Predicting global atmospheric ice nuclei distributions and their impacts on climate

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Knowledge of cloud and precipitation formation processes remains incomplete, yet global precipitation is predominantly produced by clouds containing the ice phase. Ice first forms in clouds warmer than −36 °C on particles termed ice nuclei. We combine observations from field studies over a 14-year period, from a variety of locations around the globe, to show that the concentrations of ice nuclei active in mixed-phase cloud conditions can be related to temperature and the number concentrations of particles larger than 0.5 μm in diameter. This new relationship reduces unexplained variability in ice nuclei concentrations at a given temperature from ∼10^3 to less than a factor of 10, with the remaining variability apparently due to variations in aerosol chemical composition or other factors. When implemented in a global climate model, the new parameterization strongly alters cloud liquid and ice water distributions compared to the simple, temperature-only parameterizations currently widely used. The revised treatment indicates a global net cloud radiative forcing increase of ∼1 W m^{-2} for each order of magnitude increase in ice nuclei concentrations, demonstrating the strong sensitivity of climate simulations to assumptions regarding the initiation of cloud glaciation.

aerosol indirect effects | climate forcing | ice nucleation

The formation of ice in clouds is of vital importance to life on Earth, as ice formation is one of the key processes initiating precipitation. In addition, since ice nucleation is tied to the action of specific aerosol particles, natural and human impacts on ice nucleation in the atmosphere can lead to alteration of the energy and hydrological cycles (1). Ice nucleation in clouds occurs via two primary pathways: homogeneous freezing of liquid particles below about −36 °C and heterogeneous ice nucleation, triggered by "ice nuclei" that possess surface properties favorable to lowering the energy barrier to crystallization. Once ice is formed, some circumstances may favor generation of ice from preexisting ice particles, or secondary ice formation (2). Heterogeneous ice nucleation remains an enigmatic topic involving multiple mechanistic processes (3) that sometimes defy ready investigation or description. Despite the lack of a complete understanding of heterogeneous ice formation processes, a variety of techniques have been developed and used to detect the presence of and quantify the number concentrations of atmospheric ice nuclei as a function of temperature (4). These measurements show that, although generally representing only 1 in 10^5 of ambient particles in the free troposphere (5), ice nuclei (IN) can nevertheless exert an influence on cold cloud microphysical processes that is disproportionate to their low number concentrations. For example, the concentrations of IN needed to explain observed precipitation rates range from as small as 10^{-3} per standard liter at −10 °C (6) to more typical estimates of a few IN per standard liter at −20 °C (7).

The sensitivity of precipitation initiation from, and the climate forcing of, tropospheric clouds that include regions with temperatures below the freezing level to the abundance of IN has led to the proposal of an "ice indirect effect" of aerosols on climate (8). While envisioned originally for the aerosol perturbation due to anthropogenic emissions alone, we will refer to the ice indirect effect as the impact of any scenario of altered formation and emission of ice nucleating aerosols from combustion, mechanical or biological processes following changes triggered by human activities, or due to atmospheric temperature changes. This is depicted in Fig. 1. More IN in midlevel clouds ($-5 > T_{cloud} > -30$ °C) lead to increased ice crystal concentrations that grow at the expense of liquid water. This is believed to lead to more precipitation, but a shorter cloud lifetime. Increased IN are thereby depicted to reduce the net cooling impact of such clouds on the planet and vice versa (8). Nevertheless, we must note that the sign and magnitude of the impact remains in question, because cloud modeling studies find that IN increases may lead as well to precipitation decreases, and their impact cannot necessarily be isolated from impacts of increases in all types of aerosols on the warm phase (>0 °C) regions of clouds (9), nor the dynamical context in which clouds form (10). The sign and magnitude of the effects of increased IN on high (cirrus) clouds is similarly uncertain, but the impact is likely to be a decrease in the net planetary warming of this cloud type. While counterintuitive on first glance, and strongly dependent on the dynamical forcing, the lowering of ice crystal concentrations in cirrus ice clouds is predicted to occur under increased IN influence due to earlier-onset ice formation that strongly competes with (and may even prevent) the ubiquitous homogeneous freezing process that occurs nearly spontaneously over a narrow and higher relative humidity range on more abundant populations of water-bearing aerosol particles such as sulfates at temperatures below about −36 °C (11). Cirrus may form more readily, and be more widespread under such IN influence, but the lower ice concentrations lead to sedimentation and thus shorter lifetime. Converse changes can be expected if IN decreases occur. Errors in representing IN in numerical models can thus also be expected to propagate into predicted differences in cloud microphysical properties and cloud forcing of climate.

