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Numerical Representations of Marine Ice-Nucleating Particles in Remote Marine Environments Evaluated Against Observations

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Key Points:
- Observed marine boundary layer ice-nucleating particle concentrations were successfully predicted using marine and dust parameterizations
- Sea spray aerosol was the dominant source of simulated ice-nucleating particle populations up to 3–5 km over the Southern Ocean
- Mineral dust aerosol was a critical component of model-predicted ice-nucleating particle populations present above 5 km

Supporting Information:
- Supporting Information S1

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Abstract The abundance and sources of ice-nucleating particles, particles required for heterogeneous ice nucleation, are long-standing sources of uncertainty in quantifying aerosol-cloud interactions. In this study, we demonstrate near closure between immersion freezing ice-nucleating particle number concentration (n_{INPs}) observations and n_{INPs} calculated from simulated sea spray aerosol and dust. The Community Atmospheric Model with constrained meteorology was used to simulate aerosol concentrations at the Mace Head Research Station (North Atlantic) and over the Southern Ocean to the south of Tasmania (Clouds, Aerosols, Precipitation, Radiation, and atmospherIc Composition Over the southeRN ocean campaign). Model-predicted n_{INPs} were within a factor of 10 of n_{INPs} observed with an off-line ice spectrometer at Mace Head Research Station and Clouds, Aerosols, Precipitation, Radiation, and atmospherIc Composition Over the southeRN ocean campaign, for 93% and 69% of observations, respectively. Simulated vertical profiles of n_{INPs} reveal that transported dust may be critical to n_{INPs} in remote regions and that sea spray aerosol may be the dominate contributor to primary ice nucleation in Southern Ocean low-level mixed-phase clouds.

Plain Language Summary The clouds over remote oceans are often comprised of supercooled liquid droplets, but global models struggle to represent the complex processes that control ice formation in these clouds. One poorly understood, but critical, aspect controlling the liquid-ice partitioning in these clouds is the abundance of particles that catalyze ice crystal formation, or ice-nucleating particles (INPs). Observations show that INPs are extremely rare in remote marine environments and are dominated by an oceanic source. However, current global models do not account for these uniquely low INP concentrations and their marine source. We used observations of INPs from two previous field campaigns to evaluate INP concentrations estimated from a global climate model that incorporates particles from sea salt, marine organic matter, and mineral dust. Our results constitute an early evaluation of the potential of present-day global atmospheric models to successfully predict INP concentrations in the lowest atmospheric level that feeds clouds over the ocean. Extrapolating our approach to higher altitudes, the model suggests mineral/soil dust particles from long-range transport may also be a critical INP source for marine clouds.

1. Introduction

Global climate models simulate large positive shortwave radiative biases over the Southern Ocean, which have been attributed to excessive ice clouds and too few liquid clouds when compared to satellite observations (Bodas-Salcedo et al., 2014). Aerosol-cloud interactions may be an important contributor to these biases through the action of immersion freezing ice-nucleating particles (INPs), or particles required for ice crystal formation in mixed-phase clouds. Modeling studies suggest that INPs associated with sea spray aerosol (SSA), produced at the ocean surface via bubble bursting, are important for INP populations over Southern Ocean (Burrows et al., 2013; Vergara-Temprado et al., 2017), though the ice-nucleating efficiency of SSA is significantly lower than terrestrial aerosol (DeMott et al., 2016). While studies have demonstrated that simulated Southern Ocean clouds are sensitive to primary ice nucleation (Tan & Storelvmo, 2016; Vergara-Temprado et al., 2018), most climate models do not account for differing ice nucleation potential of all atmospheric aerosol types, though atmospheric ice nucleation has been linked to simulated dust and black carbon (e.g., Wang et al., 2014). Huang et al. (2018) reported that the relative importance of the INPs associated with SSA compared to dust depends strongly on the approach used to estimate marine

INPs. The ability of models to predict INP number concentrations (n_{INPs}) in remote marine environments has not been directly evaluated against time-matched observed n_{INPs}. This evaluation is needed in order to build greater confidence in the ability of models to credibly simulate n_{INPs} and thus investigate remote mixed-phase cloud properties.

Laboratory studies have identified two categories of INPs that may be associated with the organic component of SSA, including a regularly occurring ice nucleation active molecule type (e.g., long-chain fatty acids; DeMott et al., 2018) and an episodic heat-labile microbial type (e.g., intact or fragmented diatoms; McCluskey, Hill, Sultana, et al., 2018). Two marine INP parameterizations have been reported in the literature. Wilson et al. (2015) identified a temperature-dependent relationship between total organic carbon mass and ice-nucleating entity (INE; Vali et al., 2015) number concentrations in sea surface microlayer (SML; the upper 50-100 μm of the ocean; Cunliffe et al., 2011) samples collected from the North Atlantic and Arctic ocean basins. Representing marine organic INPs, this relationship (W15) was applied to simulated marine organic carbon aerosol concentrations to estimate atmospheric n_{INPs}. McCluskey, Ovadnevaite, Rinaldi, et al. (2018) derived a marine INP parameterization based on SSA surface area and temperature from observed n_{INPs} in pristine air masses originating from the North Atlantic. While the episodic marine microbial INP type was briefly observed during the study period, this parameterization (M18) was intended to represent the regularly occurring ice nucleation active marine molecule category only.

