Cirrus Parcel Model Comparison Project. Phase 1: The Critical Components to Simulate Cirrus Initiation Explicitly

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ABSTRACT

The Cirrus Parcel Model Comparison Project, a project of the GCSS [Global Energy and Water Cycle Experiment (GEWEX) Cloud System Studies] Working Group on Cirrus Cloud Systems, involves the systematic comparison of current models of ice crystal nucleation and growth for specified, typical, cirrus cloud environments. In Phase 1 of the project reported here, simulated cirrus cloud microphysical properties from seven models are compared for “warm” (−40°C) and “cold” (−60°C) cirrus, each subject to updrafts of 0.04, 0.2, and 1 m s$^{-1}$. The models employ explicit microphysical schemes wherein the size distribution of each class of particles (aerosols and ice crystals) is resolved into bins or the evolution of each individual particle is traced. Simulations are made including both homogeneous and heterogeneous ice nucleation mechanisms (all-mode simulations). A single initial aerosol population of sulfuric acid particles is prescribed for all simulations. Heterogeneous nucleation is disabled for a second parallel set of simulations in order to isolate the treatment of the homogeneous freezing (of haze droplets) nucleation process. Analysis of these latter simulations is the primary focus of this paper.

Qualitative agreement is found for the homogeneous-nucleation-only simulations; for example, the number density of nucleated ice crystals increases with the strength of the prescribed updraft. However, significant quantitative differences are found. Detailed analysis reveals that the homogeneous nucleation rate, haze particle solution concentration, and water vapor uptake rate by ice crystal growth (particularly as controlled by the deposition coefficient) are critical components that lead to differences in the predicted microphysics.

Systematic differences exist between results based on a modified classical theory approach and models using an effective freezing temperature approach to the treatment of nucleation. Each method is constrained by critical freezing data from laboratory studies, but each includes assumptions that can only be justified by further laboratory research. Consequently, it is not yet clear if the two approaches can be made consistent. Large haze particles may deviate considerably from equilibrium size in moderate to strong updrafts (0.2−1 m s$^{-1}$) at −60°C. The equilibrium assumption is commonly invoked in cirrus parcel models. The resulting difference in particle-size-dependent solution concentration of haze particles may significantly affect the ice particle formation rate during the initial nucleation interval. The uptake rate for water vapor excess by ice crystals is another key component regulating the total number of nucleated ice crystals. This rate, the product of particle number concentration and ice crystal diffusional growth rate, which is particularly sensitive to the deposition coefficient when ice particles are small, modulates the peak particle formation rate achieved in an air parcel and the duration of the active nucleation time period. The consequent differences in cloud microphysical properties, and thus cloud optical properties, between state-of-the-art models of ice crystal initiation are significant.

Intermodel differences in the case of all-mode simulations are correspondingly greater than in the case of homogeneous nucleation acting alone. Definitive laboratory and atmospheric benchmark data are needed to improve the treatment of heterogeneous nucleation processes.

1. Introduction

The Cirrus Parcel Model Comparison Project (CPMCP) is a project of the international Global Energy and Water Cycle Experiment (GEWEX) Cloud System Study Program (GCSS; Randall et al. 2000) Working Group on Cirrus Cloud Systems (WG2). The primary goals of the CPMCP are to assess the current state of cirrus microphysical modeling and to identify cirrus model sensitivities to present knowledge of nucleation and microphysics at low temperatures. The CPMCP also gives the Idealized Cirrus Model Comparison Project
Simulations of cloud initiation processes and the evolution of hydrometeor size distribution using explicit microphysical schemes date back to the 1940s and 1950s (e.g., Howell 1949; Mordy 1958). However, the direct application of explicit microphysical schemes to simulate cirrus clouds did not begin until the 1970s (Heymsfield 1975). Interest in cirrus clouds grew in response to the recognition of the extensive area coverage of cirrus and their potential effects on the global radiation budget (e.g., Liou 1986; Ramanathan et al. 1989). In one study, Sassen and Dodd (1988) compared lidar data with parcel model results to infer the homogeneous nucleation rate of ice in supercooled cloud droplets within the temperature range between $-34.3^\circ C$ and $-37.3^\circ C$. In another, Heymsfield and Sabin (1989) conducted a numerical study of homogeneous freezing of ammonium sulfate solution droplets at water-subsaturated conditions between $-40^\circ C$ and $-50^\circ C$. They reported that the predicted number concentration of ice crystals depended on temperature, parcel cooling rate, and cloud condensation nuclei (CCN) distributions. Furthermore, results of parcel model simulations aided in the development of a parameterization of the homogeneous freezing of supercooled cloud droplets and CCN solution droplets for use in regional-scale cloud models via a bulk microphysical scheme (DeMott et al. 1994). These simulations relied on an explicit microphysical scheme, in which the number distribution of ice particles is resolved into size bins, in contrast to the use of bulk parameterizations. The parcel model approach was also adopted in evaluating the potential impact of volcanic aerosols on cirrus microphysics (Jensen and Toon 1992), and in the study of orographic clouds. For example, model results were compared with in situ measurements of orographic clouds (Jensen et al. 1998; Lin et al. 1998), and the dominant nucleation mode under various background conditions was investigated (DeMott et al. 1997; Spice et al. 1999). To simulate the formation and evolution of contrails, Kärcher (1998) developed a model describing ice nucleation and growth in contrails that is similar to cirrus parcel models in many respects. In contrails, the competition between homogeneous freezing of aqueous aerosol particles and heterogeneous nucleation of soot particles emitted by jet engines plays an important role.

Many studies have ignored heterogeneous nucleation as a mechanism for cirrus formation, primarily due to the current lack of knowledge regarding the abundance and activation of ice nuclei (IN) in the upper troposphere. However, it is becoming apparent that the conditions observed with cirrus formation (Heymsfield and Miloshevich 1995; Heymsfield et al. 1998) do not satisfy the threshold conditions required by the homogeneous freezing of sulfate particles (DeMott 2002). This differs from conclusions that were initially inferred from the model findings of orographic wave clouds by Heymsfield and Miloshevich (1993). Sassen and Benson (2000) indicated that heterogeneous ice nucleation can strongly modulate the cirrus formation process, but only if IN concentrations and activations are similar to the most favorable conditions measured at the earth’s surface. Hence, heterogeneous nucleation must ultimately be treated in some manner. This project attempts to clarify the range of effects to be expected from incorporating various available heterogeneous nucleation schemes that are either based on data or expectations.

A systematic comparison of cirrus parcel models is necessary because of the sensitivity of cloud microphysical and radiative properties to the cloud initiation processes. While qualitative agreement between cirrus parcel models has been frequently demonstrated, the quantitative assessment has not yet been made. The questions we address in the present study are as follows. What is the range of model responses to a prescribed forcing? What are the key model components causing discrepancies in the results? And what kind of bias would these discrepancies introduce if such microphysical schemes were adopted in multidimensional models?

Seven parcel modeling groups from the United Kingdom, Germany, and the United States participated in the intercomparison, which took place at the GISSWG2 workshop in Geesthacht, Germany, in 1999. A brief description of the participant models is given in section 3.

Participants were asked to submit results of a series of relatively simple calculations involving an ascending parcel lifted by a superimposed updraft without mass and heat exchange with the environment. The primary focus was on the homogeneous nucleation process operating in isolation (HN-ONLY). A secondary focus was the combination of the homogeneous and heterogeneous ice nucleation mechanisms included in cirrus parcel models (ALL-MODE). A detailed description of the project is given in section 2. Cirrus initiation requirements and cloud properties are presented and the implications of the model results discussed in sections 4 and 5 followed by a conclusion in section 6. Notation is given in the appendix.

2. Simulation protocols

We focus on the nucleation regimes of the warm and cold cases studied in the WG2 ICMCP (Starr et al. 2000), and test the models under a range of imposed updraft conditions ($0.04, 0.2$, and $1$ m s$^{-1}$). All parcel models were run in the “closed” Lagrangian mode; that is, neither particle fallout nor mixing with the environment was allowed. Nucleation and ice crystal growth were forced through an externally imposed constant rate
of lift $W$ with consequent adiabatic cooling. The parcel cooling rate is formulated as

$$\frac{dT}{dt} = -\frac{g}{C_p}W + \dot{Q}_{\text{HI}}.$$  

(1)

where $\dot{Q}_{\text{HI}}$ is the latent heat release due to diffusional growth of the hydrometeors. The environmental pressure was specified as

$$\frac{d \ln P}{dz} = -\frac{g}{R_a[T_{\text{in}} - \Gamma(z - z_{\text{in}})]},$$  

(2)

where the environmental lapse rate $\Gamma$ approximates the ice pseudo-adiabatic lapse rate for the assumed conditions. The background haze particles\(^2\) are assumed to be aqueous sulfuric acid particles with a number concentration of 200 cm\(^{-3}\), lognormally distributed with a pure $\text{H}_2\text{SO}_4$ mode radius of 0.02 \(\mu\)m, a distribution width $\sigma$ of 2.3, and the $\text{H}_2\text{SO}_4$ density of 1.841 g cm\(^{-3}\). The initial conditions for the 14 runs are tabulated in Table 1. In each of the HN-ONLY runs (warm cases: Wh004, Wh020, and Wh100; cold cases: Ch004, Ch020, and Ch100), only the homogeneous nucleation mode is allowed to form ice crystals; the other mode is turned off. Both nucleation modes are switched on in ALL-MODE runs (Wa004, Wa020, and Wa100; Ca004, Ca020, and Ca100).