Studies of the magnitude of climate forcing exerted by IN have relied on using either one of a few existing, highly simplified parameterizations of IN number concentrations or have developed speculative relationships between modeled particle types and IN. As examples of the former, the widely used parameterizations of Fletcher (7) and Meyers et al. (12) relate IN number concen-

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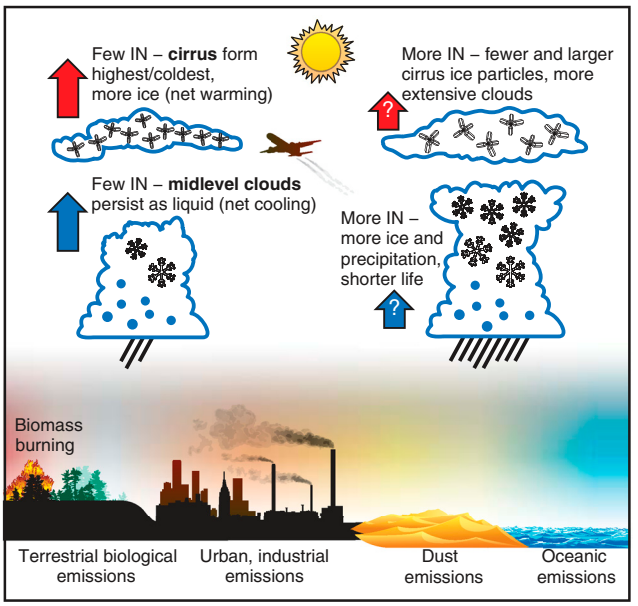

Fig. 1. Schematic diagram of the effect of ice nuclei from various possible aerosol sources on midlevel precipitating clouds and cirrus ice clouds. The likely but uncertain change in the magnitude of the general cooling impact (blue arrows) of midlevel clouds and warming impact (red arrows) of high cirrus clouds in response to increases in the relative number concentrations of IN is indicated (see text for further description).

trations solely to ambient temperature and ice saturation ratio, respectively. Although these parameterizations were based on observations available at the time, they ignore the temporal/spatial variability of IN as these may relate to total ambient particle number concentrations. As an example of the second approach, some studies (13) have used laboratory observations of IN activity of specific particle types to extrapolate to IN concentrations in the atmosphere. This method can lead to unrealistically high predicted IN number concentrations unless the model predictions are constrained by observational data (14). Indeed, IN number concentrations at a single temperature, but in different locations and time periods, have been observed to span more than 3 orders of magnitude, and thus it is difficult to know how to choose an appropriate constraint from such observations. IN concentrations applied in cloud and climate models must be accurate to within at least a factor of 10, as excursions larger than these lead already to significant differences in cloud microphysical and radiative properties, including precipitation efficiency (15).

Given recent attention to the role of the ice phase in climate and the hydrological cycle, it is evident that an improved model description of IN number concentrations, and of how these respond to anthropogenically and naturally induced changes in the ambient aerosol, is urgently needed. Here we propose a remedy to the current lack of an observationally based, yet simple, parameterization of IN number concentrations. We combine data from nine field studies occurring at a variety of locations over 14 years. We further show, using simultaneous measurements of total ambient aerosol size distributions, that a correlation exists between observed IN concentrations and the number concentrations of particles larger than 0.5-μm diameter that reduces the spread of potential errors in predicting IN concentrations at a given temperature from a factor of ~1,000 to ~10. This improvement leads to significantly more realistic and well-constrained descriptions of aerosol-ice formation relationships on relatively small time and spatial scales.

Results

Parameterization of IN Data. Ice nuclei and total ambient aerosol data were collected in nine separate studies in locations as distinct as the Arctic and the Amazon Basin. The majority of the data were collected via aircraft measurements at or near cloud levels. We focus on measurements applicable to mixed-phase cloud conditions for which liquid cloud droplets are sustained against consumption by nucleating and growing ice crystals, a critical step in the formation of precipitation (16). We therefore isolate IN concentrations measured at relative humidity with respect to water exceeding 100%, where contributions from all known heterogeneous ice nucleation mechanisms are possible, and at temperatures between −9 and −35 °C. IN data were included only if ambient aerosol size distributions were available over the same period. Other conditional aspects of sampling are described in *Methods*.

