Supplement to:

Sea ice as a source of sea salt aerosol to Greenland ice cores: a model-based study

R. H. Rhodes et al.

1. Summit aerosol data processing

Summit aerosol data from 2003-2006 AD are used because [Na] is not reported for other years. These data have low temporal resolution, particularly during winter when difficult weather conditions meant sampling was not always possible. Aerosol was collected on 8 impactor stages. We screened the data for each stage by removing data points > 2*standard deviation above the mean. Data for the 8 stages were then combined, resampled to monthly, and monthly values were stacked to produce representative monthly means. No uncertainty bars are plotted on Fig. 3 because some monthly mean values reflect data from only one year. Summit aerosol [Na] displayed on Fig. 3 are within the range of past measurements reported by Mosher at al. (1993) (mean 0.0043 μg m^{-3}).

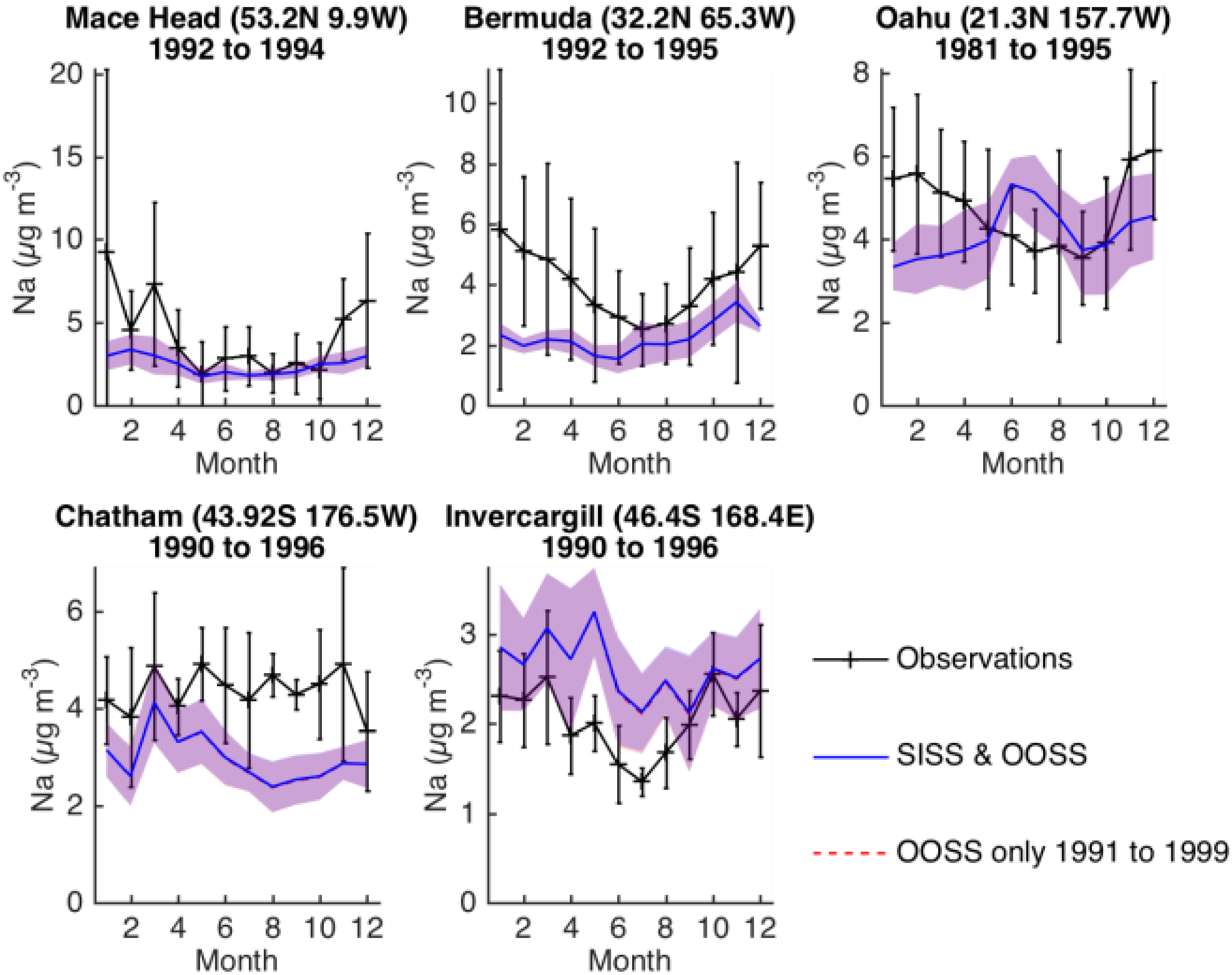

Figure S1. Sea salt Na aerosol concentrations simulated by p-TOMCAT base simulation for 1991–1999 AD compared to observations at coastal low- and mid- latitude locations. Observations and model results are mean monthly values ± 1 σ. Aerosol observations are from the AEROCE-SEAREX network (Savoie et al., 2002).

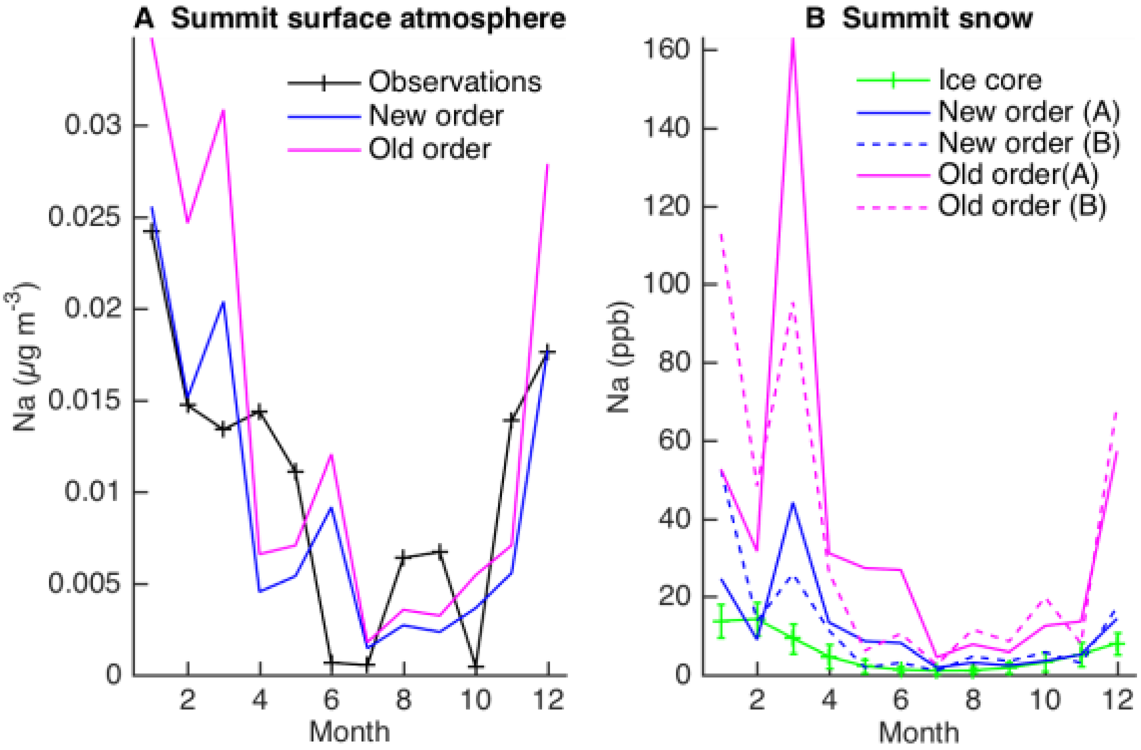

Figure S2. Effect of changing the order of events in p-TOMCAT from deposition-emissions-mixing (old order) to emissions-deposition-mixing (new order) on monthly [Na] concentrations of the surface layer of the atmosphere (A) and snow (B) at Summit, Greenland for 1997 AD. On panel B two different options for simulated sea salt concentrations are displayed: A) [Na] calculated using p-TOMCAT precipitation output in Eq. (5), B) [Na] calculated using ice core constant annual accumulation rate in Eq. (5).

