

We would like to thank all three referees for their helpful comments and insightful suggestions.

Reply to comments by Anonymous Referee #1

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 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?

- The reported uncertainties in the measurements of Na^+ mass concentrations are 11% (relative uncertainty, $0.01 \mu\text{g}/\text{m}^3$) at Barrow (Quinn et al., 2000), 5-10% ($0.01 \mu\text{g}/\text{m}^3$) at Alert and Zeppelin (WMO/GAW, 2003), 5-11% at Neumayer (Weller et al. 2008), and 5% at Dumont d'Urville (Legrand et al. 2012). This should have been included in the original manuscript, thank you for pointing out this omission. As observed wintertime Na^+ concentrations are $\sim 0.3\text{-}1 \mu\text{g}/\text{m}^3$, factors of 2-10 disagreement are well outside the observational uncertainties. We have added a discussion of measurement uncertainties in section 2.3:

“The Na^+ mass concentrations are determined by ion chromatography with uncertainties of 5%-11% ($0.01 \mu\text{g}/\text{m}^3$ in absolute 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.

-We have clarified this in the revised manuscript:

“The first two of these mechanisms lead to depletion of the sulfate to sodium ratio relative to bulk sea water as mirabilite ($\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$) precipitates from brine at temperatures below -8°C during sea ice and frost flower formation (Alvarez-Aviles et al., 2008).”

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 is 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?

- We agree with the referee that an alternative explanation for the model overestimate of summertime SSA could be related to an underestimate in the removal of SSA during summer. This point has been noted in the revised manuscript. There is support for our cold temperature suppression assumption from the laboratory study by Mårtensson et al. (2003). They examined marine aerosol production from seawater at -2°C and 5°C, finding a 50-60% decrease in the number of particles produced for $r > 0.1 \mu\text{m}$ when temperatures decrease from 5°C to -2°C. This is consistent with our assumption of suppression at very cold temperatures.

As noted by the referee, this modification to open ocean emissions does not affect our wintertime results, as the five polar sites are distant from open ocean, with very small amounts of open ocean SSA transported to those sites. For example, at Barrow and Alert wintertime SSA concentrations due to the open ocean are $< 0.1 \mu\text{g}/\text{m}^3$, more than an order of magnitude lower than observations (1-3 $\mu\text{g}/\text{m}^3$).

We have added the following:

“This is consistent with the laboratory study of Mårtensson et al. (2003), who report a 50-60% decrease in aerosol production (for $r > 0.1 \mu\text{m}$) when seawater temperature decreased from 5°C to -2°C. We note that another potential explanation for the summertime overestimate in SSA mass concentrations is inefficient wet removal from low-intensity summer precipitation in GEOS-Chem (Croft et al., 2016).”

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?

- The dry deposition velocity for aerosols over snow and ice is assumed to be 0.03 cm/s in the model, which is based on the estimates from measurements in the Arctic. As discussed in the manuscript (p10, L2), for cold clouds (mixed-phase and ice clouds with temperature $< 258 \text{ K}$) SSA is only removed by below-cloud precipitation and we assume no in-cloud scavenging in the model. If in-cloud scavenging were to be an efficient process for SSA, it would lead to decreases in our calculated SSA mass concentrations and require a larger source from sea ice to match observed concentrations. Previous studies using the GEOS-Chem model found reasonable agreement with ground-based and aircraft observations of sulfate, ammonium, black carbon, and organic aerosols

during winter and spring (Fisher et al., 2011; Wang et al., 2011). This suggests that the removal processes during winter and spring are reasonably represented in the model. This is now noted in the revised manuscript.

“Detailed comparisons of GEOS-Chem black carbon and organic aerosol (Wang et al., 2011) as well as sulfate and ammonium aerosol (Fisher et al., 2011) to ground-based and aircraft observations over the Arctic during winter and spring suggest that transport and removal processes are reasonable captured by the model. “

5) P4, L18-19: Please consider clarifying here how the sodium bins are related to the SSA bins in your parameterization. Are these sodium bins additional tracers in the model? As well, please specify where you mean by ‘see below’.

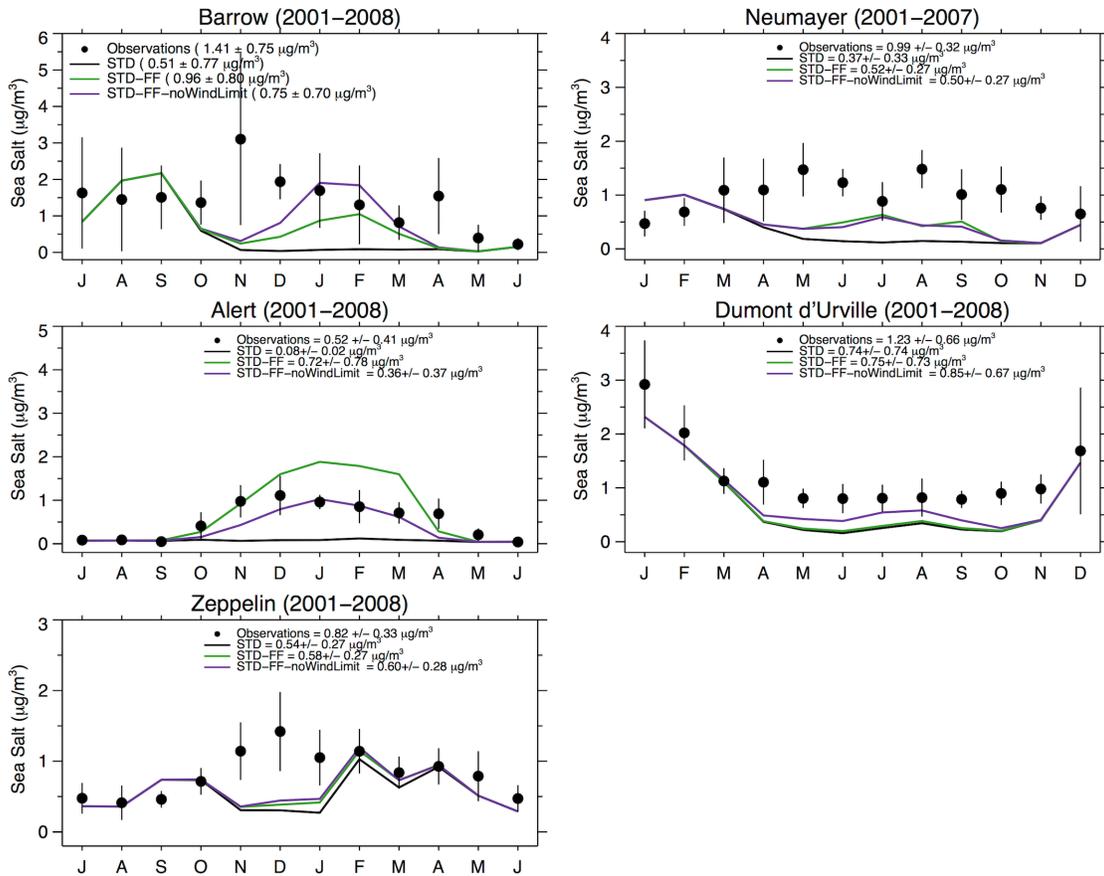
- We agree that this was unclear and have clarified this in the revised manuscript:

“For this work, we track SSA mass in two size bins: accumulation mode ($r_{\text{dry}} = 0.01\text{--}0.5 \mu\text{m}$) and coarse mode ($r_{\text{dry}} = 0.5\text{--}4 \mu\text{m}$), except in the comparison to in situ mass concentrations of SSA for which we use $r_{\text{dry}} = 0.01\text{--}0.3 \mu\text{m}$ and $r_{\text{dry}} = 0.3\text{--}3 \mu\text{m}$ (see section 2.3). “

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?

- We recognize that there is likely to be a distinction between frost flower formation under low windspeed and lofting at high windspeed. Given the uncertainty and contradictory results in the literature regarding frost flower formation and lofting (see our section 1), we have conducted a frost flower simulation removing the wind speed limit and reverting to the formulation proposed by Xu et al. (2013). This also enables us to address this referee’s comment #15 about the more recent study of Xu et al. (2016). We compare in the figure below our original frost flower simulation (STD-FF green line) to the one with no wind speed limit (STD-FF-noWindLimit purple line). As we use the same scaling factor as Xu et al. (2013), the new emissions from frost flowers are slightly lower than in our STD-FF simulation. We find good agreement at Alert for Dec-Mar, and at Barrow in January-February, but for all the other stations our results remain unchanged, with very small influence of frost flowers at Zeppelin, Neumayer and Dumont d’Urville, and therefore we find that frost flower emissions are insufficient to explain the wintertime SSA enhancements in the observations at these sites. Thus overall our conclusions remain unchanged.

For simplicity, we replace our original FF simulation with this new updated one, without any windspeed limit. The figures and text have been changed to reflect this.



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?

- This mass ratio for Na^+ to SSA (3.256) is based on the standard seawater mass ratio at a salinity of 35 psu. There can be some small changes to the ionic composition of SSA from frost flowers and snow, but these are small (<0.5%). Based on the chemical composition of frost flowers reported in Rankin et al. (2000), this ratio is 3.237 for frost flowers. Krnavek et al. (2012) find that this mass ratio is 3.278 for snow on thick first year sea ice and 3.24 for those on thin ones. Therefore, we think it is reasonable to use a constant factor (≈ 3.256) to convert observed Na^+ to SSA for comparison with the model SSA.

We have added this to the revised manuscript:

“For comparison between the GEOS-Chem model and the observations, we convert observed Na^+ mass concentrations to SSA mass concentrations using a factor of 3.256

based on the mass ratio of Na^+ in seawater (Riley and Chester, 1971). For frost flowers, this ratio is 3.237 (Rankin et al., 2000). Krnavek et al. (2012) find a ratio of 3.24-3.278 for snow on first year sea ice. As this ratio varies by less than 0.5% for these different SSA sources, we use a constant factor of 3.256.”

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?

- Yes, this indeed requires some clarification. These bins are dry sea salt aerosol radii. For simplicity, we now use one single set of size bins ($r_{\text{dry}}=0.01\text{-}0.3\ \mu\text{m}$ and $0.3\text{-}3\ \mu\text{m}$) for comparison to the ground sites and ICEALOT cruises and justify this in the revised text. Most previously published studies use ($0.01\text{-}0.5\ \mu\text{m}$; $0.5\text{-}4\ \mu\text{m}$), so this is why we use these bins in Table 1.

“The reported aerodynamic cut-off diameters of the measurements are $1\ \mu\text{m}$ and $10\ \mu\text{m}$ at Barrow and during ICEALOT (Quinn et al., 2002), and $7\text{-}10\ \mu\text{m}$ at the other sites (Wagenbach et al., 1998; WMO/GAW, 2003; Weller et al., 2008). In order to compare to model simulations, we need to convert these aerodynamic diameters to dry geometric radii. This conversion depends on aerosol density, relative humidity during sampling, and whether the particle is spherical (Seinfeld and Pandis, 2006). For example a $10\ \mu\text{m}$ aerodynamic diameter could correspond to a dry geometric radius of $2.3\ \mu\text{m}$ (80% RH, $1.2\ \text{g}/\text{cm}^3$ pure NaCl solution, factor of 2 growth between dry and 80% RH), $3\ \mu\text{m}$ (dry cubical NaCl particle, $2.2\ \text{g}/\text{cm}^3$, Lewis and Schwartz, 2004), $3.8\ \mu\text{m}$ (30% RH, ammonium sulfate and sea salt aerosol, $1.7\ \text{g}/\text{cm}^3$, Quinn et al., 1996). Thus for comparison to observations we conduct a simulation with two size bins: $r_{\text{dry}} = 0.01\text{-}0.3\ \mu\text{m}$ and $r_{\text{dry}} = 0.3\text{-}3\ \mu\text{m}$.”

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?

- Given the simplified nature of the blowing snow simulation, a number of potential factors could explain this offset, such as seasonal and spatial variability in snow mobility and/or salinity. Exploring these factors is beyond the scope of our manuscript.

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 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 predicts 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.

- It is indeed possible that both blowing snow and frost flowers contribute together to SSA concentrations. We have included a new figure in the supplemental material (Fig S5) where we compare a simulation with all 3 sources (open ocean, frost flowers and blowing snow) to observations. The resulting simulation overestimates observations, especially at Barrow and Alert, where modeled SSA is a factor of 2-3 too high in January-March. At the other 3 sites the influence of frost flowers is small, so the differences are not as large.

We have added this following discussion in P9, L1-3.

“It is possible that both blowing snow and frost flower emissions act together. However, when we add the contributions from both sources, we find that modelled SSA mass concentrations are a factor of 2-3 too high compared to observations at Barrow and Alert (Fig. S5). In particular, the frost flower simulation leads to a peak in SSA in February at Barrow, which is not observed.”

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?

- As described in Section 2.1, the sea ice in MERRA is set as a boundary condition from the NOAA Optimal Interpolation (OI) v2 gridded dataset (Reynolds et al., 2002). This dataset is based on SSMI satellite observations, so should have a reasonable representation of sea ice cover. This has been clarified in section 2.1:

“The sea ice concentration boundary conditions in MERRA are derived from the weekly product of Reynolds et al. (2002), which is based on Special Sensor Microwave Imager (SSM/I) instruments on Defense Meteorological Satellite Program (DMSP) satellites.”

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).

-We use the normalized mean bias ($NMB = (\overline{Model} / \overline{Obs} - 1) \times 100$) here as we assume that the observations are the absolute truth for evaluating the performance of different model simulations. We are using multi-year (7-8 years) observations, which helps decrease the influence of a few daily/weekly points.

13) P9, L1-2: Blowing snow and frost flowers are noted to have a larger impact on the 0.01-0.5 μm 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?

- The physical mechanisms for SSA emissions from open ocean emissions (breaking waves and bubble bursting) are different than emissions from blowing snow and frost flowers (saltation of fallen snow and suspension) or frost flowers (saltation of broken frost flower crystals and suspension). The original crystalline form of snow particles/frost flower fragments are expected to be shattered by repeated impacts with the ground and other particles during saltation. For blowing snow, the size of the dry SSA particle depends on the original snow particle size, snow salinity, and number of SSA particles produced per snow particle (Yang et al., 2008). The blowing snow particle size distribution is constrained by observations of blowing snow events (Mann et al., 2000). As discussed in the text, there are few observations of salinity of snow on sea-ice so we used uniform values of salinity for the entire Arctic and Antarctic snow salinity. This is certainly something that can be improved as more measurements become available. Assumptions of the number SSA particles produced per snow particle are highly uncertain, and we include a sensitivity study in the supplement (Fig. S2).

We have added the following in the revised manuscript:

“This difference in size distributions is related to the different physical mechanisms for SSA emissions from open ocean emissions (breaking waves and bubble bursting) compared to blowing snow (saltation of fallen snow and suspension) or frost flowers (saltation of broken frost flower crystals and suspension). The original crystalline form of snow particles/frost flower fragments are expected to be shattered by repeated impacts with the ground and other particles during saltation. Sublimation of ice from these particles leads to relatively small SSA compared to bubble bursting in the open ocean.”

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?