The goals of this study are (1) to improve the model estimates of immersion freezing INPs by accounting for INPs associated with SSA simulated by a global atmospheric model and (2) to construct the simulated distribution as well as spatiotemporal variability in INP number and source. Calculated n_{INPs} based on model-simulated aerosol were evaluated against n_{INPs} observed from air masses from the North Atlantic Ocean (McCluskey, Ovadnevaite, Rinaldi, et al., 2018) and the Southern Ocean (McCluskey, Hill, Humphries, et al., 2018).

2. Methods

2.1. Model

The Community Atmosphere Model (CAM5; Neale et al., 2010) is the atmospheric component of the Community Earth System Model version 1 (Hurrell et al., 2013). In this study, we use the CAM5 at a horizontal resolution of 1.9° latitude by 2.5° longitude, with 30 vertical layers (surface to 2.26 hPa) to simulate aerosol as input to immersion freezing INP parameterizations (section 2.2). Without modifications to the cloud microphysical scheme, we assessed the representation of aerosol species and INP parametrizations. Aerosol species are emitted, advected, and participate in atmospheric transport, aerosol microphysics (nucleation, condensation, and coagulation), cloud microphysical processes, sedimentation, and dry deposition following the Modal Aerosol Module 4-mode scheme (Liu et al., 2016), which is an extension of the model's default 3-mode scheme (Liu et al., 2012). Aerosol number and mass are predicted for four lognormal modes, namely, Aitken, accumulation, coarse, and primary carbon. Mass mixing ratios of aerosol species, including dust, sea salt, particulate organic matter, black carbon, sulfate, secondary organic aerosol, and marine organic matter, are predicted as internal mixtures within the individual modes. All volumetric concentrations reported herein have been adjusted to standardized pressure (101.345 kPa) and temperature (273.15 K). Sea salt is emitted based on sea surface temperature and wind speed, where wind speed serves as a proxy for the parameterization of whitecap area (Liu et al., 2012). For particles with geometric diameter smaller than 2.8 μm, the particle number flux is estimated based on laboratory-generated aerosol parameterized by Mårtensson et al. (2003). Particle number flux for geometric diameters greater or equal to 2.8 μm is predicted following Monahan et al. (1986). Marine organic matter associated with biological productivity is parameterized as a fraction of total Aitken and accumulation mode SSA mass, following OCEANFILMS (Burrows et al., 2018). OCEANFILMS is a mechanistic parameterization that utilizes monthly mean surface distributions of ocean organic constituents, derived from ocean biogeochemical model simulations, to determine concentrations of macromolecules and the resulting surface coverage of bubble films (Burrows et al., 2014). Dust emissions are dynamically simulated as a function of wind speed and are dependent on the threshold surface wind friction velocity and soil type (Mahowald et al., 2006), following the Dust Entrainment and Deposition Model (Zender et al., 2003).

2.2. INP Parameterizations

Three parameterizations were applied to estimate n_{INPs} off-line and are described in this section and in Table S1 in the supporting information. INPs associated with dust aerosol were estimated from the number concentration of dust particles larger than 500 nm and temperature following DeMott et al. (2015). D15 was empirically derived from laboratory and field data collected at temperatures less than −20 °C. The Wilson et al. (2015) parameterization (W15) was used to estimate INPs associated with marine organic aerosol based on temperature and the total organic carbon mass estimated assuming an organic matter to organic carbon ratio (OM:OC) of 2. Varying OM:OC over the range of values reported in the literature (1.2 to 2.6; Burrows et al., 2013) does not significantly change the conclusions of this study (Text S1 and Figure S3). Finally, ice nucleation site densities (INP per square meter of aerosol) associated with total SSA (sea salt and marine organic matter) surface area were determined following McCluskey, Hill, Sultana, et al., 2018; M18).

2.3. Observations

We compare n_{INPs} calculated from model-simulated aerosol with n_{INPs} measured at the Mace Head Research Station (MHD; 53.32°N, 9.90°W; McCluskey, Ovadnevaite, Rinaldi, et al., 2018). Observed variables used in this study included immersion freezing n_{INPs}, aerosol mass concentrations, wind direction, and wind speed during August 2015. M18 was derived from pristine air mass measurements at this location, and W15 was derived from SML samples collected from the North Atlantic and Arctic Oceans during summertime.