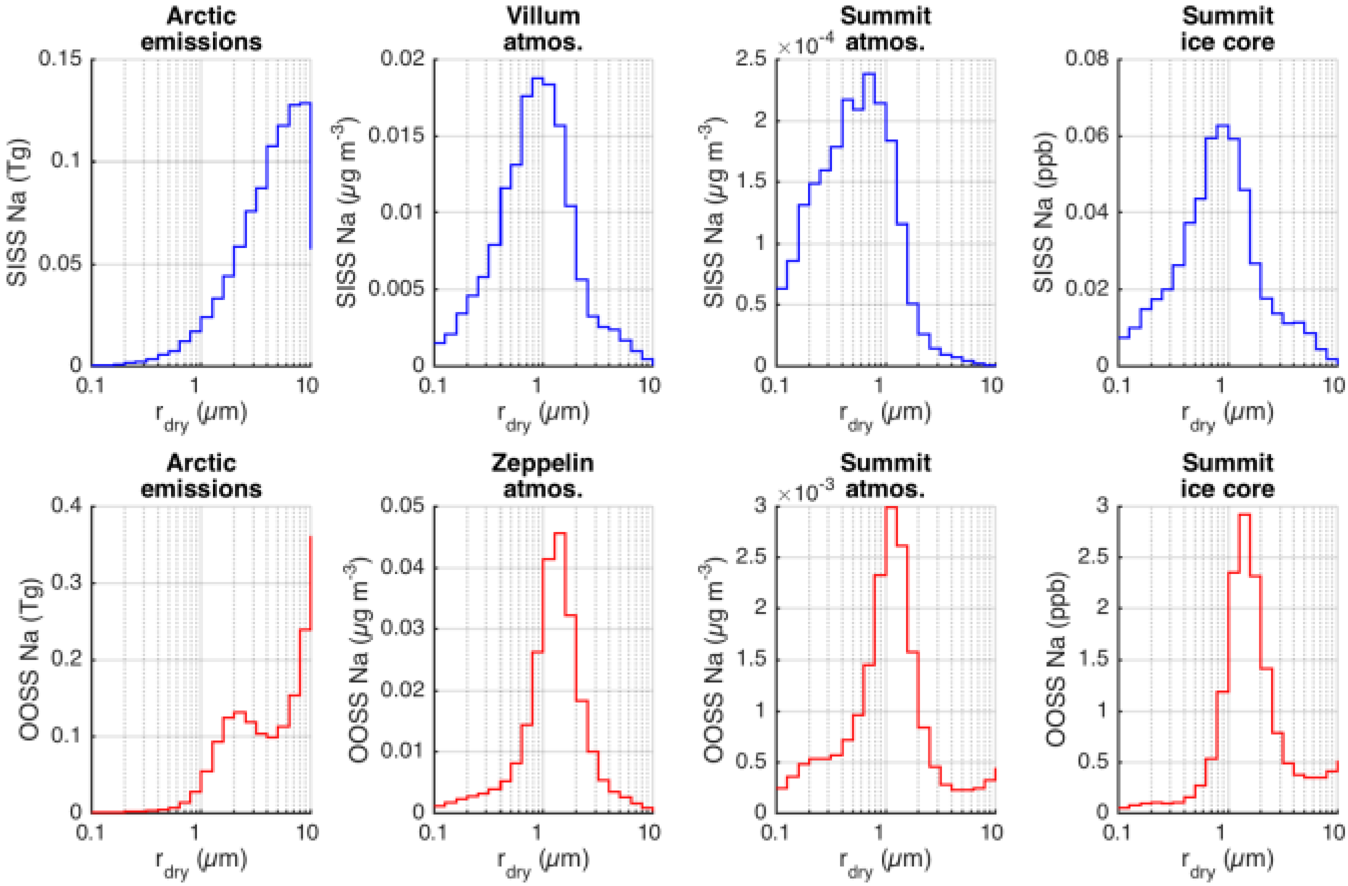

Figure S3. Particle size spectra of SISS (blue) and OOSS (red) when emitted compared to size spectra of sea salt particles in the surface atmosphere at coastal (close to sea ice or open ocean: Villum and Zeppelin) and inland (Summit) locations, and size spectra of sea salt particles deposited on Greenland ice sheet. All spectra represent mean values of DJF 1997 AD.