- Excellent point. This statement is indeed based on what is assumed in the GEOS-Chem model for scavenging. We clarify this in the text and add references to DeMott et al. (2016) and Wise et al. (2012), and note that in-cloud scavenging is not currently included in GEOS-Chem.

“Recent laboratory studies have shown that SSA could act as ice nuclei by deposition freezing (Wise et al., 2012) and immersion freezing (DeMott et al., 2016), and might thus undergo in-cloud scavenging in mixed and ice clouds. This process is not currently included in GEOS-Chem.”

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

- Thank you for bring this work to our attention. Both the magnitude of emissions and spatial distribution of our frost flower simulation are similar to the results Xu et al. (2016) for the Arctic. This has been added in the revised manuscript:

“Our emissions from frost flowers over the Arctic (0.21 Tg/yr) are consistent the accumulation model emissions reported by Xu et al. (2016) (0.24 Tg for November-February, their Table 2). We also find a similar geographic distribution.”

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.

- Yes, we agree with the referee that there are substantial uncertainties in the parameterizations and have clarified the text, as well as added sensitivity simulations (wind threshold for frost flowers) in addition to the sensitivity simulation for the blowing snow particle number. We now note in the revised text:

“One key uncertainty in our simulations is snow salinity. Indeed, SSA emissions from blowing snow have a near-linear dependence on the salinity of snow. Thus a doubling of the assumed salinity would lead to a doubling in SSA emissions from blowing snow. Furthermore, we assume a uniform salinity of snow over both first-year and multi-year sea ice. This likely overestimates the contribution of blowing snow SSA over the western Arctic, which is dominated by multi-year sea ice. More extensive observations of surface snow salinity at multiple locations over both first-year and multi-year sea ice can help further refine these assumptions. Sampling of SSA size distributions during blowing snow events can help determine the number of particles per snowflake, which we determined empirically in this study. This number will not affect to total SSA emissions, but will change the relative importance of submicron and supermicron SSA emissions.”

We have added a sensitivity simulation with/without wind conditions of frost flower emission is detailed in the supplementary material.

The overestimate of SSA in spring by the blowing model may be due to the seasonal variability of the surface snow salinity which is not taken into account in this study. This should be further constrained with observations of surface snow salinity at different location and times which are limited at this moment.

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

- Our implementation of blowing snow follows Yang et al. (2008) with modification of salinity, age of snow and number of particles. And the implementation of frost flower emission follows the emission scheme in Xu et al. (2013). Details on implementation of these sources in the model can be found in section 2.2, Yang et al. (2008), Xu et al. (2013) and references therein.

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

-Yes, this is indeed a very good point. We have added the following statement:

“Improved process-based understanding of these emissions would also lead to better constraints on the potential climatic impact of wintertime SSA on clouds, in particular mixed-phase and ice clouds, which have a strong influence on downward longwave radiative forcing. Indeed, recent studies have shown the role of SSA as ice nuclei (Wise et al., 2012; DeMott et al., 2016). Thus over the Arctic and Antarctic regions, where the abundance of other ice nuclei such as dust or black carbon are low, SSA from sea ice sources could influence the formation, radiative forcing, and precipitation of mixed-phase and ice clouds.”

Reply to comments by Anonymous Referee #2

This is a well-written paper that explores in a model the relative influence of three sources of sea salt aerosol in the polar regions. Although the blowing snow source has been explored in a model with a similar parameterisation in another model, this is the first time that all 3 sources have been tested in a similar setup. The paper compares model output with aerosol data at a number of polar sites, with rather impressive results in terms of concentration, seasonality and episodicity. It is able to conclude on the importance of the blowing snow source in wintertime, and provides reasonable evidence to dismiss the frost flower source as a significant player in most circumstances. Overall, it is a good paper, clear and well-argued, and certainly worth publishing in ACP. It provides a basis for exploring other aspects of the influence of different sources of sea salt aerosol. My only substantial quibble with the authors is that set store by the fact that they get the concentrations right and that they are testing the balance between the sources. However this ignores the fact that they have had to take several decisions (such as the number of salt particles per snowflake, the salinity of the snow, and the scaling factor (Page 5, line 29), which are essentially tunings (ie they chose them in order to match the data). I think the paper should be a little clearer in recognising this, and in admitting that the relative strength of the different sources is influenced by this rather strongly.

- We have added more discussion of our assumptions in the revised text and have recognized more clearly how these assumptions impact our conclusions. In particular, we have added the following in our conclusions:

“The SSA parameterizations for blowing snow and frost flowers have several intrinsic assumptions, such as the salinity of snow and the scaling factor for frost flowers, which influence the relative magnitudes of these two sources in polar regions. The geographic distribution, seasonal cycle, and daily variability of these sources, however, is controlled by sea ice extent and meteorological parameters (winds and temperature). In this study, we showed that the temporal and geographical variability of SSA observations at five polar sites is more consistent with blowing snow than with frost flowers. Based on this comparison, we conclude that blowing snow is likely to be the dominant source of SSA in polar winter, although frost flowers cannot be entirely ruled out. In particular, they may contribute indirectly to SSA emissions by salinating wind-blown snow (Obbard et al., 2009).”

Detailed comments

Page 1, line 18. Here and elsewhere in the paper the authors refer to submicron aerosol, meaning the range which elsewhere they describe as the 0.01-0.50 μm radius range. It would be helpful if they would clearly state this usage, perhaps on page 4, line 17, where after describing the accumulation mode they could add “which we refer to as sub-micron based on its diameter”.

- This has been clarified in the manuscript:

“For this work, we track SSA mass in two size bins: accumulation mode ($r_{\text{dry}} = 0.01\text{--}0.5$

μm) and coarse mode ($r_{\text{dry}} = 0.5\text{--}4 \mu\text{m}$), except in the comparison to in situ mass concentrations of SSA for which we use $r_{\text{dry}} = 0.01\text{--}0.3 \mu\text{m}$ and $r_{\text{dry}} = 0.3\text{--}3 \mu\text{m}$ (see section 2.3). In the rest of the manuscript we will refer to the accumulation and coarse mode SSA aerosol as submicron and supermicron SSA based on their diameters.”

Page 4, mid. I don't quite understand the description that for the blowing snow they treat just two size ranges. My understanding was that the Yang parameterisation that they are following uses many more size bins than that, and relies on this for many of its characteristics. Could the authors explain what they mean? Presumably they maintain different sizes in calculating the mass flux with respect to the number of snowflakes, so in what respect do they not use different size ranges and what are the likely impacts?

- We use the same size distribution for snow particles as reported in Yang et al. (2008), but use a different salinity and number of SSA particle per snowflake. As we track SSA mass in GEOS-Chem in two size bins, we integrate this size distribution and calculate the corresponding SSA emissions for the two size bins.

Page 5, line 5-6. I don't understand at all why they choose a lower salinity in the Antarctic, or why the higher Antarctic precipitation is relevant to that. It reads as if they think the snowfall somehow dilutes the salinity but this of course makes no sense as the salinity is more likely a function of snowpack thickness, which is likely lower in Antarctica. Please explain –at the moment this just looks like a correction factor chosen at random.

-This point was not clearly described in the original manuscript and we have clarified this in the revised version. Yes indeed, it is the snow thickness that controls the salinity of the snow on sea ice. Antarctic is surrounded by ocean, so precipitation occurs more frequently over Antarctic sea ice. As a result, snowpack tends to be thick. The Arctic Ocean is surrounded by land and precipitation is relatively rare, hence the snowpack is thinner on Arctic sea ice.

“Snowpack on Antarctic sea ice is thicker than over Arctic sea ice. Indeed, Antarctica is surrounded by the Southern Ocean which brings moisture, while the Arctic is surrounded by land with wintertime precipitation that is 3 times lower than over Antarctic sea ice (Huffman et al., 2001). As the salinity of snow decreases with snowpack thickness, we assume that the salinity of snow on Antarctic sea ice is 0.03 psu, a factor of 3 lower than over the Arctic following Yang et al. (2010). ”

Page 5, line 28. I think you mean “geometric mean diameter” and “geometric standard deviation”. Geometric diameter doesn't seem meaningful.

- Yes, this was a mistake. We have corrected this in the revised manuscript.

Page 7, line 8. Please explain what M and O are; as written your explanation of what NMB is is unclear. I assume it's the percentage mismatch between model and data – why not call it model-data mismatch? The word “bias” seems wrong when you are simply comparing alternative partial sources to the data.

- The normalized mean bias is often used as a metric for model evaluation against observations. As pointed out by the referee, our notation was unclear so we have changed the equation to the following: “ $(\text{NMB} = (\overline{\text{Model}} / \overline{\text{Obs}} - 1) \times 100)$ ”.

Page 9, line 22-25. I assume that you are saying that, for the same size and place the lifetime is the same whatever the source (this must be true), but that the lifetime is longer because the blowing snow sourced aerosol tends to form when it's colder. It might be clearer if you explain it more in this way.

- Yes, the referee is correct. We have changed the wording in the revised manuscript to clarify this point:
“Open ocean SSA form over lower latitude warmer regions, while sea ice SSA emissions occur at higher latitudes under much colder conditions, with less efficient removal processes in mixed-phased and ice clouds. The current parameterization in GEOS-Chem assumes that in-cloud scavenging of SSA does not occur in cold clouds ($T < 258 \text{ K}$) (Wang et al., 2011), thus wintertime sea-ice generated SSA are only removed by below-cloud scavenging (which is slow for accumulation mode aerosols) and dry deposition.”

Page 9, line 24. Just a question from my ignorance: wouldn't we expect sea salt to become an efficient ice nucleus below its eutectic (ie about 250K), when it would become a solid?

- The statement in the manuscript refers to what is currently assumed in GEOS-Chem. Recent laboratory studies have shown that sea salt can be act as an ice nuclei by deposition freezing or immersion freezing. This has been added to the revised manuscript:
“The current parameterization in GEOS-Chem assumes that in-cloud scavenging of SSA does not occur in cold clouds ($T < 258 \text{ K}$) (Wang et al., 2011), thus wintertime sea-ice generated SSA are only removed by below-cloud scavenging (which is slow for accumulation mode aerosols) and dry deposition. Recent laboratory studies have shown that SSA could act as ice nuclei by deposition freezing (Wise et al., 2012) and immersion freezing (DeMott et al., 2016), and might thus undergo in-cloud scavenging in mixed and ice clouds. This process is not currently included in GEOS-Chem. “

Fig 1 caption. Please explain here as well what NMB is: the reader should not have to read a quite difficult bit of text to understand the figure.

-We have made the change in the manuscript.

Fig 3 caption. I struggled to understand the text about the coloured circles. Do you mean that the larger circles (which are anyway hard to see) represent the ship's position between 15 and 19 April. If so, why not say this. If not, it needs a new explanation.

-The large circles indicate the locations of the ship between 15 and 19 April, and the color of the circle indicates the SSA mass concentrations observed on ICEALOT. We have added the clarification in the caption:

“The larger circles near Svalbard correspond to the location of the ship on 15-19 April, and they are color-coded based on observed SSA mass concentrations (same color scale as the model).”

Fig 4 and 5 caption, just for clarity please add “submicron” in the phrase “spatial distributions of wintertime submicron SSA”.

-We have made the change in the manuscript.

Supplement, section 1. Much of this text duplicates what is already written on page 4, para 1.

-We have removed the duplicated text in the Supplement.

Reply to comments by Anonymous Referee #3

This manuscript describes model-measurement comparison between GEOS-Chem with various sea-salt aerosol (SSA) sources and measurements from circum-Arctic field sites and a research ship cruise. The manuscript compares three models of SSA sources, open ocean, blowing snow, and frost flowers. Through the improved agreement between the model using the ocean + blowing snow model, the authors conclude that blowing snow is the dominant wintertime SSA source. The model using frost flowers is unable to match the observed seasonal cycles, and thus is indicated to be incorrect, and the model using only open ocean sources underpredicts SSA. The writing and logic of the manuscript are good, and the metrics for comparison are well defined and appropriate. Therefore, I support publication in ACP, with minor revisions.

General Comments:

1) The modeling in this manuscript uses a simple model for blowing snow based upon purely windspeed. However, experimental evidence (e.g. Sturm and Stuefer, 2013) shows that winds speed (alone) is insufficient to explain blowing snow fluxes fully.

Sturm, M. and Stuefer, S.: Wind-blown flux rates derived from drifts at arctic snow fences, *J. Glaciol.*, 59, 21–34, doi: 10.3189/2013JoG12J110, 2013.

This experimental observation could be a partial explanation of deviations between the model and high time resolution data shown in Figure 2. However that citation also is not able to give a simple single equation for blowing snow, so is not a solution to this challenge, and the approach adopted by the authors is reasonable given the complexity.

-We agree with the referee that other factors, in addition to windspeed, are likely to affect the blowing snow flux. We choose to use the Yang et al. (2008) parameterization, which itself is based on the blowing snow sublimation parameterization of Déry and Yau (2001), as this seems to be a well-established formulation.

2) The manuscript uses mass concentration data of SSA as the metric for model-measurement comparison. However, it would be useful to describe the rough size distribution of the modeled SSA and potentially some comparison between the model and observations. Table 1 shows this information, but it is only briefly discussed and it would be valuable to enhance the discussion. In addition, because SSA could be a source of cloud condensation nuclei, conversions of these numbers to number densities would also be valuable.

- We have added the following discussion in p5, L20:

“Our simulation yields blowing snow SSA emissions with 38% of SSA mass in the submicron range (0.01–0.5 μm) and 62% in the supermicron range (0.01–0.5 μm) for the Arctic. As we assume a lower salinity in the Antarctic, more of the blowing snow emissions are in the submicron range (60%) in that region.”

As we do not track number concentrations in the model, we cannot comment on the resulting CCN concentrations. Based on this referee's comment and the other referees as well we have added more discussion on the potential role of SSA as ice nuclei.

Page 1, line 18: The manuscript later defines "Over the Arctic..." as >60 degrees. these statements (in the abstract) should also include the definition.

-We have made the change in the manuscript.

Page 2, line 2: I would say "waters are mostly covered by sea ice."

-We have made the change in the manuscript.

Page 2, line 11: It would be more accurate here to describe studies that support SSA formation from frost flowers, as well as ones that don't support frost-flower SSA. The section later (at the bottom of this page and top of next) contains the references that are relevant.

- Good point. We have moved the paragraph describing studies that don't support SSA formation from frost flowers higher up.

Page 4, line 28: I believe that other measurements of Arctic surface snow could be compared to the 0.1 PSU concentration. Toom-Sauntry and Barrie (2002) measured fresh snowfall, and Krnavek et al. (2012) have fairly extensive data sets.

Toom-Sauntry, D. and Barrie, L. A.: Chemical composition of snowfall in the high Arctic: 1990 – 1994, *Atmos. Environ.*, 36, 2683–2693, doi:10.1016/S1352-2310(02)00115-2, 2002.

Krnavek, L., Simpson, W. R., Carlson, D., Domine, F., Douglas, T. A., and Sturm, M.: The chemical composition of surface snow in the Arctic: Examining marine, terrestrial, and atmospheric influences, *Atmos. Environ.*, 50, 349–359, 2012.

-We thank this referee to pointing out these studies. We have added a discussion of these measurements in the revised manuscript.