The second set of observations was collected onboard the Commonwealth Science and Industrial Research Organisation RV *Investigator* from 13 March to 15 April in 2016 as part of the Clouds, Aerosols, Precipitation, Radiation, and atmospherIc Composition Over the southeRN ocean campaign (CAPRICORN; McCluskey, Hill, Humphries, et al., 2018). Observed variables used in this study included immersion freezing n_{INPs}, aerosol surface area, wind speed, and wind direction. Both field studies included measurements of n_{INPs} active at temperatures from 0 to −27 °C using an ice spectrometer (Hiranuma et al., 2015), which is an off-line sampling method using collection on open-faced filters intended to capture the full aerosol size distribution. During the MHD and CAPRICORN study, 27 and 12 samples were collected, respectively, with air sampling durations ranging from 6 to 63 hr (see McCluskey, Ovadnevaite, Rinaldi, et al., 2018 and McCluskey, Hill, Humphries, et al., 2018).

2.4. Model Simulation and Evaluation

A CAM5 simulation was performed from 1 January 2015 to 31 July 2016 with constrained meteorology (Ma et al., 2013). The Modern Era Retrospective-Analysis for Research and Applications reanalysis data set (Rienecker et al., 2011) was used for nudging, after vertically regridding from the 56 original vertical levels to the 30 CAM5 vertical levels. Horizontal winds were nudged to Modern Era Retrospective-Analysis for Research and Applications reanalysis data by Newtonian relaxation, using a uniform nudging time scale of 6 hr at all vertical levels (Zhang et al., 2014), in order to match observed atmospheric dynamics and transport. For MHD, the nearest model grid box was compared to observations (Figure S1). For CAPRICORN, aerosol properties were spatially averaged over the region of the ship campaign (Figure S2). Model-simulated aerosols were output at each 30-min model dynamical time step, then averaged over the INP filter collection period. The simulated aerosol quantities were used as input for the INP parameterizations (section 2.2; Table S1) to determine model-estimated n_{INPs} associated with SSA (M18 or W15) and dust (D15) at INP measurement temperatures.

Modified normalized mean bias (B_n) and fraction gross error (E_f) were determined for each parameterization:

$$\mathrm{B_n} = \frac{2}{\mathrm{N}} \sum \frac{\mathrm{f_i} - \mathrm{O_i}}{\mathrm{f_i} + \mathrm{O_i}}, \qquad \mathrm{E_f} = \frac{2}{\mathrm{N}} \sum \left| \frac{\mathrm{f_i} - \mathrm{O_i}}{\mathrm{f_i} + \mathrm{O_i}} \right|.$$

B_n describes the mean fractional difference between the modeled estimates (f) and observations (O); E_f measures the model absolute error. These metrics are impacted by both the model's ability to simulate the aerosol quantities used for calculating INPs and the INP parameterizations. We also calculated the percentage of model-estimated n_{INPs} that are within a factor of 2 and 10 of the observed values to describe the variance of agreement.

