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: $f(SST)=0.3 + 0.1\times SST - 0.0076\times SST^2 + 0.00021\times SST^3$. Most of the observations used to derive this empirical expression were for SST>5°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°C, which lead to f(SST)=0.3-0.6. A simulation in which we assume f(SST)=0.25 for SST<5°C results in improved agreement with observed summertime SSA mass concentrations. 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 $\mu g\ m^{-3}$, a factor of 2-3 lower than observed concentrations of 0.5-1.3 $\mu g\ m^{-3}$ 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.

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

Figure S3 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. S3a) observed concentrations of SSA vary between 0.3 and 2 μg m^{-3} between November and early June, while during the rest of the year observed concentrations remain below 0.1 μg 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. S3b), 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 SO_4^{2-}/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).

4 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.

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	0.41
Dry deposition (Tg/yr)	4.0	804	808	0.93	1.6	2.5	0.23
Wet deposition (Tg/yr)	49	1262	1311	3.0	2.2	5.2	0.22
Lifetime (days)	2.1	0.44	0.48	4.2	0.6	2.4	5.6
Burden (Gg)	297	2477	2774	45	6.0	51	6.8
Surface concentration (μg m^{-3})	0.6	5.7	6.3	0.078	0.020	0.098	0.02

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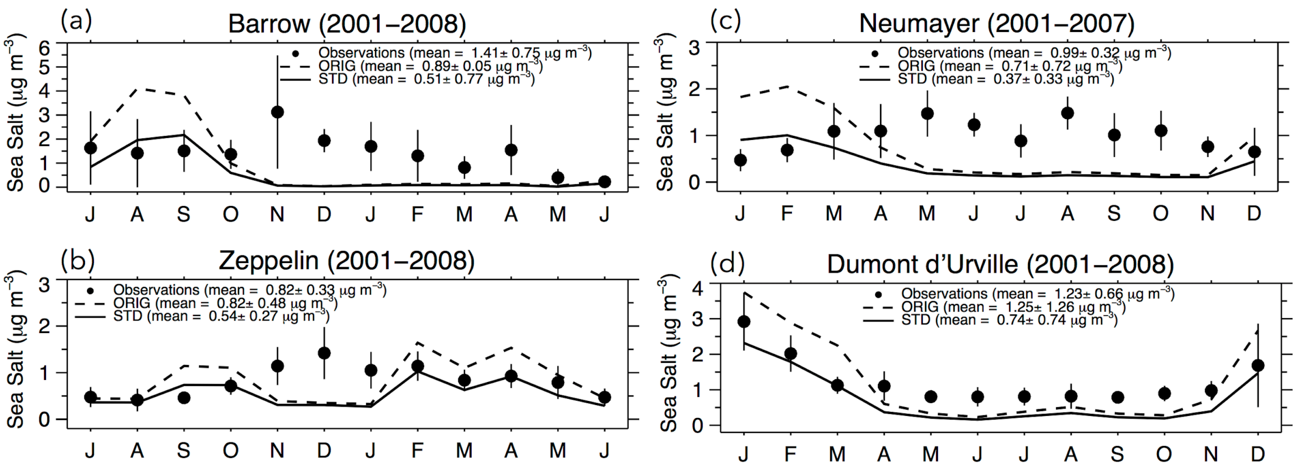

Figure S1: Monthly mean surface SSA mass concentrations at (a) Barrow, (b) Zeppelin, (c) Neumayer and (d) Dumont d'Urville. The observed mean concentrations are indicated with filled black circles, while the lines are for the GEOS-Chem model. The original GEOS-Chem simulation (ORIG) is shown with a black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