Many of the participant models used an empirical parameter $\lambda$ (to be discussed further in section 3a) to account for additional suppression ($\lambda > 1$) or enhancement ($\lambda < 1$) of the nucleation (freezing) temperature. Recent laboratory studies (Koop et al. 1998; Chen et al. 2000) implied a value close to 2 for $\text{H}_2\text{SO}_4$ solution. Participants were asked to run two additional simulations, Wh020L and Ch020L, by setting $\lambda = 2$, or tailoring the nucleation rate calculation so that the predicted freezing is in agreement with $\lambda = 2$. This comparison served as a tool to diagnose the relative importance of nucleation rate versus the treatment of haze and ice particle growth in determining the source of model differences in the predicted ice particle number concentration $N_i$.

Heterogeneous nucleation could be the dominant ice formation mode in cirrus in slow updraft conditions (e.g., DeMott et al. 1997; Spice et al. 1999; Sassen and Benson 2000). However, significant differences exist in the treatment of heterogeneous ice nucleation processes. Thus, major intermodel differences might be expected in the predicted cirrus microphysical properties. Definitive laboratory and atmospheric benchmark data are only now being developed. Despite the still primitive nature of this subject, it is important to compare the presently used schemes and assess their features.

The assumed shape of ice crystals may also affect the simulations. In order to simplify the comparison, participants assumed ice crystals to be spherical with constant ice density of 0.9 g cm\(^{-3}\). The bulk ice density assumption may be an acceptable approximation for the small crystal sizes present in most of the simulations (e.g., Heymsfield and Sabin 1989), but it is expected that the simulation results will be sensitive to this assumption. Participants also assured that time steps were small enough and that the aerosol and ice crystal size distributions were adequately resolved to minimize numerical artifacts. For the present calculations, ice particle aggregation and direct radiation effect on diffusional growth are ignored.

The depth of parcel lift used in this study was set to ensure that parcels underwent complete transition through the nucleation regime to a stage of approximate equilibrium between ice mass growth and vapor supplied by the specified updrafts. This was deemed to provide the best comparison of the models and their sensitivities to critical parameters.

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\(^2\) A similar aerosol number distribution was used in Jensen et al. (1994a).

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**Table 1. Simulation identifiers and initial conditions.** \(\text{RH}_{\text{ini}} = 100\%\) in every simulation.

<table>
<thead>
<tr>
<th>Simulation identifiers</th>
<th>Height (z_{\text{ini}}) (km)</th>
<th>Pressure (P_{\text{ini}}) (mb)</th>
<th>Temperature (T_{\text{ini}}) (°C)</th>
<th>Updraft (W) (m s(^{-1}))</th>
<th>Lapse rate (\Gamma) (°C m(^{-1}))</th>
</tr>
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<tr>
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<td></td>
<td></td>
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<tr>
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<td>340</td>
<td>-40</td>
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<tr>
<td>Cold cases*</td>
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<tr>
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<td>170</td>
<td>-60</td>
<td>0.04</td>
<td>0.0097</td>
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<td>0.0097</td>
</tr>
<tr>
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<td>170</td>
<td>-60</td>
<td>0.20</td>
<td>0.0097</td>
</tr>
</tbody>
</table>

* Here, $\lambda$, if any, is determined by the participants.

** Here, $\lambda = 2$. Note that model J incorporates parameters adjusted to the laboratory data of Koop et al. (1998) in all $\lambda$-fixed and HN-ONLY cases.
3. Model descriptions

A brief summary of the model characteristics is given in Table 2. Hereafter, we will refer to these models as the C (Cotton), D (DeMott), J (Jensen), K (Kärcher), L (Lin), S (Sassen), and X (X. Liu) models, respectively, as denoted in the table. According to the method in which particles are handled, these models are classified as either particle-tracing models or bin models (Young 1993). In the particle-tracing models, the evolution of each individual particle is traced (models S and X). In the bin models (models C, D, J, K, and L), particles are grouped according to particle size or mass to expedite numerical integration. Individual particle histories are lost in the grouping process. Among the seven models, J and L have been incorporated into fully coupled multidimensional models (Jensen et al. 1994a,b; Lin 1997). The major components in parcel models, from a numerical standpoint, are the time-integration scheme and size-coordinate scheme. From the standpoint of cloud physics, the major components are the specification and treatment of the saturation vapor pressure, homogeneous and heterogeneous nucleation schemes, diffusional growth of haze and ice particles, and haze particle size and solution concentration (Fig. 1). The latter three components are highlighted in this section.

a. Homogeneous nucleation of unactivated solution particles

Much progress has been made in estimating the homogeneous nucleation rate of ice in pure water droplets $J_w$ by theoretical, laboratory, and field measurement studies (e.g., Sassen and Dodd 1988; DeMott and Rogers 1990; Pruppacher 1995; Jeffery and Austin 1997). The nucleation rate $J_w$ can be formulated theoretically or fitted parametrically to droplet temperature only (e.g., Heymsfield and Sabin 1989; Jeffery and Austin 1997; Khvorostyanov and Sassen 1998b). The estimate of the nucleation rate of ice in solution droplets $J_{\text{haze}}$, however, remains an active research area. This is critically important for cirrus modeling because many cirrus clouds form at temperatures colder than $-40^\circ\text{C}$ (Sassen 2002). In this temperature regime, haze particles freeze at relative humidities below saturation with respect to the liquid phase, where liquid water exists only as a component of solution droplets.

Model J computes $J_{\text{haze}}$ using the equation (Pruppacher and Klett 1997)

$$J_{\text{haze}} = 2N_s \left( \frac{\sigma_{\text{s-s}}}{\rho h} \right)^{\frac{1}{2}} \exp \left[ - \frac{(\Delta F_{\text{act}} + \Delta F_{\text{germ}})}{B T} \right],$$  \hspace{1cm} (3)

where $\sigma_{\text{s-s}}$, $\Delta F_{\text{act}}$, and $\Delta F_{\text{germ}} = (4\pi\sigma_{s}/3)(2\sigma_{s}/T\ln(T_{s}/T) + (1/2)(T_s + T)R_s \ln a_s)$ are the surface tension across the ice-solution interface, the activation energy, and the ice-germ formation energy, respectively. All notation is defined in the appendix. Solution effects, primarily compositional effects on $\sigma_{\text{s-s}}$, serve to raise the critical germ radius and the required germ formation energy. In order to use (3), laboratory data on $\sigma_{\text{s-s}}$ and $\Delta F_{\text{act}}$ as functions of temperature and composition are needed. Seldom are both quantities available from direct measurements for a given solution. Typically, direct data on one parameter is used and the other is constrained by measurements of $J_{\text{haze}}$. Thus, in model J, recent direct data on $\sigma_{\text{s-s}}$ was incorporated and $\Delta F_{\text{act}}$ was inferred from laboratory measurements of $J_{\text{haze}}$ for H$_2$SO$_4$ haze particles following the method of Tabazadeh et al. (2000).

The $T_{\text{eff}}$ approach (models C, D, L, S, and X) attempts to directly link measured $J_{\text{haze}}$ to nucleation rates of equivalent-sized pure water droplets via a parameter called the effective freezing temperature. $T_{\text{eff}}$ is defined as

$$T_{\text{eff}} = T + \lambda \Delta T_m,$$  \hspace{1cm} (4)

as introduced by Sassen and Dodd (1988) such that

$$J_{\text{haze}} = J_w(T_{\text{eff}}).$$  \hspace{1cm} (5)

In (4), $\Delta T_m$ is the equilibrium melting point depression (positive valued), which is a function of solution concentration, and $\lambda$ is an empirical coefficient to account for additional suppression/enhancement of nucleation temperature due to nonideal interaction between ions and condensed water. When using the $T_{\text{eff}}$ method, the nucleation rate is extremely sensitive to $\Delta T_m$ and $\lambda$; thus, these two factors must be specified with care. The values of $\lambda$ are constrained by, and in fact have their source in, laboratory data as first described by Sassen and Dodd (1988). Chen et al. (2000) noted that, for H$_2$SO$_4$ solution, $\lambda = 2$ agreed with their own data and those presented by Koop et al. (1998). Although Sassen and Dodd (1988) noted that an average $\lambda$ for different solutions was around 1.7, the values for specific solutions may range from less than 1 to about 2.5 (e.g., Cziczo and Abbatt 1999; Chelf and Martin 2001; DeMott 2002). The existence of relation (4) as a descriptor of laboratory data on the freezing of a large variety of solution droplet types has never been explained theoretically from first principles.

Model K adopted the theory developed by Koop et al. (2000) to describe $J_{\text{haze}}$ as a function of $\Delta a_w$, the difference between the water activity in a solution $a_w$ and the water activity in the solution in equilibrium with ice. This theory implies that the required RH$_w$ for homogeneous nucleation is not species dependent, at least for micron-sized particles in the equilibrium condition. It can be shown that $\Delta T_m$ can be uniquely determined by $a_w$ (e.g., Pruppacher and Klett 1997). Given temperature and solute weight percent (wt%), the theory of Koop et al. (2000) may be interpreted as a $T_{\text{eff}}$ scheme in which $\lambda$ is species independent and $\lambda$ is close to 2. This theory was established from laboratory studies (mostly using the emulsion technique) of homogeneous nucleation in micron-sized droplets of 18 different (14

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Table 2. Participant cirrus parcel models.

