. 2.3 and include it in our model evaluation in Sect. 3.3.

- b. L. 467: I am pretty sure there have been other relevant studies, e.g., from the MOSAiC field campaign.

MOSAiC did not directly measure SSA or Na<sup>+</sup> concentrations.

- c. L. 504: "To better constrain lead impacts on SSA and reduce uncertainty in the SSA size distribution, additional ground observations with size distribution information in the Canadian archipelago, such as off the northern coast of Baffin Island and the eastern coast of Victoria Island, would be beneficial." I recommend that the authors more thoroughly check to see what data in this area are already available. The NETCARE campaign, for example, took place in that region.

Th model data is monthly-mean output that is not sufficient temporal resolution for a meaningful comparison against aircraft data (like NETCARE), so we prioritize long-term stations instead.

5. The abstract needs some work. Regarding, "Total monthly SSA emissions increase by 1.0-1.8% ( $\geq 60^\circ\text{N}$  latitude) and 5.8-8.4% ( $\geq 75^\circ\text{N}$ )," please state the time frame that the increase refers to (Nov-April? 2002-2008? Something else?). Please also state the information this finding is based on. Also, from reading the abstract alone, it is unclear how the studies can show that GEOS-Chem overestimates SSA concentrations at Arctic sites. The reader is left guessing whether this is based on some ground data or something else. L 126-130: This information should also be in the abstract.

To the first sentence mentioned in this comment, we clarify as follows: "Simulated total monthly SSA emissions increase by 1.1-1.8% ( $\geq 60^\circ\text{N}$  latitude) and 5.6-7.5% ( $\geq 75^\circ\text{N}$ ) for the 2002-2008 cold season." Lines 37-39

We modify the sentence in the abstract to include all information from previous lines 126-130: "Here, we create an emissions parameterization of SSA from leads by combining satellite data of lead area (the AMSR-E product) and a chemical transport model (GEOS-Chem) to quantify pan-Arctic SSA emissions from leads during the cold season from 2002-2008 and predict their impacts on atmospheric chemistry, evaluating the results of our simulated SSA against in-situ observations." Lines 31-35

We also modify the sentence for clarity: "GEOS-Chem overestimates SSA concentrations at Arctic sites compared to ground observations even when lead emissions are not included, suggesting underestimation of SSA sinks and/or uncertainties in SSA emissions from blowing snow and open ocean." Lines 40-42

6. The paragraph starting on L 75 reads like a collection of facts. It would help to re-write it to emphasize just the relevant information a reader needs to know, and to clarify how the different facts are relevant.

We revise the paragraph, lines 77-97.

7. L95: can you please clarify for the reader how snow becomes saline in the first place?

We clarify this with the following text: "Blowing snow is the result of saline snow over sea ice being swept up by wind; the snow becomes salty through the upward movement of brine from sea ice to the snow surface, incorporation of frost flowers, and the deposition of SSA derived from the nearby open ocean (Domine et al., 2004)." Lines 101-103

8. L 101, "Incorporating..." please clarify how incorporating blowing snow SSA emissions into models has a significant impact on atmospheric chemistry

The sentence is revised for clarification: "Incorporating blowing snow SSA emissions into models has shown how missing sources of SSA in the Arctic can have a significant impact on atmospheric chemistry; for example, Huang et al. (2020) show bromine released by blowing snow impacts modeled springtime bromine activation and ozone depletion events. The strong observational evidence that leads contribute to cold season SSA and the impact of blowing snow SSA on modeled Arctic atmospheric chemistry suggests there is a need to assess the potential impacts of lead emissions, which are currently missing from global chemistry and climate models." Lines 108-114

9. L142: "This method of detection can only be applied to the Arctic freezing season (November-April) due to surface melt of the sea ice May-October." Please state why that is.

