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Key Points:

- Simulated satellite-era interannual variability in Greenland ice core sodium concentrations is driven by meteorology not sea ice
- Sodium budget of coastal Arctic ice cores is dominated by sea salt from the sea ice surface not the open ocean
- Interannual variability of sodium records from coastal Arctic cores is driven by aerosol emission strength, not transport or deposition

Supporting Information:

- Supporting Information S1
- Data Set S1
- Data Set S2
- Data Set S3

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Sea Ice Versus Storms: What Controls Sea Salt in Arctic Ice Cores?

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Abstract The sea ice surface is thought to be a major source of sea salt aerosol, suggesting that sodium records of polar ice cores may trace past sea ice extent. Here we test this possibility for the Arctic, using a chemical transport model to simulate aerosol emission, transport, and deposition in the satellite era. Our simulations suggest that sodium records from inland Greenland ice cores are strongly influenced by the impact of meteorology on aerosol transport and deposition. In contrast, sodium in coastal Arctic cores is predominantly sourced from the sea ice surface and the strength of these aerosol emissions controls the ice core sodium variability. Such ice cores may therefore record decadal to centennial scale Holocene sea ice changes. However, any relationship between ice core sodium and sea ice change may depend on how sea ice seasonality impacts sea salt emissions. Field-based observations are urgently required to constrain this.

Plain Language Summary To better understand the variability of Arctic sea ice and its interaction with climate, we need to reconstruct its history back beyond the time range of observations. As the sea ice surface is a major source of sea salt aerosol in the polar regions, one possibility is to use sea salt sodium records in Arctic ice cores as tracers of sea ice change. However, there are other factors that influence ice core sodium, in addition to the strength of the sea ice source of aerosol. The most important is meteorology, or storminess, that impacts how aerosol is transported and deposited. We adopt a new approach to tackling this problem, using a numerical model that replicates the emission, transport, and deposition of sea salt to simulate Arctic ice core sodium records. This is used to investigate the competing controls of sea ice versus storminess. Our results suggest that sodium records of coastal Arctic ice cores, away from central Greenland, offer the greatest potential for Holocene sea ice reconstruction. However, this study also highlights the need for more field observations to improve understanding of aerosol emission over the sea ice.

1. Introduction

Predicting the future evolution of Arctic sea ice is a fundamental goal of climate science. To examine how Arctic sea ice reacted to rapid climate changes that have occurred in the past, we can consult paleoclimate archives that extend beyond the 35-year satellite record. Several different marine chemical species present in polar ice cores have been proposed as tracers of Holocene sea ice conditions, including sea salt sodium (Na; Severi et al., 2017), methane sulfonic acid (Criscitiello et al., 2013; Rhodes et al., 2009), and bromine (Maselli et al., 2017; Spolaor et al., 2016). Here we focus on sea salt Na.

Evidence from field- and model-based studies indicates that sea salt aerosol can be sourced from salty blowing snow lofted from the surface of the sea ice (Huang & Jaeglé, 2017; Jourdain et al., 2008; Rhodes et al., 2017; Wagenbach et al., 1998; Yang et al., 2008). This sea ice sea salt (SISS) contributes to the winter-late spring peaks in sea salt concentration observed in both Arctic aerosol measurements (e.g., Quinn et al., 2002) and Greenland ice core records (e.g., Banta et al., 2008). Simulations suggest that the sea ice surface produces an order of magnitude more aerosol than the same unit area of open ocean under the same conditions (Yang et al., 2008). However, two major problems currently preclude the use of Arctic ice core Na concentrations as tracers of past sea ice extent:

1. It is difficult to differentiate between open ocean sea salt (OOSS) and SISS in the atmosphere or snowpack. A chemical fingerprint of SISS is fractionation of the sea salt sulfate ($ssSO_4^{2-}$) to Na^+ ratio relative to the seawater value, which results from the precipitation of mirabilite ($Na_2SO_4 \cdot 10H_2O$) within or on top of sea ice (Rankin et al., 2002; Wagenbach et al., 1998). In the present-day Arctic anthropogenic sulfate pollution masks other sulfate contributions (Goto-Azuma & Koerner, 2001) meaning that the relative proportions of