<table>
<thead>
<tr>
<th>Organization investigator</th>
<th>U.K. Met Office (UKMO) Cotton (C)</th>
<th>Colorado State University (CSU) DeMott (D)</th>
<th>Ames Research Center (ARC) Jensen (J)</th>
<th>German Aerospace Center (DRL) Kärcher (K)</th>
<th>Goddard Space Flight Center (GSFC) Lin (L)</th>
<th>University of Utah Sassen (S)</th>
<th>University of Michigan X. Liu (X)</th>
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</thead>
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<td>10 W⁻¹ (W in cm s⁻¹)</td>
<td>Adjusting 10⁻¹ to 0.02</td>
<td>5 W⁻¹ (W in cm s⁻¹)</td>
<td>0.02</td>
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<td>r_{eq}</td>
<td>r_{eq} or \frac{dr}{dt}</td>
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<td>r_{eq}</td>
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<td>No</td>
<td>Yes</td>
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<td>Yes</td>
</tr>
<tr>
<td>\lambda</td>
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<td>—</td>
<td>1.0</td>
<td>1.7</td>
<td>2.0</td>
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<td>Immersion freezing nucleation</td>
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</table>

a Discrete vs continuous binning indicates the assumption that particles have exactly the same size in a bin or allowing a certain size distribution of particles in a bin.

b Using the moving-center structure as defined in Jacobson (1999).

c Note that r_{eq} vs \frac{dr}{dt} denotes either using the equilibrium-sized haze approximation or computing the diffusional growth of haze particles explicitly.

d The native value of λ assumed in the model.

e ε_{eq} (using the Clausius–Clapeyron equation) error is less than 0.5%; ε_{ Magnus} (using the Magnus equation) 3%, 0.5% error with respect to Goff–Gratch at 208 K, 228 K, respectively.

f β is the deposition coefficient.

g A detailed description of this model will be published elsewhere.
inorganic and 4 organic) aqueous solutions. As shown in Fig. 1b of Koop et al. (2000), on average, $\Delta a_{w}$ is the primary controlling factor for homogeneous nucleation, but the data points of $a_{w}$ under critical freezing conditions spread within a range of $\pm 0.025$. It is important to note that a difference in $a_{w}$ of a magnitude of 0.05 may result in a difference in $\log J_{haze}$ of a magnitude of $\sim 12$. This magnitude of difference in $\log J_{haze}$ is comparable to a change in $T_{eff}$ of about 7–8 K. Such a change in $T_{eff}$ can be achieved by varying $\lambda$ (Fig. 2).

In either case described above, calculations depend critically on the use of laboratory data describing properties of solution particles at cirrus temperatures. The homogeneous nucleation rates for H$_2$SO$_4$ solution drop-

![Fig. 2. Melting point depression ($\Delta T_{m}$) of H$_2$SO$_4$ solution as a function of solute wt% from the tabular data of Lide (1998). The solid and unfilled diamond markers denote tabular data points from pp. 8–81 and pp. 15–22, respectively. Also plotted are curves of $\lambda \Delta T_{m}$ for $\lambda = 1, 1.5, 1.7,$ and 2.0. The solid and unfilled right triangle (circle) markers indicate inferred $\lambda \Delta T_{m}$ from model J (K) at $T = -65^\circ C$ and $-45^\circ C$, respectively.]

*Fig. 1. The flowchart of a typical cirrus parcel model.*
lets computed by these methods are compared as functions of temperature in Fig. 3. In this figure, the same equilibrium-sized haze particle data provided by Paul DeMott are used in each model where $\lambda$ is set to 2 and the fitted $\Delta T_m$ by DeMott et al. (1997) is adopted. The curves for models using the $T_{\text{eff}}$ approach agree fairly well, for example, $1^\circ\text{C}$ difference in temperature where $J_{\text{haze}} V = 1 \text{ s}^{-1}$ for a 5% weight percent solution droplet. This is not surprising because, as long as these schemes use an identical value of $\lambda$ and the same formula to compute $\Delta T_m$, the differences in nucleation rate are solely determined by the differences in the formulae used to calculate $J_{\alpha}$. These calculations suggest that, at the critical freezing condition ($J_{\text{haze}} V = 1 \text{ s}^{-1}$), the formulation differences in $J_{\alpha}$ among the $T_{\text{eff}}$ group at most contribute an absolute 1% difference in the critical solute wt%.

Compared with the $T_{\text{eff}}$ models, model K predicts a smaller $J_{\text{haze}} V$, especially as wt% increases. Note that in order to plot the curves for model K, we have to first compute $a_\alpha$ of the given solute wt% using the formula provided by model K. Therefore, the curves denoting model K contain information not only about the specification of $J_{\text{haze}}$, but also about the specification of $a_\alpha$.

The classical approach to determining $J_{\text{haze}}$, as in model J, can be interpreted as a $T_{\text{eff}}$ scheme with varying $\lambda$. Given the same temperature, the classical approach would obtain smaller $J_{\text{haze}} V$ compared to the $T_{\text{eff}}$ approach when the solution is dilute (e.g., $T = -45^\circ\text{C}$, wt% $= 5$). However, the opposite is found when the
solution is more concentrated (e.g., \( T = -45^\circ\text{C}, \text{wt}\% = 15 \)). For the same solute \text{wt}\%, model J produces larger \( J_{\text{nuc}} V \) than models using the \( T_{\text{eff}} \) approach when the temperature is warm (e.g., \( T = -35^\circ\text{C}, \text{wt}\% = 5 \)), but produces less as temperature decreases (e.g., \( T = -45^\circ\text{C}, \text{wt}\% = 5 \)). Therefore, the intrinsic \( \lambda \) in the classical approach varies inversely with solution concentration and temperature (Fig. 2). Furthermore, the inferred \( \Delta T_m \) from model J at these two temperature regions does not approach zero as solute \text{wt}\% decreases to zero because model J tends to underestimate \( J_{\text{nuc}} \) in dilute solutions in comparison to other models. This divergence may be a potential problem when \( T \) is close to or warmer than \(-40^\circ\text{C}\).

For a given temperature, \( J_{\text{nuc}} V \) is highly sensitive to the haze particle solute \text{wt}\% (Fig. 3). This suggests that modeling the evolution of particle solute \text{wt}\% is a critical component of the models. However, the gradient of \( J_{\text{nuc}} V \) to solute \text{wt}\% in the \( T_{\text{eff}} \) models and model K is much greater than model J. For instance, at \(-45^\circ\text{C}, J_{\text{nuc}} V \) computed by model J ranges roughly from \( 10^{-8} \) to \( 1 \text{ s}^{-1} \) for solute \text{wt}\% from 15\% to 5\%; whereas, the \( T_{\text{eff}} \) approach models render a range from \( 10^{-21} \) to \( 10^{-15} \text{ s}^{-1} \) for the same solute \text{wt}\% range. In the next section, we will show that this difference in the sensitivity of \( J_{\text{nuc}} V \) to solution concentration is a dominant factor leading to the systematic differences in the freezing haze size distributions.

It is not known if the behavior predicted by the classical nucleation theory as treated in model J is a consequence of the way that theory was constrained by a single laboratory dataset, or if it reflects a real expected consequence of solution effects on freezing. Laboratory measurements usually focus on the \( J_{\text{nuc}} V = 1 \text{ s}^{-1} \) region. Nucleation rate data over a wide range of values, particularly data points beyond the critical freezing condition, are needed for solutions of similar composition to address this issue (e.g., Chen et al. 2000).

The intermodel range of \text{wt}\% corresponding to a given \( \Delta T_m \) might be used to estimate the range in the triggering \( \text{RH}_m \) condition, to a first-order basis. If \( \lambda \) is the only varying component, it is expected that homogeneous nucleation is first triggered in the model with the smallest \( \lambda \) because particles freeze at a greater solute \text{wt}\%, which entails a lower \( \text{RH}_m \) condition. It is shown from Fig. 2 that this \text{wt}\% range increases with \( \lambda \Delta T_m \). Meanwhile, the value of \( \lambda \Delta T_m \) necessary to trigger nucleation increases with a decrease in temperature.

As a consequence, the nucleation triggering \( \text{RH}_m \) becomes smaller and more sensitive to \( \lambda \) as temperature decreases. This will be further demonstrated in the model results (section 4).

b. Heterogeneous nucleation

Little constraint was imposed on formulating heterogeneous nucleation because theoretical and experimental understanding is still quite poor. For the ALL-MODE simulations, both parameterized and explicit heterogeneous nucleation calculations are used. Models C and L employ ice saturation ratio dependent parameterizations of activated IN as

\[
N_{\text{IN}} = A(S_i - 1)^\alpha, \tag{6}
\]

following Spice et al. (1999); and

\[
N_{\text{IN}} = \exp[\alpha + 100\beta(S_i - 1)], \tag{7}
\]

following Meyers et al. (1992), respectively. Note that the values of the coefficients mentioned in this section are listed in the appendix. Hence, number concentrations of ice crystals produced by heterogeneous nucleation are controlled solely by the peak \( S_i \) in the simulations.

