Sensitivity of the global distribution of cirrus ice crystal concentration to heterogeneous freezing

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[1] This study presents the sensitivity of global ice crystal number concentration, \( N_c \), to the parameterization of heterogeneous ice nuclei (IN). Simulations are carried out with the NASA Global Modeling Initiative chemical and transport model coupled to an analytical ice microphysics parameterization. Heterogeneous freezing is described using nucleation spectra derived from theoretical considerations and empirical data for dust, black carbon, ammonium sulfate, and glassy aerosol as IN precursors. When competition between homogeneous and heterogeneous freezing is considered, global mean \( N_c \) vary by up to a factor of twenty depending on the heterogeneous freezing spectrum used. IN effects on \( N_c \) strongly depend on dust and black carbon concentrations and are strongest under conditions of weak updraft and high temperature. Regardless of the heterogeneous spectrum used, dust is an important contributor of IN over large regions of the Northern Hemisphere. Black carbon however exhibits appreciable effects on \( N_c \) when the freezing fraction is greater than 1%. Compared to in situ observations, \( N_c \) is overpredicted at temperatures below 205 K, even if a fraction of liquid aerosol is allowed to act as glassy IN. Assuming that cirrus formation is forced by weak updraft addressed this overprediction but promoted heterogeneous freezing effects to the point where homogeneous freezing is inhibited for IN concentrations as low as 1 L\(^{-1}\). Chemistry and dynamics must be considered to explain cirrus characteristics at low temperature. Only cloud formation scenarios where competition between homogeneous and heterogeneous freezing is the dominant feature would result in maximum supersaturation levels consistent with observations.


1. Introduction

[2] The role of cirrus clouds in a changing climate system constitutes a major source of uncertainty in anthropogenic climate change assessment and prediction [Baker and Peter, 2008; Cantrell and Heymsfield, 2005; Seinfeld, 1998]. Cirrus clouds form by homogeneous freezing of deliquesced aerosol and heterogeneous freezing of ice nuclei (IN) [Pruppacher and Klett, 1997]. Analysis of ice crystal residues from field campaigns shows that both freezing mechanisms interact during cirrus formation [e.g., DeMott et al., 2003; Haag et al., 2003; Prenni et al., 2007], suggesting that IN can strongly affect cloud ice crystal concentration and size distribution [Barahona and Nenes, 2009a; DeMott et al., 1994; Gierens, 2003; Kärcher and Lohmann, 2003; Spichtinger and Gierens, 2009a, 2009b].

[3] Global modeling studies have shown that heterogeneous IN emissions can impact the global distribution of ice crystal concentration, \( N_c \), resulting in a potentially large climatic effect. Lohmann et al. [2004] performed simulations considering either pure homogeneous or pure heterogeneous freezing (both from sulfate aerosol) as limits of variability induced from heterogeneous IN effects. Compared to homogeneous freezing, heterogeneous freezing resulted in lower \( N_c \), higher precipitation rates, and smaller ice water paths. Hendricks et al. [2005] studied the effect of aircraft emissions of black carbon on \( N_c \), assuming that the freezing mechanism shifted from homogeneous to heterogeneous when the grid cell black carbon concentration, \( bc_{bc} \), exceeded a threshold value (around 0.5 cm\(^{-3}\)); considering IN effects reduced \( N_c \) between 10% and 40% at the midlatitudes of the Northern Hemisphere. A similar approach was used by Lohmann et al. [2008] to study the effect of IN from dust on \( N_c \); it was found that considering IN effects decreased the shortwave cloud forcing associated with cirrus clouds by 2.7 W m\(^{-2}\). Competition between homogeneous and heterogeneous freezing of dust and black carbon IN during cloud formation was considered by Penner et al. [2009] using the parameterizations of Liu and Penner [2005] and Kärcher et al. [2006] to describe ice crystal
production. The aerosol indirect forcing from black carbon IN emissions ranged between ~0.72 and 0.04 W m$^{-2}$.

[4] At atmospherically-relevant conditions, dust, soot, and biogenic particles can act as IN [DeMott et al., 2003; Fridlind et al., 2004; Pratt et al., 2009; Prenni et al., 2009; Uno et al., 2009]. The large seasonal and geographical variability of aerosol and the limited understanding of heterogeneous nucleation challenge the prediction of IN concentrations [Baker and Peter, 2008; Cantrell and Heymsfield, 2005; Lin et al., 2002; Waliser et al., 2009]. Most of the uncertainty in predicting the impact of aerosol emissions on cirrus clouds is associated with estimating the fraction of the aerosol that freezes heterogeneously, i.e., the IN concentration, $N_{IN}$ [Baker and Peter, 2008; Cantrell and Heymsfield, 2005; Lin et al., 2002]. Typically, heterogeneous freezing is treated as an extension of homogeneous nucleation [Kärcher and Lohmann, 2003] or using classical nucleation theory (CNT) [Khvorostyanov and Curry, 2004].

Empirical correlations [e.g., DeMott et al., 1998; Meyers et al., 1992; Phillips et al., 2008] are also heavily used.

[5] Cloud studies have shown that $N_c$ can be very sensitive to the assumptions made in the calculation of $N_{IN}$ [e.g., Barahona and Nenes, 2009b; Eidhammer et al., 2009; Hendricks et al., 2005]. Lin et al. [2002] compared $N_c$ from several parcel models using different parameterizations for $N_{IN}$, leading to 2 orders of magnitude difference amongst models. Monier et al. [2006] used several empirical expressions [DeMott et al., 1997, 1998; Meyers et al., 1992] and CNT methods to describe heterogeneous nucleation within the same cloud parcel model. It was found that a factor of 10 variation in $N_{IN}$ translated into a factor of three difference in calculated $N_c$ from the combined effects of homogeneous and heterogeneous freezing. A similar approach was followed by Eidhammer et al. [2009] who found several orders of magnitude variation in $N_c$ when empirical parameterizations and CNT-derived approaches where used to calculate $N_{IN}$.

[6] The large sensitivity of $N_c$ to $N_{IN}$ reported in parcel model studies has not been reflected in global circulation model (GCM) studies to date. This is largely because cirrus formation parameterizations used in GCM studies [e.g., Kärcher et al., 2006; Liu and Penner, 2005] are highly constrained, considering specific $N_{IN}$ parameterizations with a prescribed dependency on $T$, $p$, $s_r$, and aerosol concentration. This limitation can be relaxed using the Barahona and Nenes [2008, 2009a, 2009b] framework, which predicts $N_c$ using any form of $N_{IN}$ parameterization (theoretical or experimental) and unravel the dependency of $N_c$ on thermodynamic, chemical and dynamic factors that drive cloud formation. In this study, this parameterization is incorporated within the NASA Global Modeling Initiative (GMI) framework [Liu et al., 2005; Rotman et al., 2001] to study the sensitivity of $N_c$ that would form in cirrus clouds using several common parameterizations of $N_{IN}$. The sensitivity of our findings to the meteorological features is also considered, by carrying out simulations using meteorological fields from the NASA Goddard Institute for Space Studies GCM (GISS) and the NASA former Global Data Assimilation Office (DAO) GCMs.

2. Model Description

[7] The Global Modeling Initiative (GMI) is a state-of-the-art modular 3-D chemical and transport model developed for assessment calculations of anthropogenic effects on climate [Liu et al., 2007b; Rotman et al., 2001]. GMI utilizes off-line meteorological fields to calculate advective and convective transport of chemical species, transformation, and removal by wet and dry deposition [Rotman et al., 2001]. Here we use fields derived from the NASA Goddard Institute for Space Studies (GISS) and from the NASA former Data Assimilation Office (DAO) GCMs. Each of the archived data sets spans over 1 year and represents the period from March 1997 to February 1998 archived as 6 h averages. The GISS field has a resolution of 23 vertical levels, from the surface to 0.02 mb [Rind and Lerner, 1996]. DAO has a vertical resolution of 46 vertical levels, which extend from the surface to 0.15 mb. The horizontal resolution in both meteorological fields is 4° × 5°.

[8] The GMI aerosol module is coupled to the GMI-CTM advection core and includes primary emissions, chemical production of sulfate, gravitational sedimentation, dry deposition, wet scavenging, and hygroscopic growth [Liu et al., 2005, 2007b]. Anthropogenic and natural aerosol and precursor emissions include SO$_2$, organic matter, black carbon, oceanic DMS, dust and sea salt. Model prognostic variables include dimethylsulfide, sulfur dioxide, sulfate aerosol, hydrogen peroxide, black carbon (biomass burning and fossil fuel), and organic matter. Mineral dust and sea-salt mass are predicted in four size bins: 0.01–0.63 μm, 0.63–1.26 μm, 1.26–2.5 μm, and 2.5–10 μm [Liu et al., 2007b]. Explicit aerosol microphysics is not considered and aerosol size distributions are assumed to follow those derived from observations (shown in Table 1).