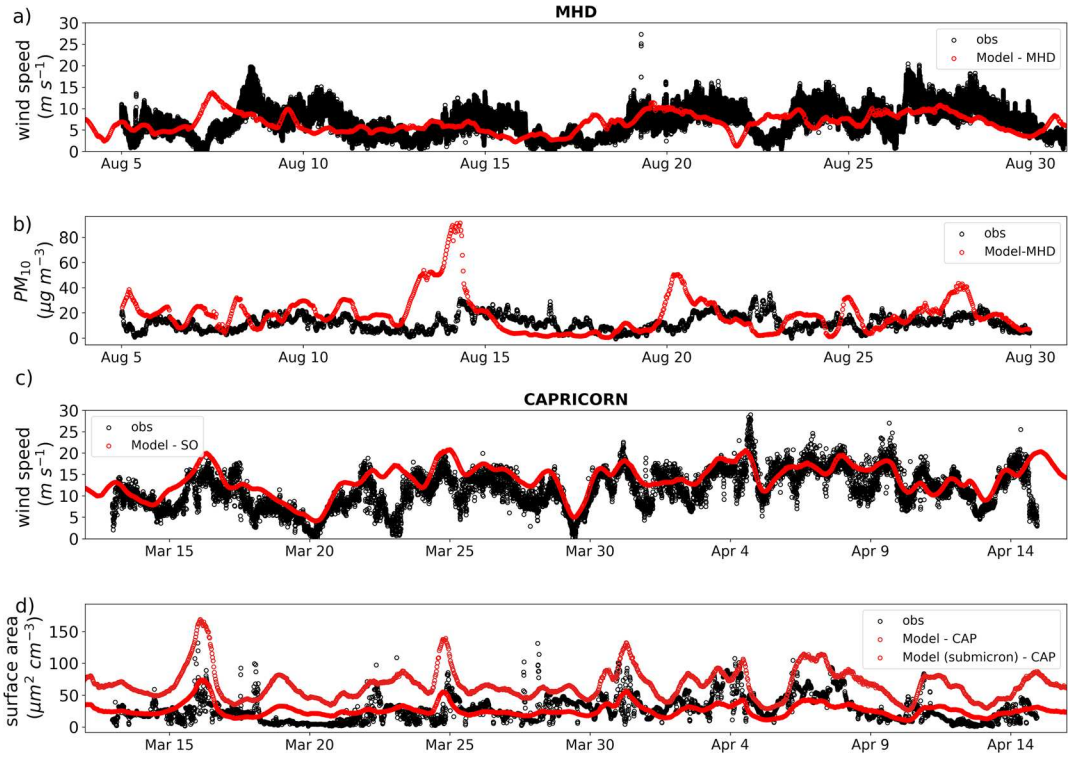

Figure 1. Timeline of observed (black) and modeled (red; a) wind speed and (b) PM_{10} at Mace Head Research Station (MHD) and (c) wind speed and (d) aerosol surface area during Clouds, Aerosols, Precipitation, Radiation, and atmospherIc Composition Over the southeRN ocean campaign (CAPRICORN). In (d), total (dark red) and submicron (bright red) aerosol surface area are shown.

3. Results

3.1. Simulated Winds and Aerosol

Wind speeds from the nudged simulations were in good agreement with observed values, shown in Figures 1a and 1c. The model captured the overall temporal variability in mass concentrations of particles with diameters smaller than 10 μm (PM_{10}) at MHD (Figure 1b), with two periods of significantly higher simulated PM_{10} compared to observations. Differences may be due to errors in simulated emissions, transport, or removal processes. Simulated Aitken and accumulation mode aerosol (which we denote as "submicron" hereafter) surface area agreed well with observed surface area for CAPRICORN (Figure 1d). Including simulated coarse mode sea salt produced a high bias in the aerosol surface area (Figure 1d), which may be associated with a model bias or a bias in the observations due to an upper limit in the sampled aerosol sizes. This is discussed further in section 3.3.

3.2. Simulated INPs

We evaluate the model prediction accuracy at different temperatures by comparing model-predicted and observed n_{INPs} (Figure 2), and B_n in 2° temperature bins (Figure 3). Elevated n_{INPs} were observed at MHD during a terrestrial organic aerosol event (McCluskey, Ovadnevaite, Rinaldi, et al., 2018), corresponding to two temperature spectra that are underpredicted in the MHD model-observation comparisons. These INPs were likely organic soils that are not accounted for in this study. W15 (Figure 2a) and M18 (Figure 2c) overpredicted ($B_n = 1.60$) and underpredicted ($B_n = -0.60$) n_{INPs} compared to MHD observations, respectively, across all temperatures (Figure 3a). Predicted n_{INPs} associated with mineral dust (D15; Figure 2e) at MHD were lower than observed n_{INPs} ($B_n = -1.54$) across all temperatures.

During CAPRICORN, estimates of n_{INPs} using W15 were higher than observed n_{INPs} ($B_n = 1.88$). M18 had better agreement, with 69% of simulated n_{INPs} within a factor of 10 of observations, although biased high