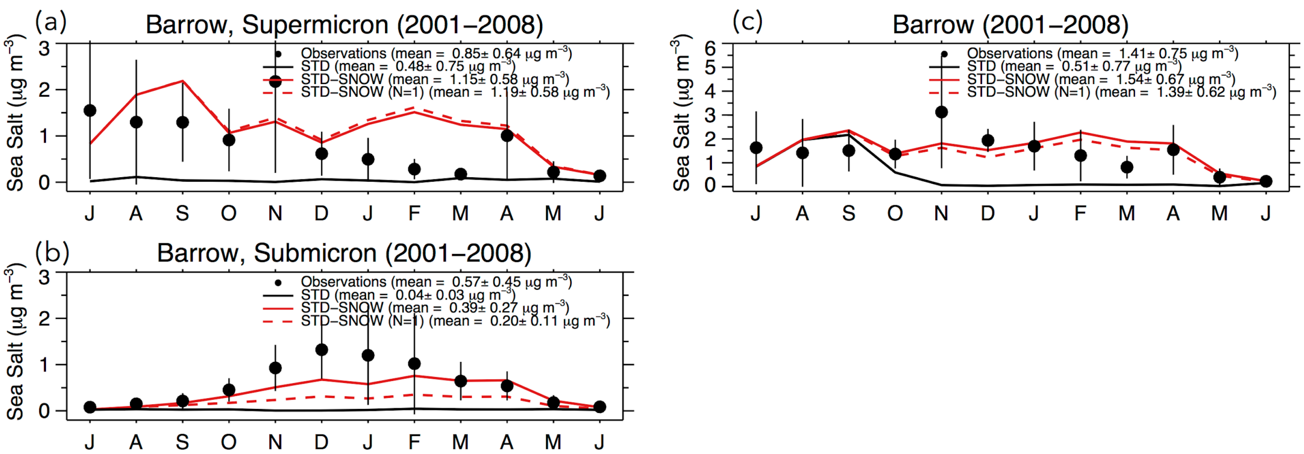

Figure S2: Monthly mean super-micron (a), sub-micron (b) and total (c) surface mass concentrations of SSA at Barrow (2001-2008). Note that the seasonal cycles are centered over local winter. The observed mean concentrations are indicated with black circles, while lines are for the concentrations in GEOS-Chem (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.

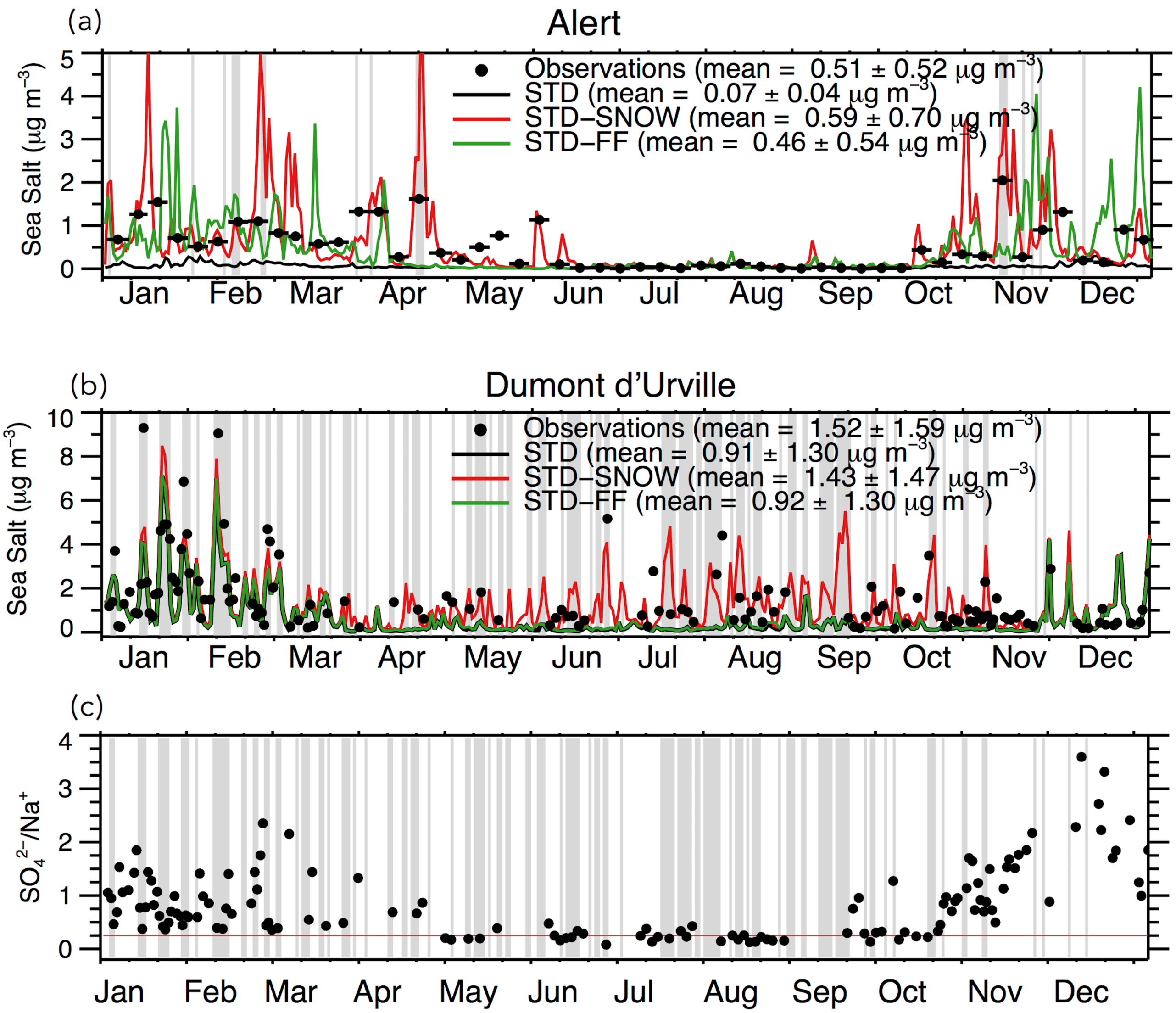

Figure S3: Weekly/daily variations in SSA mass concentrations at Alert (a) and Dumont d'Urville (b) for 2001. Observations are shown with black circles, while the GEOS-Chem simulations are indicated with lines (STD: black, STD-SNOW: red, and STD-FF: green). Also shown in the daily variations of sulphate to sodium ratio as observed at Dumont d'Urville (c) for 2001. The red line indicates the SO_4^{2-}/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.