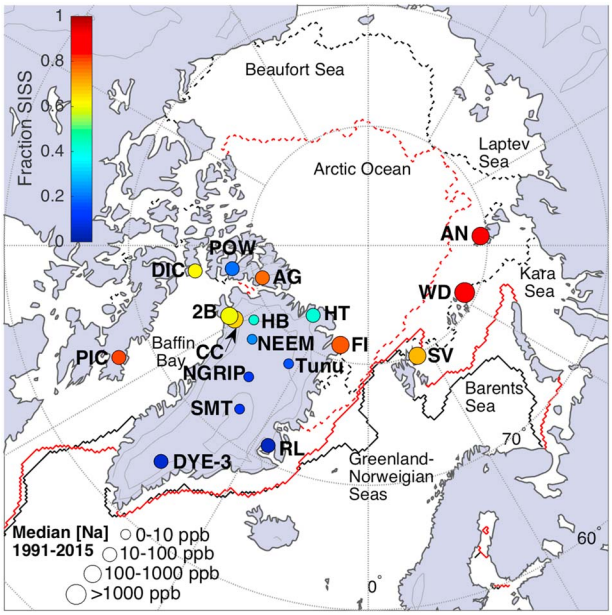

Figure 1. Simulated fraction of annual Na in snowfall that is sea ice sea salt (SISS) (not open ocean sea salt (OOSS)) at Arctic ice core locations for BASE simulation 1991–2015 C.E. Symbol size is proportional to simulated median Na concentration. Ice core labels: PIC = Penny Ice Cap; DIC = Devon Ice Cap; 2B = 2Barrel; CC = Camp Century; POW = Prince of Wales; SMT = Summit; HB = Humboldt; RL = Renland; AG = Agassiz; HT = Hans Tausen; FI = Flade Isblink; SV = Svalbard; WD = Windy Dome; AN = Akademii Nauk. Please see Table S2 for references to individual ice core studies. The solid lines mark maximum sea ice extent, and the dashed lines mark minimum sea ice extent in 1991 (black) and 2012 (red). One thousand-meter topographic contours are shown over land.

SISS and OOSS in aerosol or snow cannot be estimated using either the $ssSO_4^{2-}$ to Na^+ ratio (Jourdain et al., 2008) or the sulfur stable isotopic ratio ($\delta^{34}S$) of samples (Seguin et al., 2014).

2. The dominant control on Na variability in Arctic ice cores is unclear. Which is more important—changing sea ice extent or storminess? Although the amount of SISS emitted to the atmosphere is high likely to vary with sea ice area, emitted SISS must then be transported inland and deposited. The amount of SISS that is finally deposited is influenced by factors that can be broadly termed meteorology, such as the strength of transport winds, as well as the amount and frequency of precipitation (Fischer, 2001; Hutterli et al., 2007; Levine et al., 2014).

Here we address these two problems using a global chemical transport model. We then consider whether or not the recent decrease in Arctic sea ice (Figure 1) may be detectable in Arctic ice core Na records.

2. Materials and Methods

2.1. Chemical Transport Model

We use the Cambridge p-TOMCAT chemical transport model to represent the emission, transport, and deposition of OOSS and SISS (Legrand et al., 2016; Levine et al., 2014; Rhodes et al., 2017). p-TOMCAT has been tuned to sea salt aerosol measurements at Arctic sites (Rhodes et al., 2017). Recent modifications enable p-TOMCAT to simulate monthly Na concentrations in deposited snow, and BASE simulation of Rhodes et al. (2017) has been validated for its representation of Greenland ice core Na concentrations and interannual variability for 1991–1999 C.E.

We extend the BASE simulation time range to 1991–2015 C.E (Text S1 and Table S1 in the supporting information). Briefly, p-TOMCAT is a global model with a spatial resolution of 2.8° × 2.8° across 31 vertical sigma-pressure levels. It is driven by 6-hourly ERA-Interim wind, temperature, and humidity fields (Dee et al., 2011). Sea ice fractions are derived from the Hadley Centre Sea Ice and Sea Surface Temperature data set (Rayner, 2003) and sea ice assumed to be multiyear if it was present the previous September. OOSS emissions occur via bubble bursting (Gong, 2003; Jaeglé et al., 2011), and SISS emissions occur via sublimation of snow particles (Yang et al., 2008) lofted from the surface of sea ice during blowing snow events (e.g., Nishimura & Nemoto, 2005; Savelyev et al., 2006).