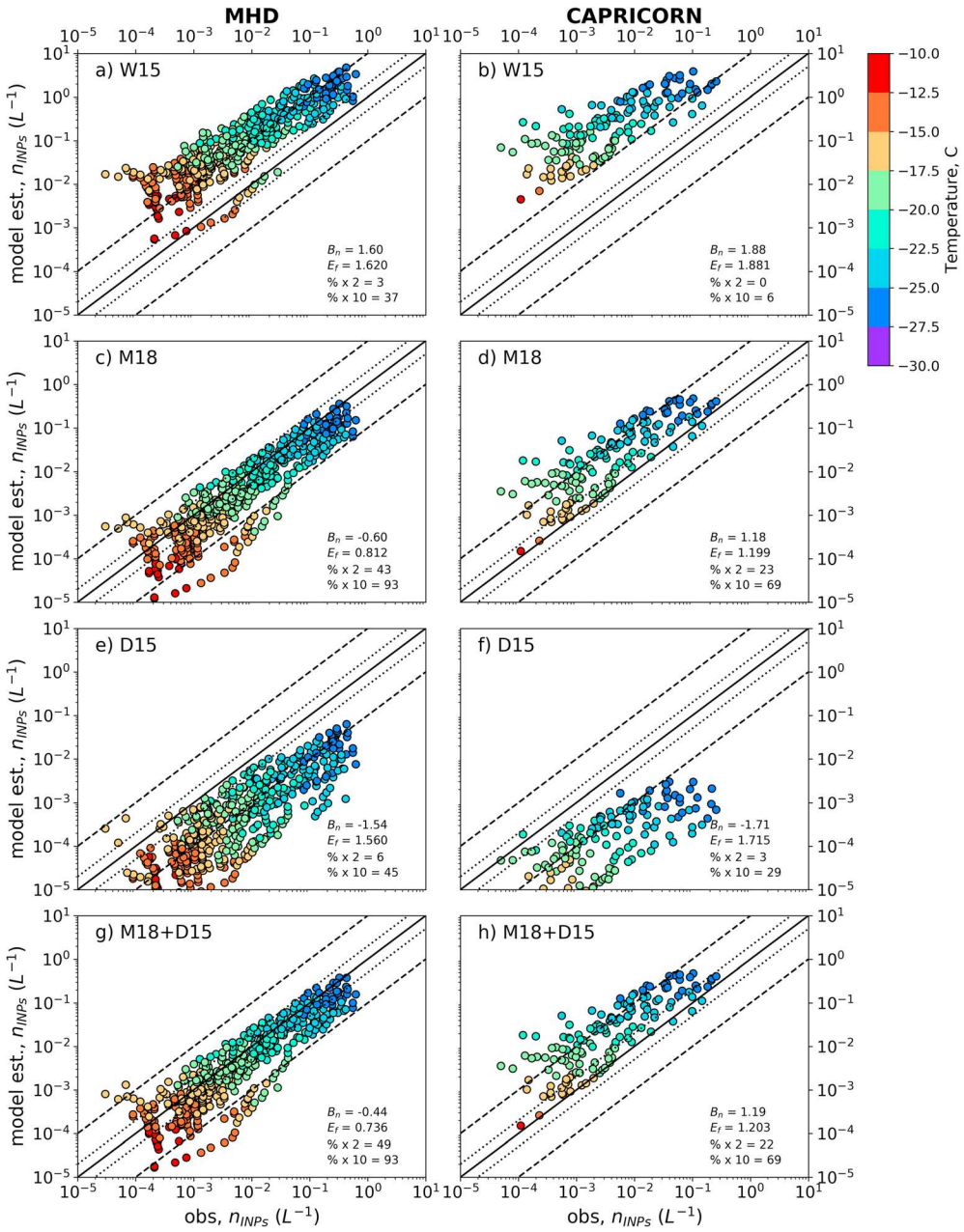

Figure 2. Number concentrations of ice-nucleating particles (n_{INPs}) estimated from simulated marine organic carbon (W15; a and b), sea spray aerosol surface area (M18; c and d), mineral dust (D15; e and f), and combined contributions from sea spray aerosol and mineral dust (M18 + D15; g and h) compared to observed n_{INPs} at Mace Head Research Station (MHD; left column) and Clouds, Aerosols, Precipitation, Radiation, and atmospherIc Composition Over the southeRN ocean campaign (CAPRICORN; right column). The marker color indicates the ice nucleation temperature. In each panel, the solid line correspond to the 1:1 line and the modified normalized mean bias (B_n), fractional gross error (E_f), and percentage of total points within a factor of 2 (dotted lines) and a factor of 10 (dashed lines) are listed.

($B_n = 1.18$). n_{INPs} predicted from D15 were significantly lower than observed n_{INPs} ($B_n = -1.71$) across all temperatures. Predictive skill was best at warmer temperatures, but the number of available observations is limited ($N = 2$) for temperatures warmer than −15 °C due to low n_{INPs} (below detection limit), and thus, the model bias is likely underestimated by this metric. A previous evaluation of simulated surface level dust concentrations against observations indicates that dust concentrations are underpredicted at southern high latitudes (Huneeus et al., 2011), potentially leading to an underestimation of simulated