We clarify this by adding: "This method of detection can only be applied to the Arctic freezing season (November-April) due to surface melt of the sea ice modifying the sea ice emissivity from May-October, which affects the lead detection algorithm." Lines 151-153

10. L153: Please specify more clearly how this information on MODIS is relevant

We clarify the relevance of this information throughout the paragraph: "We use the AMSR-E lead area product for this study as it avoids cloud interference when detecting leads and provides nearly consistent daily resolution. A limited quantitative validation by Röhrs and Kaleschke (2012) of one day (March 21, 2006) of the AMSR-E product against Moderate Resolution Image Spectroradiometer (MODIS) showed 50% of the total lead area visible in 500 m MODIS images was detected in the AMSR-E product. Leads greater than 3 km in size ("large leads") were detected with certainty for the AMSR-E product (Röhrs and Kaleschke, 2012), so our results effectively estimate emissions from large leads only." Lines 162-168

11. L172: Probably worth mentioning here that in the wintertime Arctic, there isn't a lot of rain deposition or convective precipitation. Also probably worth mentioning that precipitation is notoriously hard to predict correctly in the Arctic. Please comment on how this latter fact might influence your findings.

We clarify this with the following corrections: "For gas and aerosol species, wet deposition (both rain and snow) includes washout and rainout in convective and large-scale stratiform precipitation (Amos et al., 2012; Liu et al., 2001; Wang et al., 2014). From November to April in the Arctic, wet deposition is mainly in the form of snow (Screen and Simmonds, 2012)." Lines 188-191

In this study we do not predict precipitation but instead use a reanalysis product MERRA-2 which includes observations. We clarify what MERRA-2 is with the following description: "GEOS-Chem and HEMCO are driven by Modern-Era Retrospective Analysis for Research and Applications (MERRA-2) (Gelaro et al., 2017) meteorological fields from the NASA Global Modeling and Assimilation Office (GMAO), which is reanalysis meteorological data assimilated from various observational sources (i.e., satellite, aircraft campaigns, and ground stations) providing variables such as temperature, wind, precipitation, and humidity." Lines 175-179

12. L185-191: It is not clear why this information is in the manuscript. Also, please state the reasoning for choosing the Jaegle et al. (2011) parameterization instead of the Nilsson et al. (2001) or Ioannidis et al. (2022) parameterizations.

We remove these lines from this section and add discussion of Nilsson et al. (2001) to the new section "Uncertainties." (lines 137-149)

We also include justification for using the Jaegle et al. (2011) function: "The Jaeglé et al. (2011) function is empirically derived to best match global observations in GEOS-Chem."

And "we choose the Jaeglé et al. (2011) open ocean function for our lead emissions parameterization as it is the standard SSA emission function in GEOS-Chem that has been previously evaluated across global oceans"

13. Section 2.3: Please clarify what years the samples at Utqiāġvik, Zeppelin, and Alert were taken. Were they taken during the full time period of the study?

We clarify with the following text: "These observations are available for the time period of this study (November-April from 2002-2008, except for Pallas station, 2003-2008)." Lines 248-250

14. In Section 3, for Table 1: relative increases are more meaningful when placed in context of what they are relative to, so I recommend adding in monthly total SSA emissions to this Table. For example, a 1% increase in SSA emissions of 10 mg/m<sup>2</sup>/day might be more meaningful than an 8% increase in emissions of 0.1 mg/m<sup>2</sup>/day. This information is sort of present in Figure 3, but the information is presented later and in different units. So right now when a reader first sees Table 1, they are left wondering whether leads are really most important in January than November when freeze is still happening. Also, I would think that in January when sea ice is thicker and more compact, leads are possibly less common (not a sea ice expert here). Is that the case?

We include the addition of the absolute standard and lead emissions (in Gg) in Table 1 for both  $\geq 60$  and  $\geq 75^{\circ}\text{N}$ . We revise the text to clarify: "We focus Figs. 2 and 4 on the month of January as an example. January it is tied for highest lead emissions for latitudes  $60^{\circ}\text{N}$  and greater and second highest for latitudes  $75^{\circ}\text{N}$  and greater, and also has the second largest multi-year average lead area (see Fig. S.3b in SI)" (lines 270-273).

See also the discussion of the dominant factors determining lead emissions in lines 312-316 which we revise slightly: "Monthly total lead emissions and lead area have low correlation ( $R^2=0.13$ , see Fig. S.3), indicating the variance in monthly total lead emissions is dominated by the nonlinear dependencies on wind speed and sea surface temperature (Eq. S.1 in SI), as the lead emissions are calculated with the Jaegle et al. (2011) wind speed and sea surface temperature source function."