The rate of SISS emission from multiyear sea ice is set to half the first-year sea ice rate in the BASE simulation. In previous work this scenario produced better correspondence between p-TOMCAT simulations and available Arctic sea salt aerosol observations than scenarios in which multiyear sea ice contributed either no aerosol at all or the same amount as first-year ice (Rhodes et al., 2017). We reasonably expect there to be a distinction in aerosol production between first-year and multiyear sea ice because aerosol emissions from blowing snow (as parameterized) increase with snow salinity (Yang et al., 2008). Limited measurements of snow on Arctic sea ice (Krnavek et al., 2012) suggest the salinity of snow on thick, multiyear sea ice is lower than that of snow of thin, first-year sea ice. These observations are further supported by the knowledge that brine rejection causes the salinity of the sea ice itself to decrease with time (Cox & Weeks, 1974) and that thicker sea ice is less likely to be submerged and/or flooded by highly saline seawater (Massom et al., 2001).

2.2. Simulation of Arctic Snow Na Concentrations

We use the BASE simulation results to investigate how ice core Na concentrations and SISS:OOSS values change in response to the recent decline in Arctic sea ice. Given that SISS emissions via blowing snow are suppressed over sea ice designated as multiyear (section 2.1), we perform two additional experiments for

2000–2015 C.E. to examine the impact of the changing ratio of first-year to multiyear sea ice on our simulated ice core Na records:

ALLSI: SISS emission strength is equal across all sea ice.
FYSI: SISS emissions only occur above first-year ice.

2.3. Sensitivity Tests

To constrain the relative influence of sea ice conditions and meteorology on Arctic ice core Na concentrations, we conduct sensitivity tests. p-TOMCAT is run repeatedly over the 2000–2015 C.E. interval, holding either meteorology that influences transport and deposition of aerosol (ERA-Interim wind, temperature, and humidity fields and the model-calculated precipitation) or sea salt (OOSS and SISS) emission rate constant. As p-TOMCAT is not a coupled model, sea ice does not evolve in response to meteorology and vice versa, so we ensure that OOSS and SISS emissions reflect sea ice conditions by prescribing them (Text S2).

Three different experiments are performed for 2015 C.E.:

CONTROL: Meteorology and sea salt emissions both from current year (n.b. not the same as BASE simulation [section 2.1] because emissions are prescribed, Text S2).
VARYEMIS: Meteorology from 2000 C.E. and sea salt emissions from current year.
VARYMET: Sea salt emissions from 2000 C.E. and meteorology from current year.
For both VARYEMIS and VARYMET, we calculate the normalized root mean square deviation (NRMSD) relative to CONTROL.

2.4. Air Mass Back Trajectories

To verify the likely source regions of sea salt aerosol deposited at Arctic ice core sites, we use the Hysplit4 software (Draxler & Hess, 1998) to perform air mass back trajectories (Text S4). Back trajectories were initiated every 5 days in winter–spring 1991–2015 C.E. and allowed to travel for 3 days.

3. Results

3.1. Origin of Sea Salt in Arctic Snow: Sea Ice Surface or Open Ocean?

We first examine the proportion of SISS relative to OOSS that makes up the Na deposited at Arctic ice core sites in our BASE simulation 1991–2015 C.E. As the proportion of SISS relative to OOSS currently deposited at Arctic ice core locations cannot be determined through laboratory measurements, we seek to select ice core locations where SISS dominates the Na budget.

Our BASE simulation predicts that SISS comprises a small fraction ($<$0.25) of the annual Na budget of high elevation ($>$2,000 m), inland Greenland ice cores, such as NEEM, NGRIP, and Summit (Figure 1). Simulated SISS contributions are equally low at high elevation, southerly Greenland ice core locations, Renland (4%), and Dye-3 (5%), adjacent to limited winter sea ice cover. At some northerly Greenland ice core locations, within 100 km of coastlines bordered by sea ice, SISS becomes the dominant contributor to the Na budget. This is true for Flade Isblink, 2Barrel, and Camp Century (Figure 1). However, at Hans Tausen, just 50 km from multiyear sea ice north of Greenland, SISS accounts for only 41% of Na.