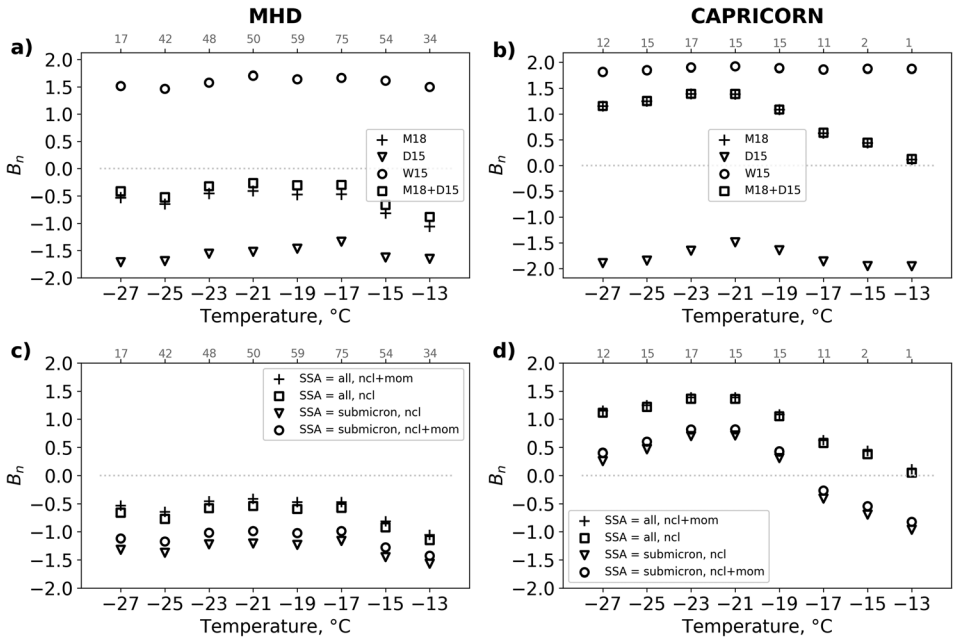

Figure 3. (a and b) Modified normalized mean bias (B_n) of n_{INPs} estimated using the M18 (crosses), D15 (triangles), W15 (circles), and M18+D15 (squares) compared to observed n_{INPs} at Mace Head Research Station (MHD; a) and Clouds, Aerosols, Precipitation, Radiation, and atmospherIc Composition Over the southeRN ocean campaign (CAPRICORN; b). (c and d) B_n calculated for n_{INPs} estimated with M18 + D15 compared to observed n_{INPs} at MHD (c) and CAPRICORN (d) using different portions of the modeled sea spray aerosol (SSA): (crosses) all modes of sea salt (ncl) and marine organic matter (mom); (squares) all modes of only ncl, (circles) submicron modes (i.e., Aitken and accumulation modes) of ncl and mom; and (triangles) submicron modes of only ncl. B_n was calculated for 2° temperature bins; the number of model-observation pairs within each temperature bin is provided along top axis.

dust INPs. However, McCluskey, Hill, Humphries, et al. (2018) noted that low atmospheric radon concentrations indicated little to no terrestrial influence during the CAPRICORN measurements.

For both studies, W15 overestimates n_{INPs} associated with marine sources, and uncertainty in the OM/OC ratio does not explain the bias (Figure S3). W15 was derived from SML samples and their corresponding concentrations of INEs per unit of organic carbon mass. Physiochemical selectivity in the transfer of INEs and organic carbon into the aerosol phase (e.g., Aller et al., 2017; Wang et al., 2017) was not accounted for in W15. While M18 has no explicit dependence on the organic carbon content of SSA, the parameterization was empirically derived from observations of ambient SSA, and thus, the effects of chemically selective emission and atmospheric processing are implicitly accounted for in M18. When combining contributions from both SSA and mineral dust (M18 + D15; Figures 2g and 2h), n_{INPs} were underpredicted ($B_n = -0.44$) at MHD and overpredicted ($B_n = 1.19$) at CAPRICORN. The high bias in model-estimated n_{INPs} for CAPRICORN is discussed more in section 3.3. Regardless of these limitations, we have demonstrated that parameterizations for SSA (M18) and mineral dust (D15) applied to simulated aerosol can estimate n_{INPs} within a factor of 10 of recently observed n_{INPs} from the Southern Ocean region that have not been previously available. Consistent with previous modeling studies guided by historical observations (e.g., Burrows et al., 2013), we find that SSA is often the dominant INP source in both remote marine environments.

3.3. Influence of Simulated Aerosol Size and Composition on INP Populations

Both the amount and the composition of organic components included in the SSA are a strong function of particles size (e.g., O'Dowd et al., 2004; Wang et al., 2017). The INP efficiency of SSA may be sensitive to the contributions of organic molecules (e.g., lipids) that OCEANFILMS aims to simulate. Direct measurements of the size-dependent organic fraction of SSA were not available during these studies. Here, we test the sensitivity of simulated marine INP populations to simulated SSA size (submicron vs. coarse mode) and OCEANFILMS-predicted organic mass fraction.

M18-estimated n_{INPs} were recalculated using the aerosol surface area from (1) all sizes of sea salt and marine organic matter, (2) all sizes of sea salt, (3) submicron sea salt and marine organic matter, and (4) submicron sea salt, shown in Figures 3c and 3d. Timelines of these simulated SSA surface area calculations show that marine organic matter contributes more to the simulated SSA surface area at MHD compared to CAPRICORN (Figure S4). Lowest B_n is achieved for MHD by including sea salt and organic components in all aerosol modes. B_n was minimized for CAPRICORN when only submicron aerosols are considered, where B_n was improved from 1.19 to 0.54 and 93% of model-estimated n_{INPs} were within a factor of 10 of observed n_{INPs} (Figure S5). Since M18 was developed from MHD observations, discrepancies between the MHD and CAPRICORN may point to differences in the detailed biological state and wind speeds, which may impact the organic compositions of SSA during the two campaigns. Additionally, simulated coarse mode sea salt contributes a greater fraction of simulated sea salt surface area during CAPRICORN compared to MHD (Figure S6). These results suggest n_{INPs} are sensitive to how these aerosol quantities are represented and future studies are needed to better quantify SSA size distributions and detailed organic composition.