2.1. Ice Crystal Number Concentration Parameterization

[9] The number concentration of ice crystals that would nucleate in cirrus is calculated using the analytical framework of Barahona and Nenes [2008, 2009a, 2009b] referred hereinafter as BN09. Competition between homogeneous and heterogeneous freezing is explicitly considered, while the dependency of $N_c$ on the conditions of cloud formation (i.e., $T$, $p$), updraught velocity, deposition coefficient, and

### Table 1. Dry Number Size Distributions for Sulfate, Dust, and Black Carbon Aerosol

<table>
<thead>
<tr>
<th>Aerosol</th>
<th>Number Fraction</th>
<th>$D_g$ (μm)</th>
<th>$\sigma_g$</th>
<th>Density (g cm$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sulfate</td>
<td>1</td>
<td>0.04</td>
<td>2.3</td>
<td>1.7</td>
</tr>
<tr>
<td>Dust</td>
<td>0.152</td>
<td>0.02</td>
<td>2.3</td>
<td>2.6</td>
</tr>
<tr>
<td></td>
<td>0.727</td>
<td>0.09</td>
<td>1.6</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.121</td>
<td>0.55</td>
<td>2.5</td>
<td></td>
</tr>
<tr>
<td>Black carbon (Fuel and biomass burning)</td>
<td>1</td>
<td>0.14</td>
<td>1.5</td>
<td>1.5</td>
</tr>
</tbody>
</table>

$D_g$ and $\sigma_g$ are the geometric mean diameter and standard deviation, respectively [Lin et al., 2002; Liu et al., 2007b; Penner et al., 2009].
soluble and insoluble aerosol concentrations is resolved. \( N_c \) is given by

\[
N_c = \begin{cases} 
N_{\text{hom}} + N_{\text{het}}(s_{\text{hom}}) & ; N_{\text{het}}(s_{\text{hom}}) < N_{\text{lim}} \\
N_{\text{het}}(s_{\text{max}}) & ; N_{\text{het}}(s_{\text{hom}}) \geq N_{\text{lim}},
\end{cases}
\]

where \( s_{\text{max}} \) is the maximum supersaturation that develops in the cirrus, \( s_{\text{hom}} \) is the homogeneous freezing threshold [Koop et al., 2000], and \( N_{\text{hom}} \) and \( N_{\text{het}} \) are the number of ice crystals forming from homogeneous and heterogeneous freezing, respectively. \( N_{\text{lim}} \) is the IN concentration that completely inhibits homogenous nucleation (calculated below) and sets the limit between combined heterogeneous-homogeneous freezing and pure heterogeneous freezing only. \( N_{\text{lim}} \) is given by

\[
N_{\text{lim}} = \frac{N^*}{\sqrt{\Delta^*_{\text{char}} \left| \frac{d ln N_{\text{het}}}{d s_{\text{hom}}} \right|}} \left( 1 + s_{\text{hom}} \right)^{\frac{1}{\nu_{\text{lim}}}},
\]

where \( N^* \) and \( \lambda \) are functions of cloud formation conditions (i.e., \( T, \rho, V, \) and deposition coefficient) defined in the notation section, and

\[
\Delta^*_{\text{char}} = \Delta_{\text{char}} \left[ \frac{4}{3} \Delta_{\text{char}} + 2(s_{\text{ch}} - \Delta_{\text{char}}) \right],
\]

where \( \Delta_{\text{char}} = \min \left( \frac{d ln N_{\text{het}}(s_{\text{ch}})}{d s_{\text{hom}}} \right)^{-1}, s_{\text{ch}} \) is a characteristic of the IN population related to the slope of the IN spectrum at supersaturation \( s_{\text{ch}} \).

[10] For the special case of a monodisperse IN population with freezing threshold \( s_{\text{hom,mono}} \), equation (2) simplifies to [Barahona and Nenes, 2009a],

\[
N_{\text{lim}} = \frac{2 \alpha V}{\beta \rho_i \pi} \left( s_{\text{hom}} + 1 \right) \left( \frac{\Gamma_1 D_{\text{lim}} + \Gamma_2}{D_{\text{lim}}} \right),
\]

where \( D_{\text{lim}} = -\gamma + \sqrt{\gamma^2 + \frac{2}{\Gamma_1 \alpha V \Delta_{\text{char}}} \left| \frac{d ln N_{\text{het}}}{d s_{\text{hom}}} \right|} \), \( \gamma = \frac{\Gamma_2}{\Gamma_1} \), and \( \Delta_{\text{char}} = s_{\text{ch}} - s_{\text{hom,mono}} \).

[11] For \( N_{\text{het}}(s_{\text{hom}}) < N_{\text{lim}} \), \( s_{\text{max}} \) is equal to the homogeneous freezing threshold \( \frac{N_{\text{lim}} N_{\text{hom}}}{N_{\text{het}}(s_{\text{max}})} \). For \( N_{\text{het}}(s_{\text{hom}}) \geq N_{\text{lim}} \), \( s_{\text{max}} \) is below \( N_{\text{hom}} \) and heterogeneous freezing does not occur (\( s_{\text{max}} \) for this case is calculated below). For \( s_{\text{max}} = N_{\text{hom}} \), it is assumed that only heterogeneous freezing takes place, which however does not introduce substantial error in \( N_c \) [Barahona and Nenes, 2009a]. The heterogeneous contribution to \( N_c \) is equal to \( N_{\text{het}}(s_{\text{max}}) \) when only heterogeneous freezing is active and equal to \( N_{\text{het}}(s_{\text{hom}}) \) when homogeneous and heterogeneous freezing are both active. The functional form of \( N_{\text{het}}(s_{\text{ch}}) \) is discussed in section 2.2. The homogeneous contribution to \( N_c \) is given by

\[
N_{\text{hom}} = \begin{cases} 
N_o e^{-f_c} (1 - e^{-f_c}) & ; f_c < 0.6 \\
N_o \left[ 1 + \exp \left( - \frac{9 - 2f_c}{7} \right) \right]^{-1} & ; f_c \geq 0.6,
\end{cases}
\]

where \( N_o \) is the number concentration of the supercooled liquid droplet population and \( f_c \) is the droplet freezing fraction for cirrus clouds formed in situ [Barahona and Nenes, 2008, 2009a] (calculated below). The original expression in BN09 [Barahona and Nenes, 2008] for \( N_{\text{hom}} \) was developed for maximum homogeneous freezing fractions below 25% (which accounts for most conditions of cirrus cloud formation). To account for high homogeneous freezing fractions where \( N_{\text{hom}} \) is limited by the available sulfate aerosol concentration (as for example in convective clouds and sensitivity studies), the second term in equation (5) (for \( f_c \geq 0.6 \)) is added to the result of Barahona and Nenes [2008, cf. equation (30)]. This term is derived by fitting a sigmoidal function for \( \frac{N_{\text{het}}(s_{\text{ch}})}{N_{\text{lim}}} \) to parcel model simulations [Barahona and Nenes, 2008], in agreement with published observations for convective clouds and cloud studies [Barahona and Nenes, 2008; Phillips et al., 2007]; \( f_c = 0.6 \) is chosen as the value where both \( N_{\text{hom}}(f_c) \) and \( s_{\text{lim}} \) in the high and low freezing fraction expressions are equal. \( f_c \) depends on the active freezing mechanism and is calculated below.

[12] When homogeneous and heterogeneous freezing are active, heterogeneously frozen crystals deplete water vapor [Barahona and Nenes, 2009a; DeMott et al., 1994; Kärcher et al., 2006] and weaken the homogeneous freezing pulse. In this case, \( s_{\text{max}} = s_{\text{hom}} \), \( N_{\text{IN}} = N_{\text{het}}(s_{\text{hom}}) \), and \( f_c \) depends on \( N_{\text{IN}} \) and the freezing characteristics of the IN population as,

\[
f_c = f_{c,\text{hom}} \left\{ 1 - \left( \frac{N_{\text{het}}(s_{\text{hom}})}{N_{\text{lim}}} \right)^{\frac{3}{2}} \right\}^{\frac{3}{2}},
\]

where \( f_{c,\text{hom}} = \frac{\rho_i}{\rho_0} \frac{N_{\text{hom}}}{N_{\text{het}}} \left( \frac{2 \alpha V s_{\text{max}}}{\pi f_{\text{hom}}} \right)^{\frac{1}{2}} \) is the droplet freezing fraction in absence of IN, i.e., for pure homogeneous freezing [Barahona and Nenes, 2008]. Other symbols are defined in the notation section.

[13] If \( N_{\text{IN}} \) is high enough, heterogeneously frozen crystals may deplete enough water vapor so that \( s_{\text{max}} < s_{\text{hom}} \), inhibiting homogeneous freezing [Barahona and Nenes, 2009a; Gierens, 2003]. In this regime, \( N_c = N_{\text{het}}(s_{\text{max}}) \) (equation (1)), and \( s_{\text{max}} \) is given by the solution of

\[
\frac{N_{\text{het}}(s_{\text{max}})}{N^*} = \frac{1}{\sqrt{\Delta^*_{\text{char}} s_{\text{max}}}} \left( 1 + s_{\text{max}} \right)^{\frac{1}{\nu_{\text{max}}}},
\]

where \( \lambda = \frac{\Gamma_1}{\Gamma_2} \sqrt{\frac{1}{\alpha V}} \). Other symbols are defined in the notation section.

2.2. Heterogeneous IN Spectra

[14] The freezing spectrum function \( N_{\text{het}}(s_{\text{ch}}) \) gives the number concentration of IN at conditions of \( T, \rho, \) and \( s \), accounting for the contribution of individual insoluble aerosol species and different freezing modes [Barahona and Nenes, 2009b]. \( N_{\text{het}}(s_{\text{ch}}) \) can be obtained from theoretical considerations, field campaign, and laboratory data [e.g., Barahona and Nenes, 2009b; Field et al., 2006; Khvorostyanov and Curry, 2009; Meyers et al., 1992; Möhler et al., 2006; Phillips et al., 2008; Welti et al., 2009]. BN09 is developed so that any expression for \( N_{\text{het}}(s_{\text{ch}}) \) (experimental or theoretical) can be used without loss of accuracy.