In the wider Arctic, our results suggest that the Na records of ice cores from low elevation ($<$2,000 m), localized ice domes, situated close to sea ice cover, are dominated by SISS. The annual fraction of Na that is SISS ranges from 0.63 at Devon Ice Cap to 0.90 at Windy Dome (Figure 1 and Table S2). The only exception to this is Prince of Wales Icefield in the Canadian Arctic, which our simulations suggest receives only 18% SISS. Previous studies suggest that the strong contribution of OOSS at this location results from the proximity of the North Water polynya (Criscitiello et al., 2016; Wasiuta et al., 2006).

3.2. What Drives Interannual Variability in Na?

Our sea ice versus storm sensitivity tests indicate that the influence of meteorology on aerosol transport and deposition is the dominant control on interannual variability in ice core Na at central Greenland sites (Tunu, Summit, and NEEM, Figure 2). For each site, VARYMET results closely follow the CONTROL results (Figure S1 in the supporting information), with NRMSDs between the two of 21 and 30% (Figure 2a). The VARYEMIS results typically diverge much more from the CONTROL results, with NRSMD values of 77–94% (Figure 2b). If only SISS Na is considered, the result does not materially change, the NRMSD between VARYMET and CONTROL

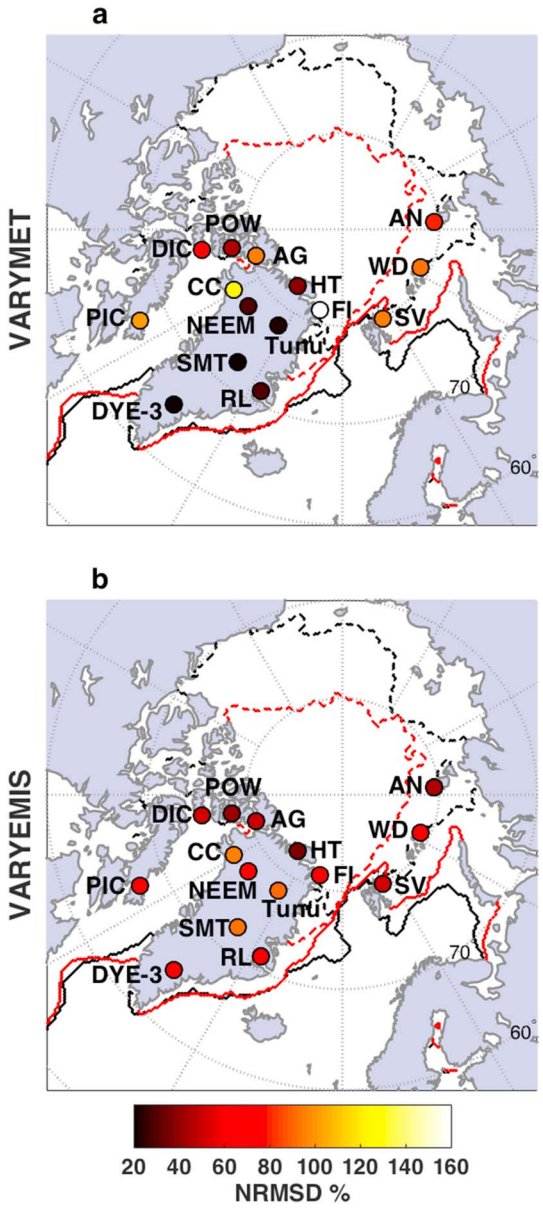

Figure 2. Relative influence of meteorology and sea salt aerosol emissions strength on Na concentrations in Arctic ice cores 2000–2015 C.E. The colored circles display normalized root mean square deviation (NRMSD) between monthly snow Na of (a) CONTROL and VARYMET and (b) CONTROL and VARYEMIS, both normalized to mean snow Na of CONTROL. The lower values (dark red colors) indicate that the varying factor (a: meteorology and b: OOSS & SISS emissions) has large influence on Na. The red and black lines show sea ice extent as Figure 1. See Figure S3 for equivalent maps for sea ice sea salt and open ocean sea salt individually and Figure S4 for equivalent maps for January to April only.

is still lower than between VARYEMIS and CONTROL at each site (56–84% versus 82–111%), suggesting meteorology is the dominant factor determining SISS Na (Figure S2).