At relatively high peak \( S_i \) conditions, the parameterized \( N_{\text{IN}} \) by these two formulas may exceed the range of \( 10^{-4} \) to \( 0.1 \text{ cm}^{-3} \) measured by Rogers et al. (1998) from aircraft; for example, \( N_{\text{IN}} \) becomes 1 \text{ cm}^{-3} when \( S_i \) is 1.38 [Eq. (6)] or 1.58 [Eq. (7)]. Although most of the flights were spent at temperatures between \(-35^\circ\text{C} \) and \(-60^\circ\text{C}, Rogers et al. (1998) processed aerosols at conditions from about \(-15^\circ\text{C} \) to \(-40^\circ\text{C} \) and from ice saturation to \(-15\% \) water supersaturation. The difference between the environmental and processing temperatures may introduce some uncertainty in interpreting the measured \( N_{\text{IN}} \). Nevertheless, the formulas [Eqs. (6) and (7)] are expected to represent a maximum heterogeneous nucleation impact.

Models D and S treat heterogeneous freezing in an explicit fashion. Haze particles of the given \( \text{H}_2\text{SO}_4 \) aerosol distribution are subject simultaneously to heterogeneous and homogeneous nucleation. The unfrozen haze particles are tracked during the entire simulation. Model D assumes that all heterogeneous IN are freezing nuclei contained within 10\% of the CCN population at sizes above 0.1 \text{ m}. Furthermore, these particles are assumed to contain 50\% insoluble matter (bulk density of 1.9 \text{ g cm}^{-3}), the rest being \( \text{H}_2\text{SO}_4 \). These choices are based upon observations from DeMott et al. (1998) at temperatures down to \(-35^\circ\text{C} \). The average freezing nuclei spectrum from that study was employed and extrapolated to cirrus temperatures in this study. Thus, the fraction of IN from each haze category \( k \) (>0.1 \text{ m}) were predicted according to

\[
F_k = \frac{a}{F_{>0.1\mu m}}(T_0 - T_{\text{eff}})\beta, \tag{8}
\]

where \( F_{>0.1\mu m} \) is the fraction of CCN with size greater than 0.1 \text{ m} compared to the total CCN. This treatment was expected to yield the most conservative (minimal) estimate of heterogeneous IN concentrations in cirrus.

Model S assumes that the ice particle formation due to heterogeneous nucleation follows a \( T_{\text{eff}} \) modified Fletcher function given by Khvorostyanov and Sassen (1998a) as

\[
\frac{dN_{\text{IN}}}{dt} = \sum_k A_k B_k \exp(-A_k z) \exp[B_k(T_0 - T_{\text{eff}})] dT, \tag{9}
\]
This equation introduces a height-dependent term into the concentration of IN in the upper troposphere. A larger than normal value of $B^\star$ [0.8 compared to 0.4–0.6 of the “typical cirrus” in Sassen and Benson (2000)] is arbitrarily used to simulate the condition in which heterogeneous nucleation competes most favorably. Note that model S, using the particle tracing scheme, calculates the evolution of a total of 20 000 individual particles, corresponding to a parcel of volume 100 cm$^3$ at the starting point of simulation. Therefore, the minimum detectable change in $N^r$ is $\sim 0.01$ cm$^{-3}$. Parcels of greater volume, which would allow for the detection of smaller changes in $N^r$, are recommended for future research.

c. Condensation and deposition coefficients

Condensation and deposition coefficients are critical parameters to assess the growth rate of aerosol particles, newly activated cloud droplets and ice crystals. The condensation coefficient is defined as the ratio of the number of vapor molecules absorbed by water over the number of vapor molecules impinging on the water surface. Analogously, the deposition coefficient may be defined for the ice surface. Laboratory experiment and field measurement derived values of these two parameters scatter over a range from about 0.01 to 1.0 (for condensation coefficient: Table 5.4 of Pruppacher and Klett (1997); Bonacci et al. (1976); Shaw and Lamb (1999); Marek and Straub (2001); Li et al. (2001); for deposition coefficient: Table 5.5 of Pruppacher and Klett (1997); Bonacci et al. (1976); Haynes et al. (1992); Heymsfield and Miloshevich (1998)).

Although consensus about the reason for the divergence of reported values has not been established, guidelines can be found from these published data. In general, the condensation coefficient declines with increasing temperature and pressure, and decreases with time possibly due to the dynamic surface tension (Bonacci et al. 1976; Marek and Straub 2001). Contamination may also affect the condensation coefficient significantly. Since aerosol particles and newly activated droplets are not pure water, the condensation coefficient for pure water is probably not the best choice to suit our purpose.

Recall that this CPMCP is limited to idealized nonfaceted ice crystal growth. In general, the deposition coefficient also declines with increasing temperature (Haynes et al. 1992; Kossacki et al. 1999). Impurity may similarly affect the deposition coefficient. For a faceted crystal, the deposition coefficient at a given location on a crystal surface depends on temperature and the spatially varying surface supersaturation (Wood et al. 2001).

It will be shown in later sections that the model predicted $N^r$ is extremely sensitive to these coefficients, especially the deposition coefficient. We advise that future studies choose these coefficients with care.

d. Haze particle size and solution concentration

The volume of any haze particle is given by

$$ V = m_s \left( 1 + \frac{1000}{M_s \rho_s} \right), $$

where $M$, $m_s$, and $\rho_s$ are the molality (mol of solute per 1000 g of water), solute mass, and solution density, respectively, in cgs units. Within the temperature and moisture ranges considered in this study, the volume of equilibrium-sized haze particles of a given solute mass varies within a factor of 10. Following Eq. (10) and Fig. 3, the sensitivity of $J_{eq}$ to $V$ is far less than to solution molality, temperature, and $\lambda$. However, if haze volume or radius is the prognostic or diagnostic variable in a model and the water mass of haze is derived by $m_w = \rho_s V - m_s$, solution density is not a trivial issue. The difference in $\rho_s$ would significantly affect the obtained solute wt%. Measurements of the H$_2$SO$_4$ solution density for temperatures below 0°C were not available until the work by Myhre et al. (1998). They showed that extrapolation of existing $\rho_s$ data, which were measured above 0°C, matches well with their measurements. Participants of this project used different formulae for solution density, explicitly or implicitly$^1$ (Fig. 4).

Upper troposphere haze particles in the typical cirrus initiation conditions are in equilibrium with the environment except (i) when temperature is warmer than $-42^\circ$C and the required RH$_w$ to trigger homogeneous nucleation is close to, or greater than, 100% (Heymsfield and Sabin 1989); and (ii) in a strong updraft associated with a cold environment (to be discussed more in the following). When equilibrium is ensured, the haze particle radius $r$ can be obtained by iteration either using (e.g., models D and L)

$$ S_w = a_w \exp \left( \frac{2\sigma_{\text{vs}}}{\rho_s R_s T_r} \right), $$

where $a_w = \exp(-\nu \phi W M_s / m_a M_w)$, or the dilute solution approximation (ln $S_w = S_w - 1$; $\Phi_w = 1$; $m_a \gg m_w$; $\rho_w \approx \rho_s$, e.g., models C, K, S, $^4$ and X)

$$ S_w = 1 - \frac{3m_w M_w M_s}{4\pi\rho_s r^5} + \frac{2\sigma_{\text{vs}}}{\rho_s R_s T_r}. $$

Using a temperature-independent $a_w$ formula (Chen 1994), the haze equilibrium radius in model L is not

$^1$ In model X, $\rho_w \approx \rho_s + (\rho_d - \rho_s)(r/r_d)^{3}$, where the subscript $d$ denotes dry aerosol. The temperature dependency of $\rho_d$ has been taken into account such that $\rho_d$ becomes less than 1 g cm$^{-3}$ below 0°C.

$^4$ Model S calculates the equilibrium haze radius using Eq. (50) of Fukuta and Walter (1970) and a Van’t Hoff coefficient of 3. Model S renders equilibrium haze solute wt% close to that of model C, which sets $\phi = 1$ and $\nu = 3$. Model K uses the parameterization of the water vapor saturation pressure as proposed by Luo et al. (1995), but with slightly modified parameters resulting in values of the water activities close to models C, S, and X.
very sensitive to temperature within the cirrus temperature range (−45°C to −65°C) considered in this project, especially for haze with solute mass greater than $10^{-15}$ g. In other words, Kelvin’s effect (curvature effect) is controlled by the value of solute mass and is not sensitive to a 10% change in either temperature nor haze surface tension $\sigma_{sw}$ within this temperature range. Using a temperature-dependent $\alpha_w$ formula (Clegg and Brimblecombe 1995), the required $S_w$ for the equilibrium condition in models D and J fluctuates within ±0.005 in this temperature range.

Figure 5 compares solute wt% computed by models C [Eq. (12)] and L [Eq. (11)] as solid and dashed lines, respectively. The obtained equilibrium haze solute wt% by model C can be a few percent smaller (larger) than that obtained by model L in high (low) ambient humidity RH$_w$ conditions. The two families of curves gradually diverge as RH$_w$ decreases to less 85%. Note that substantial differences in solute wt% exist even at RH$_w$ ~ 95%, probably because the assumption that $\Phi_v = 1, \nu = 3$ in the dilute approximation deviates significantly from the $\alpha_w$ used by models D and L.