See also newly added Figure S.3(b), which shows the multi-year monthly average lead areas.

15. L. 255: "Total emissions are resolution independent" Why is that? Wouldn't there be more relevant information at a higher model resolution?

To clarify, we revise the paragraph within the methods which explains the resolution independence of the emissions: "We first calculate SSA emissions at the highest resolution of HEMCO ( $0.5^\circ \times 0.625^\circ$ ), which is the native resolution of MERRA-2. Two sets of emissions are calculated: (1) the standard emissions only (i.e., open ocean and blowing snow SSA emissions, the "standard" case); (2) SSA emissions with lead emissions added ("standard + leads" case). Each set of emissions are then implemented separately into GEOS-Chem "offline" to ensure total SSA emissions are properly scaled and distributed and not influenced by the resolution-dependence of the wind speed (Lin et al., 2021). GEOS-Chem is run at the highest global horizontal ( $2^\circ$  latitude  $\times 2.5^\circ$  longitude) and vertical (72 vertical levels) resolution." Lines 214-221

16. L 274: "Poleward..." can you speculate as to why this is? Presumably in April at lower latitudes melt is already occurring in some places, but what is going on in January?

We clarify the text as follows: ". The smaller magnitude of standard emissions later in the cold season poleward of  $60^\circ$  N make lead emissions relatively more important, with the largest percent increase  $\geq 60^\circ$  N in SSA emissions due to leads occurring in April. Poleward of  $75^\circ$  N, the lead emissions represent a larger fraction of the standard emissions, resulting in higher percent increases due to leads (~4-6% higher than for  $\geq 60^\circ$  N). Absolute lead emissions peak in December for  $\geq 75^\circ$  N latitude, which is also the month with the highest percent increase due to leads  $\geq 75^\circ$  N, and decrease more than twofold by April. Controlling factors of the lead emissions are discussed in the next paragraph." Lines 296-303

17. Supplement L28: Which value for theta did the authors use? They only say the recommended value.

We clarify with the following text in the Supplement: "and  $\theta$  is an adjustable parameter controlling the shape of the size distribution of submicron (recommended value of  $\theta = 30$ , which is used in our study)."

18. L.287: Please clarify the logic here instead of referencing section 2.2. Why would monthly total lead emissions and lead area having a low correlation mean that variance in monthly total lead emissions is dominated by the nonlinear dependencies on wind speed and sea surface temperature?

We clarify this with this modification: "Monthly total lead emissions and lead area have low correlation ( $R^2=0.13$ , see Fig. S.3), indicating the variance in monthly total lead emissions is

dominated by the nonlinear dependencies on wind speed and sea surface temperature (Eq. S.1 in SI), as the lead emissions are calculated with the Jaegle et al. (2011) wind speed and sea surface temperature source function" lines 312-316

19. L. 324: It is important to say whether this "slight decreasing trend" is statistically significant.

Based on the SD, it doesn't look like it is. If so, the authors should take that part of the sentence out. Same with the statement, "Changes in SSA mass concentration are also higher poleward of 75°N."

The  $R^2$  of the SSA line  $\geq 75^{\circ}\text{N}$  is 0.7376, so it is not statistically significant. We remove this part of the sentence.

We change the phrasing of the second statement for clarification: "Changes in monthly mean SSA mass concentrations are also higher for poleward of 75°N."

20. Figs. 5 and 6: As stated before, I don't find the percent increase due to leads very meaningful. Please either convincingly explain what scientific process this metric is meaningful for, or remove the figure, or relate it more clearly to something like absolute concentrations.

We add the absolute increase to the figures. We retain the percent increase to quantify the relative importance of leads with respect to other processes.

21. Paragraph starting on L. 435: It reads strangely to have this much text referencing a figure in the supplement. I recommend either moving the supplement figure to the main text, or moving the paragraph to the supplement, and just summarizing the paragraph in a sentence or two in the main text.

We move the figure to the main text (Figure 8).

22. L. 474: "We find that lead SSA emissions occur primarily in regions where other SSA emissions sources are very low, mainly within the Canadian archipelago and the eastern Greenland Sea." From Fig. 2, the lead emissions are higher in the Nares St. and in the Bering Strait than over the Canadian archipelago.