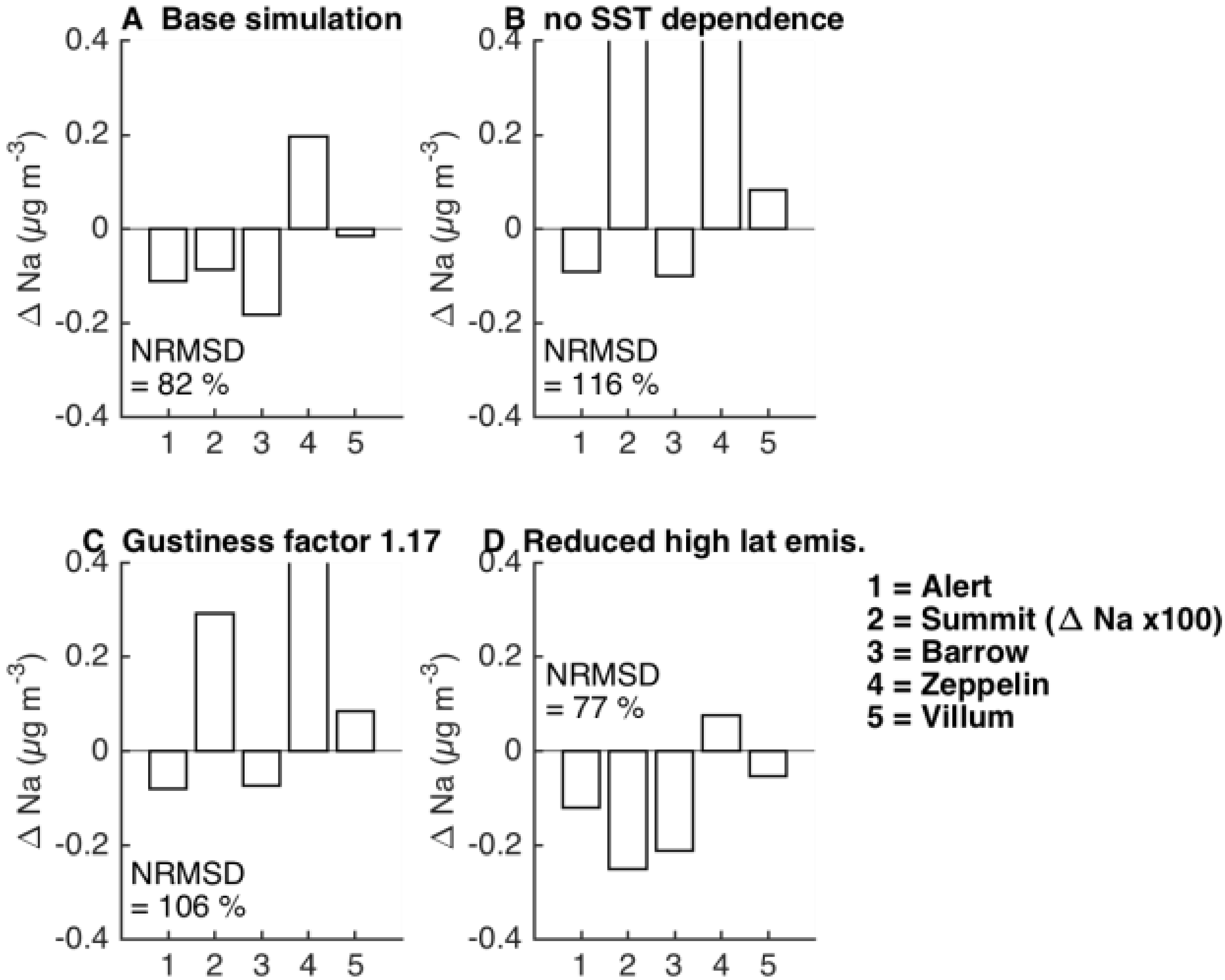

Figure S4. Sensitivity of p-TOMCAT Na aerosol simulations for 1997 AD at 5 Arctic locations to parameters associated with OOSS emissions. A) Base simulation uses Gong et al. (2003) parameterisation of OOSS emission via bubble bursting with modification to account for dependence of OOSS emissions on SST (Jaeglé et al., 2011); B) Uses OOSS emissions scheme with no SST dependence, as Legrand et al. (2016); C) As base simulation but with surface winds increased by a gustiness factor, as Levine et al. (2014); D) As base simulation with additional modifications to reduce OOSS emissions at SSTs <5°C and prevent any OOSS emissions in grid squares with <50% water coverage in the polar regions (Huang and Jaeglé, 2017). Each panel (A-D) displays the mean difference between monthly model results and observations (ΔNa) for each site. Positive [negative] values indicate that p-TOMCAT over- [under-] estimates aerosol Na concentration. The normalised root mean square difference (NRMSD) between model simulations and aerosol data is calculated for each of the 5 sites and the mean NRMSD across all 5 sites is displayed on each panel.