“This profile is consistent with a salinity source from the underlying sea ice and little influence from atmospheric deposition. Toom-Sauntry and Barrie (2002) find that freshly fallen snow itself tends to have low salinity (<0.01 psu). For simplicity, we assume the same salinity for surface snow on first-year and multi-year sea ice, although we recognize that in reality the surface snow may be less salty on multi-year sea ice due to less efficient upward transport of brine. Indeed, Krnavek et al. (2012) reported that the ion concentrations of surface snow sampled in the Alaskan Arctic display large variability depending on sea ice type: 0.01 psu for snow on multiyear sea ice, 0.1 psu for snow on thick first year sea ice and 0.8 psu for snow on thin first year sea ice. “

Page 5, line 9: "ease" instead of "easiness"

-We have made the change in the manuscript.

Page 7, around line 12: It would be useful to mention that modeled spatial maps will be presented later.

-We have made the change in the manuscript.

Page 8, line 30: The wording here is a bit confusing, because the normal conditions used for modeling frost flower formation include open water and cold temperatures. In this work, page 5, line 26 indicates that frost flowers are suppressed by the high winds (or are covered by drifting snow), which is the origin of the statement. Please reword this section to indicate clearly that the high winds (rather than open water and cold temperatures) are the reason for "inhibition of frost flowers".

-We have removed this statement from our manuscript, as based on referee #1's comments we now use a frost flower simulation with no wind inhibition. The wind inhibition simulation is included in the supplementary material.

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Wintertime enhancements of sea salt aerosol in polar regions consistent with a sea-ice source from blowing snow

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Abstract. Sea salt aerosols (SSA) are generated via air bubbles bursting at the ocean surface, as well as by wind mobilization of saline snow and frost flowers over sea-ice covered areas. The relative magnitude of these sources remains poorly constrained over polar regions, affecting our ability to predict their impact on halogen chemistry, cloud formation and climate. We implement a blowing snow and a frost flower emission scheme in the GEOS-Chem global chemical transport model, which we validate against multi-year (2001-2008) in situ observations of SSA mass concentrations at three sites in the Arctic, two sites in coastal Antarctica, as well as during a cruise in the Arctic (ICEALOT, 2008). A simulation including only open ocean emissions underestimates SSA mass concentrations by factors of 2-10 during winter-spring for all ground-based and ship-based observations. When blowing snow emissions are added, the model is able to reproduce observed wintertime SSA concentrations, with the model bias decreasing from a range of -80% to -34% for the open ocean simulation to -2% to +9% for the simulation with blowing snow emissions. We find that the frost flower parameterization cannot fully explain the high wintertime concentrations and displays a seasonal cycle decreasing too rapidly in early spring. Furthermore, the high day-to-day variability of observed SSA is better reproduced by the blowing snow parameterization. Over the Arctic ($>60^{\circ}\text{N}$) (Antarctic, $>60^{\circ}\text{S}$), we calculate that submicron SSA emissions from blowing snow account for 1.0 ~~(2.5)~~ (2.5) Tg yr^{-1} , while frost flower emissions lead to 0.25 ~~21~~ 21 Tg yr^{-1} (0.14) 25 Tg yr^{-1} compared to 0.78 1.0 Tg yr^{-1} from the open ocean. Blowing snow emissions are largest in regions where persistent strong winds occur over sea ice (East of Greenland, over the central Arctic, Beaufort Sea, as well as the Ross and Weddell Seas). In contrast, frost flower emissions are largest where cold air temperatures, and open leads ~~and mild winds~~ are co-located (over the Canadian Arctic Archipelago, coastal regions of Siberia, and off the Ross and Ronne ice shelves). Overall, in situ observations of mass concentrations of SSA suggest that blowing snow is likely to be the dominant SSA source during winter, with frost flowers playing a much smaller role.

1 Introduction

Breaking waves over the open ocean are recognized as the main mechanism for the global production of sea salt aerosol (SSA) (Lewis and Schwartz, 2004; de Leeuw et al., 2011 and references therein). Observations of SSA in polar regions, however, exhibit several characteristics that are not consistent with this canonical open ocean source. Indeed, submicron or

total SSA mass concentrations at Arctic (Sirois, et al., 1999; Quinn et al., 2002) and Antarctic sites (Wagenbach et al., 1998; Weller et al., 2008; Jourdain et al., 2008; Udisti et al., 2012) often exhibit a maximum during local winter, when polar ocean waters are mostly covered by sea-ice. Furthermore, the ionic composition of SSA observed at polar sites during winter shows a systematic depletion of the sulfate to sodium mass ratio relative to bulk sea water (Wagenbach et al., 1998; Rankin et al., 2000; Jourdain et al., 2008; Hara et al., 2012; Jacobi et al., 2012; Seguin et al., 2014). Finally, Arctic and Antarctic ice core records display factors of 2.5-4 increase in SSA deposition fluxes during glacial periods relative to warmer interglacial period (Wolff et al. 2006; Fischer et al., 2007; Abram et al., 2013).

To explain these seasonal and glacial-interglacial variations, frost flowers has been proposed as a new source of SSA (Wagenbach et al., 1998; Rankin et al. 2000, 2002; Wolff et al., 2003; Shaw et al., 2010). They are highly saline ice crystals that can rapidly form on freshly freezing sea ice (Martin et al., 1995; Domine et al., 2005; Roscoe et al., 2011). Frost flowers wick up brine from the sea ice and can be lofted in the atmosphere by surface winds to become SSA (Rankin et al. 2000; Domine et al., 2004; Xu et al. 2013 and references therein). The seasonality of frost flower formation and their sulfate to sodium ratios are similar to those of observed SSA in polar regions (Rankin et al., 2002; Rankin and Wolff, 2003; Wolff et al., 2003; Alvarez-Aviles et al., 2008; Beaudon and Moore, 2009; Seguin et al., 2014). Field observations have cast some doubt on the role of frost flowers as SSA, noting that frost flowers are rigid and difficult to break (Domine et al., 2005; Alvarez-Aviles et al., 2008). In particular, Obbard et al. (2009) observed no mechanical breakage of frost flowers in winds up to 6 m s⁻¹ over the Hudson Bay. Furthermore, laboratory experiments performed by Roscoe et al. (2011) demonstrated that no aerosol were produced when frost flowers were exposed to winds speeds up to 12 m s⁻¹. Their result is consistent with electron microscope imaging by Yang et al. (2017), which show that evaporating frost flowers form a cohesive chunk of salt that is unlikely to be a source of SSA.

Another hypothesis is that blowing snow, often observed over sea ice-covered regions (Nishimura and Nemoto, 2005; Savelyev et al., 2006), could act as a direct source of SSA (Simpson et al., 2007a; Yang et al., 2008). The snow over sea ice becomes salty by upward migration of brine from the sea ice to the snow surface, incorporation of frost flowers, as well as SSA deposition from the adjacent open ocean (Domine et al., 2004). The first two of these mechanisms lead to depletion of the sulfate to sodium ratio relative to bulk sea water as a result of the precipitation of mirabilite (Na₂SO₄·10H₂O) precipitates from brine at temperatures below -8°C during sea ice and frost flower formation (Alvarez-Aviles et al., 2008). Once lifted by wind, these salty snow particles can produce SSA via sublimation (Yang et al., 2008).

Many questions remain on the formation, composition, occurrence, and mobility of frost flowers and salty blowing snow. Most studies examining these two sources have focused on their potential role as an indirect source of gas-phase bromine resulting in ozone depletion events during late winter and early spring, with conflicting results as to which source would be most important. Kaleschke et al. (2004) developed a one-dimensional thermodynamic model to calculate frost flower coverage. They found that more than 66% of forward trajectories from areas with high frost flower coverage intercepted

regions with enhanced BrO over the Arctic and Antarctic during polar sunrise. Similarly, Jones et al. (2006) showed that ozone depletion events observed at Halley station in Antarctica were associated with air masses having recent contact with newly forming sea ice. However, using backtrajectories to examine the origin of enhanced BrO abundance measured at Barrow, Alaska, Simpson et al. (2007a) found that saline snow and ice on first-year sea ice was a more likely source of bromine than frost flowers. ~~Another line of evidence casting doubt on the role of frost flowers are field observations noting that frost flowers are rigid and difficult to break (Domine et al., 2005; Alvarez-Aviles et al., 2008). In particular, Obbard et al. (2009) observed no mechanical breakage of frost flowers in winds up to 6 m s^{-1} over the Hudson Bay. Furthermore, laboratory experiments performed by Roseoc et al. (2011) demonstrated that no aerosol were produced when frost flowers were exposed to winds speeds up to 12 m s^{-1} .~~ Yang et al. (2010) implemented a blowing snow bromine source in the p-TOMCAT chemistry-transport model. With this blowing snow source they were able to successfully simulate bromine explosion events retrieved from the Global Ozone Monitoring Experiment (GOME).

To our knowledge, the only modeling studies examining blowing snow and frost flowers as direct sources of SSA are the work of Xu et al. (2013, 2016), Levine et al. (2014), and Legrand et al. (2016). Xu et al. (2013) developed an empirical frost flower formulation in the WRF-Chem model. They found that adding frost flower emissions doubled the surface concentrations of Na^+ at Barrow, in better agreement with observations. However, their study was limited to two days during winter 2009. Their work was expanded in Xu et al. (2016), where the same frost flower emission scheme was implemented in the Community Earth System Model (CESM) for the year 2000 and compared to SSA observations at Barrow and Alert. They found that the frost flower simulation lead to improved agreement Barrow, but overestimated observations at Alert by 150%. Levine et al. (2014) found that the local winter peak in Na^+ mass concentrations at polar sites was attributable to blowing snow, but they were not able to constrain the relative sources of open ocean and blowing snow because rates of SSA wet deposition were tuned regionally in the p-TOMCAT model. Using the p-TOMCAT model, Legrand et al. (2016) report that 50-70% of wintertime Na^+ concentrations at two East Antarctic sites were due to sea ice emissions from blowing snow. However, the model overestimated observed Na^+ concentrations by factors of 2-3 and was not able to reproduce the observed seasonal cycle.

In this study, we implement a blowing snow and a frost flower parameterization in the GEOS-Chem global chemical transport model. We evaluate the ability of these two sources to reproduce multi-year (2001-2008) in situ measurements of Na^+ mass concentrations at three Arctic sites (Barrow, Alaska; Alert, Canada; Zeppelin, Svalbard), two coastal Antarctic sites (Neumayer and Dumont d'Urville), as well as Na^+ measurements obtained during the International Chemistry Experiment in the Arctic Lower Troposphere (ICEALOT) cruise during spring 2008. We then examine the relative contributions of open ocean, blowing snow and frost flower sources to the distribution of SSA over polar regions.

2 Model simulations and observations

2.1 The GEOS-Chem chemical transport model

We use the GEOS-Chem global 3-D chemical transport model (Bey et al., 2001) driven by the Modern Era Retrospective-Analysis for Research and Applications (MERRA; Rienecker et al., 2011) meteorological fields. The MERRA fields have a native horizontal resolution of 1/2° latitude by 2/3° longitude with 72 vertical levels. We regrid these fields to a 2°×2.5° horizontal resolution and vertical levels above 80 hPa are merged to retain 47 vertical levels in total for computational expediency. The temporal resolution of MERRA data is 3 hours except for surface variables and mixing depths, which have a 1-hour resolution. The sea ice concentration boundary conditions in MERRA are derived from the weekly [sea-surface temperature analysis](#) product of Reynolds et al. (2002), [which is based on Special Sensor Microwave Imager \(SSM/I\) instruments on Defense Meteorological Satellite Program \(DMSP\) satellites. The weekly products are](#) linearly interpolated in time to each model time step. In this study we use GEOS-Chem v10-01 (<http://www.geos-chem.org>).

The GEOS-Chem open ocean SSA simulation is described in Jaeglé et al. (2011). Ocean emissions are based on the wind speed-dependent source function of Gong (2003) and Monahan et al. (1986), with an empirical dependence on sea surface temperature (SST) derived by Jaeglé et al. (2011) via comparisons to open ocean cruise observations of coarse mode SSA mass concentrations. This SST dependence leads to a decrease in SSA emissions of a factor of 2.6 as SST decreases from 25°C to 5°C, consistent with the factors of 2-3 measured in laboratory experiments for particles with a radius greater than 0.5 µm (Bowyer, 1984, 1990; Woolf et al., 1987; Mårtensson et al., 2003; Sellegri et al., 2006). Most of the in situ observations of SSA mass concentrations used to derive the polynomial SST dependence (Fig. 6 in Jaeglé et al., 2011: $f(\text{SST})=0.3 + 0.1 \times \text{SST} - 0.0076 \times \text{SST}^2 + 0.00021 \times \text{SST}^3$) were for SST > 5°C. In this work, we find that the SST dependence results in a factor of 2 overestimate of summertime SSA observations at coastal polar sites with SST ranging from -2°C to 5°C. This indicates that the suppression at cold SST might not be strong enough. We thus modify the expression derived in Jaeglé et al. (2011) to impose $f(\text{SST})=0.25$ for SST < 5°C (see Fig. S1 in the Supplement). [This is consistent with the laboratory study of Mårtensson et al. \(2003\), who report a 50-60% decrease in aerosol production \(for \$r > 0.1 \mu\text{m}\$ \) when seawater temperature decreased from 5°C to -2°C. We note that another potential explanation for the summertime overestimate in SSA mass concentrations is inefficient wet removal from low-intensity summer precipitation in GEOS-Chem \(Croft et al., 2016\).](#)

[Within GEOS-Chem, we assume that open ocean emissions occur only in gridboxes covered by more than 50% water. We thus neglect emissions from bubble bursting in leads within sea ice. This is based on observations that the small fetch of leads results in SSA production which is an order of magnitude lower compared to open ocean \(Nilsson et al. 2001\). Including SSA emissions over leads results in a 1% increase in SSA emissions over polar regions \(>60°\). Even if we were to assume that leads were as efficient as the open ocean in producing SSA, this would only result in a 10% increase in SSA emissions.](#)

Dry deposition of SSA over land accounting for particles growth under high humidity conditions follows the size-segregated scheme described in Zhang et al. (2001). The dry deposition velocity over the ocean is calculated based on the Slinn and Slinn (1980) deposition model for natural waters. Over snow and ice surfaces, Fisher et al. (2011) implemented a dry deposition velocity of 0.03 cm s^{-1} based on the measurements of Nilsson and Rannik (2001). The wet deposition scheme includes convective updraft scavenging, rainout and washout from precipitation (Liu et al., 2001), as well as snow scavenging (Wang et al., 2011). For this work, we ~~use~~track SSA mass in two-SSA size bins: accumulation mode ($r_{\text{dry}} = 0.01\text{--}0.5 \text{ }\mu\text{m}$) and coarse mode ($r_{\text{dry}} = 0.5\text{--}4 \text{ }\mu\text{m}$), except in the comparison to in situ mass concentrations of ~~Na⁺SSA~~ for which we use $r_{\text{dry}} = 0.01\text{--}0.3 \text{ }\mu\text{m}$ and $r_{\text{dry}} = 0.3\text{--}3 \text{ }\mu\text{m}$ (see ~~below~~)-section 2.3). In the rest of the manuscript we will refer to the accumulation and coarse mode SSA as submicron and supermicron SSA based on their diameters.