Ice nuclei number concentrations as a function of temperature are shown in Fig. 2 for all sampling periods. These data demonstrate the large variability in IN concentrations at a single temperature and emphasize the small subset of total atmospheric aerosol concentrations that IN represent. For example, total aerosol concentrations range from <100 cm^{-3} in some remote regions to tens of thousands cm^{-3} in urban locations and regions of new particle formation, compared with an upper limit of ~500 L^{-1} for IN. We also show in Fig. 2 the inverse dependence of IN on temperature associated with three previously proposed parameterizations that do not link IN to aerosol properties, as well as an exponential fit vs. temperature alone for the compiled dataset. These highlight the wide disagreement in predicted IN concentrations, particularly at temperatures warmer than −25 °C, despite all of these relationships being observationally based. The curve of Cooper (17) was developed using observations of average ice crystal concentrations measured in clouds for which secondary ice formation processes were determined not to be active. An examination of the data used to develop that relationship shows that the IN number concentrations at any temperature encompassed 3 orders of magnitude. Similar variability exists in our data, as well as in the datasets used to develop the other curves shown.

Variability in IN concentration at any temperature $T(n_{\mathrm{IN},T})$ must relate to spatiotemporal changes in IN source particle types, and it seems possible that these changes may mirror changes in distinct physicochemical categories of the overall atmospheric aerosol population.

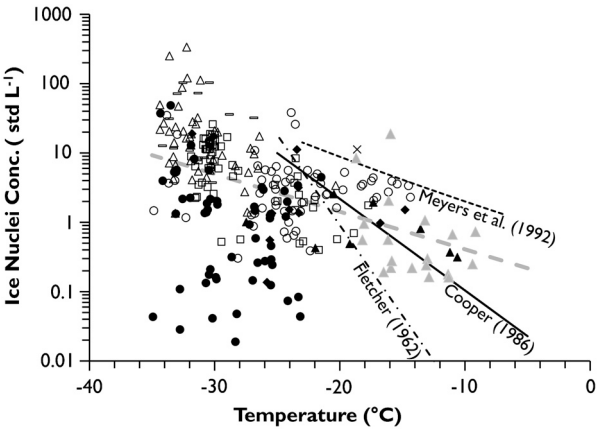

Fig. 2. IN number concentration (at STP) active at water saturation or above vs. temperature. Projects (see *SI Text*) are WISP-94 (gray triangle), Alliance Icing Research Study—2 (X), AMAZE-08 (square), Cloud Layer Experiment-10/Canadian Cloudsat/CALIPSO Validation Project (open circle), Ice in Clouds Experiment—Layer Clouds (solid circle), Ice Nuclei SPECTroscopy-1 (–), Ice Nuclei SPECTroscopy-2 (diamond), Mixed-Phase Arctic Cloud Experiment (black triangle), and Pacific Dust Experiment (open triangle). Parameterizations described in the text are labeled and are plotted over the experimental measurement range on which they were based. The dashed gray line is a T-dependent fit to all data [$0.117\exp(-0.125^{*}(T_K - 273.2))$; $r^2 = 0.2$].

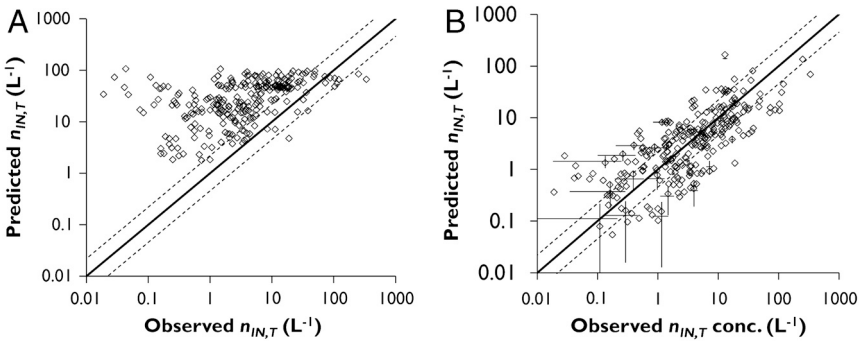

Fig. 3. Comparison of predicted vs. observed IN concentrations. In *A*, the points correspond to the Meyers et al. (12) relation calculated at water saturation. Other parameterizations are compared in the SI. In *B*, predictions are based on the parameterization put forth in this paper that depends on both $n_{aer,0.5}$ and *T*. Uncertainties (1 standard deviation) are shown on selected data points in *B*. Dotted lines outline a range of a factor of 2 about the 1:1 line (solid line) in both panels.