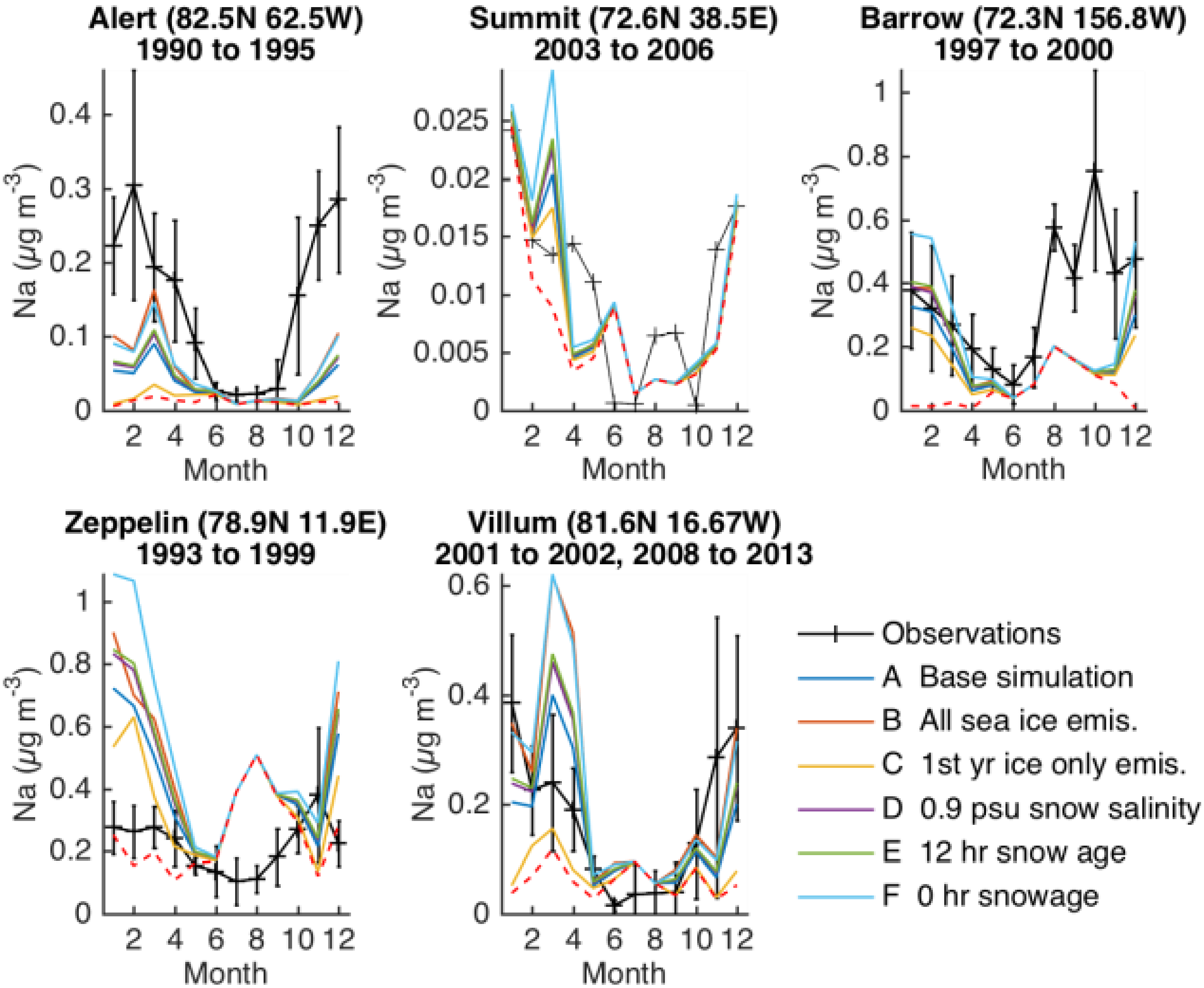

Figure S5. Results of sensitivity tests to investigate the impact of changing parameters associated with SISS emissions in p-TOMCAT for 1997 AD. In the base simulation, SISS emissions are reduced by 50% over multi-year sea ice relative to 1st year sea ice, mean snow salinity is 0.6 psu, and snow age is 24 hr. OOSS contribution remains constant for each test (dashed red line).