The GEOS-Chem SSA simulation was evaluated by Jaeglé et al. (2011) against in situ measurements of SSA from 6 open ocean cruises (mean normalized bias of +33%) and 15 ground-based stations (mean normalized bias of -5%) as well as aerosol optical depth (AOD) from MODIS and AERONET. Detailed comparisons of GEOS-Chem black carbon and organic aerosol (Wang et al., 2011) as well as sulfate and ammonium aerosol (Fisher et al., 2011) to ground-based and aircraft observations over the Arctic during winter and spring suggest that transport and removal processes are reasonably captured by the model.

2.2 Implementation of blowing snow and frost flower parameterizations in GEOS-Chem

We implement SSA emissions from blowing snow following the parameterization of Yang et al. (2008, 2010), with a few modifications. The SSA production from blowing snow is a function of relative humidity, temperature, age of snow, snow salinity and wind speed. The wind needs to be strong enough ($>$ about 7 m s^{-1}) to ~~lift up~~saltate and suspend snow particles from the sea ice surface. The size distribution of ~~lofted~~suspended blowing snow particles follows a two-parameter gamma distribution (Yang et al., 2008 and references therein). We set a uniform surface snow salinity of 0.1 psu (practical salinity unit) over Arctic sea ice based on observations of surface snow salinity (Mundy et al., 2005; Krnavek et al., 2012). This is an order of magnitude lower than the salinity assumed in Yang et al. (2008, 2010). Their salinity was based on bulk snow measurements (Massom et al., 2001), which overestimate surface snow salinity because of the rapid decreases of salinity with height above the ice surface. For example, Mundy et al. (2005) found that springtime snowpack over first year sea ice in the central Canadian Arctic displayed a salinity of 7.6 ± 3.3 psu in the bottom snow layers and decreased to 0.26 ± 0.37 psu and 0.11 ± 0.25 psu in the middle and surface snow layers. This profile is consistent with a salinity source from the underlying sea ice and little influence from atmospheric deposition. Toom-Sauntry and Barrie (2002) find that freshly fallen snow itself tends to have low salinity (<0.01 psu). For simplicity, we assume the same salinity for surface snow on first-year and multi-year sea ice, although we recognize that in reality the surface snow may be less salty on multi-year sea ice due to less efficient upward transport of brine. ~~We assume a salinity of 0.03 psu for surface snow on Antarctic sea ice, following the approach of Yang et al. (2008, 2010) based on observations showing that wintertime precipitation over the Arctic is about~~

~~1/3 of that over the Antarctic (60–70°S) (Huffman et al., 2001). Indeed, Krnavek et al. (2012) reported that the ion concentrations of surface snow sampled in the Alaskan Arctic display large variability depending on sea ice type: 0.01 psu for snow on multiyear sea ice, 0.1 psu for snow on thick first year sea ice and 0.8 psu for snow on thin first year sea ice. Snowpack on Antarctic sea ice is thicker than over Arctic sea ice. Indeed, Antarctica is surrounded by the Southern Ocean which brings moisture, while the Arctic is surrounded by land with wintertime precipitation that is 3 times lower than over Antarctic sea ice (Huffman et al., 2001). As the salinity of snow decreases with snowpack thickness, we assume that the salinity of snow on Antarctic sea ice is 0.03 psu, a factor of 3 lower than over the Arctic following Yang et al. (2010).~~

~~2001)~~ Based on the time between precipitation events in the MERRA fields, we estimate a mean snow age of 3 days for the Arctic and 1.5 days for the Antarctic. In comparison, Yang et al. (2008) used a globally uniform snow age of 3 days, while Levine et al. (2014) assumed 5 days. Our younger snow age over Antarctic sea ice (1.5 days) increases the ~~easiness~~ of lifting snow particles and SSA production increases by 40% compared to a 3 day snow age. We also assume that 5 SSA particles are produced per snowflake. This fractionation can occur when snowflakes are broken by strong winds and abraded into smaller particles with round corners (Mellor, 1965), or when SSA particles experience cracking during sublimation under low relative humidity as observed in the laboratory experiments of Wise et al. (2012). The number of particles produced per snowflake (N) does not change the total SSA production substantially, but it influences the size distribution of SSA particles produced from these lofted snow particles. We choose the value of N=5 based on wintertime observations of supermicron and submicron SSA at Barrow (see Fig. S2 in the Supplement). This leads to a doubling of submicron SSA production compared to the assumption of N=1 in Yang et al. (2008). Our simulation yields blowing snow SSA emissions with 38% of SSA mass in the submicron range (0.01–0.5 μm) and 62% in the supermicron range (0.01–0.5 μm) for the Arctic. As we assume a lower salinity in the Antarctic, more of the blowing snow emissions are in the submicron range (60%) in that region. Overall, our modifications to the Yang et al. (2008, 2010) parameterization lead to SSA emissions from blowing snow that are an order of magnitude lower than in Yang et al. (2010).

We implement a frost flower SSA source in GEOS-Chem following the parameterization of Xu et al. (2013, 2016), which is based on the potential frost flower (PFF) coverage derived by Kaleschke et al. (2004). Frost flowers are formed on very young sea ice once ambient air temperatures are cold enough ($< -20^\circ\text{C}$). Below that threshold, PFF increases rapidly as temperature decreases. We set a limit of 10 cm for the thickness of newly formed sea ice beyond which we assume that frost flower crystals can no longer be formed. Based on the thermodynamic model of Kaleschke et al. (2004), it takes 1-2 days for sea ice to reach a thickness of 10 cm for air temperatures of -40 to -20°C . Following Xu et al. In addition, we assume (2013), we assume that SSA from frost flowers have a lognormal size distribution with a geometric mean diameter of 0.015 μm and a geometric standard deviation of 1.9, hence all frost flower emissions occur in the submicron range ($r_{\text{dry}} \leq 0.5 \mu\text{m}$). We use the same scaling factor of $5 \times 10^6 \text{ m}^2 \text{ s}^{-1}$ as Xu et al. (2013) for our frost flower source. We also conducted a sensitivity simulation (Fig. S3), in which we assumed that frost flowers can only form under mild wind speed conditions ($< 5 \text{ m s}^{-1}$), as

strong winds inhibit frost flower formation and bury existing frost flowers with snow (Perovich and Richeter-Menge, 1994; Rankin et al., 2000). ~~2000). Following Xu et al. (2013), we assume that SSA from frost flowers have a lognormal size distribution with a geometric diameter of 0.015 μm and a standard deviation of 1.9, hence all frost flower emissions occur in the submicron range ($r_{\text{dry}} \leq 0.5 \mu\text{m}$). We use a scaling factor of $5 \times 10^6 \text{ m}^2 \text{ s}^{-1}$ for our frost flower source. This is larger than the $10^6 \text{ m}^2 \text{ s}^{-1}$ scaling factor used in Xu et al. (2013), but because of our modifications related to ice thickness and wind speed we estimate that the magnitude of our frost flower emissions is a factor of 5–10 lower than in Xu et al. (2013). Our choice of this scaling factor is based on reproducing wintertime observations of SSA at Alert, where we predict the largest influence from frost flowers (see section 3).~~

We conduct three simulations. Our standard simulation (STD) includes only the open ocean source of SSA. In a second simulation (STD-SNOW), we add the blowing snow source to the STD simulation. A third simulation (STD-FF) adds frost flower emissions to the STD simulation.

2.3 In situ observations

We use in situ observations of Na^+ mass concentrations from 5 polar sites: Barrow, Alaska (71.3°N, 156.6°W; 11m a.s.l.; Quinn et al., 2002); Alert, Nunavut, Canada (82.5°N, 62.5°W; 210m a.s.l., WMO/GAW, 2003); Zeppelin Mountain, Svalbard, Norway (78.9°N, 11.9°E; 475m a.s.l, WMO/GAW, 2003); Neumayer (70.7°S, 8.3°W; 42m a.s.l; Weller et al., 2008) and Dumont d'Urville (66.7°S, 140°E; 43m a.s.l.; Legrand et al., 2012). These observations are available for 2001–2008 (except for Neumayer station, 2001–2007). At Barrow Na^+ mass concentrations are available for both submicron and supermicron aerosol, while all the other sites measure total mass concentrations. ~~For comparison between the GEOS-Chem model and the observations, we convert Na^+ mass concentration to SSA mass concentrations using a factor of 3.256 (Riley and Chester, 1971) and use two size bins in the model ($r_{\text{dry}} = 0.01–0.3 \mu\text{m}$ and $r_{\text{dry}} = 0.3–3 \mu\text{m}$), which approximately correspond to the cut off diameters of the measurements (Wagenbach et al., 1998; Quinn et al., 2002; WMO/GAW, 2003; Weller et al., 2008). The Na^+ mass concentrations are determined by ion chromatography with uncertainties of 5%–11% (0.01 $\mu\text{g}/\text{m}^3$ in absolute uncertainty).~~ The aerosol sampling frequency ranges from daily (Zeppelin, Dumont d'Urville, submicron at Barrow) to weekly (Alert, Neumayer, Barrow supermicron). In winter months, the coastlines near these sites are mostly covered by sea ice.

We also use the submicron Na^+ mass concentrations measured aboard the R/V *Knorr* during the International Chemistry Experiment in the Arctic Lower Troposphere (ICEALOT) cruise in March–April 2008 (<http://saga.pmel.noaa.gov/Field/icealot>). The research cruise took place over the North Atlantic Ocean and the ice-free Arctic Ocean (41°–81°N). ~~To compare these submicron observations to GEOS-Chem we use a model size bin of $r_{\text{dry}} = 0.01–0.4 \mu\text{m}$ as in Jaeglé et al. (2011).~~

For comparison between the GEOS-Chem model and the observations, we convert observed Na⁺ mass concentrations to SSA mass concentrations using a factor of 3.256 based on the mass ratio of Na⁺ in seawater (Riley and Chester, 1971). For frost flowers, this ratio is 3.237 (Rankin et al., 2000). Krnavek et al. (2012) find a ratio of 3.24-3.278 for snow on first year sea ice. As this ratio varies by less than 0.5% for these different SSA sources, we use a constant factor of 3.256.

- 5 The reported aerodynamic cut-off diameters of the measurements are 1 μm and 10 μm at Barrow and during ICEALOT (Quinn et al., 2002), and 7-10 μm at the other sites (Wagenbach et al., 1998; WMO/GAW, 2003; Weller et al., 2008). In order to compare to model simulations, we need to convert these aerodynamic diameters to dry geometric radii. This conversion depends on aerosol density, relative humidity during sampling, and whether the particle is spherical (Seinfeld and Pandis, 2006). For example, a 10 μm aerodynamic diameter could correspond to a dry geometric radius of 2.3 μm (80% RH,
- 10 1.2 g/cm³ pure NaCl solution, a factor of 2 growth between dry and 80% RH), 3 μm (dry cubical NaCl particle, 2.2 g/cm³, Lewis and Schwartz, 2004), or 3.8 μm (30% RH, ammonium sulfate and sea salt aerosol, 1.7 g/cm³, Quinn et al., 1996). Thus for comparison to observations we choose the mid-range estimate and conduct a simulation with two size bins: r_{dry} = 0.01–0.3 μm and r_{dry} = 0.3–3 μm.

3 Model evaluation with ground-based and ship-based in situ observations

- 15 Observations at the three Arctic sites display enhanced SSA mass concentrations of 1-3 μg m⁻³ during the cold part of the year from November to April (Fig. 1a-c). In contrast, when the sea ice retreats during summer and late fall, SSA concentrations are much lower (<0.5 μg m⁻³ at Alert and Zeppelin). This seasonality is opposite to what is expected from an open-ocean source. Indeed, we find that the STD simulation fails to capture the high wintertime concentrations at all three Arctic sites but reproduces the summer/late fall observations reasonably well. During winter at Barrow and Alert, the STD
- 20 simulation predicts very low SSA concentrations (< 0.1 μg m⁻³), while at Zeppelin, which is closer to the open Atlantic Ocean, STD mass concentrations reach 0.5-1 μg m⁻³.

At Neumayer station (Fig. 1d), SSA observations show a broad maximum of 1-1.5 μg m⁻³ during the cold months (March-September). The seasonality is opposite at Dumont d'Urville (Fig. 1e), with a summertime maximum of 2.9 μg m⁻³, as it is exposed to a longer open ocean season compared to Neumayer (Wagenbach et al. 1998). Between March and November,

25 SSA concentrations remain fairly constant around 0.8 μg m⁻³. The summertime maximum at Dumont d'Urville is captured by the STD simulation, confirming the open ocean source. However, the STD model predicts SSA concentrations <0.3 μg m⁻³ during cold months, factors of 3-10 lower than observations at both Antarctic sites.

The addition of a blowing snow source in GEOS-Chem (STD-SNOW) results in improved agreement with observations. The normalized mean bias (NMB=($\overline{M}/\overline{O}$ Model/ \overline{O} bs - 1)×100) decreases significantly at all five sites: Barrow (STD: -64%,

30 STD-SNOW:+9%), Alert (STD: -85%; STD-SNOW: +25%); Zeppelin (STD: -34%; STD-SNOW: +12%); Neumayer (STD:

-63%; STD-SNOW: -2%); Dumont d'Urville (STD: -40%; STD-SNOW: +12%). The STD-SNOW simulation captures the observed SSA seasonal cycle reasonably well, with modeled wintertime SSA mass concentrations increasing to 1-2 $\mu\text{g m}^{-3}$.

Overall, the frost flower simulation (STD-FF) displays a large geographical variability, with little influence at Dumont d'Urville and Zeppelin, but much larger influence at Barrow, Neumayer and especially at Alert, where modeled SSA concentrations reach ~~2~~1 $\mu\text{g m}^{-3}$. Indeed, the STD-FF simulation predicts very large SSA emissions over the Canadian Arctic Archipelago (see section 4) [for a detailed discussion and maps of spatial distributions](#). The NMB in the STD-FF simulation ranges from ~~-47~~49% (Neumayer) to ~~+38~~-27% (Alert) ~~(Zeppelin)~~, not displaying as large an improvement as the STD-SNOW simulation. Furthermore, the seasonal cycle of frost flower SSA concentrations decreases too rapidly during early spring compared to observations at Alert, Barrow and Neumayer.

10 We examine in more detail the daily variability in submicron SSA at Barrow for January-July 2001 (Fig. 2a). Between January and late April, the observations show large day-to-day variations with concentrations ranging from $<0.5 \mu\text{g m}^{-3}$ to 2-4 $\mu\text{g m}^{-3}$. These SSA enhancements last for 1-7 days. We find that the timing and magnitude of these events are often reproduced by the blowing snow simulation (observations: $0.98 \pm 0.9 \mu\text{g m}^{-3}$; STD-SNOW: $0.94 \pm 0.8 \mu\text{g m}^{-3}$) and are driven in part by variability in windspeed (Fig. 2b, gray shaded areas). Some events are not associated with high local winds (29
15 January - 5 February; 24-28 March), and are due to transport from nearby regions. For example, the high levels of SSA submicron concentrations seen in the blowing snow simulation on 29 January - 5 February are associated with wind-blown snow coming from sea ice in the northern Beaufort Sea. In contrast, the frost flower simulation fails to capture the variability and magnitude of observed SSA events (STD-FF: ~~0.34~~28 \pm ~~0.39~~29 $\mu\text{g m}^{-3}$). An examination of weekly SSA mass concentrations at Alert and daily concentrations at Dumont d'Urville for 2001 yields similar conclusions (see Fig. ~~S3~~S4 in
20 the Supplement).