[15] Table 2 describes the heterogeneous freezing spectra used in this study. Three empirically derived expressions are used: the spectra of Meyers et al. [1992, MY], Phillips et al.
are assumed to be monodisperse and chemically homogeneous [Barahona and Nenes, 2009a]. $N_{\text{het}}(s_i)$ is then a step function at a prespecified freezing threshold, $s_{\text{hom}} < s_{\text{het}}$:

$$N_{\text{het}}(s_i) = \begin{cases} N_{\text{dust}} + N_{\text{bc}} & s_i \geq s_{\text{het}}, \\ 0 & s_i < s_{\text{het}}. \end{cases}$$ (8)

Where $N_{\text{dust}}$, $N_{\text{bc}}$ are the number concentrations of black carbon and dust, respectively. The steep increase in $N_{\text{het}}(s_i)$ about $s_{\text{het}}$ is a behavior predicted by classical nucleation theory (CNT) [Khvorostyanov and Curry, 2004, 2009]; it is also obtained by generalizing expressions derived for homogeneous nucleation [Kärcher and Lohmann, 2003]. The “monodisperse approximation” is the main type of IN parameterization currently implemented in GCMs [e.g., Liu et al., 2007a; Lohmann et al., 2004; Penner et al., 2009] with $s_{\text{het}}$ typically treated as a free parameter constrained by observations. Lohmann et al. [2004] assumed $s_{\text{het}} = 0.3$, based on reported measurements of black carbon freezing [DeMott et al., 1999]. Liu and Penner [2005] did not directly assume a value for $s_{\text{het}}$, but rather used prescribed parameters as input to a CNT expression [Khvorostyanov and Curry, 2004], which effectively results in $s_{\text{het}} \sim 0.2$ for black carbon [Barahona and Nenes, 2009b]. Laboratory studies [e.g., Eastwood et al., 2008; Field et al., 2006; Möhler et al., 2006; Welti et al., 2009] suggest that ice nucleates on dust in the deposition mode around $s_i \sim 0.1$–0.3. Theoretical studies show that for $s_{\text{het}}$ between 0.1 and 0.3 the effect of IN on $N_e$ is not very sensitive to $s_{\text{het}}$ [Barahona and Nenes, 2009a; Gierens, 2003]. On the basis of these considerations, $s_{\text{het}} = 0.3$ is assumed in this study (Table 2).

![Figure 1](https://example.com/figure1.png)

**Figure 1.** Examples of heterogeneous freezing spectra used in this study (presented in Table 2). Conditions considered were $N_{\text{dust}} = 0.01 \text{ cm}^{-3}$, $N_{\text{bc}} = 0.5 \text{ cm}^{-3}$, $T = 210 \text{ K}$, and $p = 22000 \text{ hPa}$. For the GLASS spectrum, $T = 200 \text{ K}$ and $N_{\text{glassy}} = 50 \text{ cm}^{-3}$. The vertical line represents the onset of homogeneous freezing at $T = 210 \text{ K}$ [Koop et al., 2000].

[2007, BKG], and Phillips et al. [2008, PDA] (Figure 1). MY and BKG depend only on $s_i$ and have a similar functional form. BKG yields about tenfold lower $N_{\text{IN}}$ than MY for the same $s_i$; this is because MY is derived from surface measurements and therefore represents an upper limit in $N_{\text{IN}}$. PDA is derived from several field campaign data sets; apart from $s_i$ and $T$, the contribution of individual aerosol species (dust, black carbon and organics) and freezing modes (i.e., immersion and deposition) to $N_{\text{het}}(s_i)$ is provided. PDA uses BKG as a “background” IN spectrum from which the individual contribution of dust, black carbon, and organic aerosol species are scaled according to their surface area distribution. In this study, PDA is applied assuming the Phillips et al. [2008] background size distribution for dust and black carbon.

[16] Theoretically derived $N_{\text{het}}(s_i)$ are also used in this study. The simplest $N_{\text{het}}(s_i)$ scheme is the so-called “monodisperse IN” (MONO) approximation, in which IN

### Table 2. Heterogeneous Nucleation Spectra Used in This Study

<table>
<thead>
<tr>
<th>$N_{\text{het}}(s_i)$</th>
<th>Description</th>
<th>Type</th>
</tr>
</thead>
<tbody>
<tr>
<td>HOM</td>
<td>No heterogeneous freezing allowed [Barahona and Nenes, 2008; Koop et al., 2000]</td>
<td>Semiempirical</td>
</tr>
<tr>
<td>MONO</td>
<td>Monodisperse IN with $s_{\text{het},\text{MONO}} = 0.3$ (equation (8)) [Barahona and Nenes, 2009a]</td>
<td>Theoretical</td>
</tr>
<tr>
<td>CNT-BN</td>
<td>Equation (9) with $s_{\text{dust}} = 0.2$, $s_{\text{bc}} = s_{\text{om}}$, $f_{\text{s_dust}} = 0.011$ ($\theta_{\text{dust}} = 16^\circ$), $f_{\text{s_bc}} = 0.039$ ($\theta_{\text{bc}} = 40^\circ$) [Chen et al., 2008], and $s_{\text{hom}}$ from Barahona and Nenes [2008]</td>
<td>Semiempirical</td>
</tr>
<tr>
<td>MY</td>
<td>Meyers et al. [1992]</td>
<td>Empirical</td>
</tr>
<tr>
<td>BKG</td>
<td>Phillips et al. [2007]</td>
<td>Empirical</td>
</tr>
<tr>
<td>PDA</td>
<td>Phillips et al. [2008]</td>
<td>Empirical</td>
</tr>
<tr>
<td>GLASS</td>
<td>Murray et al. [2010]. The fraction of deliquesced aerosol in glassy state was assumed as $N_{\text{glassy}} / N_o = 0.8 \text{ min} \left( \frac{212 - T}{32} \right)$ for $T &lt; 212 \text{ K}$ and $N_{\text{glassy}} / N_o = 0$ for $T &gt; 212 \text{ K}$ [Murray, 2008; Murray et al., 2010]</td>
<td>Empirical</td>
</tr>
</tbody>
</table>
polydisperse IN, the relative concentration of monodisperse
IN classes is approximately equal to the ratio of their hetero-
genous nucleation rates at $s_i$ (calculated using CNT [Khvorostyanov and Curry, 2004]). This result in a semi-
empirical freezing spectrum derived from CNT (referred to as CNT-BN) but constrained with observations. When applied
to dust and black carbon, it takes the form

$$N_{het}(s_i) \approx \min \left\{ \frac{s_i}{s_{h,dust}} \exp \left[-k_{hom,fh,dust}(s_{h,dust} - s_i) \right], 1 \right\} N_{dust}$$

$$+ \min \left\{ \frac{s_i}{s_{h,bc}} \exp \left[-k_{hom,fh,bc}(s_{h,bc} - s_i) \right], 1 \right\} N_{bc},$$  \hspace{1cm} (9)

where $s_{h,dust}$ and $s_{h,bc}$ are the values of $s_i$ at which the dust and black carbon freezing fraction is unity, respectively
(other symbols are defined in the notation section), and can be constrained using measurements of nucleation rate [e.g.,
Chen et al., 2008]. On the basis of published laboratory studies, we assume $s_{h,dust} = 0.2$ [e.g., Eastwood et al., 2008;
Field et al., 2006; Möhler et al., 2006; Wetti et al., 2009]. As black carbon tends to freeze in both the immersion and
deposition modes, $s_{h,bc}$ is highly variable and depends on the IN concentration, the water activity in the deli-
quesced aerosol, and the particle history [Dymarska et al., 2006; Kärcher et al., 2007; Möhler et al., 2005;
Popovicheva et al., 2008; Zobrist et al., 2008b]. Laboratory results however indicate that when water saturation is
reached, black carbon is an active IN in the condensation mode [DeMott et al., 1999; Dymarska et al., 2006]. Thus,
it is assumed that $s_{h,bc} = s_{i,sat}$, where $s_{i,sat}$ is the super-
saturation with respect to ice at liquid water saturation
(e.g., $s_{i,sat} = 0.51$ at $T = 230$ K, and $s_{i,sat} = 0.85$ at $T = 200$ K) [Murphy and Koop, 2005].

19 Besides dust and black carbon, glassy organic aerosol
[Murray et al., 2010] have been proposed as a source of IN
in the upper troposphere. Laboratory measurements indicate
that the vitreous transition temperature of organic aerosol
solutions can be as high as 210 K [Murray, 2008; Murray et al., 2010; Zobrist et al., 2008a]. Liquid droplets contain-
ing organic solutes can thus become "glassy" at cirrus conditions.
The fraction of deliquesced aerosol in glassy state depends on composition (e.g., organic fraction) and
temperature but is still uncertain. To test the potential sensi-
tivity of $N_c$ to this type of IN, the fraction of deliquesced aerosol in glassy state is assumed to depend linearly on $T$, from
zero at $T \geq 210$ K to 80% for $T \leq 185$ K. The freezing fraction of glassy aerosol is then calculated using the hetero-
genous freezing spectrum reported by Murray et al. [2010].

Solid ammonium sulfate can also act as IN [Abbatt et al.,
2006; Shilling et al., 2006]. For this, the liquid aerosol must pass by a state of very low relative humidity, effloresce,
and then freeze before deliquescence can take place [Shilling et al., 2006]. Recent studies [Jensen et al.,
2010] suggest that this is preferred at very low temperature
($T < 200$ K) where the saturation ratio with respect to liquid water remains low even if the environment is
supersaturated with respect to ice [Jensen et al., 2010]. At warmer conditions, most air parcels pass through states of
high relative humidity before reaching the upper troposphere
[Wiacek et al., 2010] and solid ammonium sulfate IN may be
less common. It is however very difficult to estimate the
fraction of ammonium sulfate particles that act as IN in the
upper troposphere, as it depends on the history of individual
parcels. Thus, the effect of ammonium sulfate IN is left for
future work.

2.3. Implementation of Ice Crystal Concentration
Parameterization Within GMI

20 In this study we assume that $N_c$ that can form in cirrus
is equal to the nucleated ice crystal concentration. Cloud
fraction and relative humidity are not available in GMI; this
is not a limitation however, as BN09 uses the cloud-scale
updraft velocity to calculate the maximum in-cloud $s_{max}$ and $N_c$. The spatial distribution of cirrus is analyzed in
section 3.2 using climatology data. To obtain the maximum
sensitivity of ice crystal concentration to heterogeneous
freezing, ice crystal sedimentation and sublimation are not
considered (the consequences of this assumption are not
discussed in section 4).