Closer to the Greenland coast, ice core sites such as Renland, Humboldt, and 2Barrel still show lower NRMSDs for VARYMET than VARYEMIS, suggesting a dominance of meteorology on Na variability (Figure 2 and Table S2). However, the difference between the two NRMSD is less than for inland sites. Camp Century, which is located close to 2Barrel in NW Greenland, shows a lower NRMSD for VARYEMIS, suggesting that emission strength has a greater influence here, but both NRMSDs are high (>90%).

To obtain VARYEMIS NRMSDs that are significantly lower than VARYMET values, we have to move closer to the coast, to relatively low elevation, localized ice domes located close to the sea ice. This is almost accomplished at Hans Tausen on the northern tip on Greenland which sees very similar NRMSDs for VARYMET and VARYEMIS. Southeast from there at Flade Isblink the difference between NRMSDs for VARYMET (184%) and VARYEMIS (72%) is marked, suggesting that emission strength dominates over meteorology in the control of ice core Na variability. Moving around the Arctic basin into the Siberian Arctic, the same is true for ice cores on Svalbard, at Windy Dome, and Akademii Nauk. The influence of emissions strength on Na concentrations is also greater than the influence of meteorology for Canadian Arctic ice cores such as Agassiz and Devon Ice Cap.

These patterns are largely repeated if just SISS Na is considered (Figure S3) or if only Na variability during months of peak sea ice (January–April) is examined (Figure S4). Taken together, our results indicate that emission strength is more important than meteorology in determining the Na content of ice cores from coastal ice domes in the Arctic, opposite to our findings for central Greenland cores.

4. Discussion

4.1. High Arctic Ice Core Na Records

Our simulations predict that the Na concentration records of seven coastal Arctic ice cores are (1) dominated by SISS rather than OOSS and (2) more sensitive to changes in emission strength than meteorology (Figures 1 and 2 and Table S2). The seven cores are Penny Ice Cap, Devon Ice Cap, Svalbard, Akademii Nauk, Agassiz, Windy Dome, and Flade Isblink.

We propose that these ice cores have the best chance of preserving a Na signal of Holocene Arctic sea ice change and test this by searching for a signal of the recent decadal-scale decline in Arctic sea ice cover in our simulations (Text S3; Bertler et al., 2005). We regress simulated ice core annual mean Na concentrations from the BASE simulation against the annual mean sea ice area of each grid cell in the Arctic region (Figure S5). Annual mean Na values are used because seasonal values are often difficult to resolve in these high Arctic cores (e.g., Criscitiello et al., 2016). Most of the ice cores show only limited areas of significant ($p < 0.05$) regression between Na and sea ice in distal locations where sea salt aerosol is unlikely to be sourced from. The lack of positive correlation between sea ice area and the simulated ice core Na records could be due to the nature of the recent Arctic sea ice change. A marked decrease in area has occurred in minimum (September) sea ice, whereas winter (February) sea ice area has remained fairly