In Fig. 3 we demonstrate the improvement in predictive ability of a parameterization of $n_{IN,T}$ that links IN number concentrations and total aerosol number concentrations exceeding a selected diameter d in microns ($n_{aer,d}$), in this case $d = 0.5$. This parameterization approach (Fig. 3*B* and Fig. S1) predicts 62% of the observed points within a factor of 2 and aligns the overall dataset along the 1:1 predictive line, in strong contrast to approaches that consider temperature dependence alone (Fig. 3*A*). The Meyers et al. (12) relation used as the temperature-dependent example here predicts only 16% of the observed points within a factor of 2 and is biased above the 1:1 line. The predictions of other temperature-dependent IN relations are shown in *SI Text* (Fig. S2). Based on our prior studies quantifying the compositions of the detected IN (18–21), we expect that the remaining unaccounted-for variability can be attributed to variations in chemical composition that are not captured by this parameterization. Nevertheless, a greatly improved degree of constraint on IN number concentrations is achieved, which is required to model the initiation of ice formation and to thereby represent the main microphysical features of liquid and ice phase distributions in low- and midlevel supercooled clouds in global climate model simulations.

Global Climate Simulations. We focus here on the key differences in annually and zonally averaged fields from two global climate simulations, one using the Meyers et al. (12) parameterization for IN, and the other using the newly proposed relationship. Differences in the two simulations should reflect the influence of the improved IN measurement database used and give inferences to the sensitivity of climate to the magnitude of IN change represented via incorporating dependence on aerosols.

The simulation using the IN parameterization developed herein predicts large increases (10–30 $g\,m^{-2}$) in annually averaged liquid water path (Fig. S3*A*), and stronger (5–10 $W\,m^{-2}$ more negative) shortwave cloud forcing at storm track latitudes and at higher latitudes (Fig. S3*B*) that is not completely balanced out by positive changes in longwave cloud forcing, compared to simulations with the Meyers et al. parameterization (Table 1). These changes are a consequence of reduced IN number concentrations in regions with low predicted number concentrations of non-sea-salt particles larger than 0.5-μm diameter; the lower IN concentrations inhibit liquid conversion to ice in mixed-phase clouds in these regions (i.e., the left-hand scenario for midlevel clouds in Fig. 1). Consequently, cloud cover increases of 5%–10% at high latitudes, and reduction in the annual zonal mean downwelling shortwave radiation reaching the surface also occur.

Although uncertainties in the observations listed in Table 1 are not well known and could be quite large, we note that the global net values of all cloud and radiative quantities are moved closer to the average observational values with the developed parameterization (Table 1). We thus conclude that the parameterization is a significant improvement in the representation of aerosol impacts on cold clouds, especially mixed-phase clouds at mid to high latitudes. A possible drawback of the proposed parameterization is that it requires specified or prognosed fields of aerosol number concentrations for particles with diameters larger than 0.5 μm; however, many current models now include these fields, and the addition of prognostic aerosols and their coupling to clouds is recognized as a requirement for future climate models (1).

Discussion

The apparent relation of ice nuclei number concentrations to the number concentrations of certain subsets of the atmospheric aerosol has been recognized for more than 40 years (20, 22–24). All of these studies noted either linear or power law relations between ice nuclei and aerosol number concentrations within restricted size ranges, generally those particles larger than 0.3-μm diameter. Recent measurements suggest that such relations are stronger for a lower size limit of 0.5 μm, but are inconsistent when the lower size limit is 0.1 μm (24, 25). Reasons for this relate to the average size of IN and their typical compositional sources. For example, the mode diameter of particles found at the centers