21 Calculation of $N_c$ using the BN09 parameterization
requires the knowledge of $T, p, V$, and the concentration
of individual aerosol species (i.e., deliquesced droplets, dust,
and black carbon). $T$ and $p$ were assumed to be those of the
grid cell (subgrid cell fluctuation in cooling rate is also con-
sidered as described below). Following Chen and Penner
[2005, and references therein], the total number of deli-
quesced aerosol available for homogeneous freezing $N_c$ is
assumed equal to the sulfate aerosol number (calculated from
the sulfate mass by using a lognormal size distribution
function derived from observations; Table 1). Since $N_{hom}$ is
rarely limited by the available aerosol (i.e., $N_{hom} \ll N_c$)
[Kärcher and Lohmann, 2002; Seifert et al., 2004] this
assumption is not expected to introduce a significant bias in
$N_c$. A similar approach is used to calculate $N_{dust}$ and $N_{bc}$;
prescribed dust and black carbon size distribution functions
are shown in Table 1 (the consequences of assuming a fixed
size distribution for dust and black carbon is assessed in
section 4). In agreement with theoretical studies and field
campaign data, the vapor-to-ice deposition coefficient, $\alpha_\text{v}$, was set to 0.1 [e.g., Barahona and Nenes, 2008; Gayet et al.,
2004; Hoyle et al., 2005; Jensen et al., 2008; Khvorostyanov
et al., 2006; Liu and Penner, 2005; Monier et al., 2006].

22 The cloud scale updraft velocity $V$ sets the rate of
expansion cooling during cloud formation and is consid-
erably different from the grid scale value resolved in a large-
scale model. Subgrid scale variations in $V$ are associated with
turbulence and gravity waves [Comstock et al., 2008; Haag
and Kärcher, 2004; Hoyle et al., 2005; Joos et al., 2008;
Kärcher and Ström, 2003; Kim et al., 2003; Lohmann and
Kärcher, 2002], the effect of which on $N_c$ can be accounted
for by averaging over the probability distribution function
(PDF) of updraft velocities $P_V(V)$,

$$N_c = \frac{\int_V N_c[V, s_{max}(V)] P_V(V) dV}{\int_V P_V(V) dV}. \hspace{1cm} (10)$$
PDF averaging is also applied to calculate grid cell average $N_{\text{IN}}, s_{\text{max}}$, and $N_{\text{het}}$. According to equation (10), the grid cell in-cloud $N_c$ results from a weighted average of cirrus formation events where homogeneous and heterogeneous freezing take place (Figure 2). Thus, for a given $N_{\text{IN}}$, there are always a fraction of cirrus formation events (associated with the lowest updrafts) for which homogeneous freezing is completely inhibited.

In this study, $P_{\chi}(V)$ is assumed to be a normal distribution with zero mean and standard deviation $\sigma_{\chi}$ of 25 cm s$^{-1}$ [Gayet et al., 2004], reflective of the gravity wave activity and small scale turbulence in the upper troposphere [Bacmeister et al., 1999; Haag and Kärcher, 2004; Jensen and Pfister, 2004; Kärcher and Ström, 2003]. The sensitivity of $N_c$ to $\sigma_{\chi}$ is addressed by assuming a linear variation of $\sigma_{\chi}$ with temperature from 25 cm s$^{-1}$ at 238 K to 1 cm s$^{-1}$ at 198 K [Wang and Penner, 2009] and constant outside this range, suggesting that the influence of gravity wave motion on cirrus formation decreases with altitude and is driven by large-scale dynamics (and therefore weak updrafts) at the tropopause level [e.g., Jensen et al., 2008; Khvorostyanov et al., 2006; Spichtinger and Gierens, 2009a, 2009b]. $V_{\text{min}}$ and $V_{\text{max}}$ are set to 1 and 50 cm s$^{-1}$, respectively, in agreement with field measurements [Comstock et al., 2008; Herzog and Vial, 2001; Khvorostyanov et al., 2006; Lawson et al., 2008].

3. Results and Discussion

The global distributions of $N_c$, $s_{\text{max}}$, and $N_{\text{IN}}$ are calculated by solving equations (1)–(7) and (10) using $p$ and $T$, $N_{\text{dust}}$, $N_{bc}$, and $N_o$ (provided by GMI) and $P_{\chi}(V)$ as described in section 2.2. $N_{\text{IN}}$ is calculated as $N_{\text{het}} (s_{\text{max}})$ (equation (10)) when only heterogeneous freezing is active and as $N_{\text{het}} (s_{\text{hom}})$ when both homogeneous and heterogeneous freezing are active. Sections 3.1–3.6 focus on the influence of dust and black carbon on $N_c$. The effect of glassy aerosol on $N_c$ at low temperature is analyzed in section 3.7.

3.1. Global Distribution of IN Concentration

Figure 3 shows the annual zonal average of $N_{\text{IN}}$, for the different IN spectra of Table 2. The spatial distribution of $N_{\text{IN}}$ depends strongly on the freezing spectra employed to describe heterogeneous freezing. The lowest global mean $N_{\text{IN}}$ (for all grid cells with $N_c > 0$ and $T < 235$ K) was found for BKG and PDA (~$3$ L$^{-1}$) and the highest for MONO (~$2.4$ cm$^{-3}$). As MY and BKG do not account for the spatial variation of dust and black carbon concentration, $N_{\text{IN}}$ is quite uniform across the globe and only increases near the tropical tropopause level in response to an increase in $s_{\text{max}}$ associated with decreasing $T$. The dependency of $N_{\text{IN}}$ on the dust and black carbon concentration is considered using the MONO, PDA, and CNT-BN spectra (section 2.2). For levels below 300 hPa, $N_{\text{IN}}$ is larger in the Northern Hemisphere than in the Southern Hemisphere, decreasing with altitude. The PDA spectrum yields $N_{\text{IN}}$ below $10^{-3}$ cm$^{-3}$ for most of the upper troposphere, being even lower than with the BKG spectrum. This is likely because $N_{\text{dust}}$ and $N_{bc}$ predicted by the GMI model [Liu et al., 2007b] are below the background levels assumed by Phillips et al. [2008]. When using the MONO spectrum, black carbon is the predominant IN precursor (as $N_{bc}$ is typically much higher than $N_{\text{dust}}$ [Liu et al., 2007b]). CNT-BN and PDA however predict a much higher freezing fraction for dust than for black carbon, and in some regions (around 30°N) $N_{\text{dust}}$ dominates $N_{\text{IN}}$; these features are further analyzed in section 3.3. $N_{\text{IN}}$ from glassy aerosol (GLASS) is typically below $10^{-3}$ cm$^{-3}$ and reaches about $10^{-2}$ cm$^{-3}$ at the tropical tropopause level (TTL), where their effect is most prominent (section 3.7).

3.2. Global Distribution of Ice Crystal Number Concentration

The variation in $N_{\text{IN}}$ from application of the freezing spectra of Table 2 (glassy aerosol IN is only significant at low $T$ and its effect is analyzed in section 3.7) results in a factor of 20 variation in global mean $N_c$ (Figure 4). Considering only homogeneous freezing (HOM) produces high $N_c$ (greater than 10 cm$^{-3}$) near the tropical tropopause level (TTL) which decreases with increasing $T$. When heterogeneous freezing is considered, usage of the MY spectrum results in the lowest $N_c$ (0.12 cm$^{-3}$) whereas the MONO spectrum gives the highest $N_c$ (2.5 cm$^{-3}$). All the spectra presented in Table 2 (except for MONO) resulted in average $N_c$ below the level for pure homogeneous freezing (HOM); this means that competition between homogeneous and heterogeneous freezing could occur globally. The MONO spectrum resulted in global mean $N_c$ higher than in HOM, suggesting that $N_{\text{IN}}$ is high enough to completely prevent homogeneous freezing over large regions of the globe. Using BKG and PDA resulted in global mean $N_c$ close to the HOM case (0.58 cm$^{-3}$), whereas using CNT-BN resulted in a global mean $N_c$ about 35% below HOM.
It is important to identify regions of the globe where cirrus could be susceptible to competition effects from IN. Figure 5 presents the frequency and visible cloud optical depth (COD) of cirrus, obtained from ISCCP climatology [Rossow and Schiffer, 1999]. Cirrus are more frequently observed in the tropics than in the midlatitudes and more common over the continents than over the oceans (except in the tropics). COD is about 0.8–1.4 in the northern midlatitudes and in the southernmost latitudes of the Southern Hemisphere, and slightly lower in the tropics and the midlatitudes of the Southern Hemisphere (∼0.4–1). Cirrus form typically at around 8 km in the midlatitudes and above 12 km (hence very low $T$) in the tropics [Dowling and Radke, 1990; Liou, 2002]. Thus, the effect of IN on the spatial distribution of $N_c$ is analyzed at two different vertical levels, $p = 281$ hPa and $p = 171$ hPa. At the $p = 281$ hPa...
hPa level, competition effects from IN on \( N_c \) are strongest, as this is the minimum height where cirrus could form (i.e., \( T \) is less than 235 K over the entire year) and aerosol concentrations (i.e., \( N_{\text{IN}} \)) are highest (for the CNT-BN, PDA and MONO spectra). Cirrus formation at \( p = 171 \) hPa exhibits less pronounced IN competition, a consequence of decreasing aerosol concentration with height and lower \( T \).