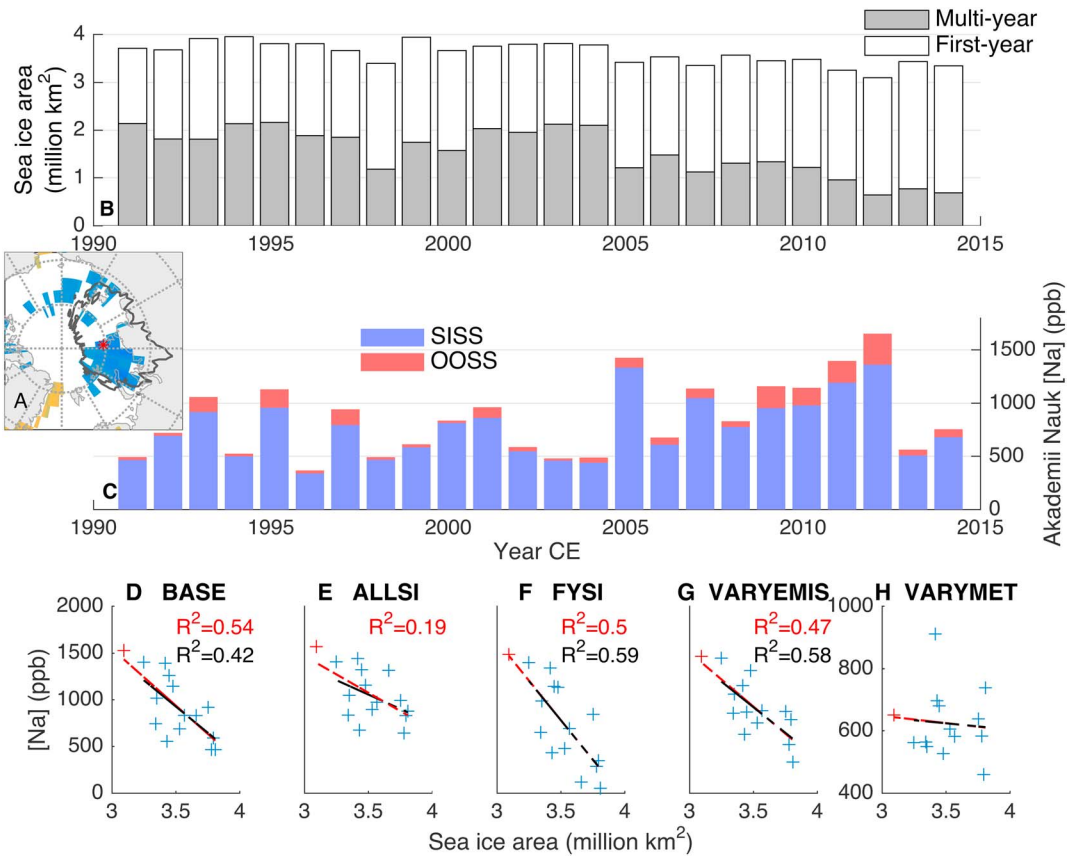

Figure 3. Relationship between sea ice and Na concentrations in the Akademii Nauk ice core, as simulated by p-TOMCAT. Sea ice and Na concentrations are annual means calculated for July to June. (a) The map displays grid cells with significant ($p < 0.05$) negative regression (blue colors) between sea ice area and Na of ice core (located at red asterisk). The black contour denotes likely origin of sea salt aerosol arriving at location (see Figure 4 caption). Sea ice area across this region (60–87°N, 60–150°E, 7.7 million km^2 total area) is compared to simulated Akademii Nauk Na concentrations. See Figure S5 for equivalent maps for other ice core sites. (b) Change in multiyear and first-year sea ice area over time. (c) BASE simulation Na concentrations (sea ice sea salt (SISS) and open ocean sea salt (OOSS)) over time. The scatterplots display correlation between sea ice area and ice core Na 2000–2015 C.E. for experiments: (d) BASE, (e) ALLSI, (f) FYSI, (g) VARYEMIS, and (h) VARYMET. The 2012–2013 C.E. result (red cross) is included in red linear fit and excluded from black linear fit. Coefficients of determination (R^2) displayed are significant at $p < 0.05$.

consistent (Figure 1) until very recently. As most of the SISS is deposited during the winter months when sea ice is at its maximum extent (Rhodes et al., 2017), a modest winter sea ice change may be difficult to detect in ice core Na. There are very few regions of significant regression between maximum (winter) sea ice area and the simulated ice core Na time series (Figure S6).

Contrary to expectations, the simulated ice core Na records from Akademii Nauk, and to a lesser extent Penny Ice Cap, show negative correlation with local sea ice area. Akademii Nauk Na has significant inverse relationship ($R^2 = 0.54$, $p < 0.002$) with sea ice area (Figure 3d) across a broad region encompassing areas of the Laptev, Kara, and Barents Seas (Figure 3a). Particularly from 2000 C.E. onward, a gradual decrease in sea ice area is accompanied by a decadal-scale increase in ice core Na (Figures 3b and 3d). The Na increase is not fuelled by additional OOSS input—the SISS:OOSS remains more or less stable (Figure 3c). Instead, the Na increase can be linked to the changing proportion of sea ice that is first-year (annual) relative to multiyear (perennial). Over the 1991–2015 C.E. interval, the fraction of sea ice that is first-year rises from 0.43 in 1991–1995 C.E. to 0.70 in 2010–2015 C.E. (Figure 3b). As first-year sea ice produces twice as much SISS aerosol as multiyear sea ice in our SISS emission parameterization (section 2.1), SISS emission increase causes elevated Na levels at Akademii Nauk despite the decrease in total sea ice area.