Table 1. Global annual mean values

	LWP, $g\,m^{-2}$	IWP, $g\,m^{-2}$	SWCF, $W\,m^{-2}$	LWCF, $W\,m^{-2}$	FSDS, $W\,m^{-2}$	CLDTOT, %
Meyers et al.	40.5	5.6	−49.5	19.2	186.4	55.8
This study	46.0	4.5	−51.8	20.2	183.5	57.2
OBS (ref)	48 (40) (SSM/I, ocean)		−54.2*(ERBE)	30.4*(ERBE)		66.8* (ISCCP D2)
	79 (41) (SSM/I, ocean)		−47.1*(CERES2)	29.9*(CERES2)		

Liquid water path (LWP), ice water path (IWP), shortwave cloud forcing (SWCF), longwave cloud forcing (LWCF), surface downwelling shortwave radiation (FSDS), and cloud coverage (CLDTOT) in CAM3 simulations using the different IN parameterizations and based on satellite observations (OBS).

*Satellite retrieved observations are from the NCAR CAM-3 database of global analyses results (http://www.cgd.ucar.edu/ccr/CPT/climatology/observations.html) maintained by the Climate Process Team. Abbreviations stand for the Earth Radiation Budget Experiment (ERBE), the Clouds and the Earth's Radiant Energy System compilation 2 (CERES2; 2000–2005), the International Satellite Cloud Climatology Project D level 2 dataset (ISCCP D2), and the Special Sensor Microwave/Imager (SSM/I).

of single ice crystals has been reported to be ~0.5 μm (26). We have measured a similar mode size for IN collected and analyzed in many of the same studies represented in Fig. 2 (24). Furthermore, the predominant role of mineral dust particles as IN (20) and apparent ice crystal nuclei (27) suggests an expected influence of particles with diameters larger than 0.5 μm, a size range for which mineral and soil dusts often dominate in the free troposphere (28). Biological aerosol particles occur at all atmospheric aerosol sizes, but some of the best-known biological organisms possessing ice nucleating surface structures, such as bacteria, are one to a few microns in overall size (29). Finally, while black carbon can be present in the atmospheric aerosol at sizes as small as 0.05 μm, the theory of active sites for ice nucleation suggests that sizes >0.1 μm are usually required for soot, or in fact any particle type to act as IN (30). We thus conclude that there are sound, physically based reasons for the observed correspondence between IN concentrations and the total number concentrations of particles larger than 0.5 μm in our datasets. However, one important exception is sea-salt aerosols, which although being present in higher number concentrations at supermicron sizes in the marine boundary layer, are not known to have any role in ice nucleation in the temperature regimes considered here. Therefore, aerosols with a strong marine influence were not included in our parameterization development, nor were sea-salt particles used to drive the parameterization in the global model.

Recently, Phillips et al. (31) introduced a model parameterization framework for ice nucleation that empirically accounts for variations in ice nuclei number concentrations associated with composition, aerosol surface area, relative humidity, and, indirectly, temperature. The parameterization concept is based on the theoretical expectation that ice nucleation rate should scale with particle surface area, and separately evaluates the rates for mineral dust, black carbon, and organic particles, which include bacteria. This approach captures most of the features and details desired for predicting ice nucleation in cloud and climate models, absent explicit treatment of different ice nucleation mechanisms. A number of input parameters are required, and specialized observational data are needed for the evaluation of the various scaling factors used in the parameterization; the factors presently are based on data from just two field studies (31). The validity of this approach has been demonstrated in selected cases, but remains to be validated for the array of aerosol compositional and cloud scenarios in the atmosphere. Further, there remain knowledge gaps in defining the required input parameters and their variability for different compositions. It may be some time before the establishment of an adequate particle-by-particle database linking ice nuclei activation properties (i.e., dependence on thermodynamics) to physical and chemical characteristics of particles. The parameterization proposed here may be regarded as a simplified version of the Phillips et al. (31) scheme that can be readily and immediately applied in many existing cloud and climate models, and in which the number concentrations of particles larger than 0.5-μm diameter are proxies for aerosol surface areas. Although variations in IN with particle composition are neglected in our scheme, the simpler approach captures most of the variability in IN that is large enough to affect the formation and phase of clouds at temperatures down to −35 °C. An additional advantage to this approach is that the state of the science for measurement of ice nuclei number concentration appears ahead of our ability to fully describe or prescribe from a theoretical standpoint the many chemical complexities that can affect the action of specific ice nucleation mechanisms.