[28] Figure 6 shows global maps of \( N_c \) normalized with \( N_c \) from pure homogeneous freezing, \( N_{c,\text{HOM}} \), and \( N_{c,\text{HOM}} \) is used to express relative changes in total crystal concentration from the effect of IN (and is preferred over using \( \frac{N_c}{N_{\text{IN}}+N_{\text{HOM}}} \), since \( N_c \) can be greatly affected by IN even if the contribution of heterogeneous freezing to \( N_c \) is very small [Barahona and Nenes, 2009a]). At \( p = 281 \) hPa, usage of empirical IN spectra that only depend on \( s_c \) (BKG and MY) predict a uniform global distribution of \( \frac{N_c}{N_{c,\text{HOM}}} \) with slight variations occurring from variations in \( T \) (e.g., Figure 4). At this level, \( N_c \) for the CNT-BN, PDA, and MONO spectra is controlled by the spatial distribution of dust and black carbon concentration (e.g., Figures 3 and 8). For the PDA spectrum, heterogeneous effects are confined to the midlatitudes of the Northern Hemisphere and are the strongest \( \left( \frac{N_c}{N_{c,\text{HOM}}} \sim 0.5 \right) \) in regions of high dust concentration in north of Africa, South East Asia, and the west coast of North America \( (N_{\text{dust}}, \text{between } 0.1 \text{ and } 0.5 \text{ cm}^{-3}, [Liu \text{ et al., } 2007b]) \). The high frequency of cirrus (and the relatively large COD, Figure 5) in these regions (except in North Africa) indicates that IN effects may have a significant impact on regional climate. This is also evident when using the CNT-BN spectrum, yielding \( \frac{N_c}{N_{c,\text{HOM}}} \sim 0.6 \) in the same regions. Usage of CNT-BN however results in low \( \frac{N_c}{N_{c,\text{HOM}}} \) over the Caribbean, Central America, and the remote Pacific Ocean, caused by the high \( N_{\text{bc}} \) [Liu et al., 2007b] in these regions. The relatively low frequency of cirrus in these regions (except near the tropics, Figure 5) and their relatively low COD however suggest that IN effects (at the \( p = 281 \) hPa level) on regional climate may not be as significant as in the Northern Hemisphere. The strongest effect of IN on \( N_c \) is found with the MONO spectrum, where \( \frac{N_c}{N_{c,\text{HOM}}} > 1 \) over most of the Northern Hemisphere. \( \frac{N_c}{N_{c,\text{HOM}}} \) for MONO is however close to unity over large areas of the southern hemisphere and the tropics. This however does not indicate a weak IN effect but complete inhibition of homogeneous freezing, and \( N_{\text{IN}} \) close to \( N_{c,\text{HOM}} \), as analyzed below (Figure 7).

[29] Comparison between the \( \frac{N_c}{N_{c,\text{HOM}}} \) contours at the 281 and 171 hPa vertical levels (Figure 6) show that \( \frac{N_c}{N_{c,\text{HOM}}} \) increases with height for the PDA, CNT-BN, and BKG spectra and decreases for the MONO and MY spectra. For PDA and CNT-BN this results from a decrease in \( N_{\text{dust}} \) and \( N_{\text{bc}} \) with height that limits competition effects. With the MONO spectrum, the predominance of heterogeneous freezing results in a direct correlation of \( N_c \) (hence \( \frac{N_c}{N_{c,\text{HOM}}} \)) with \( N_{\text{IN}} \). The decrease in \( \frac{N_c}{N_{c,\text{HOM}}} \) with height seen with the MY spectrum results from a slightly higher \( N_{\text{IN}} \) caused by the lower \( T \) (and higher \( s_{\text{max}} \) ) at the 171 hPa level than at \( p = 281 \) hPa (Figure 3). \( N_{\text{IN}} \) predicted by BKG also increases with decreasing \( T \) \( \frac{N_c}{N_{c,\text{HOM}}} \) however still increases as competition effects are weak and homogeneous freezing is more vigorous at lower \( T \) [Barahona and Nenes, 2008]. At the \( p = 171 \) hPa level, the MONO and MY spectra result in an appreciable effect of IN emissions on \( N_c \) in the Southern Hemisphere and the tropics. Using CNT-BN results in \( \frac{N_c}{N_{c,\text{HOM}}} \sim 0.6 \sim 0.8 \) over most of the Southern Hemisphere, but it is close to 1 in the tropics (except over the Atlantic Ocean). For the other spectra of Table 2 (BKG and PDA), \( N_{\text{IN}} \) is too low to impact \( N_c \), which is agreement with in situ observations [Haag et al., 2003]. This would indicate that IN spectra that predict high black carbon freezing fraction would tend to overestimate IN effects at the high levels of the upper troposphere, affecting mainly tropical regions, where cirrus are more frequent (Figure 5).

[30] The active freezing mechanisms can be deduced from \( \frac{N_c}{N_{\text{HOM}}} \). When \( \frac{N_c}{N_{\text{IN}}} < 1 \), homogeneous and heterogeneous freezing actively contribute to \( N_c \); when \( \frac{N_c}{N_{\text{IN}}} > 1 \), only heterogeneous freezing takes place (equation (6)). Figure 7 presents the annual average \( \frac{N_c}{N_{\text{HOM}}} \) for the spectra of Table 2...
Figure 6. Annual average ice crystal concentration obtained in the GMI model at (top) $p = 281$ hPa and (bottom) $p = 171$ hPa, normalized with $N_c$ from pure homogeneous freezing (HOM).
at the $p = 281$ hPa level. For the MONO spectrum, $\frac{N_{IN}}{N_{lim}} > 1$, which indicates that $N_{IN}$ is high enough to inhibit homogeneous freezing over most of the globe. Figure 3 indicates that this is the case up to very high levels in the atmosphere. If MONO reflected atmospheric IN, cirrus would show a marked difference in $N_c$ between the northern and the southern hemisphere, resulting from the interhemispheric differences in $N_{dust}$ and $N_{bc}$ (e.g., Figure 6, bottom). This behavior was seen in the global modeling study of Penner et al. [2009], whom used an IN framework derived from classical nucleation theory. For the CNT-BN spectrum, $\frac{N_{IN}}{N_{lim}} > 1$ in the Northern Hemisphere (although lower than for MONO). In the midlatitudes of the southern hemisphere (30°S to 60°S), $\frac{N_{IN}}{N_{lim}} \sim 1$ and $\frac{N_{IN}}{N_{HOM}} \sim 0.1$ (Figure 6, top), indicating strong competition between homogeneous and heterogeneous freezing; heterogeneous IN impacts would however not manifest given the low frequency of cirrus in these regions (Figure 5). At the southernmost latitudes, $N_{IN}$ decreases ($\frac{N_{IN}}{N_{lim}} \sim 0.2$ and $\frac{N_{IN}}{N_{HOM}} > 0.7$) due to the scarcity of dust and black carbon in these regions. At higher levels ($p < 281$ hPa), $N_{IN}$ predicted by the CNT-BN spectrum decreases (Figure 3) and competition effects vanish in the Southern Hemisphere and the tropics ($\frac{N_{IN}}{N_{lim}} < 0.2$, not shown). As $N_{IN}$ predicted by the PDA and BKG spectra is generally low (Figure 3), competition effects are weak over most of the globe (i.e., $\frac{N_{IN}}{N_{lim}} \ll 1$) except near dust sources in the Northern Hemisphere; PDA results in $\frac{N_{IN}}{N_{lim}}$ up to 0.4, depleting $N_c$ to about half of the homogeneous freezing value ($\frac{N_{IN}}{N_{HOM}} \sim 0.5$). Competition effects are very strong for the MY spectrum being $\frac{N_{IN}}{N_{lim}} \sim 1$ around the globe, which results in very low $\frac{N_{IN}}{N_{HOM}}$ (about 0.1–0.2, Figure 6).

3.3. The Role of Dust and Black Carbon as IN Precursors

[31] As dust and black carbon emissions originate from different sources [Liu et al., 2007b], it is important to distinguish their relative contribution to the IN population. Figure 8 shows $\frac{N_{IN}}{N_{HOM}}$ for the CNT-BN and PDA spectra considering either only black carbon ($N_{dust} = 0$) or dust ($N_{bc} = 0$) as IN precursor. Generally, ice nucleates on dust at low $s_i$ (~0.1–0.3) [Eastwood et al., 2008]; dust also has a higher freezing fraction than black carbon (i.e., it is a more “efficient” IN). Therefore lower $N_{dust}$ than $N_{bc}$ is needed to impact homogeneous freezing. The number concentration of black carbon is however higher in the upper troposphere ($N_{bc} \gg N_{dust}$) [Liu et al., 2007b] so, even if it is not a very efficient IN, heterogeneous freezing of black carbon still contribute appreciably to $N_{IN}$.

[32] Using CNT-BN and assuming dust as the only IN precursor produces a global annual mean $N_{IN}$ (at the $p = 281$ hPa vertical level) around 0.05 cm$^{-3}$, about 0.1 cm$^{-3}$ in most of the Northern Hemisphere and as high as 0.3 cm$^{-3}$ near dust sources (not shown). The spatial variability of dust concentration results in near pure homogeneous freezing in Tropical cirrus ($\frac{N_{IN}}{N_{HOM}} \sim 1$ and $\frac{N_{IN}}{N_{lim}} \ll 1$) and strong competition between freezing mechanisms in the Northern midlatitudes.
(\(\frac{N}{N_{c,\text{HOM}}} \sim 0.1\) and \(\frac{N_{\text{bc}}}{N_{\text{lim}}} \sim 1\)). Regions of \(\frac{N}{N_{\text{lim}}} \sim 0.5\) result from dominant heterogeneous freezing \((N_{\text{HOM}} > N_{\text{lim}})\) near dust sources in the northern hemisphere, and partially inhibited homogeneous freezing \((N_{\text{HOM}} < N_{\text{lim}})\) in the midlatitudes of the Southern Hemisphere. Using the PDA spectrum results in mean \(N_{\text{HOM}}\) around 0.001 cm\(^{-3}\) (and up to 0.01 cm\(^{-3}\) near dust sources). Given the simulated \(N_{\text{dust}} \sim 0.1\) cm\(^{-3}\) at \(p = 281\) hPa (not shown), PDA on average predicts that ice nucleates on about 1% of the dust aerosol (as opposed to \(\sim 100\%\) predicted by CNT-BN). Thus, only in regions of high \(N_{\text{dust}}\) (around 0.5 cm\(^{-3}\)) are IN numerous enough to impact \(N_c\).