Figure 3a shows submicron SSA mass concentrations measured aboard the R/V *Knorr* during the ICEALOT experiment in March-April 2008. The first part of the cruise took place over the North Atlantic, where the largest enhancements in SSA mass concentrations (1-2 $\mu\text{g m}^{-3}$ on 25-26 March and 29 March) were due to open ocean SSA emissions and are reproduced by the STD simulation. As the R/V *Knorr* travelled towards the Norwegian, Barents and Greenland Seas (3-20 April), the
25 STD simulation predicts very low SSA concentrations ($<0.2 \mu\text{g m}^{-3}$) and can no longer reproduce the observed concentrations (0.5-2 $\mu\text{g m}^{-3}$). The STD-FF simulation produces enhancements that are too weak, but the STD-SNOW simulation captures some of these enhancements, in particular on 6-7 April (the R/V *Knorr* was along the Norwegian coast) and 15-19 April (near the coast of Svalbard). During both periods, Gilman et al. (2010) report concurrent decreases in observed O_3 and in the acetylene to benzene ratio, indicative of destruction of surface O_3 by Br and oxidation of acetylene by
30 both Br and Cl. Figure 3c shows that based on our STD-SNOW simulation, a major blowing snow event developed on 15-19 April over the central Arctic, poleward of 80°N . At that time the R/V *Knorr* was positioned within a few kilometres off the sea ice edge and the observed O_3 decreased from 43 ppbv to 1.5 ppbv (Gilman et al., 2010). The STD-SNOW simulation

predicts an increase in SSA concentrations of up to $3 \mu\text{g m}^{-3}$ (Fig. 3a, shaded gray area) reproducing the timing of the observed enhancement, but overpredicting the observed concentrations by a factor of 2–3. During this event the FLEXPART Lagrangian particle dispersion model predicts that 50–60% of the air masses intercepted by the ship were exposed to multi-year sea ice and 50–40% to first year sea ice (Gilman et al., 2010). Thus, the model overestimate could be due to our assumption of uniform snow salinity of 0.1 psu over both first and multi year sea ice, while in reality the salinity of snow on multi-year sea ice is likely lower. $1.5 \mu\text{g m}^{-3}$ (Fig. 3a, shaded gray area) reproducing the timing and magnitude of the observed enhancement.

Overall, we find that the blowing snow source can explain the large wintertime enhancements in observed SSA mass concentrations over both the Arctic and Antarctic regions. Furthermore, the STD-SNOW simulation captures the episodic nature of the observed enhancements. The frost flower source reproduces some of the observed enhancements over the Arctic, but is not able to match the high SSA concentrations over coastal Antarctica, and does not have a strong enough day-to-day variability. It is possible that both blowing snow and frost flower emissions act together, however our simulations. However, when we add the contributions from both sources, we find that modelled SSA mass concentrations are a factor of 2–3 too high compared to observations at Barrow and Alert (Fig. S5). In particular, the frost flower simulation leads to a peak in SSA in February at Barrow, which is not observed. Our simulations thus suggest that the dominant influence is from blowing snow.

4 Emissions and distributions of SSA over polar regions

Table 1 summarizes the annual SSA budgets over the Arctic and Antarctic as calculated in GEOS-Chem for the year 2005 poleward of 60° latitude (see Table S1 in the Supplement for the global budgets). We find that annual SSA emissions vary by 10–30% for 2004–2008, but the overall seasonality and spatial distribution of emissions are similar from year to year. The total (0.01– $4 \mu\text{m}$) blowing snow source is 2.6 Tg yr^{-1} for the Arctic and 4.2 Tg yr^{-1} for Antarctica. We find that the larger blowing snow source over Antarctica, despite the lower snow salinity, is a result of faster winds over Antarctic sea ice. Furthermore, the younger age of snow assumed over Antarctic sea ice (1.5 days compared to 3 days over Arctic sea ice) contributes to 30% of the difference in blowing snow emissions between the Arctic and Antarctic. In contrast, we find that the frost flower emissions are weaker slightly stronger over the Antarctic (0.425 Tg yr^{-1}) than the Arctic (0.25 Tg yr^{-1}); because Antarctic coastal regions where cold temperature and leads coexist often have 21 Tg yr^{-1} due to strong katabatic winds, which inhibit over the formation of frost flowers in our simulation Antarctic. The open ocean accounts for 30 Tg yr^{-1} over the Arctic and 40 Tg yr^{-1} over Antarctica. Examining submicron SSA ($r_{\text{dry}} = 0.01\text{--}0.5 \mu\text{m}$), we see that this is the size range where blowing snow (Arctic: 1.0 Tg yr^{-1} ; Antarctic: 2.5 Tg yr^{-1}) and frost flower (0.25 21 Tg yr^{-1} ; 0.425 Tg yr^{-1}) emissions have their largest impact relative to the open ocean (0.78 Tg yr^{-1} ; 1.0 Tg yr^{-1}). Therefore, in This difference in size distributions is related to the different physical mechanisms for SSA emissions from open ocean emissions (breaking waves

and bubble bursting) compared to blowing snow (saltation of fallen snow and suspension) or frost flowers (saltation of broken frost flower crystals and suspension). The original crystalline form of snow particles/frost flower fragments are expected to be shattered by repeated impacts with the ground and other particles during saltation. Sublimation of ice from these particles leads to relatively small SSA compared to bubble bursting in the open ocean. In the following sections we

5 focus on the seasonality and spatial distribution of submicron SSA emissions (Fig. 4 and 5).

4.1 Arctic

Figure 4a shows the seasonal evolution of our three SSA sources over the Arctic (>60°N). SSA emissions from the open ocean maximize in September-October as a result of strong winds combined with minimum sea ice extent. During winter months, SSA emissions from the open ocean are largest over the ice-free North Atlantic Ocean, extending towards the

10 Barents Sea (Fig. 4b). SSA emissions from blowing snow reach their maximum in December-April (Fig. 4a) with the largest emissions occurring over sea ice covered regions with the strongest winds (Fig. 4c): East of Greenland, over the central Arctic and the Beaufort Sea. The modeled blowing snow SSA surface mass concentrations reach 2-3.5 $\mu\text{g m}^{-3}$ over these regions (Fig. 5c). We find that atmospheric transport leads to inland incursions of blowing snow SSA over northern Canada, Alaska, and Siberia (Fig. 5c).

15 Frost flower emissions maximize in December-March and are ~~2-3~~4 times smaller than blowing snow emissions during these months (Fig. 4a). We find that frost flower emissions are highly localized with the strongest emissions over the Canadian Arctic Archipelago (Fig. 4d), where surface concentrations of SSA reach ~~2-3~~5 $\mu\text{g m}^{-3}$ (Fig. 5d), explaining the large influence we noted at Alert and Barrow (Fig. 1). Weaker emissions occur over coastal Siberia and in leads located within the central Arctic sea ice. In our simulation, the location of frost flower emissions largely depends on the simultaneous

20 occurrence of very cold air temperatures (~~<-20°C~~) and open leads, ~~and mild winds (<5 m s⁻¹)~~. Other regions in the Arctic have cold temperatures ~~and open leads~~ during winter, however they ~~tend to be associated~~ are mostly covered by sea ice with strong winds, which prevent the formation of limited open leads areas. Our emissions from frost flowers over the Arctic (0.21 Tg yr⁻¹) are consistent the accumulation model emissions reported by Xu et al. (2016) (0.24 Tg for November-February, their Table 2). We also find a similar geographic distribution.

25 We note that the mean lifetime of both blowing snow and frost flower submicron SSA is 6-7 days in the Arctic, nearly twice as long as open ocean SSA (Table 1). ~~We assume~~ Open ocean SSA form over lower latitude warmer regions, while sea ice SSA emissions occur at higher latitudes under much colder conditions, with less efficient removal processes in mixed-phased and ice clouds. The current parameterization in GEOS-Chem assumes that in-cloud scavenging of SSA does not occur in cold clouds (T<258 K) ~~as they are not efficient ice nuclei~~ (Wang et al., 2011), thus wintertime sea-ice generated

30 SSA are only removed by below-cloud scavenging (which is slow for accumulation mode aerosols) and dry deposition. Recent laboratory studies have shown that SSA could act as ice nuclei by deposition freezing (Wise et al., 2012) and

immersion freezing (DeMott et al., 2016), and might thus undergo in-cloud scavenging in mixed and ice clouds. This process is not currently included in GEOS-Chem.

4.2 Antarctic

In the southern hemisphere polar regions, open ocean SSA emissions display a weak seasonal cycle due to persistent strong winds over the Southern Ocean (Fig. 4e). During austral winter, emissions from the open ocean are strongest at $\sim 50^\circ\text{S}$ leading to modeled surface SSA concentrations of $1\text{--}3\ \mu\text{g m}^{-3}$ (Fig. 4f and 5f). Blowing snow emissions maximize in June–October (Fig. 4a) and are strongest over the sea ice of the Ross and Amundsen Seas, because of the strong katabatic winds flowing off the Antarctic Plateau, as well as strong winds in the Indian Ocean sector (Fig. 4g). In these regions, modeled submicron SSA concentrations from salty snow reach $1\text{--}3\ \mu\text{g m}^{-3}$, explaining the increase of $1\text{--}2\ \mu\text{g m}^{-3}$ seen at Neumayer and Dumont d’Urville (Fig. 4e and Fig. 1d–e). The model predicts that frost flower emissions are concentrated near the Ross ~~and~~ Ronne ~~and Amery~~ ice shelves and along coastlines (Fig. 4h), accounting for $1\text{--}2\ \mu\text{g m}^{-3}$ surface submicron SSA over these regions (Fig. 5h). Neumayer thus receives influence from frost flowers formed off the Ronne ice shelf (Fig. 5h and 1d), ~~in contrast to while~~ Dumont d’Urville ~~where minimal frost flower~~ has a weaker influence ~~occurs from frost flowers forming along the local coastline~~ ($<0.1\ \mu\text{g m}^{-3}$).

Spatially, we find that the locations of blowing snow and frost flower emissions are complementary to each other due to the different requirements of ~~wind ($>7\ \text{m s}^{-1}$ for blowing snow compared to $<5\ \text{m s}^{-1}$ for frost flowers) and~~ sea state (sea ice compared to open leads).

5 Discussion and conclusions

In this work, we implement two new SSA emission schemes in the GEOS-Chem chemical transport model: a blowing snow parameterization following the work of Yang et al. (2008, 2010) and a frost flower parameterization based on Xu et al. (2013) and Kaleschke et al. (2004). We find that the GEOS-Chem simulation with open ocean emissions fails to capture the elevated SSA mass concentrations observed at five coastal stations in the Arctic and Antarctic during winter (2001–2008) and during the ICEALOT research cruise in March–April 2008. When blowing snow emissions are added, the model is able to reproduce the wintertime observed SSA levels as well as their large day-to-day variability driven by wind speed. We find that the frost flower parameterization cannot fully explain the high wintertime concentrations and displays a seasonal cycle decreasing too rapidly in early spring. Furthermore, our frost flower simulation cannot reproduce the large daily variability of observed SSA. ~~Our results thus suggest that blowing snow is the dominant source of SSA in polar winter, although frost flowers cannot be entirely ruled out. In particular, they may contribute indirectly to SSA emissions by calinating wind-blown snow (Obbard et al., 2009).~~

Over the Arctic, we estimate that annual blowing snow emissions of submicron SSA are 1.0 Tg yr^{-1} , compared to 0.8 Tg yr^{-1} from the open ocean. Over the Antarctic, these emissions are 2.5 Tg yr^{-1} for blowing snow and 1.0 Tg yr^{-1} for the open ocean. Blowing snow emissions are mostly controlled by wind speed and are thus larger over the Antarctic due to the strong katabatic winds off the Antarctic Plateau and strong westerlies over the Southern Ocean. Frost flower SSA emissions are $0.2521 \text{ Tg yr}^{-1}$ over the Arctic ($0.1425 \text{ Tg yr}^{-1}$ for the Antarctic) and depend on the co-location of cold air temperatures, ~~mild winds~~ and open leads.

The parameterizations for blowing snow and frost flowers have several intrinsic assumptions, such as the salinity of snow and the scaling factor for frost flowers, which will affect the relative magnitudes of these two sources in polar regions. The geographic distribution, seasonal cycle, and daily variability of these sources, however, are controlled by sea ice extent and meteorological parameters (winds and temperature). In this study, we showed that the temporal and geographical variability of SSA observations at five polar sites is more consistent with blowing snow than with frost flowers. Based on this comparison, we conclude that blowing snow is likely to be the dominant source of SSA in polar winter, although frost flowers cannot be entirely ruled out. In particular, they may contribute indirectly to SSA emissions by salinating wind-blown snow (Obbard et al., 2009).

These polar sources of SSA are subject to substantial uncertainties due to the limited observations available. ~~Our assumption of~~ One key uncertainty in our simulations is snow salinity. Indeed, SSA emissions from blowing snow have a near-linear dependence on the salinity of snow. Thus a doubling of the assumed salinity would lead to a doubling in SSA emissions from blowing snow. Furthermore, we assume a uniform salinity of snow over both first-year and multi-year sea ice. This likely overestimates the contribution of blowing snow SSA over the western Arctic, which is dominated by multi-year sea ice. More extensive observations of surface snow salinity at multiple locations over both first-year and multi-year sea ice can help further refine these assumptions. Sampling of SSA size distributions during blowing snow events can help determine the number of particles per snowflake, which we determined empirically in this study. This number will not affect to total SSA emissions, but will change the relative importance of submicron and supermicron SSA emissions. There is insufficient knowledge on frost flower occurrence, growth and their mobilization by winds. In particular, the role of favourable wind conditions, as well as the ice thickness for frost flower to grow, are highly uncertain, and thus the predicted locations of frost flower emissions in our simulation are also uncertain.

Reducing these remaining uncertainties would help constrain how sea ice emissions of SSA affect the chemistry of the polar atmosphere by acting as a source of halogens, leading to ozone and mercury depletion events (Barrie et al., 1988; Fan and Jacob, 1992; Simpson et al., 2007b; Schroeder et al., 1998; Steffen et al., 2008). Improved process-based understanding of these emissions would also lead to better constraints on the potential climatic impact of wintertime SSA, ~~which could increase cloud droplet number concentrations and hence increase downward longwave radiative forcing (Xu et al., 2013).~~ on clouds, in particular mixed-phase and ice clouds, which have a strong influence on downward longwave radiative forcing.

Indeed, recent studies have shown the role of SSA as ice nuclei (Wise et al., 2012; DeMott et al., 2016). Thus over the Arctic and Antarctic regions, where the abundance of other ice nuclei such as dust or black carbon are low, SSA from local sea ice sources could influence the formation, radiative forcing, and precipitation of mixed-phase and ice clouds.

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Table 1: Arctic (>60°N) and Antarctic (>60°S) SSA budgets for the open ocean, blowing snow, and frost flower sources for the year 2005.

