

Interactive comment on “Wintertime enhancements of sea salt aerosol in polar regions consistent with a sea-ice source from blowing snow” by Jiayue Huang and Lyatt Jaeglé

Anonymous Referee #1

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General Comments:

In this study, parameterizations for sea salt aerosol (SSA) emissions from blowing snow and frost flowers are implemented in the GEOS-Chem global chemical transport model. Model to measurement comparisons for SSA mass concentrations are presented for three Arctic and two Antarctic sites, as well as for an Arctic cruise. The authors conclude that blowing snow is a dominant SSA source during winter in the polar regions, with a smaller contribution from frost flowers. The paper is well written and addresses scientifically important questions regarding the sources for SSA in the polar regions. The related parameterizations are challenging to develop because there are several uncertainties involved. The impact of these uncertainties on the conclusions could be

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discussed and examined more explicitly as outlined in the following comments. The manuscript should be suitable for publication if the following concerns can be satisfactorily addressed

Specific Comments:

1) P1, L12: The model with open ocean emissions alone underestimates the SSA mass concentrations by factors of 2-10. What is the uncertainty in the measurement SSA mass concentrations? Is it possible that the model and measurements could agree within the measurement uncertainty?

2) P2, L17-19: Please consider adding a sentence here to describe why the first two mechanisms lead to depletion of sulfate relative to sodium through the precipitation of mirabilite.

3) P4, L9: These lines note that the simulation with open water emissions alone of SSA overestimates the summertime SSA. Is it possible that the model could have errors in the summertime removal of SSA in precipitation in the polar regions (particularly if the model neglects aerosol removal by summertime drizzle). If the summertime removal is too inefficient, are you able to justify this suppression of the emissions? Are there any previous studies that have examined emissions for temperatures below 5 C? Fig. S1 was instructive since this seems to indicate a low sensitivity to this assumption about the emissions for the polar winter. Is there a concern that this assumption might introduce a low bias to the open ocean emissions in an effort to compensate for errors in the removal scheme?

4) P4, L14-15: Are there any uncertainties related to the dry deposition parameterization? How might this affect your analysis, particularly in winter? As well, are there any uncertainties related to the wintertime removal by precipitation from mixed-phase and ice clouds and how might this affect your analysis?

5) P4, L18-19: Please consider clarifying here how the sodium bins are related to the

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SSA bins in your parameterization. Are these sodium bins additional tracers in the model? As well, please specify where you mean by 'see below'.

6) P5, L26: In the frost flower parameterization, SSA emissions are only allowed if the wind speed is $< 5 \text{ m s}^{-1}$. However p. 2, lines 31-32 suggest that higher wind speeds are needed in order to break the frost flowers. There appears to be two processes here – the frost flowers form under low wind speeds, but do the emissions actually occur at greater wind speeds? Could this assumption that the frost flowers only emit SSA at low wind speed cause a low bias in the emissions from frost flowers? How do the emissions change if the frost flowers are allowed to emit SSA at larger wind speeds than 5 m s^{-1} , and how does this affect your conclusions?

7) P6, L12: A factor of 3.256 is used to convert all observed Na^+ mass concentrations to SSA for comparison with the simulations. Is there any uncertainty in this factor that might make an apples-to-apples comparison between the measurements and model difficult?

8) P6, L12: 'use two size bins in the model' – are these bins for sea salt aerosol or sodium? Why are these bin limits different than in Table 1 and also different than P6, L21? As well, how are the observed Na^+ concentrations in the submicron and supermicron ranges apportioned between the two bins used by the model for the purpose of the model-measurement comparison? What are the size limits for the observed submicron and supermicron aerosol?

9) P6, L23: Figure 1 shows a maximum in the measured SSA mass concentrations in the Arctic in November-December, whereas the simulations have a maximum Jan-Feb. What factors contribute to this model-measurement discrepancy?

10) Figure 1: The blowing snow simulation alone does appear to improve agreement with observations. However, in reality, both blowing snow and frost flowers might be expected to contribute together to the SSA concentrations. Have you conducted simulations with both of these sources implemented at the same time? Figure 1 seems to

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suggest that if the model included both sources simultaneously, then the SSA would agree more closely with the observations in November/December in the Arctic and in April/May at Neumayer, but the model would strongly over predict the observations in subsequent winter months. Please consider adding some related discussion. Does this suggest that the simulation might over predict the blowing snow emissions? As well, Fig. 3 seems to indicate that the model over predicts the SSA during the blowing snow event near 17 April.

11) P6, L24: As noted here, the advance and retreat of sea ice affects SSA. How well does the model simulation of sea ice agree with observations?

12) P7, L8-11: Why was the normalized mean bias chosen as the metric for the model evaluation as opposed to mean fractional bias? The latter metric has the advantages of not allowing a few data points to dominate the metric and allows for some error in measurements (Boylan and Russell 2006).

13) P9, L1-2: Blowing snow and frost flowers are noted to have a larger impact on the $0.01\text{-}0.5 \text{ }\mu\text{m}$ SSA mass concentrations than the open ocean source. What is the physical mechanism for relatively greater emission into this size range? For the case of blowing snow emissions, are there any uncertainties related to how the emitted mass is distributed between the two SSA bins using the assumed size distribution? Has the model been used for sensitivity studies to examine the potential impact of related uncertainties?

14) P9, L24: 'they are not efficient ice nuclei' – if possible, please add a reference to observations that support this statement. This seems in contradiction to some studies (e.g. DeMott et al. 2016). Or do you mean to indicate that SSA is not an efficient ice nucleus in the model?

15) P10, L20: How do these frost flower simulations compare to the recent work of Xu et al. (2016)?

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16) P10, L28-30: As noted here, there are substantial uncertainties associated with these parameterizations. Since this is a model-based study, please consider whether the presentation of a few sensitivity study results related to the key uncertainties in the emissions parameterizations (salinity, size distribution for blowing snow and wind conditions for frost flower emissions) might be of help to the reader in interpreting the statement in the abstract that 'blowing snow is likely to be the dominant SSA source during the winter'. As well, this could help in interpreting the presented values for the SSA emissions from blowing snow and frost flowers since there seems to be some evidence that the blowing snow parameterization over predicts the observations.

17) Are you able to provide any recommendations to modelers about the implementation of blowing snow and frost flower parameterizations into global models?

18) P11, L9-10: For the potential impacts of wintertime SSA, would you expect any effect on mixed-phase and ice clouds?

References:

Boylan, J.W. and Russell, A. G.: PM and light extinction model performance metrics, goals, and criteria for three-dimensional air quality models. *Atmos. Environ.*, 2006; 40, 4946-4959, doi:10.1016/j.atmosenv.2005.09.087.

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Xu, L., Russell, L.M. and Burrows, S.M.: Potential sea salt aerosol sources from frost flowers in the pan-Arctic region. *J. Geophys. Res. Atmos.*, 2016; 121, 10,840–10,856, doi:10.1002/2015JD024713.

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