The curves for models S and X can be roughly estimated by connecting the pairs of same-solute-mass data points plotted in Fig. 5. It is apparent that models C, S, and X predict relatively similar equilibrium haze solute wt% when humidity is high. However, the curves diverge as humidity decreases. This trend occurs due to differences in solution density specification in the three models and demonstrates the importance of $\rho_v$ when acquiring equilibrium solution concentration from haze radius. This result has obvious consequences on $\lambda \Delta T_m$, and thus, nucleation rate. In cases where haze particles are approximately in equilibrium with the environment [discussed further in section 4a(1)], Fig. 5 may provide useful information about the nucleation triggering RH$_w$.

Multidimensional models cannot afford to iterate because iteration is extremely time consuming. Thus, further simplification is required for parcel models that are modules of multidimensional models. Model L uses fitted functions based on Eq. (11). Model J neglects Kelvin’s effect and sets $\alpha_w = S_w$, for which the equilibrium haze solute wt% becomes only a function of the environment $S_w$. As shown in Fig. 5, Kelvin’s effect results in 1% to 2% differences in solute wt% between haze containing solute weights of $10^{-15}$ and $10^{-15}$ g. This is a significant difference (Fig. 3). Hence, ignoring Kelvin’s effect enhances the freezing possibility of particles with small solute mass and leads to the broadening of the freezing haze particle size distribution.

![Figure 4. H$_2$SO$_4$ solution density as a function of solute wt%. Also shown in the figure is H$_2$SO$_4$ solution density at −45°C fitted by Myhre et al. (1998). The tabular data of CRC Handbook of Chemistry and Physics for H$_2$SO$_4$ solution density at 20°C matches well with the curve of model D.](image-url)
The equation for diffusional growth of haze particles is given by Fukuta and Walter (1970) as

$$\frac{dm_w}{dt} = \frac{4\pi r(S_w - S_w^\infty)}{R_e T} + \left(\frac{L_v}{R_e T} - 1\right) \frac{L_v}{TK} S_w^\infty,$$

where $S_w^\infty$ is obtained by substituting the instantaneous haze radius and solute wt% into Eqs. (11) or (12), depending on the model. At equilibrium, $S_w$ equals $S_w^\infty$. The heat transfer contribution term (second term in the denominator) is more than an order of magnitude smaller than the mass transfer term (first term in the denominator) for the temperature range considered here. Thus, the following discussion considers only on the effect of the mass transfer term and its sensitivity to the condensation coefficient $\beta_w$. Given $S_w$ and $r$, the diffusional growth rate decreases exponentially with temperature, reflecting the strong temperature dependence of saturation water vapor pressure. The response timescale to the deviation from equilibrium can be considerably greater than one single model time step in a cold environment. The stronger the updraft, the wider the gap between these two timescales may be. Therefore, haze particles with large solute mass become more concentrated than the corresponding equilibrium-sized particles in a cold environment and strong updraft conditions.

Haze particle growth rate is sensitive to $\beta_w$, which modifies the value of $D'$ and has to be included with caution. Both models C and S use $\beta_w = 0.05$ resulting in considerable delaying of haze growth in several simulations. In contrast, diffusional growth of haze particles does not cause noticeable effects in models K and L (e.g., Ch020L) because a much larger $\beta_w$ is used (0.5 and 0.7 for models K and L, respectively).

Figure 6 shows the haze solute wt% as a function of solute mass for model C at a height level just below $z_h$ in the 0.04 m s$^{-1}$ updraft simulation. Note that the height at which homogeneous nucleation is triggered increases with updraft speed. Large haze particles deviate from the equilibrium condition (approximately depicted by the 0.04 m s$^{-1}$ curve) as the strength of updraft increas-
es. The haze particles that are the most dilute are not necessarily the largest ones when the limit of diffusional growth is imposed (e.g., the Ch100 curve). In such cases, it is expected that nucleation starts with the haze particles of a size between the most dilute ones and the largest ones to take into account the change of $J_{\text{haze}}$ and $V$ with respect to $m_s$. In contrast, the deviation from equilibrium is not as significant in warm cases. Haze solution concentration is close to being in equilibrium with the environment in slow and moderate updraft cases (Wh004 and Wh020). Note that $\pm 1\%$ in wt$\%$ at wt$\% = 20\%$ results in approximately a $3^\circ C$ difference in $T_{\text{eff}}$ when $\lambda = 2$ (Fig. 2).

4. Results of the baseline simulations

A parcel lifted by an updraft goes through four stages (Fig. 7). In the first stage, the parcel cools adiabatically and expands. Haze particles grow, but uptake of vapor by these particles is negligible.

The nucleation regime starts as the first ice crystals form. We arbitrarily define the level at which ice particle number concentration $N_i$ reaches $10^{-3} \text{ cm}^{-3}$ as the beginning of the nucleation regime or cirrus cloud base $z_b$ (relative to the parcel initial height $z_{\text{ini}}$; Table 1). At the beginning of this stage, the RH$_i$ tendency $d\text{RH}_i/dt$ is positive but decreasing in magnitude due to vapor uptake by growing ice crystals. Homogeneous nucleation, if activated, shuts off shortly after RH$_i$ reaches its peak value. It is important to note that the homogeneous nucleation regime occurs within a relatively small range of vertical displacement of the parcel.

The next regime features a rapid decrease in RH$_i$ corresponding to the rapid increase of ice water content (IWC). RH$_i$ tendency starts from zero, falls to a minimum, then increases asymptotically back to zero. The
equilibrium regime begins when $d\text{RH}_i/dt \sim 0$. In general, the parcel’s temperature tendency $dT/dt$ is close to ice moist adiabatic in the equilibrium regime, although this may depend on the value of RH, at equilibrium (hereafter, RH$_{eq}$).

Comparison of $N_i$ generated by the different models is the centerpiece of this project (Fig. 8) when values of $N_i$ at 800 m above $z_{eq}$ are given. As illustrated in Fig. 7, the equilibrium regime is well established and little additional nucleation is occurring by this time. Temperature differences among models are negligible at this level. In the HN-ONLY cases, to a first-order approximation, the logarithm of ice crystal number concentration increases quasi-linearly with the logarithm of updraft speed within the investigated updraft range. An increase of $N_i$, with $W$ was first demonstrated by Heymsfield and Sabin (1989) and Sassen and Dodd (1989) and followed by others. This figure demonstrates that while all models manifest this positive trend of $N_i$ with $W$, the quantitative differences among models cannot be overlooked. The predicted $N_i$ by models J and C tends to form the lowest and highest bounds in the six HN-ONLY cases, respectively. There is as much as a factor of 25 difference in $N_i$ among the models. Generally, more ice crystals are produced in the cold cases. This tendency was also first reported by the aforementioned pioneer works. Jensen and Toon (1994) conjectured that the inverse trend of ice crystal growth rate with temperature was the primary cause.

The predicted $z_b$, RH$_{z_b}$, and $\Delta$RH, (peak RH, minus RH$_{z_b}$) increase with updraft speed in the HN-ONLY cases (Fig. 9). Cirrus forms in a narrower range of altitude in the warm cases ($360 < z_b < 400$ m; Fig. 9b) than in the cold cases ($325 < z_b < 410$ m; Fig. 9a). If the simulation stops at an altitude that is coincidentally between the minimum cloud-base height and the maximum nucleation-ceasing altitude of all models, the discrepancy in the predicted $N_i$ would be greatly enhanced, consistent with the research of Lin et al. (1998) on the sensitivity of cloud properties in lift-limited situations.

The corresponding range of RH$_{z_b}$ in the warm cases is also smaller than that in the cold cases. The primary factor increasing the sensitivity of the cloud-base RH, as temperature decreases in the five $T_{eff}$ models was discussed in section 3a: the range of critical freezing RH$_w$ corresponding to a given range of $\lambda$ increases with
Fig. 9. The RH, at cloud base $z_b$, and the corresponding $\Delta RH_i$, defined as the difference between peak RH, and RH, at cloud base $z_b$, for HN-ONLY simulations.

decreasing temperature. For a given updraft velocity, the order of RH, from low to high is L (1.0), C (1.5), S (1.7), X (2.0) ~ D (1.5) in the warm cases, and L (1.0), D (1.5), C (1.5), S (1.7) ~ X (2.0) in the cold cases, where the numbers in parentheses denote $\lambda$. Thus, a smaller $\lambda$ generally entails a lower cloud-base height. Other factors (e.g., differences in solution density; specification of haze particle solution concentration; saturation water vapor pressure over water) may also affect RH. For example, models C and D using $\lambda = 1.5$ obtain different RH, in the warm case because, for 94% < RH, < 99%, haze particles are more concentrated in model D (Fig. 5).

The predicted $z_b$, RH, and peak RH, in the ALL-MODE cases vary even more because of our respective unbounded choices of heterogeneous nucleation (Fig. 10). The impact of heterogeneous nucleation on lowering cloud formation altitude, peak RH, and $N_h$ is extremely sensitive to the onset conditions for nucleation and the subsequent rate of ice formation.