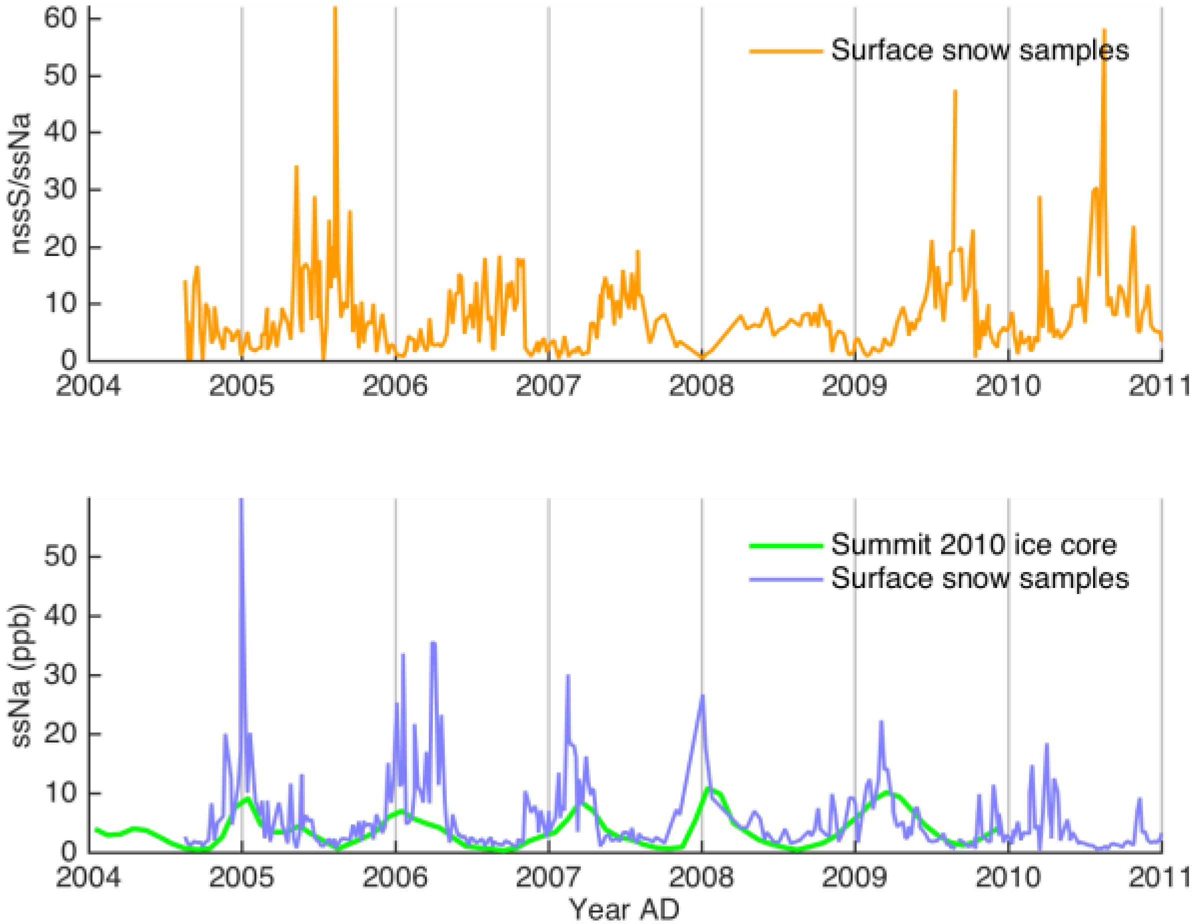

Figure S6. Comparison between ice core and surface snow sea salt measurements at Summit, Greenland. Sea salt Na (ssNa) and non-sea salt sulfur (nssS) concentrations measured in surface snow collected weekly (5 replicates in each weekly sample). Some samples are missing from 2007/2008 winter season. Summit2010 ice core ssNa data are shown for the same time interval (green). Summit2010 ice core (and other Greenland ice cores) is dated by assuming minimum nssS/ssNa values occur in mid-winter (Jan. 1st) and the surface snow chemistry data support this assumption (orange). This method typically results in peak [ssNa] in ice cores being dated as mid-winter, which is in agreement with surface snow data.