[33] If black carbon is assumed to be the only species freezing heterogeneously \((N_{\text{dust}} = 0, \text{Figure 8, "bc-only"})\) the CNT-BN spectrum predicts \(N_{\text{IN}}\) around 0.04 cm\(^{-3}\) and up to 0.07 cm\(^{-3}\) in the midlatitudes of the Northern Hemisphere (at \(p = 281\) hPa). As \(N_{\text{IN}} \sim N_{\text{lim}}\), there is strong competition between freezing mechanisms, resulting in very low \(N_c\) over most of the Northern Hemisphere. Using the PDA spectrum for this case gives a mean \(N_{\text{IN}}\) around 0.001 cm\(^{-3}\) (up to 0.01 cm\(^{-3}\) cm\(^{-3}\)) and black carbon freezing fraction around 0.01%, which is too low to impact freezing (i.e., \(\frac{N_{\text{bc}}}{N_{\text{HOM}}} \sim 1\) and \(\frac{N_{\text{bc}}}{N_{\text{lim}}} < 0.1\)).

[34] Comparison of Figures 6 and 8 shows that the combined effect of dust and black carbon on \(N_c\) is not additive and depends on their relative freezing characteristics. For CNT-BN, there are enough IN from black carbon alone to strongly reduce \(N_c\) from pure homogeneous freezing concentrations (Figure 8, “bc-only”). Most of the black carbon however does not freeze in the presence of dust (Figure 6) because \(s_{\text{max}}\) does not exceed its freezing threshold. Therefore sufficient amounts of dust prevent the freezing of supercooled droplets and black carbon. In the southern hemisphere and the tropics \(N_{\text{dust}}\) is very low and black carbon is the dominant factor controlling \(N_c\), leading to competition between homogeneous and heterogeneous freezing (which however is not significant at high altitudes, section 3.2). This is not the case for PDA, as the black carbon freezing fraction is too small to have an appreciable impact on \(N_c\) and dust is the only significant source of IN. However, the presence of low dust concentration may reduce the \(N_{\text{bc}}\) required to have an appreciable effect on \(N_c\); the very small black carbon freezing fractions predicted by the PDA spectrum may thus exert a noticeable effect of IN on \(N_c\). This is illustrated by the values of \(\frac{N_{\text{bc}}}{N_{\text{HOM}}}\) at the northern latitudes of the Atlantic Ocean (around 50°); for both the dust-only and bc-only cases, \(\frac{N_{\text{bc}}}{N_{\text{HOM}}} \approx 1\) (Figure 8, PDA), but \(\frac{N_{\text{bc}}}{N_{\text{HOM}}} \approx 0.8\) when both dust and black carbon act as IN precursors (Figure 6, top, PDA).

### 3.4. Sensitivity to Dynamical Forcing

[35] When \(\sigma_V\) is assumed to decrease with temperature (Figure 9) \(N_c\) near the tropical tropopause level (TTL, \(p \sim 100\) hPa) is much lower \((N_c \sim 1\) cm\(^{-3}\)) than when using a fixed \(\sigma_V\) \((N_c \sim 10\) cm\(^{-3}\)). In terms of the global annual mean \(N_c\) for the whole atmosphere, this variation in
leads to about a threefold decrease for all the spectra of Table 2, primarily because reductions in updraft velocity decrease \( N_{\text{hom}} \) [Barahona and Nenes, 2008; Kärcher and Lohmann, 2002], which inevitably limits \( N_c \). However, small values of \( \sigma_V \) also exacerbate the effect of IN on \( N_c \) because \( N_{\text{lim}} \) decreases with decreasing \( V \) (i.e., fewer IN are required to perturb homogeneous freezing, Figure 2) [Barahona and Nenes, 2009a]. For the CNT-BN, MONO, and PDA spectra, \( N_{\text{IN}} \) decreases with height (as \( N_{\text{dust}} \) and \( N_{bc} \) decrease); if \( \sigma_V \) decreases with \( T \), \( N_{\text{lim}} \) also drops, allowing for significant heterogeneous effects even at very low \( N_{\text{IN}} \) (~1 L\(^{-1}\); Figure 3). Because of this, using the BKG and MY spectra, where \( N_{\text{IN}} \) does not depend on aerosol concentration (therefore does not decrease with height), leads to the predominance of heterogeneous freezing at low \( T \). This behavior is further analyzed in sections 3.6 and 3.7.

3.5. Effect of Temperature on \( N_c \)

As \( N_{\text{IN}} \) is limited by the available aerosol concentration, \( N_c \) in cirrus formed by heterogeneous freezing is less sensitive to changes in \( T \) than clouds where homogeneous freezing dominates [Barahona and Nenes, 2009a; Lohmann et al., 2008]. Figure 10 compares the results of the GMI model for the runs of Figure 3 against in situ data reported

\[
N_c (\text{cm}^2)
\]

Figure 9. Similar to Figure 3 with \( \sigma_V \) decreasing from 25 cm s\(^{-1}\) (at 238 K) to 1 cm s\(^{-1}\) (for \( T \leq 198 \) K).

\[
N_c (\text{cm}^2)
\]

Figure 10. Ice crystal concentration against grid cell temperature using the spectra of Table 2. Results shown assuming (left) \( \sigma_V = 25 \) cm s\(^{-1}\) and (right) \( \sigma_V \) decreasing from 25 cm s\(^{-1}\) (at 238 K) to 1 cm s\(^{-1}\) (for \( T \leq 198 \) K). Error bars represent one standard deviation about the mean \( N_c \) (error bars are omitted if lower than a factor of two about the mean). The shaded area corresponds to a factor of two from the mean \( N_c \) observed in situ [Krämer et al., 2009].
by Krämer et al. [2009]. The extensive data of Krämer et al. [2009] is based on aircraft measurements of relative humidity and ice crystal concentration in cirrus taken during 28 flights in several field campaigns in the Arctic, at midlatitudes, and in the tropics, covering regions from 20°S to 75°N, and temperatures between 183 and 240 K [Krämer et al., 2009]. Nc measurements from Krämer et al. [2009] are in good agreement with independent studies [Gayet et al., 2004; Hoyle et al., 2005; Lawson et al., 2008] and therefore are considered representative of global cirrus. [37] Agreement of simulations to within a factor of two with observations is obtained for T > 205 K for all IN spectra except MY (which underpredicts Nc). The positive slope of Nc versus T given by observations (as opposed to the slightly negative slope predicted by the model), may be a result of observational uncertainties or artifacts (e.g., ice crystal scattering in FSSP probes [Krämer et al., 2009]) that tend to introduce positive bias (mostly within a factor of 2 [Field et al., 2003]) in observed Nc at high T. For T < 205 K and assuming a fixed σV (Figure 10, left), the model largely overestimates Nc by at least tenfold, even in cases where heterogeneous effects are the strongest (e.g., MONO and MY). Assuming that σV decreases with T (hence height) yields low Nc at low T (section 3.4), giving a much better agreement with observations for the CNT-BN, HOM, and PDA spectra than when a fixed σV is used. Nc is however underestimated for the BKG and MY spectra from an overestimation in NIN at high altitudes. For T > 205 K, the CNT-BN, BKG, HOM, and MONO spectra display roughly similar Nc (mostly within a factor of 3). However, similar Nc may result from very different interactions between homogeneous and heterogeneous freezing (Figures 6 and 7; section 3.2). The distribution of smax that results from application of each IN spectrum considered in this study can help elucidate whether lowering σV with altitude is consistent with other available cirrus microphysical characteristics.

3.6. Maximum Supersaturation Statistics

[38] The frequency distribution of maximum supersaturation achieved during cloud formation, P(smax) is shown in Figure 11, for fixed σV (right) and for σV decreasing with T (left). As homogeneous freezing occurs over a very narrow s interval [Barahona and Nenes, 2008; Kärcher and Lohmann, 2002], P(smax) for HOM is insensitive to σV and depends only on T; smax ranges mostly between 0.4 and 0.65 with a mean around 0.5. Predominance of pure heterogeneous freezing for MONO and MY is expressed by a shift in P(smax) toward low smax (mean around 0.2–0.3). Competition between homogeneous and heterogeneous freezing (e.g., BKG and CNT-BN) results in a broad P(smax) with a maximum about the homogeneous freezing threshold (smax ≈ 0.5) extending to low smax. Assuming a T-dependent σV exacerbates IN effects on smax; P(smax) therefore resides mostly below the homogeneous freezing threshold for all the IN spectra (except in PDA where NIN is too low to significantly impact smax.).