The threshold humidity for heterogeneous nucleation of ice in a water-subsaturated and ice-supersaturated environment may be much less than that for homogeneous nucleation. If the upward motion is confined within a thin layer, in which the maximum humidity that may be achieved by an ascending parcel is between the threshold humidity of the two modes, the presence of IN will determine if cirrus forms or not. If the vertical displacement of parcels is large enough to provide the necessary condition to trigger homogeneous nucleation, the presence of IN may partially or completely suppress homogeneous nucleation and change the cloud properties depending on IN characteristics (Sassen and Benson 2000). With heterogeneous nucleation, the peak RH, is lower in all cases (except Wa100 by model S, possibly due to the height dependency of the IN concentration).
Here we define the $N_i$ reduction ratio as the $N_i$ of the ALL-MODE case over that of the corresponding HN-ONLY case at 800 m (Table 3). Except for S (Ca004), activation of the heterogeneous nucleation process reduces the value of $N_i$ in the equilibrium regime. Generally speaking, the largest difference in $N_i$ between the ALL-MODE and HN-ONLY simulations takes place when the updraft speed is slow to moderate and homogeneous nucleation is completely suppressed (Fig. 8 and Table 3); the difference is small when the updraft speed is fast. Therefore, although wave clouds may be appropriate for the study of homogeneous nucleation, they do not represent cirrus very well because wave clouds are generally subject to an intense lifting and cooling. Complete suppression of homogeneous nucleation is achieved when the uptake of excess water vapor by ice crystals formed by heterogeneous nucleation sufficiently lowers the maximum humidity achieved by the parcel. Hence for a given cooling rate, a larger difference between the threshold humidity of the two modes would provide particles more time to grow and thus a more favorable condition for suppression (e.g., models C and L). The difference in $N_i$ between ALL-MODE and HN-ONLY cases by model D is only prominent in the warm and slow updraft case (Wa004). The weak influence of heterogeneous nucleation in model D is tied to two factors. First, the maximum IN concentration predicted by the formulation are no more than a few hundred per liter in the range of conditions simulated. Second, the lowering of the onset RH for ice formation is limited by solute concentration via the use of the effective freezing temperature in the immersion freezing mechanism. This directly relates to the conservative estimate of the concentrations of available heterogeneous IN made in model D.

The $N_i$ reduction ratios of model S are significantly higher in the 0.04 m s$^{-1}$ cases. The predicted $N_i$ in Ca004 is even larger than that of Ch004; $N_i$ of Ca004 increases exponentially with height from 200 m above the starting level (not shown). It is possible that the anomalous behavior for this single model run results from the large value of $B_s$ used. However, this large $B_s$ is the only value capable of producing significant heterogeneous nucleation, in view of the height-dependent term in Eq. (9). More study is required to understand this anomaly.

### Table 3. $N_i$ reduction ratio that is defined as $N_i$ of the ALL-MODE divided by $N_i$ of the HN-ONLY simulation, $N_i^{\text{ALL-MODE}}/N_i^{\text{HN-ONLY}}$.

<table>
<thead>
<tr>
<th>Model simulation</th>
<th>$N_i$ reduction ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>C</td>
</tr>
<tr>
<td>Ca004</td>
<td>0.039</td>
</tr>
<tr>
<td>Ca020</td>
<td>0.114</td>
</tr>
<tr>
<td>Ca100</td>
<td>0.791</td>
</tr>
<tr>
<td>Wa004</td>
<td>0.083</td>
</tr>
<tr>
<td>Wa020</td>
<td>0.036</td>
</tr>
<tr>
<td>Wa100</td>
<td>0.524</td>
</tr>
</tbody>
</table>

5. Further simulations and discussion

#### a. Causes of variability in the $\lambda$-fixed simulations

Recent model intercomparison projects demonstrate that the benchmark is not necessarily the median or the average of model results. With this in mind, we investigate the reasons for the aforementioned noted differences. To minimize differences in predicted nucleation rates as a source of discrepancies in the results, simulations for which participants set the coefficient $\lambda = 2$ (or an approximation of this in models J and K) were performed.

The number density of haze particles freezing in time interval $\Delta t$ is formulated as

$$
N_i^{\text{ALL-MODE}}/N_i^{\text{HN-ONLY}}
$$
Fig. 11. Ice water content, ice number concentration, ice particle formation rate $dN_i/dt$, and RH as functions of $z - z_a$, where $z_a$ is the altitude at which nucleation starts ($N_i = 10^{-3} \text{ cm}^{-3}$).

$$
\frac{dN_i}{dt} \Delta t = \sum_k N_{haze,k}(t) [1 - \exp(-J_{haze,k} V_k \Delta t)],
$$

where the summation in $k$ is the integration over haze particle size bins, and $dN_i/dt$ is the ice particle formation rate. It can be shown that the predicted $N_i$ is simply

$$
N_i = \sum_k N_{haze,k}(t = 0) \left[1 - \exp\left(-\int_0^{t} J_{haze,k} V_k \, dt\right)\right].
$$

A hierarchy of factors interact to produce differences in the predicted $N_i$. Given temperature, size and solute wt% of a haze particle, the difference in formulating the homogeneous freezing nucleation would affect the nucleation rate of a single haze particle. Formulation of haze particle evolution, together with homogeneous freezing nucleation formulation, determine the instantaneous ice particle formation rate. For a given uplift speed, the diffusional growth rate of ice crystals and the number concentration of ice crystals present (the latter determined by the aforementioned components acting in concert) controls the water vapor uptake rate, and thus, the evolution of the parcel RH, which then determines the predicted $N_i$.

1) **The level at which homogeneous nucleation is triggered**

The nucleation regimes of participant models occurred within the temperature range of $-43.2^\circ\text{C}$ to $-44.2^\circ\text{C}$ and $-63.2^\circ\text{C}$ to $-64.2^\circ\text{C}$ in the warm and cold simulations, respectively. The effect of temperature variation on nucleation rates within this 1°C range is secondary compared to the evolution of RH. Therefore, the model results are analyzed according to the $z - z_a$ level as shown in Fig. 11. As expected for the $\lambda = 2$ simulations, the intermodel range of RH was reduced to less than 2% in Wh020L and 4% in Ch020L, in comparison to 3% and 8% in simulations Wh020 and Ch020.

There are still significant differences in $N_i$ among the models. It can be seen that $N_i$ is only slightly changed by the value of $\lambda$ when comparing $N_i$ of the $\lambda = 2$ cases with the corresponding $\lambda$-unspecified cases. For cold cases, the increase in $\lambda$ results in a slight decrease in the predicted $N_i$ in models D and L, whereas the predicted $N_i$ by models C and S are little affected. In contrast, the increase in $\lambda$ results in a slight increase in $N_i$ in these four models for warm cases. Thus, $\lambda$ is not the dominant factor controlling the number concentration of ice crystals nucleated.
In simulation Wh020L, haze particles are almost in equilibrium with the environment (cf. Fig. 6). Nucleation is triggered at around RH020L = 95.5% in models C, J, K, S, and X, suggesting that aerosols—with solute wt% around 7%–9% in models C, K, S, and X, and 11% in model J—start to freeze under these conditions (Fig. 5). As shown for models D and L in Fig. 5, equilibrium-sized aerosols of solute wt% 8% ~ 9% are not formed until the ambient RH is around 97%. This is probably the reason that RH020L is about 1% higher in models D and L.

Compared to Wh020L, the sequence of RH020L in Ch020L changes among the five T_eff models. Models D and L have the lowest RH020L (about 88%) in this case, suggesting that aerosols with solute wt% at around 20%–21% start to freeze. As shown in Fig. 5, if models S and C would turn off the diffusional growth of haze particles and use equilibrium-sized particles instead, models S and C would have an RH020L lower than or similar to those of models D and L, respectively. The diffusional growth of haze particles likely delays the nucleation being triggered.

A slightly higher RH020L is required for model X to form an equilibrium-sized particle of wt% at around 20%–21% (Fig. 5). For a given solute wt%, model K predicts the smallest nucleation rate (Fig. 3), while at the same time, model K predicts the smallest solute wt% for a given RH020L. The latter prevails in the Ch020L simulation. As a result, RH020L by model K is about 1% lower than models D and L.

2) THE NUCLEATION REGIME

At the beginning of the nucleation stage in simulation Wh020L, ice particle formation rates among all the models are very close except for model J (Fig. 11). However, ∆RH020L (the difference between peak RH020L and RH020L) of models C and D is larger, so these models develop higher instantaneous nucleation rates later on and maintain the peak ice formation rate longer than the other models. As a consequence, model C produces the largest N_i by far followed by model D.

In contrast, from the start of the nucleation stage in simulation Ch020L, the N_i curves of models D, K, L, and X distinctly separate from those of models C and S. The ice particle formation rates in models C and S are more than one order greater than for the other models. This grouping incidentally coincides with the grouping according to haze particle specifications. Haze particles are in equilibrium or close to equilibrium in models D, K, L, and X, while large haze particles are more concentrated than the corresponding equilibrium values in models C and S. For models C and S, the slope of solute wt% at the large end of the haze particle size spectrum over the solute mass becomes flat or even positive (Fig. 6). Thus, nucleation does not start at the large particle end of the size spectrum, where only a few particles are present. Instead, nucleation starts with the particles that are slightly larger than the most dilute ones. These particles are much more numerous than the largest ones. Thus, nucleation is delayed, but more haze particles nucleate at the same time just above z_b.