Figure S7. Monthly mean sea salt Na concentrations simulated by p-TOMCAT for 1991–1999 AD compared to Greenland ice core data. Data are identical to Fig. 5, but p-TOMCAT simulations have not been smoothed prior to stacking. Two different options for simulated sea salt concentrations are displayed: A) [Na] calculated using p-TOMCAT precipitation output in Eq. (5), B) [Na] calculated using ice core constant annual accumulation rate (Table 1) in Eq. (5). In each case, the mean monthly ice core Na concentrations (green) are shown with uncertainty bars denoting ± 1 σ of the inter-annual variability. Model simulations for OOSS only (red dashed line) and the combined OOSS and SISS simulations (blue line) are shown with uncertainty envelopes (red and blue shading), representing ± 1 σ of the simulated inter-annual variability. For two ice core locations, three different ice core records are plotted, as indicated by the legend.

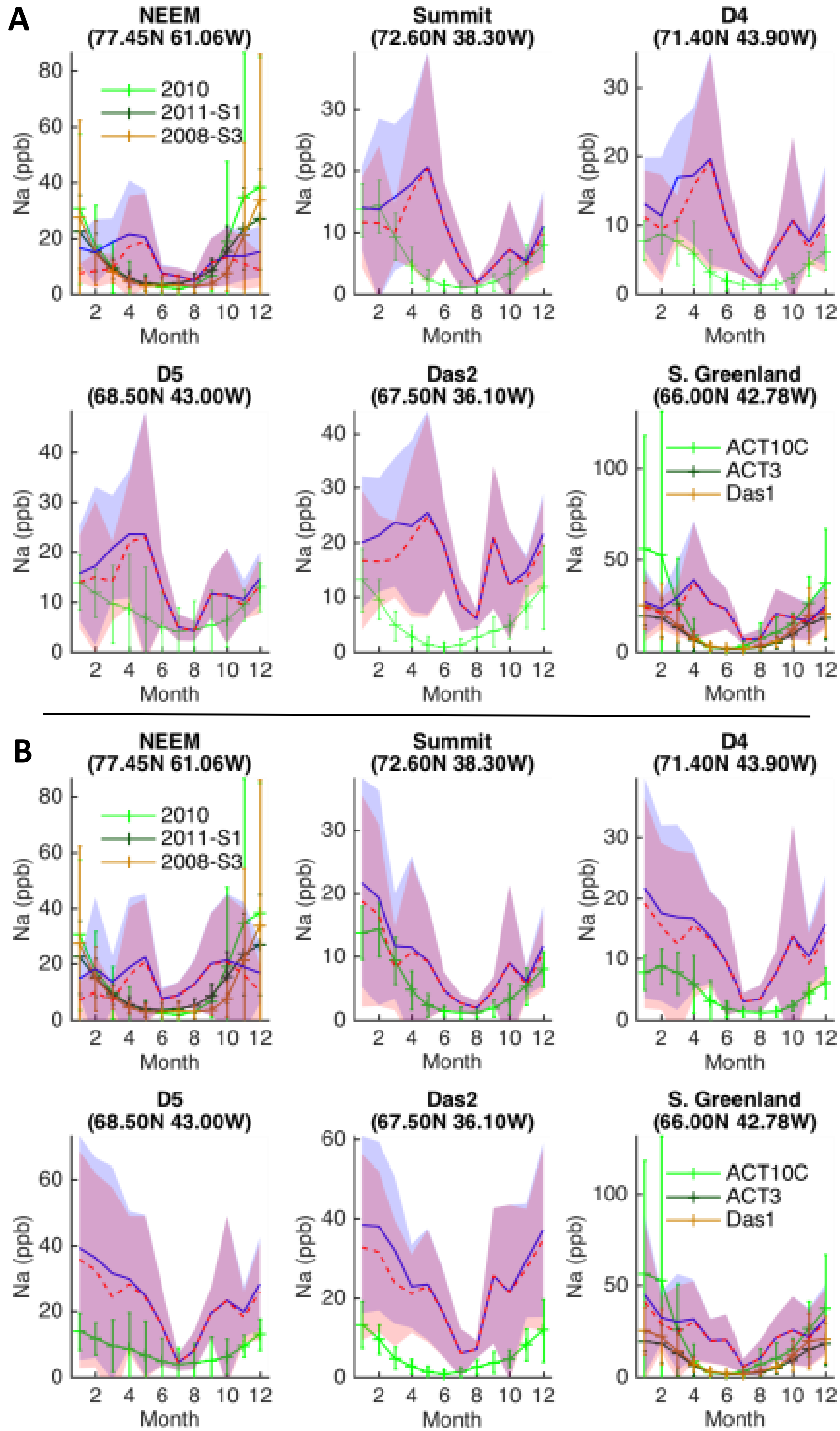

Table S1. Mean sea salt Na concentrations for 1991–1999 AD recorded in ice cores (bold) and simulated by p-TOMCAT [Na] calculated using the constant annual snow accumulation rate indicated by the ice core records (Table 2) in Eq. (5).

Ice core	Annual [Na] (ppb)	DJF [Na] (ppb)	JJA [Na] (ppb)	Seasonal cycle [Na] (ppb) ‡	DJF SISS: OOSS
Tunu	**16**			**22**	
	15	26	6	26	0.3
NEEM-2008-S3	**11**	**25**	**3**	**31**	
	17	17	11	14	0.8
Summit	**6**	**12**	**1**	**13**	
	9	17	3	17	0.2
D4	**4**	**8**	**1**	**8**	
	12	18	5	16	0.1
D5	**8**	**13**	**4**	**10**	
	23	34	10	30	0.1
Das2	**5**	**12**	**2**	**12**	
	25	37	11	33	0.2
Das1*	**11**	**23**	**2**	**23**	