[39] Observations can be used to diagnose smax as it is related to the upper limit of the relative humidity distribution in recently formed clouds [Haag et al., 2003; Heymsfield et al., 1998]. As freezing and subsequent crystal growth limit the maximum supersaturation within the cloud, smax is also associated with the predominant freezing threshold in the cirrus cloud. Unfortunately no systematic study of the global distribution of freezing thresholds (hence of P(smax)) has been carried out to date. We can however study which of the distributions presented in Figure 11 agrees with common features observed in different field campaigns. Field campaign data consistently show smax hear the homogeneous freezing threshold (i.e., between 0.45 and 0.7) [e.g., Comstock et al., 2008; DeMott et al., 2003; Gayet et al., 2004; Haag et al., 2003; Heymsfield and Sabin, 1989; Heymsfield et al., 1998; Krämer et al., 2009] at temperatures below 235 K. Haag et al. [2003] and Ström et al. [2003] have shown however that in some regions of the Northern Hemisphere, smax can be lower (~0.3) due to heterogeneous freezing, which is also consistent with theoretical considerations [Barahona and Nenes, 2009b; Kärcher and Lohmann, 2003]. The uncertainty associated with relative humidity measurements in the upper troposphere is typically about 0.1 (in absolute

Figure 11. Global annual average probability distribution of smax for T > 195 K calculated by the BN09 parameterization, for the different heterogeneous freezing spectra of Table 2. σV was assumed (left) constant equal to 25 cm s⁻¹ or (right) decreasing from 25 cm s⁻¹ at 238 K to 1 cm s⁻¹ for T ≤ 198 K.
Thus, to be consistent with these observations, $P(s_{\text{max}})$ should exhibit a mean around $s_{\text{max}} \sim 0.4$–0.6, with a broad tail toward lower $s_{\text{max}}$ values. Figure 11 shows that these features cannot be reconciled assuming weak dynamical forcing (a $T$-dependent $s_V$; Figure 10, right), or when heterogeneous IN completely inhibits homogeneous freezing during cloud formation (i.e., MY and MONO spectra). Indeed, only cloud formation scenarios where competition between homogeneous and heterogeneous freezing is the dominant feature (e.g., CNT-BN, BKG) would result in $P(s_{\text{max}})$ consistent with observations. Thus, simulations that prescribe a weakening of dynamical forcing ($s_V$) with height provide $N_c$ predictions that are closer to observations, but for the wrong reason.

3.7. Glassy Aerosol as IN at Low Temperature

At $T < 210$ K, $N_{\text{IN}}$ from glassy aerosol is high enough to impact $N_c$ and $P(s_{\text{max}})$. Figure 10 shows that using the GLASS spectrum leads to $N_c \sim 0.2$ for $T < 190$ K and fixed $s_V$. For this case, $P(s_{\text{max}})$ (for $T$ between 180 and 200 K, Figure 12) is centered around 0.5, indicating that homogeneous freezing is the predominant freezing mechanism, leading to high $N_c$ compared to observations. Thus, $N_{\text{IN}}$ is not enough to completely inhibit homogeneous freezing and only at low $V$ (typically below 5 cm s$^{-1}$) heterogeneous freezing dominates. When $s_V$ decreases with decreasing $T$ (leading to $V$ mostly around 1 cm s$^{-1}$, for $T < 200$ K) heterogeneous freezing dominates, which is evidenced by $P(s_{\text{max}})$ centered around $s_{\text{max}} \sim 0.3$ (Figure 12). This however results in $N_c$ much below observations at low $T$ (Figure 11), i.e., $N_{\text{IN}}$ is high enough to completely prevent homogeneous freezing but still much lower than observed $N_c$.

Few measurements of in-cloud relative humidity are available at low $T$; Krämer et al. [2009] reported values of relative humidity up to the homogeneous freezing threshold (and some few episodes above, also consistent with other studies [Jensen et al., 2005; Lawson et al., 2008]), which is consistent with the predominance of homogeneous freezing at the TTL. Froyd et al. [2009] reported cloud episodes with cutoff supersaturations up to 0.3, providing support to the hypothesis of predominant heterogeneous freezing at the TTL. The dynamics of the TTL may be not be passive enough to lead to a heterogeneous-only scenario (i.e., $T$ fluctuations around leading to $V$ mostly around 10–30 cm s$^{-1}$ are quite common at the TTL [Bacmeister et al., 1999; Herzog and Vial, 2001; Jensen et al., 2010; Sato, 1990]), and competition between homogeneous and heterogeneous freezing is more likely to occur.

3.8. Sensitivity of $N_c$ Distributions to Meteorological Field

The global distribution of temperature, as well as of aerosol concentration, can be greatly affected by the meteorological field employed to drive the aerosol simulation [Liu et al., 2007b]. To study how this sensitivity
would affect the global distribution of $N_c$. GMI runs were carried out using the DAO meteorological field (instead of GISS) for the HOM, CNT-BN, and PDA spectra (Figures 13 and 14). When no heterogeneous effects are considered (HOM) the global annual mean $N_c$ obtained with the DAO field (0.60 cm$^{-3}$) is very close to that obtained using GISS (0.62 cm$^{-3}$), although the spatial distribution of $N_c$ differs. Upper tropospheric ($p < 200$ hPa) $N_c$ is generally larger in DAO than in GISS and vice versa at lower levels. This results from a weaker vertical transport in DAO (compared to GISS) and slightly lower temperature (for the same vertical level) in DAO than in GISS [Liu et al., 2007b]. For the CNT-BN spectra, $N_c$ in the Northern Hemisphere (for $p > 300$ hPa) is much lower using the DAO field than obtained with the GISS field. For simulations with the PDA spectrum, the DAO field results in even weaker IN effects on $N_c$ than obtained with the GISS field (Figure 4). These features originate from differences in predicted $N_{dust}$ and $N_{bc}$ (therefore different interaction between homogeneous and heterogeneous freezing) and can be understood in terms of $N_{c,HOM}$ and $N_{c,HOM}$ [11]. The spatial distribution of $N_{c,HOM}$ at the $p = 258$ hPa level obtained with the DAO field is shown in Figure 14 for the PDA (right) and CNT-BN (left) spectra. Compared to simulations with the GISS meteorological field (Figure 6), the CNT-BN spectrum generally results in lower $N_c$ around the globe. The opposite occurs for the PDA spectrum where $N_{c,HOM} \sim 1$ (and $N_{c,HOM} \ll 1$) is considerably different from simulations with the GISS field ($N_{c,HOM} \sim 0.5$ near dust sources, section 3.2). These differences result from weaker dust transport to the upper troposphere in the DAO (compared to the GISS field). Indeed, the mean upper level ($p$ below 300 hPa) $N_{dust}$ and $N_{bc}$ predicted with DAO (0.05 and 0.8 cm$^{-3}$, respectively) are about a factor of two lower than for GISS (0.1 and 1.17 cm$^{-3}$, respectively). Thus, $N_{IN}$ is insufficient to prevent freezing of black carbon when using the CNT-BN spectrum with the DAO field. This implies that instead of pure heterogeneous freezing ($N_{c,HOM} > 1$) caused by dust for the GISS field, there is competition between freezing mechanisms ($N_{c,HOM} \sim 1$) when using the DAO field. Lower $N_{dust}$ with DAO winds also resulted in lower $N_{IN}$ when using PDA, therefore a weaker impact of dust on $N_c$ ($N_{c,HOM} \approx 1$) than simulated with GISS. However, this can partly result from the slightly lower pressure level used for comparison in the DAO field (DAO does not have the $p = 281$ hPa vertical level and $p = 258$ hPa is the lowest level for which $T < 235$ K over the entire year). At $p = 301$ hPa, DAO field simulations result in slightly lower $N_{c,HOM}$ (global mean 0.95) for PDA and slightly higher (global mean 0.27) for CNT-BN, than at $p = 258$ hPa. Such difference is still lower than the difference in $N_{c,HOM}$ between GISS and DAO; both meteorological fields however consistently overpredict $N_c$ at the TTL (Figures 4 and 13) and a significant contribution of dust to $N_{IN}$ in the Northern Hemisphere.

4. Summary and Conclusions

[44] A parameterization of cirrus cloud formation that accounts for competition between homogeneous and heterogeneous freezing was implemented in the Global Modeling Initiative (GIIM) chemical and transport model. Simulations were then carried out to study the sensitivity of ice crystal number concentration to dust and black carbon aerosol concentrations. Ammonium sulfate was assumed to deliquesce and contribute supercooled droplets that freeze homogeneously or heterogeneously if a fraction is in a glassy state (GLASS). Dust and black carbon were assumed to heterogeneously freeze, using spectra derived from empirical data (PDA, BKG, and MY) and theory (CNT-BN and MONO). The global annual mean $N_{IN}$ predicted by these formulations varied between 0.003 and 2.4 cm$^{-3}$. When included into the cirrus formation framework, this variation resulted in up to a factor of 20 difference (0.13–2.5 cm$^{-3}$) in the predicted global mean $N_c$. Although this value may likely represent an upper limit of variability (as ice crystal sedimentation and transport are not considered), it shows the large sensitivity of $N_c$ (and therefore of cloud properties) to the parameterization of $N_{IN}$. For low $N_{IN}$ (PDA spectrum), homogeneous freezing
was predominant and vice versa for high $N_{IN}$ (MONO spectrum), giving $N_c$ even higher than for pure homogeneous freezing. The lowest $N_c$ resulted when strong competition between homogeneous and heterogeneous freezing ($\frac{N_{IN}}{N_{lim}} \sim 1$) was predominant (MY spectrum).

The sensitivity of $N_c$ to IN is a strong function of the heterogeneous IN spectrum. Empirical parameterizations that only depend on supersaturation (BKG and MY) predicted a uniform $N_{IN}$ around the globe and little variation of $N_{IN}$ with height. Explicitly accounting for the dependency of dust and black carbon concentration (MONO, CNT-BN, and PDA) resulted in a highly variable spatial distribution of $N_{IN}$. In the latter, heterogeneous effects on $N_c$ were the strongest at the lowest levels of cirrus formation ($T \sim 230$ K) in the midlatitudes of the Northern Hemisphere, mostly near regions impacted by transport of dust and black carbon. In the tropics and midlatitudes of the Southern Hemisphere, it was found that IN would affect cirrus formation only if the freezing fraction of black carbon is close to unity. In general, heterogeneous effects were less significant at low temperatures as dust and black carbon concentration were low and the $N_{IN}$ needed to impact homogeneous freezing (i.e., $N_{lim}$) was higher than at warmer levels. Dust was found to be an important contributor of IN over large regions of the Northern Hemisphere. The effect of black carbon as heterogeneous IN is still controversial, as it depends on the functional form of the freezing spectrum. The simulations carried out in this study suggest that at least 1% of black carbon aerosol should freeze at cirrus levels in the Northern Hemisphere to have an appreciable effect on $N_c$. The larger $N_{IN}$ from dust and black carbon for the CNT-BN, PDA, and MONO spectra also results in a larger fraction of events dominated by heterogeneous freezing, and stronger competition between homogeneous and heterogeneous freezing in the Northern Hemisphere (Figures 6 and 7). This will lead to lower $s_{max}$ (i.e., lower freezing thresholds) in the Northern Hemisphere than in the Southern Hemisphere (particularly for $p > 200$ hPa), consistent with field campaign studies [Gayet et al., 2004; Haag et al., 2003].