The ice particle formation rate by model J is smaller than the rest. The range between RH020L and peak RH_w is also greater in simulations by model J. As shown in Fig. 12, the freezing haze number distribution in simulations by model J is much broader. These three distinct features of model J might result from a combination of the following elements. First, the homogeneous nucleation scheme of model J is less sensitive to solution concentration compared to the others. Second, the curvature effect is ignored when computing haze size. Finally, with ice particle formation rates an order of magnitude smaller than the others and with the long nucleation duration, the average size of ice crystals in model J can be much greater when nucleation stops.

The nucleation regime in model S does not last as long as in model C. Near the end of the nucleation regime, model C has the greatest N_i. Model S has slightly larger N_i than model D, despite the fact that the former has a much greater initial ice particle formation rate. The longer nucleation duration in model D eventually produces a value of N_i that is comparable to model S. The evolution of ice water content, which is the cumulative water vapor uptake, will control the peak RH020L and peak ice particle formation rate. The growth rate of small ice crystals (r < 10 μm), under the influence of the kinetic effect, is quite sensitive to the deposition coefficient β_w. Sensitivity tests on β_w have been performed to check its effect in isolation. In both models D and L, varying β_w from 0.04 to 1 [the range of β_w values specified by participants (Table 2)] results in about a factor of 4 ~ 5 and 9 ~ 12 variation in the predicted N_i for simulations Wh020L and Ch020L, respectively (Fig. 13). Nucleation begins at the same RH020L, but simulations with smaller β_w reach higher peak RH020L and consequently triggers the freezing of many more small haze particles. The predicted N_i decreases as β_w increases from 0.04 to 0.2; it only adjusts marginally to β_w when 0.2 < β_w < 1. Sensitivity tests of CCN activation on β_w manifested a similar pattern (Li et al. 2001). It is worth noting that by adjusting β_w alone, model D was able to obtain the N_i predicted by model J.

A systematic difference in the relationship between N_i and IWC is prominent among models (Fig. 14). At a given N_i, model C had the smallest IWC while model J had the most. However, the order of the predicted N_i does not match the order of β_w. For example, the predicted N_i for Ch020L, ordered from small to large, are J (1.0), K (0.5), L (0.1), D (0.04), S (0.36), X (0.1), and C (0.24); for Wh020L, J (1.0), K (0.5), S (0.36), L (0.1), X (0.1), D (0.04), and C (0.24), where the numbers in parentheses denote β_w. The mismatch suggests that the difference in β_w is a crucial, but not the only, component affecting this systematic difference. For example, com-
paring models L and X, which chose the same $b_i$, the differences in their results demonstrate the impact of haze particle specification. The change of $J_{\text{haze}}$ with time for a given particle may be written as

$$\frac{dJ_{\text{haze}}}{dt} = \frac{\partial J_{\text{haze}}}{\partial T} \frac{dT}{dt} + \frac{\partial J_{\text{haze}}}{\partial \text{wt\%}} \frac{\partial \text{wt\%}}{\partial \text{RH}} \frac{\partial \text{RH}}{dt}.$$  

(16)

We found that differences in $\partial \text{wt\%}/\partial \text{RH}$ led to the differences in the evolution of ice particle formation rates between these models.

It is conjectured that an intrinsic characteristic of the numerical component—for example, bin characteristic—would also affect the ensemble growth rate. By increasing the frequency at which the ice particle size distribution is rebinned (every 5 m to every 0.5 s), the $N_i$ predicted by model C is significantly reduced. This sensitivity is being further explored in Phase 2 of CPMCP.

b. Optical consequences

Cirrus microphysical and radiative properties are affected by the dominant nucleation mode in cloud initiation because ice particle formation rates of the two modes are expected to be distinct. Information about detailed ice crystal size distributions enables us to compute the volume absorption coefficient $b_{\text{abs}}$ as

$$b_{\text{abs}} = \sum_k Q_{\text{abs},k} \pi r_k^2 N_{i,k},$$  

(17)

where $Q_{\text{abs}}$ is the absorption efficiency obtained from Mie theory assuming spherical ice particles. The absorption efficiency at wavelength 10.75 $\mu$m increases from 0.27 for ice particles with radii of 1 $\mu$m to 1.12 for particles with $r \sim 15$ $\mu$m, and gradually decreases to $\sim 1.0$ as particles radius equals 250 $\mu$m.

As depicted in Fig. 15, on average, $b_{\text{abs}}$ increases with updraft speed. For a given $W$, the difference in $b_{\text{abs}}$ among models in the HN-ONLY simulations can be more than 100%. Except for simulation Ca100 by models C and S, $b_{\text{abs}}$ for the HN-ONLY cases is greater than that of their ALL-MODE counterparts; maximum differences exceeding 100% are found between ALL-MODE and HN-ONLY simulations. Though the uncertainty of the estimated $N_i$ is 25-fold in the extreme, the uncertainty of estimated optical properties is about two-fold for a given cooling rate.

Ice crystal size distribution information also gives us
FIG. 13. Same as Fig. 11, except this is a sensitivity test on \( \beta \) performed by model L.

a unique chance to check the calculation of cloud optical properties using effective radius. Current parameterizations of cloud optical properties usually incorporate \( \beta_{\text{abs}} \) in the form

\[
\beta_{\text{abs}} = k(r_e)IWC,
\]

where \( k \) is the mass absorption coefficient and is a function of the effective particle radius for the ice crystal distributions (e.g., Ebert and Curry 1992; Fu et al. 1998). In Fig. 15, \( r_e \) is computed according to \( r_e = \Sigma_i r_i N_i / \Sigma_i r_i N_i \) (Ebert and Curry 1992). In our simple simulations assuming spherical particles, all definitions of \( r_e \) converge (McFarquhar and Heymsfield 1998).

Note that the mass absorption coefficient decreases as the effective radius increases only when \( r_e > 20 \mu \text{m} \).

The maximum difference in IWC among the models for the cold and the warm cases is less than 15\% (not shown). It is evident that the difference in IWC cannot explain the large difference in \( \beta_{\text{abs}} \) for two simulations with similar \( r_e \). For example, the effective radii obtained by models D and S are about the same in case Ch020. However, the values of \( \beta_{\text{abs}} \), calculated according to Eq. (17), are significantly different. This difference arises from the much broader ice crystal distribution produced by model D than that produced by model S.

The differences in \( N_i \) between ALL-MODE and HN-ONLY are least when \( W = 1 \text{ m s}^{-1} \). However, the differences in \( \beta_{\text{abs}} \) (Fig. 15) for the warm cases (Wh100 and Wa100) are much greater than those for the cold cases (Ch100 and Ca100) except for model D. The differences in the width of ice crystal size distributions between Wa100 and Wh100 are more significant than those between Ca100 and Ch100. We speculated that the growth rates of ice crystals for the warm cases are much greater, such that ice crystals produced by heterogeneous nucleation near the cloud base grow to much larger sizes in comparison to ice crystals produced by homogeneous nucleation.

c. Implications for large-scale models

The \( \text{RH}_{\text{eq}} \) (see section 4) of parcel models provides large-scale models and simplified cirrus cloud models, which use a moisture-adjustment scheme to convert excess water vapor to cloud ice mass (e.g., Starr and Cox 1985; Krueger et al. 1995), with a guideline to set the threshold RH for cloud formation. It can be shown that the \( \text{RH}_{\text{eq}} \) is proportional to \( W \) and inversely related to \( N_i \). Figure 16 demonstrates the relation between these three components at 800 m above the starting point of the simulations. The \( \text{RH}_{\text{eq}} \) is well within 110\% except
for cases Wa020 and Ca020 by model L. However, the moisture adjustment timescale, the time interval between peak RH$_i$ and RH$_{eq}$, also has to be considered in order to render a complete view. The moisture adjustment timescale defined in this paper is conceptually similar to the phase relaxation time defined in Khvorostyanov and Sassen (1998a), yet it differs in that the latter is defined according to the local gradient. The moisture adjustment time almost decreases exponentially with updraft speed. For example, at $W = 0.04$, 0.2, and 1 m s$^{-1}$, the timescales are roughly 30~100 min, 6~40 min and 1~6 min, respectively (Fig. 15). Therefore, RH$_i$ higher than 110% can be sustained in regions of small to moderate cooling rates if $N_i$ is not too large. This suggests that models using time steps of around 1 h may set a nucleation RH$_i$ and a threshold RH$_i$ to simulate cold cirrus in a weak (synoptic) ascent; models using smaller time steps face a more difficult modification.

6. Conclusions

Simulation results of seven state-of-the-art cirrus parcel models are compared in Phase 1 of CPMCP. Broad qualitative agreement has been found in the simulations where-in homogeneous nucleation was the only nucleation mechanism activated. The model predicted cirrus microphysical and optical properties are strongly influenced by updraft speed. However, significant differences were found in the predicted ice crystal number concentrations $N_i$ and ice crystal size distributions. For a given cooling rate, there was up to a factor of 25 difference in $N_i$, corresponding to about a twofold difference in infrared absorption coefficient at the end of simulation (800 m above the starting point of the parcel ascent).