Modified to "mainly within the Bering Strait, Nares Strait, Wynniatt Bay in the Canadian archipelago, and the eastern Greenland Sea."

23. L. 493: Please add uncertainty estimates here.

We add uncertainty estimates: "The highest increase in multi-year average SSA mass concentrations due to leads, spatially averaged for  $\geq 75^{\circ}\text{N}$ , occurs in November ( $5.7\% \pm 5.2\%$ ) and the lowest occurs in April ( $3.7\% \pm 2.9\%$ )."

24. L. 494: "The percent increase due to leads in SSA and Br concentrations are spatially coherent." Please clarify what this means.

We clarify this with the following sentence: "Increased SSA from leads increases surface Br concentrations during the cold season in corresponding locations."

25. *The figures need some work. Here are some suggestions:*

Please note that we plan to upload separate pdf files of each figure for our final paper to improve resolution, as the embedded png files of the figures become blurry.

- a. Fig. 1: There is not enough contrast between the white background and the light blue colors. Please redo the figure so that a readers can clearly see the lead area fraction and related percentages. Maybe a rainbow color scheme instead of just a blue-based color scheme would help?

The rainbow color scheme is not color-blind or black-and-white print friendly. We change the Figure 1 colormap to the scheme in python called “inferno” as it shows the lead area fraction more clearly than the blue.

- b. Figure 1: You might consider changing the month to January from November, so people can compare lead fraction in January to the data shown in Figs. 2 and 4.

We change Figure 1 to show 01/01/2003, the first day of data in January, instead of November. The spatial distribution of these lead area percentages is more comparable to the emissions and concentrations in other figures.

- c. Fig. S1: Please increase the font size of the color bar

Corrected.

- d. Fig. 4: Please make the sites have larger point sizes and larger fonts. It's really hard to see them, and I could barely find Alert at all. Also, I think the Utqiaġvik, Alaska point is currently placed in the figure in Russia, so please check the coordinates.

We increase the size of the points/fonts of the labeled sites and change the color to white for better contrast.

- e. Figs. 4, 6, S5, and S7: Please enhance the contrast in the land border color in the figures relative to the figure colors. Right now the black thin borders cannot be easily seen, making it harder to distinguish feature locations.

We add thicker black borders for better contrast in each of these figures.

- f. Fig. 7: The current color scheme and shaded areas make it very difficult to distinguish between the Observations and the Standard run. Please fix.

Corrected.

- g. Fig. S6: Please increase font of the months in the key.

Corrected.

- h. Figs. S4, S8 are blurry. Please increase resolution.

Figure S.4 is removed, and S.8 is moved to the main text with improved resolution.

- i. Fig. S8: The color contrast between the blues and the oranges are hard to distinguish. Please fix.

Corrected (Now Figure 8 main text).

## 26. Technical comments:

We incorporate each of these technical comments in their respective lines.

- a. L 75: "lead-based SSA" would it be more accurate to say something like, "emissions of SSA from leads"?
- b. L105: "incorporates" should be changed to "incorporated"
- c. 109: "analysis" should be changed to "analyses"
- d. 123: should be "produces"
- e. L162: "(Community, 2021)" This isn't the correct citation
- f. L166: "from the NASA"
- g. L168: "wind-" not "wind"
- h. Paragraph starting on L168 should be broken up into several paragraphs
- i. L169: "sea-surface-temperature-dependent"
- j. L182: " The AMSR-E satellite data is regridded to  $0.5^\circ \times 0.625^\circ$  from  $6.25 \times 6.25$  km using a distance-weighted average remapping." This sentence seems out of place.

Now lines 154-157

- k. L183: "This is..." Please specify what "this" refers to
- l. L100 in Supplement: "updates"
- m. L105 in Supplement: "running" not "run"?
- n. L 490: "the standard concentration " of what? SSA?

Reference in this response to reviewer that is not in the manuscript: Qu, M., Pang, X., Zhao, X., Zhang, J., Ji, Q., and Fan, P.: Estimation of turbulent heat flux over leads using satellite thermal images, *The Cryosphere*, 13, 1565–1582, <https://doi.org/10.5194/tc-13-1565-2019>, 2019.