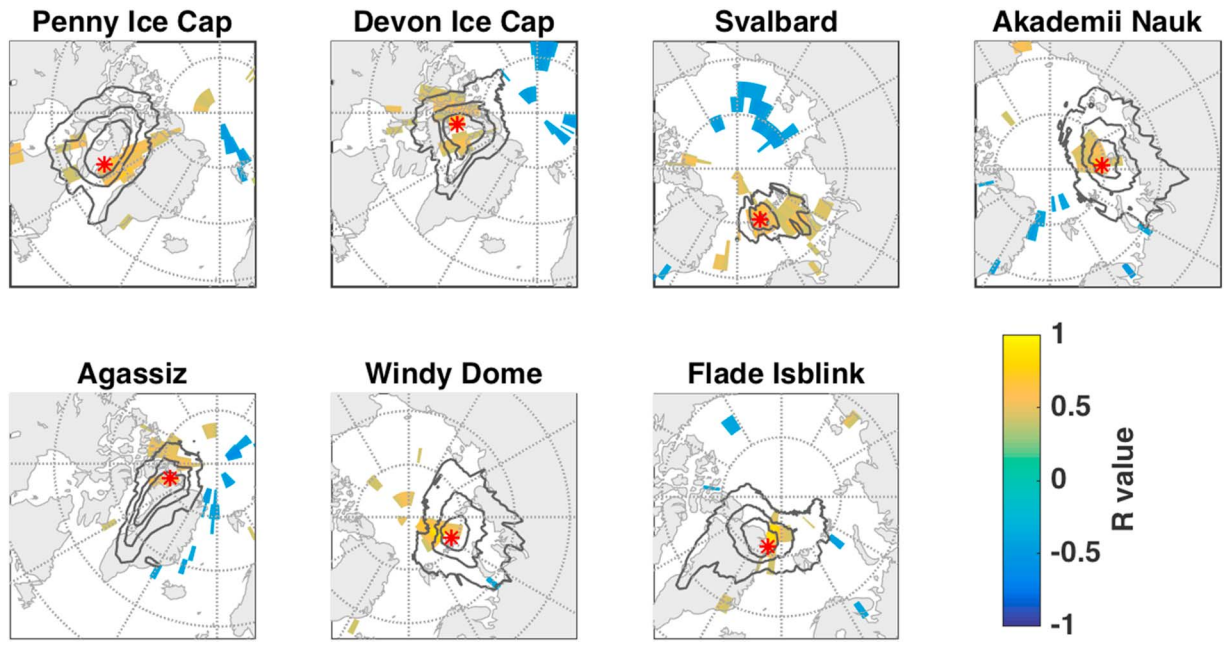

Figure 4. Regression of simulated ice core Na against sea ice sea salt (SISS) emissions for seven selected Arctic ice cores. Each map shows regression of simulated ice core mean annual Na concentration against the mean annual SISS emissions rate (Tg Na yr^{-1}) for each grid cell 1991–2015 C.E. Annual mean values are calculated for 12 months from July to June. Only areas of significant regression ($p < 0.05$) are shown. Ice core locations are marked by red asterisks. The black contours denote the fraction of air mass back trajectories from the ice core location in winter-spring that reside within 1,000 m of the surface (0.1, 0.05, and 0.02 fractions decreasing away from ice core location).

This result can be compared to the ALLSI and FYSI experiments. When all sea ice emits SISS equally (ALLSI), there is no significant relationship between sea ice area and Akademii Nauk Na (Figure 3e) once the outlier (2012–2013 C.E., year of record low sea ice) is removed. It is the first-year sea ice variability that generates the inverse relationship between Akademii Nauk Na and sea ice area, as the FYSI results show (Figure 3f).

If we examine this relationship for the VARYEMIS and VARYMET runs, the results are as expected. No significant statistical relationship between sea ice and Na is identifiable for the VARYMET run (Figure 3h), while the VARYEMIS run (Figure 3g) produces a similar relationship to the BASE simulation. This reiterates our finding that emission strength, and not transport and deposition processes driven by meteorology, controls Akademii Nauk Na concentrations.