Detailed examination of the simulations revealed that the homogeneous nucleation formulation, haze particle solution concentration, and ice crystal growth rate were critical factors affecting the water vapor uptake rate, the peak ice supersaturation achieved, and subsequently, the predicted $N_i$. The chief findings can be summarized as follows:

- The parameter $\lambda$, the value of which is currently debatable, mainly affects the required initiation humidity to trigger homogeneous nucleation. Preliminary results show that the predicted $N_i$ is marginally affected by a reasonable choice of $\lambda$.
- The relationship between solute wt% and RH$_w$ for a given solute mass has to be specified with care. Ignoring Kelvin’s effect results in the broadening of the freezing haze size distribution, which represents an enhancement of the model sensitivity to the existence of small haze particles. If particles of a given solute mass are in equilibrium with the environment, the gradient of the solute wt% with respect to RH$_w$, $\partial\text{wt}\%/\partial\text{RH}_w$, influences the evolution of the homogeneous nucleation rate $J_{\text{haze}}$ over time in the cloud initiation regime. As a consequence, the ice particle formation rate will be affected, and thus, the predicted $N_i$.
- The width of the predicted size distribution of freezing haze particles is also affected by the gradient of nucleation rate with respect to solution concentration $\partial J_{\text{haze}}/\partial\text{wt}\%$, which cannot be determined from laboratory measurements of the critical freezing condition alone. Laboratory nucleation studies performed at critical and noncritical freezing conditions are needed to construct a consistent formulation of the homogeneous nucleation rate.
- The equilibrium assumption of haze particle size is violated in strong updrafts in a cold environment. Assuming equilibrium may result in an underestimate of ice particle formation rate at the beginning of the cloud initiation stage, and thus, the predicted $N_i$. In a non-equilibrium approach, the growth rate of haze particles is sensitive to the condensation coefficient of water, the value of which in supercooled conditions over a concentrated solution is still uncertain.
- The deposition coefficient of ice, which directly af-
ffects the growth rate of small ice crystals, is a critical factor determining the predicted $N_i$. The difference in the predicted $N_i$ can be greatly reduced if the same value of $\beta_i$ is used for the models. These results highlight the need for new laboratory and field measurements to infer the correct values for this critical quantity in the cirrus regime. Inferred from Fig. 13, $(1/N_i)(dN_i/d\beta_i)$ decreases nonlinearly with increasing $\beta_i$. If the tolerance of uncertainty in $N_i$ is a factor of 2, the confidence level of $\beta_i$ has to be within a factor of 2 to guarantee the above requirement for $\beta_i$ that falls within the range between 0.04 and 0.2, at least for the simulated cases. If $\beta_i$ is between 0.2 and 1.0, the required confidence level of $\beta_i$ is greatly relaxed because the predicted $N_i$ becomes less sensitive to $\beta_i$.

In general, discrepancies among models were greater in the ALL-MODE simulations. No attempt was made to scrutinize the causes of differences in ALL-MODE simulations due to a great variety of heterogeneous nucleation
formulations. Nevertheless, it was confirmed that the expected effect of a heterogeneous nucleation process is to lower $N_i$ and the RH$_{i}$ required for cloud initiation, and increase the average size of ice crystals formed in cirrus clouds compared to the case of a singular homogeneous freezing process, especially for a mild cooling rate (weak lifting rate). Clearly, new measurements of IN activation in cirrus conditions are warranted. Moreover, it is important to come to grasp with the effects of solution droplet strength and chemistry on the activation of IN.

This study was conducted based on a single CCN distribution. The sensitivity of predicted cirrus microphysical and optical properties to an assumed CCN distribution must be addressed in order to advance this study. In Phase 2 of the CPMCP, now underway, the effects of varying input aerosol distributions (e.g., number concentration, mode radius, and distribution width) are taken into account. Sensitivity of model results to CCN composition will indirectly be made by altering the value of $\lambda$.

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APPENDIX

Notation

- $A$: Coefficient in the heterogeneous nucleation scheme used by model C ($2.7 \times 10^{10}$ m$^{-3}$)
- $A_{s0}$: Coefficient in the modified Fletcher equation used by model S ($10^{-2}$ m$^{-3}$)
\( A_i \) Coefficient in the modified Fletcher equation used by model S (7.5 \( \times \) 10\(^{-4} \) m\(^{-1} \))
\( a \) Coefficient in the immersion freezing parameterization by model D (1.3 \( \times \) 10\(^{-2} \); dimensionless)
\( a_w \) Water activity
\( B \) Coefficient in the heterogeneous nucleation scheme used by model C (10.6; dimensionless)
\( B_i \) Coefficient in the modified Fletcher equation used by model S (0.8°C\(^{-1} \))
\( b \) Coefficient in the immersion freezing parameterization by model D (11.75; dimensionless)
\( b_{obs} \) Volume absorption coefficient (m\(^{-1} \))
\( B \) Boltzmann constant
\( C_p \) Specific heat capacity for dry air at constant pressure (J kg\(^{-1} \) K\(^{-1} \))
\( D' \) Coefficient of diffusion of water vapor in air modified by kinetic effect [see Eqs. (13)–(14) of Pruppacher and Klett (1997); m\(^2\) s\(^{-1} \)]
\( e_{sw} \) Saturation water vapor pressure over a flat pure water surface (Pa)
\( F_i \) Number fraction of frozen haze particles in bin \( k \)
\( \Delta F_{aciv} \) Activation energy for diffusion of a water molecule across the water–ice boundary (J)
\( \Delta F_{gern} \) Energy of ice-germ formation (J)
\( F_{>0.1\mu m} \) CCN fraction of haze size greater than 0.1 \( \mu m \)
\( g \) Acceleration due to gravity
\( h \) Planck constant
\( IWC \) Ice water content (kg m\(^{-3} \))
\( J_{sw} \) Homogeneous nucleation rate of ice in pure water (m\(^{-3}\) s\(^{-1} \))
\( J_{haze} \) Homogeneous nucleation rate of ice in a solution droplet (m\(^{-3}\) s\(^{-1} \))
\( K' \) Coefficient of diffusion of water vapor in air modified by kinetic effect [see Eqs. (13)–(20) of Pruppacher and Klett (1997); J m\(^{-1} \) s\(^{-1} \) K\(^{-1} \)]
\( \bar{L}_{sw} \) Average latent heat of fusion of water between \( T \) and \( T_0 \) (J kg\(^{-1} \))
\( L_w \) Latent heat of condensation (J kg\(^{-1} \))
\( M \) Molality (mol per 1000 g of water)
\( M_s \) Molecular weight of solute
\( M_w \) Molecular weight of water
\( m_i \) Solute mass
\( m_w \) Water mass
\( N_i \) Number of monomers of water in contact with unit area of the ice surface (m\(^{-2} \))
\( N_{haze,k} \) Number concentration of haze particles in bin \( k \) (m\(^{-3} \))
\( N_i \) Number concentration of ice crystals (total) (m\(^{-3} \))
\( N_{i,k} \) Number concentration of ice crystals in bin \( k \) (m\(^{-3} \))
\( N_{IN} \) Number concentration of activated ice nuclei (m\(^{-3} \))
\( P \) Pressure (Pa)
\( Q_{obs} \) Absorption efficiency
\( Q_{DL} \) Diabatic heating due to latent heat release (K s\(^{-1} \))
\( R_c \) Gas constant for dry air (J kg\(^{-1} \) K\(^{-1} \))
\( R_v \) Gas constant for water vapor (J kg\(^{-1} \) K\(^{-1} \))
\( RH \) Relative humidity with respect to ice
\( RH_w \) Relative humidity with respect to water
\( r \) Radius (m)
\( r_e \) Effective radius for optical properties (m)
\( S_i \) Water vapor saturation ratio with respect to ice
\( S_w \) Water vapor saturation ratio with respect to water
\( T \) Temperature (K)
\( T_f \) Reference temperature 273.15 K
\( T_{eff} \) Effective freezing temperature (K)
\( T_m \) Equilibrium melting temperature (K)
\( \Delta T_m \) Melting point depression (K)
\( t \) Time (s)
\( \Delta t \) Time interval (s)
\( V \) Volume (m\(^3\))
\( W \) Updraft speed (m s\(^{-1} \))
\( \text{wt}\% \) Weight percent \([100\% * m_s/(m_i + m_w)]\)
\( \alpha \) Coefficient in the heterogeneous nucleation scheme used by model L (6.269; dimensionless)
\( \beta \) Coefficient in the heterogeneous nucleation scheme used by model L (0.1296; dimensionless)
\( \beta_i \) Deposition coefficient
\( \beta_{sw} \) Condensation coefficient
\( \lambda \) See section 3.
\( \Gamma \) Lapse rate (K m\(^{-1} \))
\( \rho_i \) Ice density (kg m\(^{-3} \))
\( \rho_s \) Average ice density between \( T \) and \( T_0 \) (kg m\(^{-3} \))
\( \rho_s \) Solution density (kg m\(^{-3} \))
\( \rho_w \) Water density (kg m\(^{-3} \))
\( \sigma_{aas} \) Surface tension across the air and solution interface (J m\(^{-1} \))
\( \sigma_{asa} \) Surface tension across the ice and solution interface (J m\(^{-1} \))
\( \nu \) Dissociation constant for ions in solution
\( \Phi \) Molal osmotic coefficient

Subscripts \( \text{eq}, \text{ini}, k, p \) denote equilibrium, the initial condition, category \( k \), and the peak value, respectively.

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