3.4. Vertical Distribution and Seasonality of INPs

While surface-level measurements were used to validate model-estimated n_{INPs}, knowledge regarding the abundance of n_{INPs} at cloud-relevant altitudes is needed. To investigate the simulated seasonal variations in INP populations and the contributions from marine and dust aerosol, the M18 + D15 approach for predicting n_{INPs} was applied to vertical profiles of aerosol quantities for both locations. For CAPRICORN, coarse mode sea salt was excluded from the M18 calculation for the best estimate of n_{INPs} (see section 3.3). The n_{INPs} vertical profiles, shown in Figure 4, were constructed using simulated aerosol properties, which are sensitive to model uncertainties and errors including those associated with turbulence, vertical mixing, and ventilation of the boundary layer, and removal by clouds and precipitation, which we have not attempted to quantify here. A previous evaluation of simulated dust concentrations against observations indicates that modeled surface dust concentrations near MHD were reasonable but are underpredicted in southern high latitudes (Huneeus et al., 2011). The following results highlight a need for quantifying and reducing model biases in modeled aerosol throughout the atmosphere. We note that a summer maximum in marine organic aerosol is known to occur due to biological processes (e.g., Ovadnevaite et al., 2014); this process is parameterized by OCEANFILMS, but its influence on n_{INPs} is not represented in M18.

During all seasons at MHD, daily mean variation in $n_{INPs,-25}$ spanned 2 or more orders of magnitude at all altitudes (Figure 4). A minimum in $n_{INPs,-25}$ (10^{-3} L^{-1}) was simulated for Winter months (December–February), though $n_{INPs,-25}$ up to 10 L^{-1} occurred, associated with a dust event. Below approximately 2 km, SSA is the main contributor to the simulated $n_{INPs,-25}$ population, and above approximately 2 km, the model-estimated $n_{INPs,-25}$ populations were dominated by mineral dust likely transported from distant sources. The role of mineral dust in the mixed-phase cloud regime is maximized during summer and minimized in winter. If we extrapolate D15 to warmer temperatures (e.g., −15 °C; Figure S8), dust dominates the INP population even in the boundary layer during spring months. Based on these model estimates, while SSA is important below 2 km, transported mineral dust may play a critical role in determining the INP populations in the mixed-phase cloud regime at MHD, and this impact should be constrained in future work.

During all seasons at CAPRICORN, n_{INPs} within the boundary layer are dominated by INPs from SSA, and this is consistent across all temperatures explored in this study (Figures 4, S8, and S9). n_{INPs} are less variable at a given altitude in this region compared to MHD and sporadic days with elevated dust lead to increases in n_{INPs} throughout the entire vertical profile. The role of mineral dust at higher altitudes maximizes in spring and summer months (Figures 4f and 4g) when mobilization and long-range transport of dust from Argentina, Australia, and South Africa (Prospero et al., 2002) is most prevalent, though n_{INPs} simulated for the mixed-phase cloud regime in the CAPRICORN region are lower compared to MHD. INPs associated with SSA dominate the $n_{INPs,-25}$ population up to 3–5 km throughout the year, with a wintertime (June–August) maximum, similar to MHD. Mace and Protat (2018) reported a cloud cover of 76% during CAPRICORN, 65% of which had cloud tops below 4 km. These simulated vertical distributions of $n_{INPs,-25}$ suggest that SSA is an important contributor to primary ice nucleation in Southern Ocean mixed-phase clouds.