The sensitivity of the global distribution of $N_c$ to large scale meteorological features was tested by running the GMI model using two different meteorological data sets (GISS and DAO). Differences in the vertical transport of dust and black carbon to the upper troposphere lead to significant variation in $N_{dust}$ and $N_{bc}$ (therefore on $N_{IN}$ and $N_c$). The largest sensitivity to the meteorological field resulted in regions where $N_{IN} \sim N_{lim}$ (midlatitudes of the Northern Hemisphere and near the tropics). For such conditions, a factor of two decrease in $N_{dust}$ and $N_{bc}$ at cirrus levels may change the predominant freezing regime from pure heterogeneous ($\frac{N_{bc}}{N_{lim}} > 1$) to competition between homogeneous and heterogeneous freezing ($\frac{N_{bc}}{N_{lim}} < 1$), and therefore the predicted response of cirrus to increased IN emissions. Despite this, runs with DAO and GISS meteorological fields consistently show high $N_c$ at the TTL (Figure 4) and a significant contribution of dust to $N_{IN}$ in the Northern Hemisphere.

Comparison of $N_c$ predicted with the GMI model against reported observations [Krämer et al., 2009] showed agreement for $T > 205$ K for most of the freezing spectra tested. However, at least a tenfold overprediction in $N_c$ for $T < 200$ K where $N_c$ was as high as 10 cm$^{-3}$, resulting from a high fraction of sulfate freezing homogeneously a low temperature. It was shown that if cirrus formation at low temperature forms in weak updrafts, $N_c$ is in much better agreement with observations. This however magnifies the sensitivity to heterogeneous nuclei, so that $N_{IN} \sim 1$ cm$^{-3}$ as low as 1 L$^{-1}$ was enough to affect homogeneous freezing. This results in $s_{max}$ distributions centered around values well below $s_{hom}$, in strong disagreement with available observations of freezing thresholds in cirrus clouds [e.g., DeMott et al., 2003; Haag et al., 2003; Krämer et al., 2009]. Since $s_{max}$ largely controls the steady state size of the ice crystals and the ice water content [Korolev and Mazin, 2003], incorrect predictions of $P(s_{max})$ will ideally lead to biases in predicted cirrus ice crystal size distribution. Thus, even if the bias of $N_c$ can be addressed by reducing $s_{max}$, this would likely introduce other biases in the effective radius of ice crystals and ice water path with potentially important implications for radiative forcing. Altogether this reveals a fundamental weakness in the “classical” theory of cirrus formation at low $T$.

The effect of glassy aerosol on the formation of cirrus clouds at low temperature was also studied. It was found that for $s_{max} \sim 25$ cm s$^{-1}$, insufficient IN are produced from glassy aerosol to completely prevent homogeneous freezing, thus leading to $N_c$ much higher than observed [Krämer et al., 2009]. Using $s_{max} \sim 1$ cm s$^{-1}$ at $T < 198$ K led to predominant heterogeneous freezing; the low freezing fraction of glassy aerosol however results in $N_c \sim 0.01$ cm$^{-3}$, below observed values. Murray et al. [2010] found $N_c \sim 0.05$ cm$^{-3}$ for $V \sim 3$ cm s$^{-1}$ at $T \sim 190$ K for pure heterogeneous freezing. The slightly lower $N_c$ in Figure 12 (right) results from the lower $V \sim 1$–2 cm s$^{-1}$ used in this work. The large sensitivity of $N_c$ to $s_{max}$, even in the presence of glassy aerosol, suggests that “chemical effects” (such as glassy IN) may not be the sole underlying cause for the characteristics of cirrus at low $T$. Dynamics, ice sedimentation and transport effects [Spichtinger and Gierens, 2009a, 2009b], and the existence of other sources of IN (e.g., solid ammonium sulfate) at the TTL [Abbatt et al., 2006; Jensen et al., 2010] may also play a role in explaining the characteristic of low temperature cirrus.

The results presented in this work must be interpreted as the potential of IN to alter $N_c$ whenever a cirrus cloud is formed. Cirrus cloud fraction was not calculated explicitly (i.e., grid cell cloud fraction was unity for $T < 235$ K); regions where IN effects are expected to be climatically important where however identified by focusing on areas with high cloud frequency and high cirrus optical depth (using climatological data from ISCCP). Another assumption made in this work was to consider $N_c$ at its maximum, i.e., no sedimentation or sublimation effects were accounted for. Sedimentation may structure the cirrus cloud layer and reduce the ice crystal concentration, particularly at low updraft velocity [Spichtinger and Gierens, 2009a, 2009b]. Sublimation may impact the ice crystal concentration in dry regions at high temperatures ($T > 235$ K) [Kärcher and Burkhardt, 2008]. Although important, these effects are not expected to introduce $N_c$ variability comparable to the variability introduced by $N_{IN}$. 

and will not modify the conclusions of this study. However, the ice crystal concentrations presented in Figure 10 may have a positive bias, particularly at warmer temperatures where sedimentation and evaporation are more significant. Nevertheless, the comparison of $N_c$ with available measurements ($T > 205$ K) suggests that they are usually within a factor of two of predictions.

The ice water content of cirrus clouds is also sensitive to heterogeneous IN. High $s_{\text{max}}$ (associated with homogeneous freezing) favors the diffusional growth of crystals; high $N_c$ however implies the distribution of ice mass among more particles [Korolev and Mazin, 2003]. Thus, high $N_c$ leads to small ice crystal sizes and reduced sedimentation rates. Small $N_c$ (resulting from strong competition between homogeneous and heterogeneous freezing) leads to large ice crystals but may also lead to increased sedimentation rates, which in turn reduces the ice water content of the cloud. Further research is needed to assess what process (sedimentation/nucleation) determines the cirrus cloud ice water content under atmospheric conditions.

The size distribution of different aerosol species was assumed to follow prescribed functions as presented in Table 1. This is not expected to introduce a bias for homogeneous freezing however may introduce errors in $N_{\text{dust}}$ and $N_{\text{bc}}$, and therefore in $N_c$. The freezing fraction of the dust and black carbon is also expected to be higher for larger aerosol size [e.g., Khvorostyanov and Curry, 2004; Welti et al., 2009]. Explicit aerosol dynamics is currently not considered but will be the subject of future work. Some uncertainty may also originate from the coarse GMI resolution ($4^\circ \times 5^\circ$) used in this work. Using a finer resolution may lead to a slightly different $N_c$ from differences in $T$ and aerosol concentrations, particularly when competition between homogeneous and heterogeneous freezing is important (although not enough to explain the features of Figures 10 and 11). This, however, is not likely to significantly influence the relative difference in $N_c$ from application of different heterogeneous freezing spectra. Thus, we have favored the ability to run a large number of cases over using a high resolution GCM. Nevertheless, the plethora of IN treatments is placed for the first time within the same global modeling dynamical framework. Despite the very large variability in IN concentration and relative contribution of freezing seen, ice crystal number concentrations are less variable than we had initially anticipated. However, the large sensitivity of $s_{\text{max}}$ to the prevailing mechanism clearly points out that it too needs to be sufficiently constrained for cirrus optical properties and climate forcing to be correctly represented in climate models.

### Notation

- $\alpha$ deposition coefficient of water vapor to ice.
- $g$ acceleration of gravity.
- $M_a$ molar mass of water and air, respectively.
- $M_w$ molar mass of water.
- $p$ ice saturation vapor pressure.
- $R$ universal gas constant.
- $\rho_a, \rho_i$ ice and air density, respectively.
- $s_i$ characteristic freezing threshold of a monodisperse IN population.
- $s_{\text{hom}}$ homogeneous freezing threshold.
- $s_{\text{max}}$ maximum ice supersaturation ratio.
- $s_{\text{sat}}$ value of $s_i$ at 100% relative humidity.
- $T$ temperature.
- $t$ time.
- $V$ updraft velocity.
- $D_e$ water vapor mass transfer coefficient.
- $f_c$ fraction of frozen particles at $s_{\text{hom}}$ with and without IN present, respectively.
- $f_{\text{dust}, \text{bc}}$ shape factor of the dust and black carbon, respectively.
- $\Gamma$ effective growth parameter [cf. Barahona and Nenes, 2008, equation (25)].
- $k_{\text{hom}}$ homogeneous nucleation rate coefficient at $s_{\text{hom}}$.
- $k_a$ thermal conductivity of air corrected for noncontinuum effects.
- $N^{\ast} \sqrt{2(\alpha V \Gamma_1/12)}(\beta + \frac{a}{c})^{-1}$.
- $N_{\text{dust}}$ number concentration of dust and black carbon, respectively.
- $N_{\text{bc}}$ number concentration of the supercooled liquid droplet population.
- $P$ ambient pressure.
- $\rho_a, \rho_i$ ice and air density, respectively.
- $\rho_f$ ice saturation vapor pressure.
- $R$ universal gas constant.
- $\rho_i, \rho_a$ ice and air density, respectively.
- $s_{\text{min}}$ ice crystal number concentration.
- $s_{\text{lim}}$ number concentration at $s_i$.
- $s_{\text{in}}$ ice crystal concentration from homogeneous freezing.
- $s_{\text{hom}}$ cumulative heterogeneous freezing spectrum; IN number concentration at $s_i$.
- $s_{\text{min}}$ number concentration of the supercooled liquid droplet population.
- $s_{\text{max}}$ maximum ice supersaturation ratio.
- $T$ temperature.
- $t$ time.
- $V$ updraft velocity.

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