4.2. Link to SISS Emissions

Despite the lack of positive correlation between ice core Na and sea ice at the Arctic sites identified, if we regress the simulated ice core Na records against SISS emissions instead of sea ice area, all the cores show a region of significant positive ($R > 0.5$) regression located close to the ice core site (Figure 4). Furthermore, each of these regions is encompassed by the area of air mass origin for that ice core site, as indicated by back trajectory analysis. This illustrates that the Na variability of these ice cores is related to aerosol source strength, as our sensitivity tests suggest, but that recent local sea ice change (particularly in winter) has not been great enough to substantially impact annual SISS emissions. However, the significant positive relationships between local SISS emissions and ice core Na signals suggest that a larger-scale change in sea ice conditions would impact SISS emissions and the Na records of the proximal ice cores would register a shift. The critical question is as follows: Which direction would Na concentrations shift? A sea ice change involving total collapse of year-round sea ice cover would result in decreased Na, but the Akademii Nauk results suggest that a switch from a perennial to annual sea ice regime could instead cause an increase in Na.

5. Conclusions

Based on the chemical transport model results presented, we conclude that it is highly unlikely that interannual variability in Na concentrations from inland Greenland ice cores will reflect fluctuations in sea ice extent or

that a decadal scale trend in Greenland ice core Na could be related to Arctic sea ice under present-day (late Holocene) conditions. The contribution of SISS to the Na budget is consistently low across Greenland interior (Figure 1) and variations in transport and deposition related to meteorology determine Na variability, rather than emissions strength (Figure 2). These results lend support to previous work on Holocene Greenland ice cores that demonstrate that Na variability is closely linked to large-scale atmospheric circulation patterns and their influence on aerosol transport and deposition (Fischer & Mieding, 2005; Hutterli et al., 2007).

Alternatively, our results suggest that low elevation ice domes located close to the sea ice in the high Arctic offer the best potential for preservation of interannual and/or decadal scale Na signals related to sea ice variability. These ice cores are dominated by Na sourced from the sea ice surface rather than the open ocean (Figure 1), and our sensitivity tests suggest that storminess, and its impact on aerosol transport and deposition, is of secondary importance to the strength of sea salt emissions in controlling the ice core Na signal (Figure 2). This is reflected by positive correlation between the strength of local SISS emissions and ice core Na concentration in all seven of the Arctic ice cores selected (Figure 4). Despite this, none show significant positive correlation between Na and sea ice area over 1991–2015 C.E., suggesting that the recent decline in winter Arctic sea ice was too modest to impact annual SISS emissions significantly.

However, the Akademii Nauk ice core simulation shows negative correlation between local sea ice area and Na concentration, driven by the increasing area of first-year sea ice as the sea ice regime changes from predominantly perennial to annual (Figure 3). The strength of this relationship is dictated by the parameterization of sea salt aerosol emissions from the sea ice, specifically the influence of sea ice thickness and/or age seasonality on snow salinity, which we have very few constraints on. This example highlights the critical need for coordinated field measurements of snow on sea ice (salinity, depth, and density) and its relationship to sea ice age/thickness and sea salt aerosol production.

Under our current assumption that multiyear sea ice contributes half as much aerosol as first-year sea ice, our simulations suggest that Arctic ice core Na may be sensitive to two modes of sea ice change in the Holocene: (1) retreat/expansion of winter sea ice and (2) switch between perennial and annual sea ice regimes. Combining sea salt Na with other ice core chemistry related to sea ice and associated marine productivity could prove valuable in disentangling such changes.

Acknowledgments
Data Set S1 comprises BASE simulation results (also at doi: 10.1594/PANGAEA.887987). Data Set S2 comprises CONTROL, VARYMET, and VARYEMIS simulations, and Data Set S3 comprises FYSI and ALLSI results. References to existing ice core data are provided in Table S2. This study was funded by a European Union Horizon 2020 Marie Sklodowska-Curie Individual Fellowship (658120, SEADOG) awarded to R.H.R. E.W.W. acknowledges support from the Royal Society (RP 120096 and NF171273), and X.Y. and E.W.W. both acknowledge funding from NERC (NE/J023051/1).

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