	Open ocean			Blowing Snow			Frost Flowers
	0.01-0.5 μm	0.5-4 μm	Total	0.01-0.5 μm	0.5-4 μm	Total	0.01-0.5 μm
Arctic (>60°N)							
Emission (Tg yr^{-1})	0.78	29	30	1.0	1.6	2.6	0.25 <u>21</u>
Dry deposition (Tg yr^{-1})	0.13	13	13	0.35	0.77	1.1	0.14 <u>091</u>
Wet deposition (Tg yr^{-1})	1.2	20	21	0.43	0.89	1.3	0.047 <u>050</u>
Lifetime (days)	3.5	0.36	0.48	6.6	0.72	2.6	6.9 <u>7.0</u>
Burden (Tg <u>Gg</u>)	12	32	45	14	3.3	17	3.6 <u>2.7</u>
Surface concentration ($\mu\text{g m}^{-3}$)	0.19	1.0	1.2	0.40	0.17	0.57	0.19 <u>11</u>
Antarctic (>60°S)							
Emission (Tg yr^{-1})	1.0	39	40	2.5	1.7	4.2	0.14 <u>25</u>
Dry deposition (Tg yr^{-1})	0.30	24	24	0.41	0.62	1.0	0.058 <u>082</u>
Wet deposition (Tg yr^{-1})	2.6	25	28	1.2	1.0	2.2	0.036 <u>074</u>
Lifetime (days)	3.5	0.38	0.55	4.4	0.52	2.4	6.6 <u>3</u>
Burden (Gg)	28	50	78	19	2.4	21	12 <u>.7</u>
Surface concentration ($\mu\text{g m}^{-3}$)	0.46	1.6	2.1	0.45	0.093	0.54	0.077 <u>094</u>

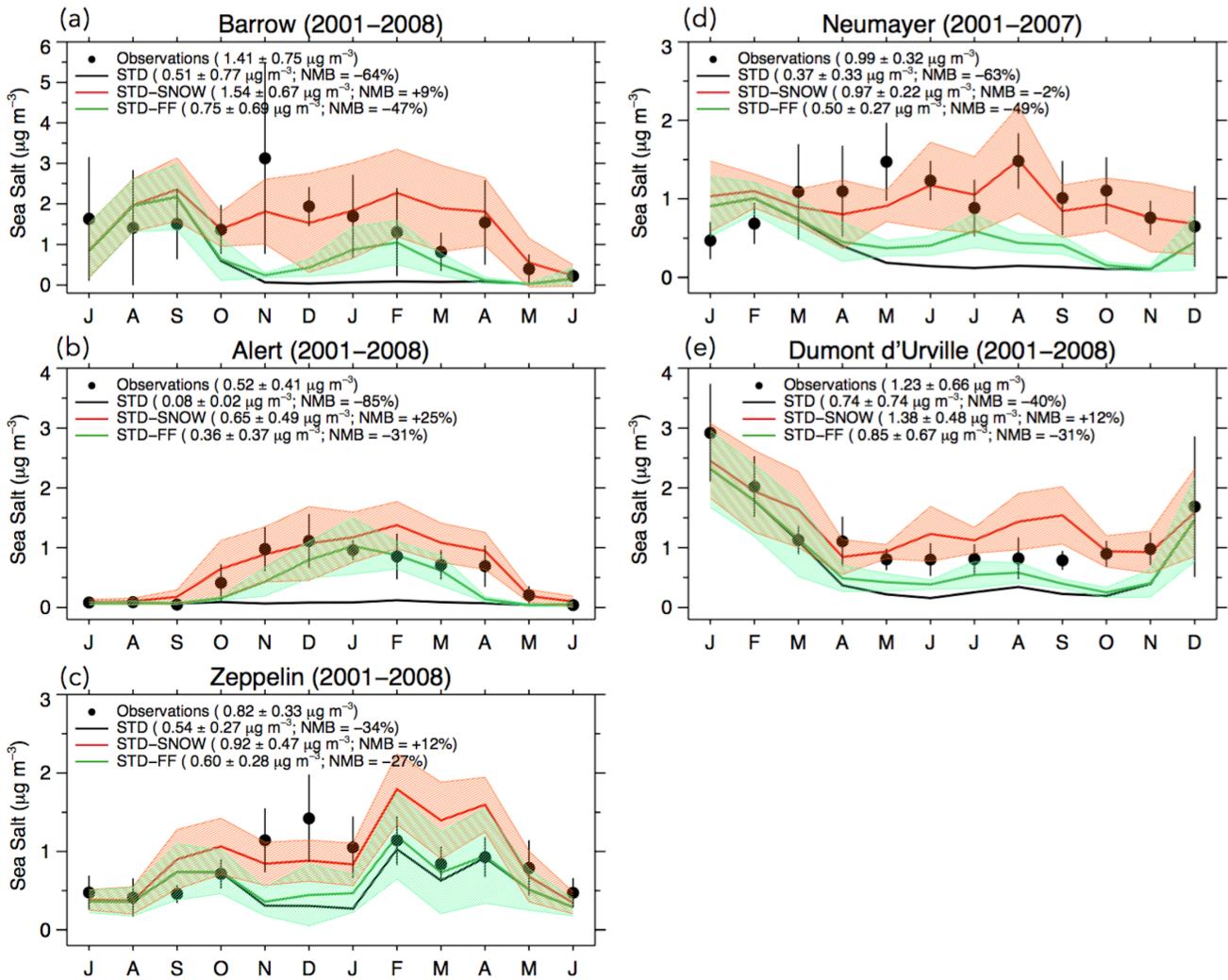


Figure 1: Monthly mean mass concentrations of SSA at Arctic (a. Barrow, b. Alert, c. Zeppelin) and Antarctic sites (d. Dumont d'Urville, e. Neumayer). All observations and model results are for 2001-2008 except at Neumayer (2001-2007). Note that the seasonal cycles are centered over local winter. The observed mean concentrations are indicated with filled black circles, while the lines are for the GEOS-Chem simulations (STD: black line, STD-SNOW: red line, STD-FF: green line). The black vertical lines and shaded areas correspond to the standard deviations of monthly means for observations and model simulations. For each individual panel, the legend lists mean concentrations and standard deviations, as well as the normalized mean bias (NMB, $\text{NMB} = \frac{\text{Model} - \text{Obs}}{\text{Obs}} \times 100$).

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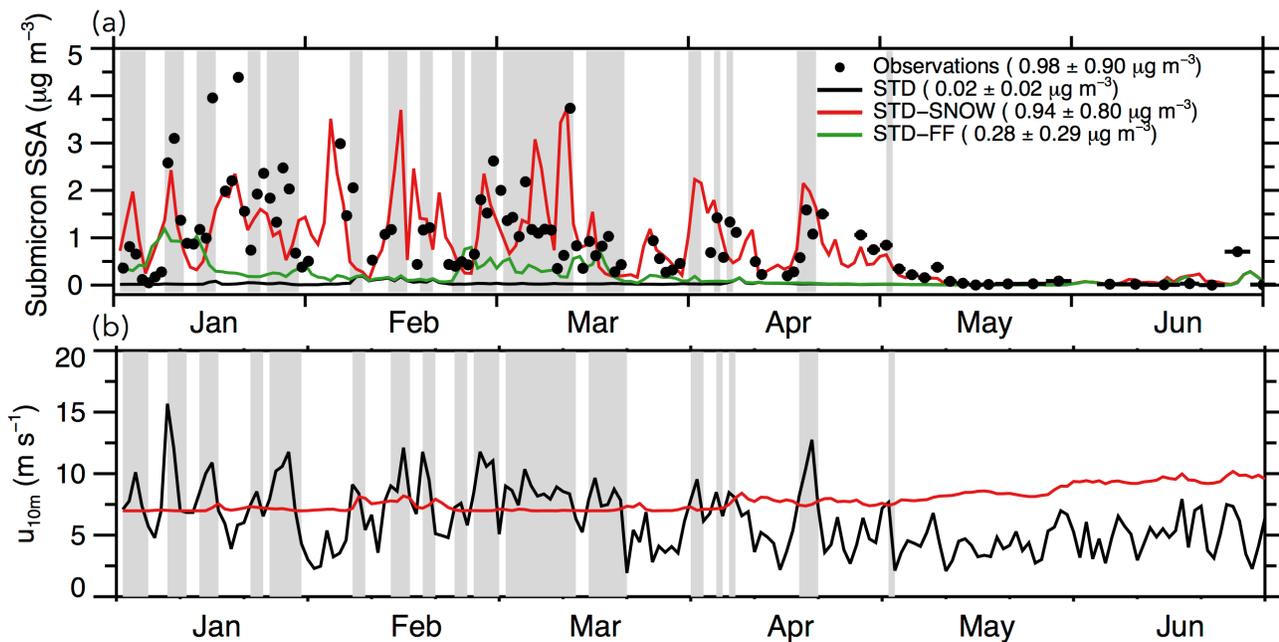
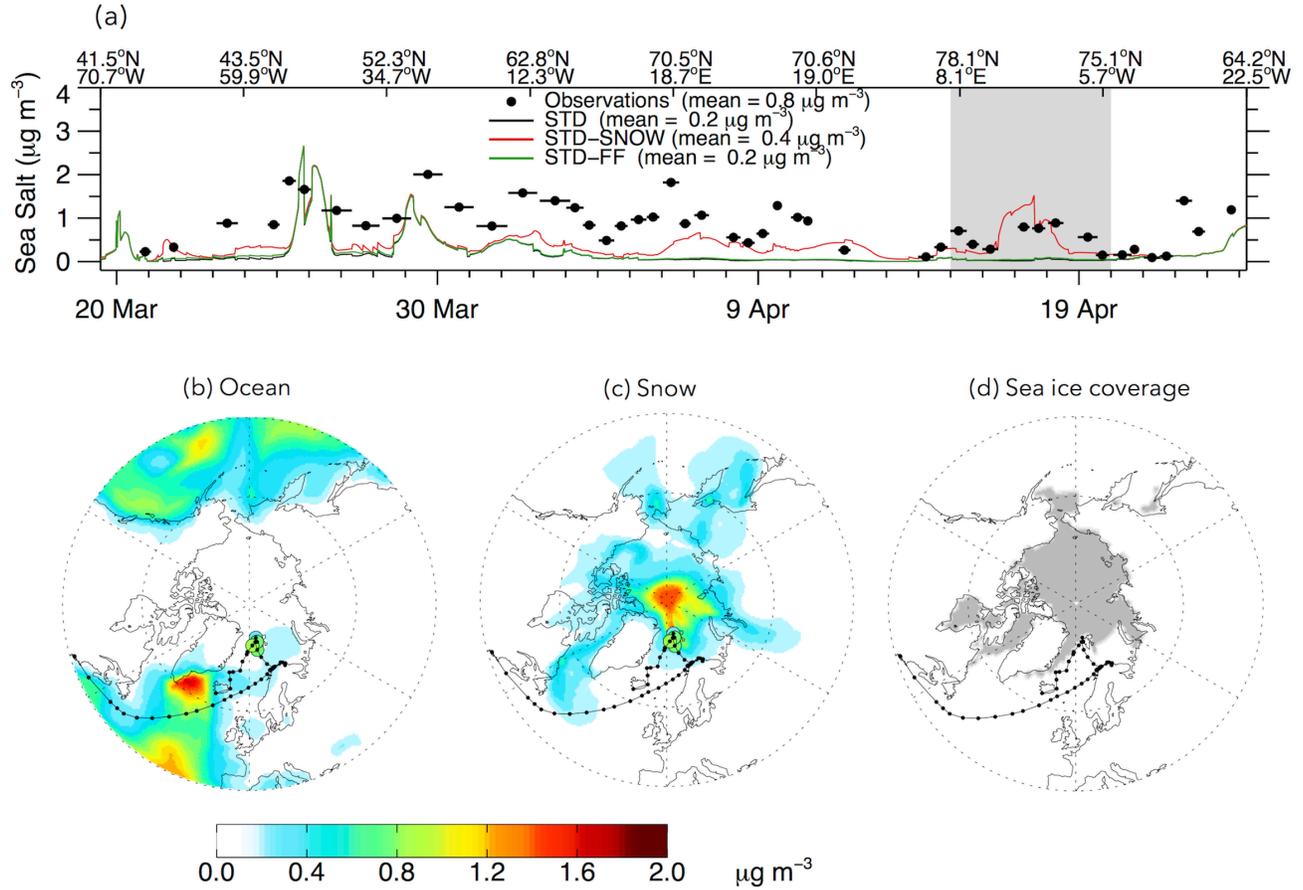


Figure 2: (a) Daily variations in submicron SSA mass concentrations at Barrow for 1 January to 30 June 2001. Observations are shown with filled black circles, while the GEOS-Chem simulations are indicated with lines (STD: black, STD-SNOW: red, and STD-FF: green). (b) MERRA 10m wind speed (u_{10m}) at Barrow. The red line indicates the wind speed threshold for blowing snow events calculated with the local MERRA 2m temperatures. Shaded gray areas indicate time periods when u_{10m} exceeds the blowing snow wind threshold.

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Figure 3: (a) Timeseries of submicron SSA mass concentrations during the ICEALOT cruise between 19 March and 24 April 2008. Observations are shown as filled black circles with horizontal bars indicating the sampling period. The GEOS-Chem simulations are indicated with lines (STD: black, STD-SNOW: red, and STD-FF: green). The 15-19 April period discussed in the text is indicated by the gray shading. The bottom panels show the spatial distribution of mean surface SSA mass concentrations for the 15-19 April period. SSA mass concentrations due to open ocean emissions are shown in panel (b) while those due to blowing snow are shown in panel (c). The ship track is indicated with the black line and dots in panels b-d. The colored larger circles of near Svalbard correspond to the location of the ship on 15-19 April, and they are color-coded based on observed SSA mass concentrations. (same color scale as the model). Panel (d) displays the MERRA sea ice extent.