	34	50	16	40	0.1
ACT10C*	**21**	**49**	**4**	**55**	
	25	36	12	29	0.1
ACT3*	**9**	**19**	**2**	**18**	
	30	44	14	35	0.1
ACT2**	**8**	**13**	**3**	**10**	
	50	64	29	43	0.1
ACT11d**	**7**	**10**	**5**	**7**	
	53	68	30	45	0.1

‡ Seasonal cycle is the maximum monthly mean [Na] minus the minimum monthly mean [Na].
* same grid square in p-TOMCAT.
** same grid square in p-TOMCAT.

Table S2. Inter-annual variability of [Na] in ice cores compared to simulated values for 1991-1999 AD. Coefficients of determination (R^2) indicate the proportion of the inter-annual variability of the ice core record that can be explained by p-TOMCAT. Statistically significant values ($p \leq 0.05$) are in bold type. Negative correlations are indicated by n/a.

	[Na] using p-TOMCAT accumulation rate			**[Na] using ice core accumulation rate**		
Ice core	Annual mean [Na]	Annual max. [Na]	Inter-annual diff. in annual mean [Na]	Annual mean [Na]	Annual max. [Na]	Inter-annual diff. in annual mean [Na]
Tunu	n/a	n/a	n/a	0.20	0.29	0.46
NEEM-2011-S1 **	0.15	n/a	0.17	0.11	n/a	0.40
NEEM-2008-S3 **	0.29	**0.58**	0.18	0.04	0.11	0.09
NEEM-2010-20m **	n/a	n/a	n/a	n/a	n/a	n/a
Summit	**0.54**	**0.43**	**0.62**	0.00	0.04	0.00
D4	0.09	0.04	0.20	0.06	0.27	0.33
D5	0.01	n/a	0.00	0.09	n/a	0.14
Das2	n/a	0.05	n/a	0.00	0.34	n/a
Das1*	n/a	n/a	n/a	n/a	0.02	0.02
ACT10C*	n/a	n/a	n/a	n/a	n/a	n/a
ACT3*	n/a	n/a	n/a	n/a	n/a	n/a
ACT2***	0.03	n/a	0.03	n/a	n/a	n/a
ACT11d***	n/a	n/a	0.01	n/a	n/a	n/a

* same grid square in p-TOMCAT

** same grid square in p-TOMCAT

*** same grid square in p-TOMCAT

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