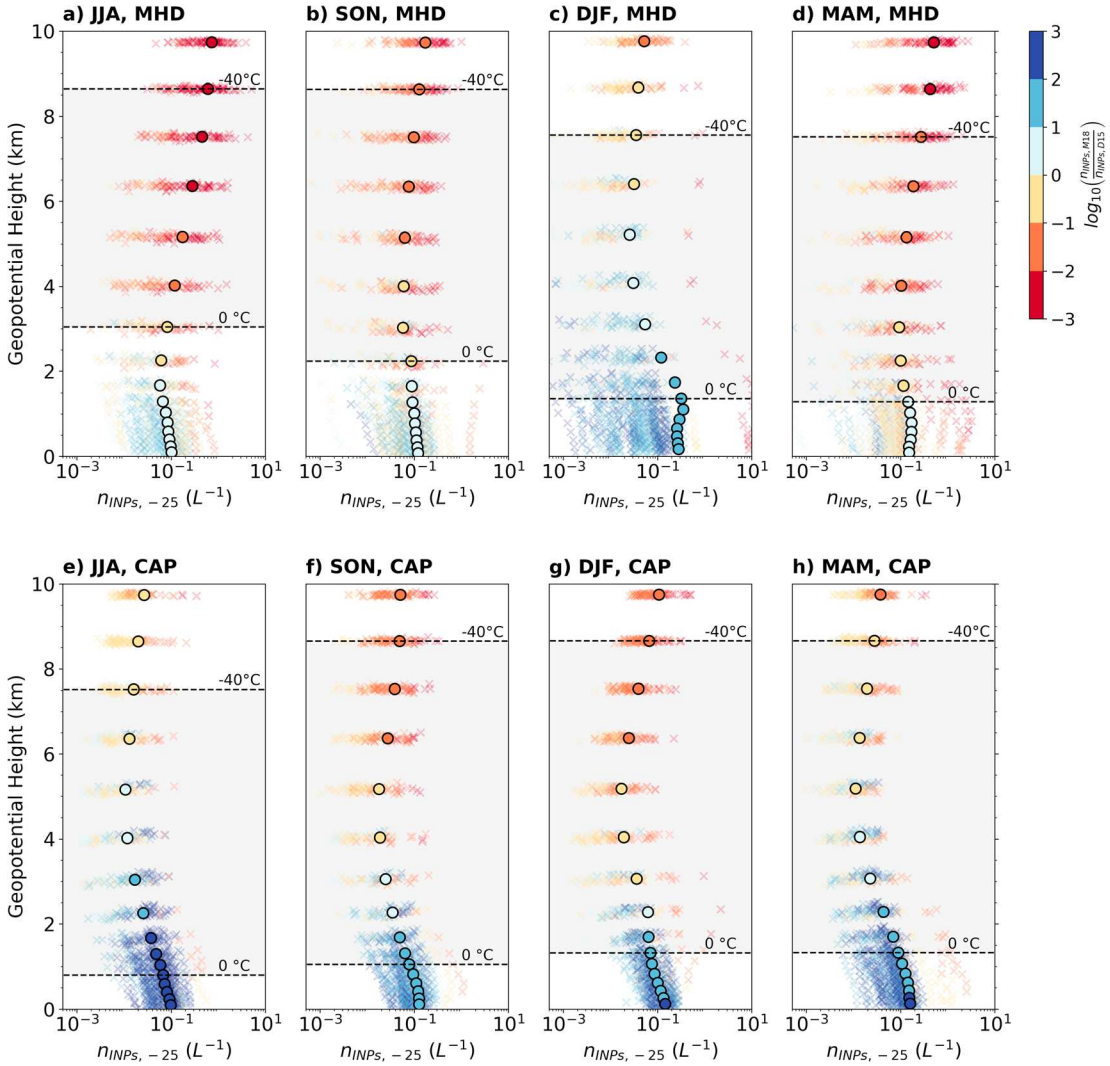

Figure 4. Seasonal (circles) and daily (crosses) mean vertical distributions of model-predicted (M18 + D15) n_{INPs} active at −25 °C for Mace Head Research Station (MHD; a–d) and Clouds, Aerosols, Precipitation, Radiation, and atmospherIc Composition Over the southeRN ocean campaign (CAPRICORN; e–h) locations for Northern Hemisphere summer (July–August [JJA]), fall (September–November [SON]), winter (December–February [DJF]), and spring (March–May [MAM]) months. Marker colors correspond to the $\log_{10}$ of M18-estimated n_{INPs} divided by D15-estimated n_{INPs}. Blue (red) shades indicate a larger contribution of sea spray aerosol (mineral dust). Shaded gray regions highlight the seasonally averaged altitude range corresponding to the mixed-phase cloud temperature range.

4. Conclusions

The CAM5 with constrained meteorology was used to simulate surface-level time-matched aerosol mass and surface area concentrations for MHD and CAPRICORN studies. Simulated aerosol quantities were used as input for three immersion freezing INP parameterizations to estimated n_{INPs}. The best model-observation closure was achieved when combining contributions estimated from SSA surface area (M18) and number concentrations of dust particles larger than 500 nm (D15). The M18 + D15 approach was biased on average 44% lower ($B_n = -0.44$) than observations at MHD. One possible explanation for this discrepancy is that an INP source was unaccounted for at this location, such as organic soil particles (McCluskey, Ovadnevaite, Rinaldi, et al., 2018; O'Sullivan et al., 2018). For CAPRICORN, M18 + D15 was biased high by an average of 119%, possibly due to large contributions of coarse mode sea salt, which contains little organic matter and few INEs, thereby increasing the SSA surface area without increasing ice nucleation active material. The bias reduced to 54% ($B_n = 0.54$) when only Aitken and accumulation mode SSA were used to calculate the SSA surface area. While more effort will be required to understand how INP efficiency of SSA varies with size-dependent aerosol composition, SSA was demonstrated to be the dominant contributor to the surface-

level modeled INP population during both studies, and model-estimated n_{INPs} were within a factor of 10 of observed n_{INPs} at MHD and CAPRICORN 93% and 69% of the time, respectively. Simulated vertical profiles of n_{INPs} using the M18+D15 approach revealed that SSA may be the dominant contributor to primary ice nucleation, especially over the Southern Ocean. These results suggest that INPs associated with SSA should be considered in numerical representations of remote mixed-phase clouds. Long-range transport of mineral dust may be critical to mixed-phase cloud level INP populations in these two locations, and observational and modeling studies regarding vertical and seasonal variability in n_{INPs} and aerosol composition over remote regions are needed. Regardless, this study illustrates a new numerical capability for predicting n_{INPs} that can be used for exploring mixed-phase aerosol-cloud interactions in remote regions.

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