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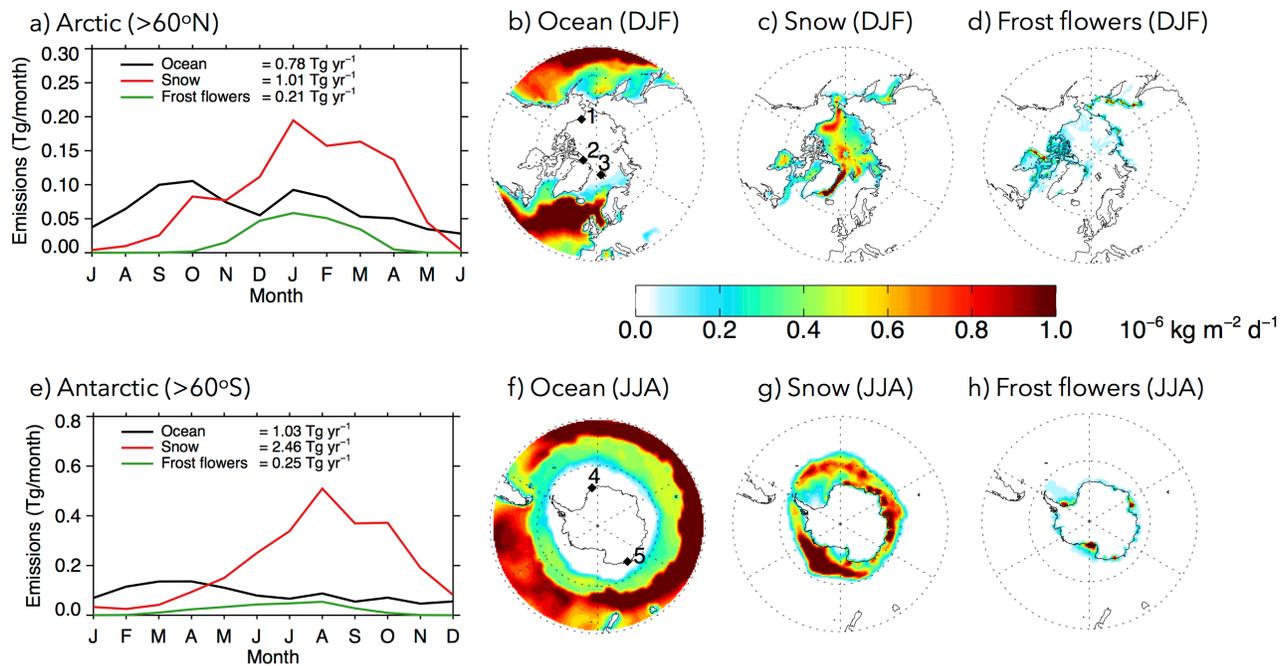


Figure 4: Seasonality of submicron SSA emissions in 2005 from open ocean, blowing snow, and frost flowers over (a) the Arctic and (e) the Antarctic for latitudes poleward of 60°. Also shown are spatial distributions of wintertime submicron SSA emissions over the Arctic (b-d) and the Antarctic (f-h). Filled diamonds in panels (b) and (f) correspond to the locations of Barrow (1), Alert (2), Zeppelin (3), Neumayer (4) and Dumont d'Urville (5).

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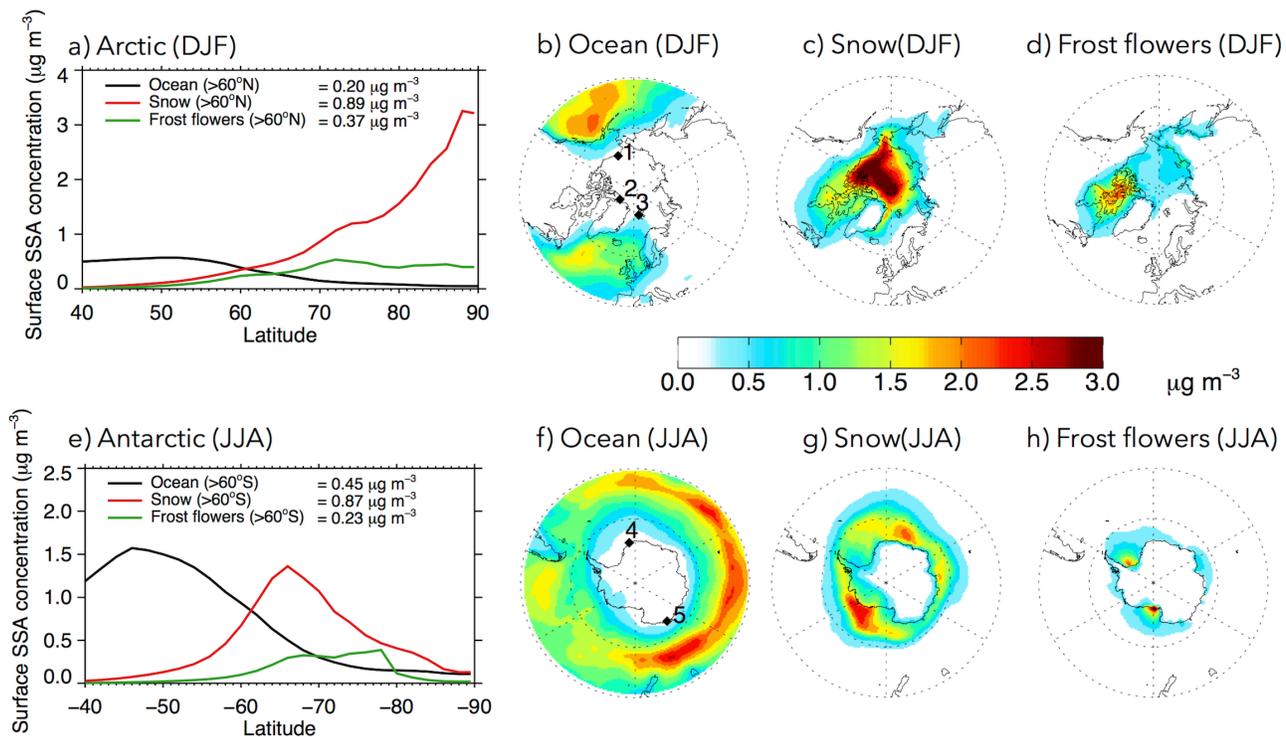


Figure 5: Surface mass concentrations of wintertime submicron SSA. Zonal mean concentrations are shown over the (a) Arctic and (e) Antarctic for the open ocean (black line), blowing snow (red line), and frost flowers (green line). The panels on the right side show the spatial distributions of wintertime surface **submicron** SSA mass concentrations over the Arctic (b-d) and the Antarctic (f-h) for each source type. Filled diamonds in panels (b) and (f) correspond to the locations of the ground stations (see Fig. 3).

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Supplement for “Wintertime enhancements of sea salt aerosol in polar regions consistent with a sea-ice source from blowing snow”

In this study, we implement a blowing snow and a frost flower emission parameterization in the GEOS-Chem global chemical transport model based on the work of Yang et al. (2008, 2010) and Xu et al. (2013). We use multi-year (2001–2008) in situ observations of SSA mass concentrations at five polar ground sites and during a research cruise over the N. Atlantic and Arctic Oceans to evaluate the relative roles of sea ice and open ocean sources of SSA in polar regions.

1 Open ocean emissions over cold polar waters during summer

Figure S1 compares observed SSA mass concentrations at Barrow, Zeppelin, Neumayer, and Dumont d’Urville to the GEOS-Chem simulation (ORIG simulation) with open ocean SSA emissions only. During warm months when local sea ice extent is at a minimum and open ocean emissions dominate SSA mass concentrations, the model overestimates observations by a factor of 2. We hypothesize that this overestimate indicates a stronger suppression of SSA emissions over cold polar waters than accounted for in the SST dependence of Jaeglé et al. (2011) used in GEOS-Chem: $f(\text{SST})=0.3+0.1\times\text{SST}-0.0076\times\text{SST}^2+0.00021\times\text{SST}^3$. Most which most of the observations used to derive this empirical expression were for $\text{SST}>5^\circ\text{C}$ (see Fig. 6 in Jaeglé et al., 2011). The SST at the 4 polar sites in Fig. S1 range from -2°C to $+5^\circ\text{C}$, which lead to $f(\text{SST})=0.3-0.6$. A simulation in which we assume $f(\text{SST})=0.25$ for $\text{SST}<5^\circ\text{C}$ results in improved agreement with observed summertime SSA mass concentrations— and does not affect SSA mass concentrations during winter. This is the standard simulation (STD) used in the main text.

2 Influence of assumed number of particles produced per snowflake on the size distributions of SSA from blowing snow

Figure S2 shows the seasonal variations of observed submicron and supermicron SSA mass concentrations at Barrow, Alaska. The observed submicron SSA concentrations at Barrow maximize in winter, while the supermicron SSA concentrations have their maximum during summer (Quinn et al. 2002). We examine how the size distribution of SSA from blowing snow is affected by assumptions on the number of particles produced per snowflake (N). For $N=1$, the blowing snow simulation predicts submicron SSA concentrations $<0.3\ \mu\text{g m}^{-3}$, a factor of 2–3 lower than observed concentrations of $0.5\text{--}1.3\ \mu\text{g m}^{-3}$ for November–March. Increasing N to a value of 5 shifts more of the blowing snow SSA mass to submicron aerosol and leads to better agreement with observed submicron SSA concentration at Barrow, without much change in the supermicron SSA. The blowing snow simulation with $N=5$ corresponds to the STD-SNOW simulation in the main text.

33 Sensitivity simulations on wind thresholds for frost flower formation

We perform a sensitivity simulation for frost flower formation, in which we impose a wind threshold of 5 m/s beyond which frost flower formation and emission are inhibited in the model: STD-FF (<5 m/s). This simulation assumes that strong winds inhibit frost flower formation and bury existing frost flowers with snow (Perovich and Richeter-Menge, 1994; Rankin et al., 2000). The frost flower emission in this sensitivity simulation has a scaling factor 5 times larger than the one used in STD-FF ($=1 \times 10^6 \text{ m}^2 \text{ s}^{-1}$, Xu et al. 2013), in order to reproduce the SSA mass concentrations observed at Alert. The resulting simulation is shown in Fig. S3. With this wind threshold, the new frost flower simulation underestimates the SSA mass concentration by factors of 2-3 at Barrow in December-January, and overestimates the SSA mass concentrations by a factor of 2 at Alert in December-March. At the other sites, the influence of frost flower remains very small, and thus cannot fully explain the wintertime SSA enhancements. Therefore, imposing a wind threshold for frost flower emission does not lead to a better model agreement and does not alter our conclusions on frost flower simulations.

4 Daily variability of SSA mass concentration observed at Alert and Dumont d'Urville

Figure S3S4 compares observed and modeled variations in SSA mass concentrations at Alert and Dumont d'Urville for the year 2001. Observations are weekly at Alert and daily at Dumont d'Urville. At Alert (Fig. S3aS4a) observed concentrations of SSA vary between 0.3 and 2 $\mu\text{g m}^{-3}$ between November and early June, while during the rest of the year observed concentrations remain below 0.1 $\mu\text{g m}^{-3}$. The blowing snow simulation (STD-SNOW) captures the timing of some of the large episodic enhancements in SSA: in mid-January, early and late April, June, and November. This variability is not well reproduced by the frost flower simulation. At Dumont d'Urville (Fig. S3bS4b), the influence of frost flowers is minor, but the blowing snow simulation reproduces to some extent the variability and magnitude of observed SSA mass concentrations. Fig. S3c shows that the $\text{SO}_4^{2-}/\text{Na}^+$ ratio at Dumont d'Urville has winter minima (<0.25), and is particularly low when elevated levels of SSA mass concentrations were observed during winter. This is consistent with the low/negative non-sea-salt SO_4^{2-} levels found in aerosol samples over polar regions in several previous studies (Jourdain et al., 2001; Rankin et al., 2000; Wagenbach et al., 1998), which is related to the sulphate depletion in snow and frost flower on sea ice (Krnavek et al., 2012; Rankin et al., 2000; Seguin et al., 2014).

5 Sensitivity simulation including both blowing snow and frost flower emissions

Figure S5 compares observed SSA mass concentrations at Barrow, Alert, Zeppelin, Neumayer, and Dumont d'Urville to a simulation including open ocean, blowing snow, and frost flowers (STD-SNOW-FF). Overall, the combined influence of blowing snow and frost flowers in the model does not yield a better model agreement with the observations (with NMB, normalized mean bias, ranging from 12% to 81%) compared to blowing snow simulation (STD-SNOW in the main text, with NMB ranging from -2% to +25%). In particular, combining the influence of blowing snow and frost flowers in the model leads

to factor of 2-3 overestimates of the SSA mass concentrations at Barrow in Jan-Mar and at Alert in Dec-Mar. At the other three polar sites, the influence of frost flowers is very small and thus the STD-SNOW-FF simulation is similar to the blowing snow simulation (STD-SNOW).

5 **6 Global SSA budgets for the open ocean, blowing snow and frost flower simulation**

Table S1 summarizes the global annual budget for SSA for the three sources considered in this study for the year 2005.

10 Table S1. Global budgets of SSA generated by the open ocean, blowing snow, and frost flowers for the year 2005.

	Open Ocean (STD)			Blowing Snow			Frost Flowers
	0.01-0.5 μm	0.5-4 μm	Total	0.01-0.5 μm	0.5-4 μm	Total	0.01-0.5 μm
Emission (Tg/yr)	52	1957	2009	3.8	3.6	7.4	0.4149
Dry deposition (Tg/yr)	4.0	804	808	0.93	1.6	2.5	0.2321
Wet deposition (Tg/yr)	49	1262	1311	3.0	2.2	5.2	0.2232
Lifetime (days)	2.1	0.44	0.48	4.2	0.6	2.4	5.61
Burden (Gg)	297	2477	2774	45	6.0	51	6.874
Surface concentration ($\mu\text{g m}^{-3}$)	0.6	5.7	6.3	0.078	0.020	0.098	0.02

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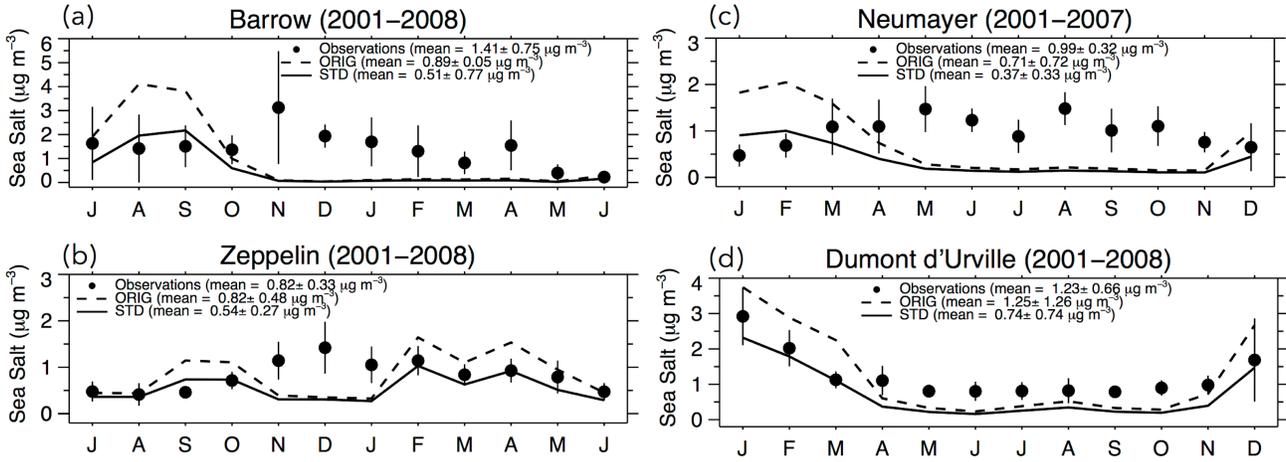
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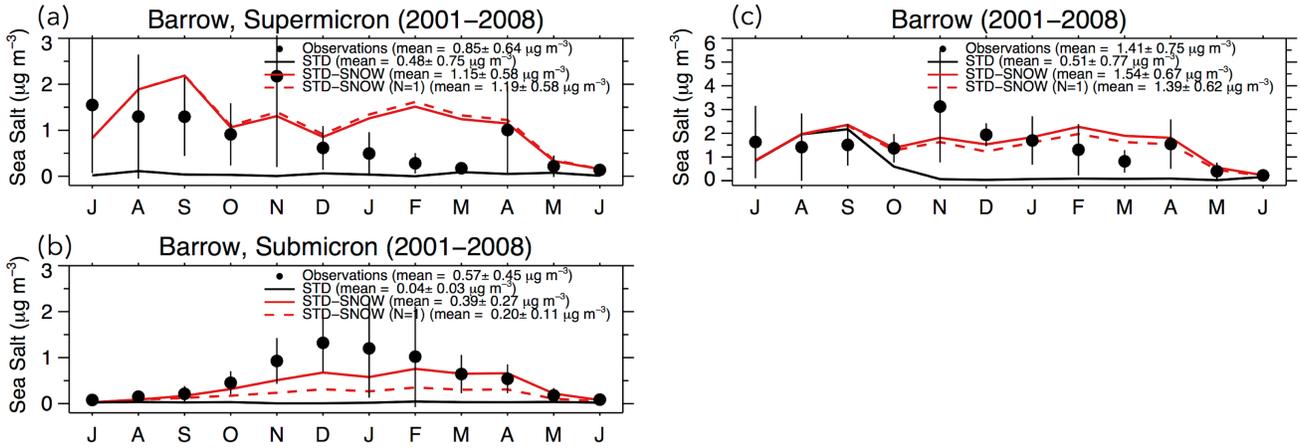
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5 **Figure S1: Monthly mean surface SSA mass concentrations at (a) Barrow, (b) Zeppelin, (c) Neumayer and (d) Dumont d'Urville. The observed mean concentrations are indicated with filled black circles, while the lines are for the GEOS-Chem model. The original GEOS-Chem simulation (ORIG) is shown with a black dashed line, while the standard simulation (STD) with enhanced SSA suppression for SST < 5°C is shown by the black line.**



5 **Figure S2: Monthly mean super-micron (a), sub-micron (b) and total (c) surface mass concentrations of SSA at Barrow (2001-2008).** Note that the seasonal cycles are centered over local winter. The observed mean concentrations are indicated with black circles, while lines are for the concentrations in GEOS-Chem (STD: black line; STD-SNOW: red line; STD-SNOW (N=1): red dashed line). The STD-SNOW simulations assumes that the number of SSA particles per snowflake is N=5, while the STD-SNOW (N=1) assumes N=1. The black vertical lines correspond to the standard deviations of monthly means for observations.