Atmospheric Implications. Based on our modeling studies, it appears just as important, for the accurate representation of cloud forcing, to properly simulate the *lack* of available IN as it is to simulate the presence of IN. As is evident from Fig. 3 (and Fig. S2), commonly applied parameterizations of ice initiation generally overestimate IN number concentrations, leading to too-frequent glaciation via ice nucleation processes alone, with subsequent too-frequent initiation of precipitation, and thus reduction of cloud cover. As a result, the contributions of mixed-phase clouds to net radiative forcing are not well represented.

The developed IN parameterization results in a global net cloud forcing change (decrease) of 1.3 $W m^{-2}$ compared to the Meyers et al. (12) scheme that represented the state of knowledge less than 20 years ago. The implied sensitivity of climate forcing is thus ~1 $W m^{-2}$ per decade increase of IN number concentration, calculated from the difference in predicted global IN concentrations between the Meyers et al. formulation and the parameterization proposed in this study, evident in Figs. 2 and 3. The strong sensitivity of climate forcing to IN suggests that long-range import of IN from dust storms, boreal biomass burning, and anthropogenic pollution could lead to feedbacks on mixed-phase clouds, impacting precipitation and other phenomena such as Arctic sea ice loss and bioproductivity (i.e., this responds to photosynthetically active radiation, a component of the shortwave radiation). However, until present-day conditions of ice initiation are better represented, in part through application of more realistic parameterizations of IN such as the one proposed here, it will be impossible to predict with any confidence how future perturbations in atmospheric aerosol concentrations may affect cold cloud formation and properties.

Methods

Datasets. We used ice nuclei and aerosol datasets collected in nine separate studies, as listed in *SI Text*. IN number concentration data are all from the Colorado State University Continuous Flow Diffusion Chamber (CFDC) (32). During aircraft measurements, the CFDC either sampled from an ambient inlet, or in one study from the outlet of a counterflow virtual impactor (CVI) (33) that passed only cloud particle residual aerosols to the CFDC. We selected time periods where the CFDC was measuring IN concentrations at relative humidity with respect to water exceeding 100%, with most data collected at water relative humidity between 101% and 104%. This water-supersaturated condition supports the expression of all heterogeneous ice nucleation processes, including deposition nucleation (vapor to ice on a particle surface), and condensation and immersion freezing (ice formation during or following the condensational growth of liquid droplets, respectively). Because these processing conditions lead to water droplet activation of most particles, including those that do not nucleate ice at the selected temperature, and because residence time is short, the CFDC technique does not directly assess whether particles are active as contact freezing nuclei (ice formation occurring from collision of interstitial IN with droplet surfaces over time in mixed-phase clouds). Although we did not specifically address parameterization of deposition nucleation, studies have shown that deposition nucleation primarily contributes to ice nucleation at temperatures below about −30 °C (31), just a few degrees warmer than the lower temperature limit of the data used here. Thus, we interpret our measured IN number concentrations to reflect the maximum ambient IN number concentrations spontaneously active (31) in mixed-phase clouds at temperatures warmer than −35 °C. Counting statistics determined that IN number concentrations lower than 0.3 L^{-1} in standard inlet sampling and 0.01 L^{-1} from the CVI inlet (the CVI enhances concentrations by >20) were below lower levels of quantification and were removed (see *SI Text*). Lower IN concentration data are also included from the Winter Icing in Storms Project 1994 (WISP-94) study, for which large volume (200 L) grab samples were collected from the aircraft in large conductive bags and then processed in the CFDC in our laboratory. Additional criteria applied to the screening of IN data were that the measurements during a selected sample period occurred within a 500-m-altitude layer and that the CFDC processing temperature and relative humidity over the period did not vary more than 5 °C or 3%, respectively. Cloudy periods were defined on the basis of other microphysical measurements and removed from consideration, except when sampling was specifically made using a CVI.