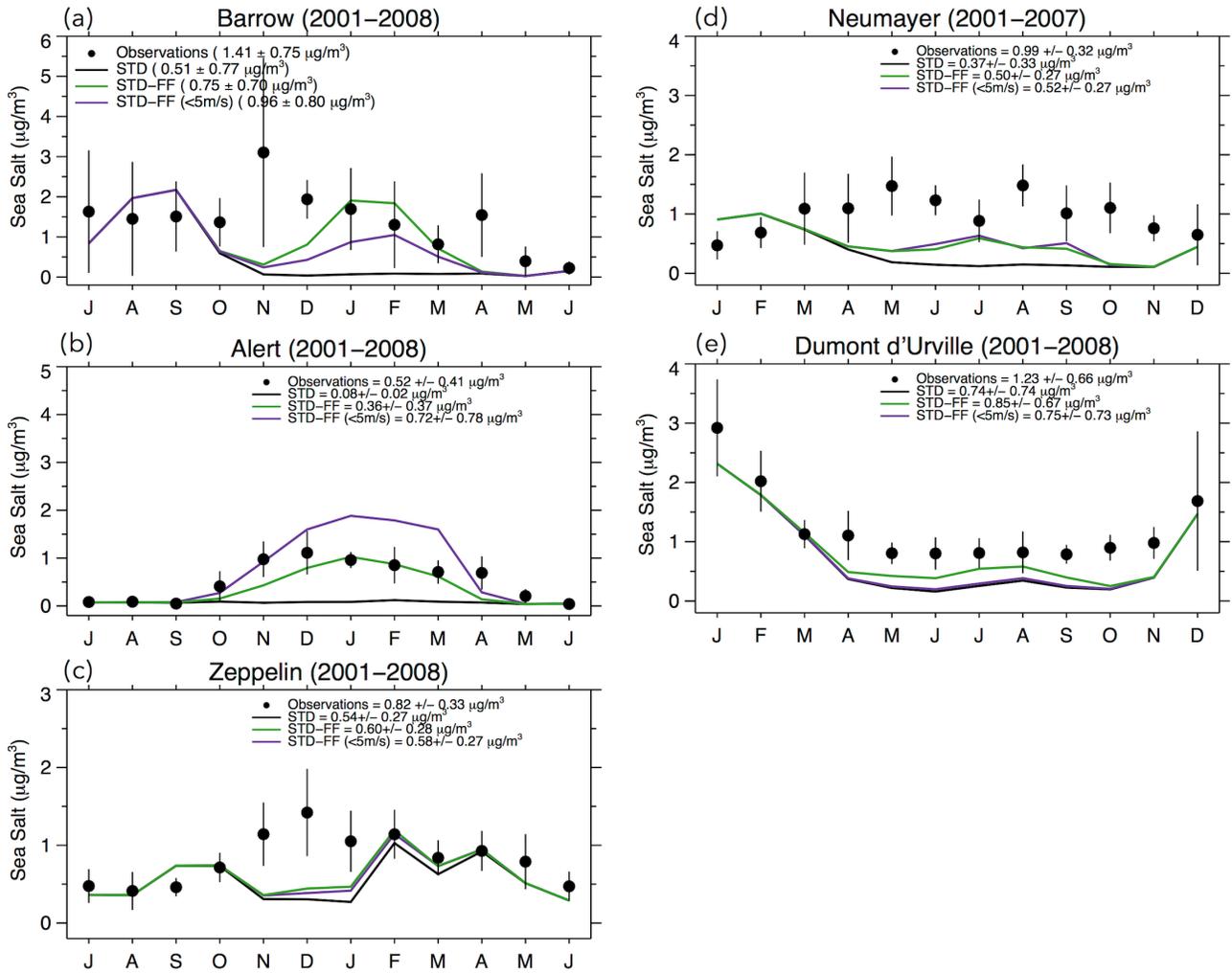


Figure S3: Monthly mean surface SSA mass concentrations at (a) Barrow, (b) Alert, (c) Zeppelin, (d) Neumayer and (e) Dumont d'Urville. The observed mean concentrations are indicated with filled black circles, while the black line is for the GEOS-Chem model with open ocean emissions (STD) and the green line is for the frost flower simulation. We also show a sensitivity simulation in which we impose a 5 m/s wind threshold beyond which frost flower emissions are suppressed: STD-FF (<5m/s) (purple line). The black vertical lines and for each individual panel, the legend lists mean concentrations and standard deviations.

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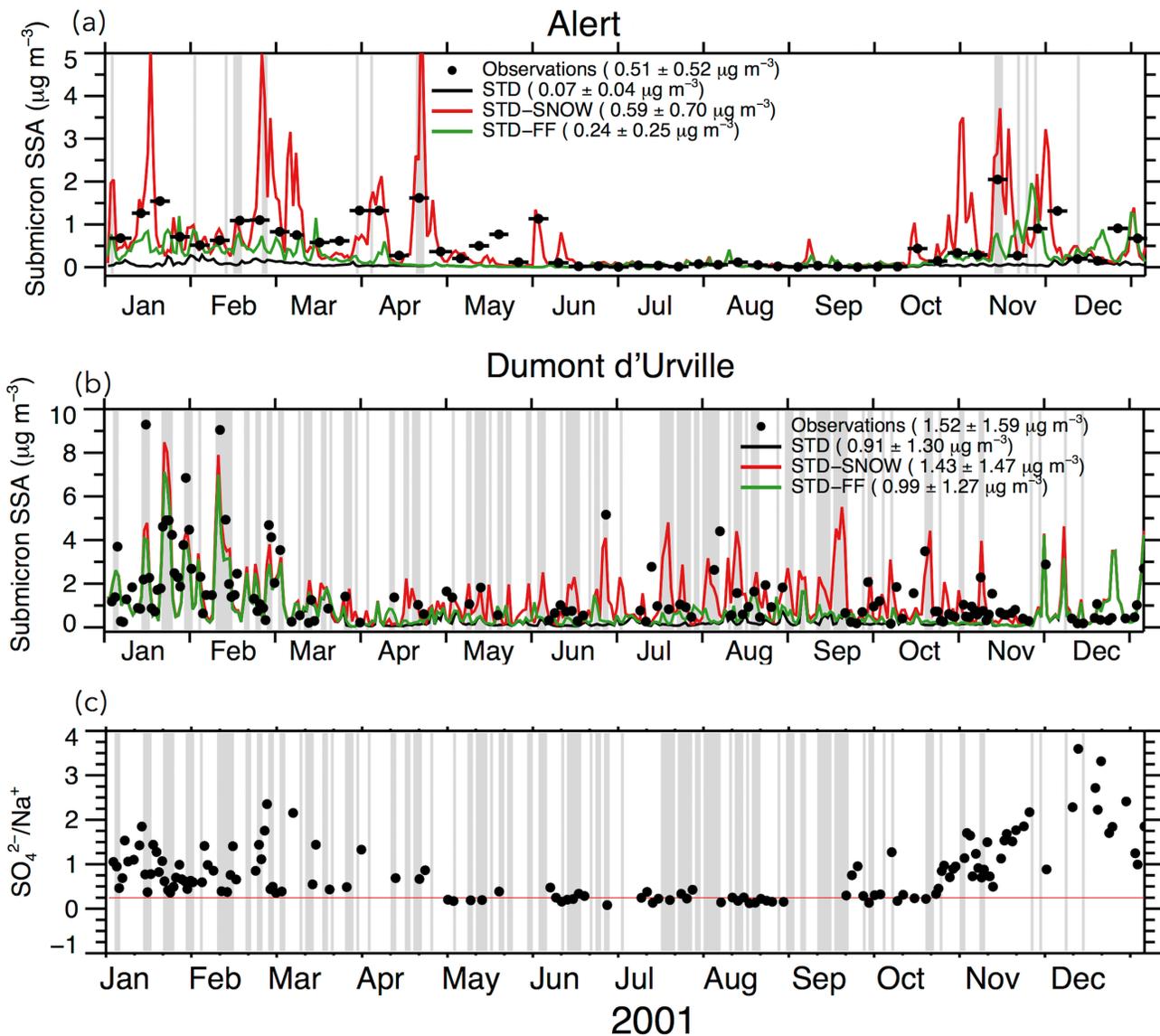


Figure S4: Weekly/daily variations in SSA mass concentrations at Alert (a) and Dumont d'Urville (b) for 2001. Observations are shown with black circles, while the GEOS-Chem simulations are indicated with lines (STD: black, STD-SNOW: red, and STD-FF: green). Also shown in the daily variations of sulphate to sodium ratio as observed at Dumont d'Urville (c) for 2001. The red line indicates the $\text{SO}_4^{2-}/\text{Na}^+$ of 0.25 in seawater. Shaded grey areas in (a-c) indicate time periods when local u_{10m} exceeds the blowing snow wind threshold.

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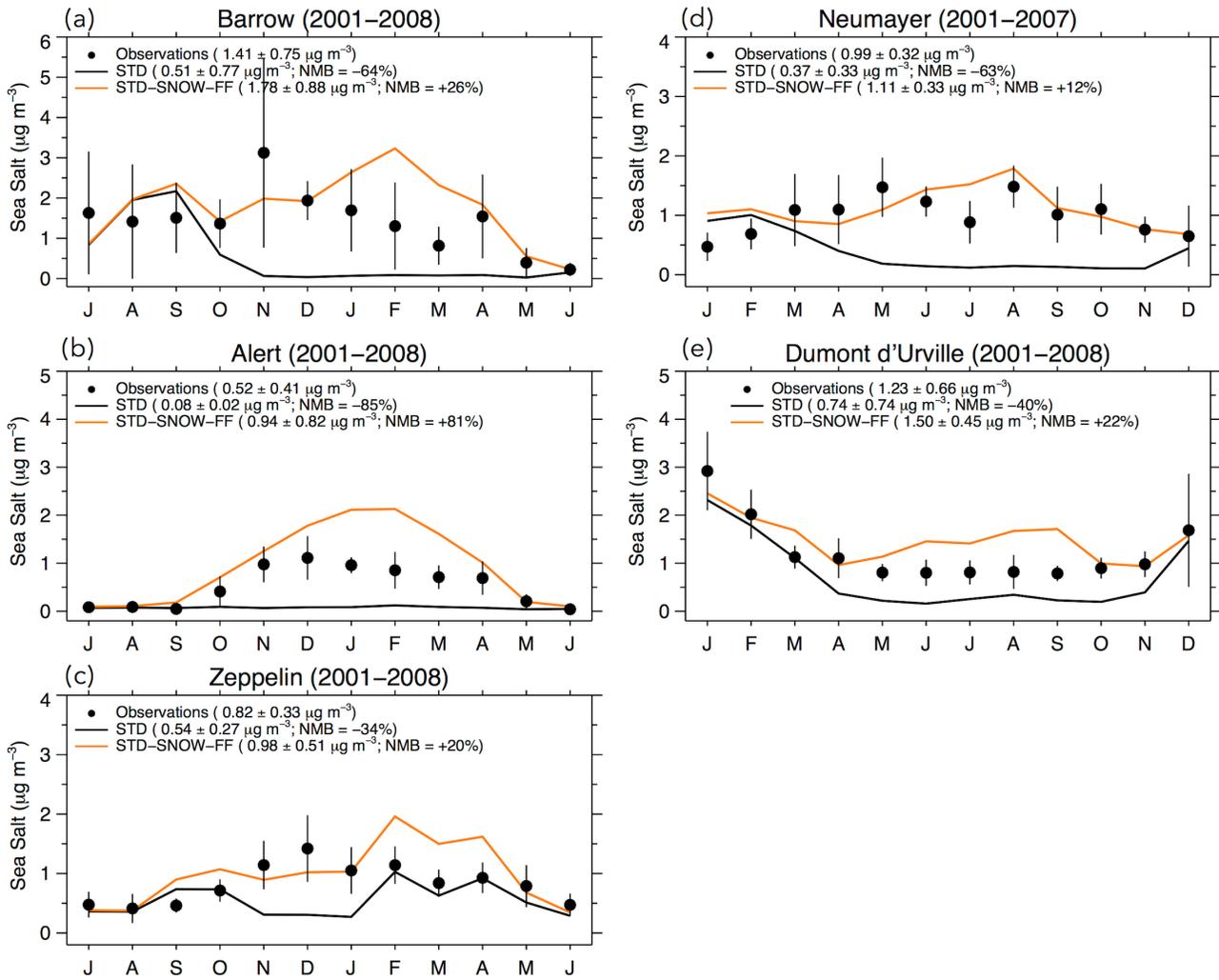


Figure S5: Monthly mean surface SSA mass concentrations at (a) Barrow, (b) Alert, (c) Zeppelin, (d) Neumayer and (e) Dumont d'Urville. The observed mean concentrations are indicated with filled black circles, while the lines are for the GEOS-Chem model. The black line is for open ocean simulation (STD), while the orange line is when all tree SSA sources are included (open ocean, blowing snow, and frost flowers). The black vertical lines and for each individual panel, the legend lists mean concentrations and standard deviations.

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