For a sampling interval to be selected, we also required simultaneous measurements of aerosol size distributions, leading to a total of 290 sampling periods ranging from 5 to 30 min (some WISP-94 periods were longer) for use in our analyses. In various projects, size distributions were measured using mobility, optical, and aerodynamic particle sizing instruments. We did not correct data from optical instruments for varying refractive index. When applicable, aerodynamic diameters were converted to physical diameters assuming an average particle density of 2.3 $g cm^{-3}$, which was

selected as appropriate for the dust particles that dominated the atmospheric aerosol during those periods. Aerosol data were averaged for the same intervals as IN. Both aerosol and IN concentrations were corrected to standard temperature and pressure conditions (STP; 273.15 K, 1013.5 mb). The choice of 0.5-μm diameter as the lower limit for summing number concentrations of "large" particles is a relatively arbitrary one of convenience, selected to limit the influence on derived relationships of high concentrations of non-IN particles in the range 0.1–0.5 μm, while retaining sufficient number concentrations of particles to reference to IN concentrations.

Our instrumental design requires use of an impactor system (32) upstream of the CFDC to limit sampled particles to aerodynamic sizes smaller than about 1.6 μm (50% cut size for a particle of density 1.0 g cm^{-3} at 1 L min^{-1} sample flow). We therefore cannot assess contributions to IN concentrations from the small number concentrations of particles with diameters >1.6 μm present in typical ambient aerosol. The failure to sample the largest particles in the present dataset is a potentially important limitation at the warmest temperatures where errors of a factor of 2 are possible at −15 °C based on the size dependence of IN number concentrations measured at the Earth's surface in a recent study (34). At higher (cloud) altitudes, where large aerosol concentrations are lower, a previous study suggests that supermicron particles contribute more than 50% to total IN number concentrations at warmer than −12 °C (23).

Parameterization. We aggregated our multistudy dataset into 3 °C temperature intervals for fitting the relation between $n_{IN,T}$ and $n_{aer,0.5}$. Pearson correlation coefficients for power law fits were >0.6 (to 0.8) (Fig. S1 and discussion in *SI Text*). Correlations for individual projects over similar temperature ranges were sometimes much higher, reflecting the secondary role of particle composition. The power law coefficients were then used to determine the overall size and temperature dependencies of IN active under mixed-phase cloud conditions, represented as follows:

$$n_{\mathrm{IN},T_k} = a(273.16 - T_k)^b (n_{\mathrm{aer},0.5})^{(c(273.16-T_k)+d)}, \qquad [1]$$

where $a = 0.0000594$, $b = 3.33$, $c = 0.0264$, $d = 0.0033$, T_k is cloud temperature in degrees Kelvin, $n_{aer,0.5}$ is the number concentration (scm^{-3}) of aerosol particles with diameters larger than 0.5 μm, and n_{IN,T_k} is ice nuclei number concentration (std L^{-1}) at T_k. This is the relation used in Fig. 3*B*. Although coefficients may change and could be considered for specific compositional categories given adequate future data, we expect the form of this parameterization to remain valid. A particular area for future evaluation and development is the temperature regime warmer than about −15 °C, where larger aerosols appear most influential as IN, where IN number concentrations are lowest, and where the compositional nature of IN may be more specialized. Our present data imply weaker sensitivity of $n_{IN,T}$ to $n_{eer,0.5}$ in this regime (see *SI Text* and Fig. S1), but we cannot rule out inadequate sampling of the largest particles as a possible source of error in this case.

Model. The National Center for Atmospheric Research (NCAR) Community Atmospheric Model-3 (CAM3) is the atmospheric component of the Community Climate System Model version 3 (CCSM3) (35). We incorporated the ice nucleation parameterization into the CAM3 version including the two-moment scheme of Liu et al. (36) to treat liquid and ice microphysics in CAM3, thereby predicting both mass and number mixing ratio of cloud particles and explicitly treating liquid and ice mass partitioning in mixed-phase clouds to account for the Bergeron–Findeisen process in which ice crystals grow at the expense of liquid drops.

An aerosol module recently implemented in CAM3 (37, 38) was used to predict aerosol mass and number concentrations of three modes (Aitken, accumulation, and coarse modes). This module treats mixtures of sulfate, black carbon, organic, dust, and sea-salt aerosols in a realistic way (i.e., internal mixtures within modes and external mixtures between modes). Thus, aerosol-size distributions can be calculated directly from model-predicted aerosol mass and number concentrations. For the aerosol number calculation used in the IN parameterization developed in this study, sea-salt contributions were excluded since these particles are not represented within our IN analyses and are not believed to be an important source for IN (39).

The CAM3 global simulations were run for a 5-yr period using climatological sea surface temperatures as input in addition to the present-day (Year 2000) aerosol emissions.

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