The Effects of Surface Kinetics on Crystal Growth and Homogeneous Freezing in Parcel Simulations of Cirrus

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ABSTRACT

The uptake of water vapor excess by ice crystals is a key process regulating the supersaturation in cold clouds. Both the ice crystal number concentration and depositional growth rate control the vapor uptake rate and are sensitive to the deposition coefficient \(a_d\). The deposition coefficient depends on temperature and supersaturation; however, cloud models either ignore or assume a constant \(a_d\).

In this study, the effects of \(a_d\) on crystal growth and homogeneous freezing of haze solution drops in simulated cirrus are examined. A Lagrangian parcel model is used with a new ice growth model that predicts the deposition coefficients along two crystal growth axes. Parcel model results indicate that predicting \(a_d\) can be critical for predicting ice nucleation and supersaturation at different stages of cloud development. At cloud base, model results show that surface kinetics constrain the homogeneous freezing rate primarily through the growth impact of small particle sizes in comparison to the mean free path. The deposition coefficient has little effect on homogeneous freezing rates, because the high cloud-base supersaturation produces \(a_d\) near unity. Above the cloud-base nucleation zone, decreasing supersaturation causes \(a_d\) to decrease to values as low as 0.001. These low values of \(a_d\) lead to higher steady-state supersaturation. Also, the low values of \(a_d\) produce substantial impacts on particle shape evolution and particle size, both of which are dependent on updraft strength.

1. Introduction

Ice crystal growth from vapor is an important microphysical process critical for the evolution of atmospheric cold clouds. Unlike liquid drops, ice crystals can grow to precipitation sizes by vapor depositional growth alone. The growth rates of crystals are determined by the particle shape and surface characteristics, which in turn control the particle dimension, ultimately affecting collection and sedimentation processes (Fukuta and Takahashi 1999; Libbrecht 2005; Avramov and Harrington 2010). On the other hand, ice crystal growth acts as a sink of water vapor, regulating the saturation state in cold clouds. Ice nucleation processes, both homogeneous and heterogeneous, are sensitive to the concentration of excess vapor; therefore, the vapor uptake rate also impacts newly formed ice particle size distributions and number concentrations (DeMott et al. 1994; Comstock 2004; Lohmann et al. 2008).

According to our traditional understanding, ice formation takes place when ice supersaturation (usually caused by dynamic forcing) reaches to a threshold value for ice nucleation, which can be either heterogeneous or homogeneous nucleation, though many studies focus on homogeneous freezing in cirrus clouds. The growth of ice crystals immediately following nucleation rapidly quenches supersaturation and brings the supersaturation back to steady state over a short time scale (Peter et al. 2006). The time required for dynamic and microphysical processes to bring about a steady-state supersaturation is called supersaturation relaxation time. The sustained high supersaturation over ice and low ice crystal concentrations discovered in low-temperature cirrus clouds observations (i.e., Lawson et al. 2008; Krämer et al. 2009) challenge our understanding of...
ice-cloud microphysics. A climate modeling study (Gettelman and Kinnison 2007) shows that, if elevated supersaturation in cirrus is allowed in a GCM, then thick cirrus are predicted to decrease, and water vapor increases in the upper troposphere; both results have a positive radiative impact on climate. The high supersaturation and low ice number observations suggested suppressed nucleation and/or suppressed growth of ice in large parts of the upper troposphere (Peter et al. 2006). A number of studies explain the low ice concentrations with the onset of heterogeneous freezing. For instance, glassy particles can serve as heterogeneous ice nuclei (Murray et al. 2010), which can then suppress homogeneous nucleation. Earlier numerical studies by DeMott et al. (1994) and DeMott et al. (1997) had simulated cirrus initiation with both homogeneous and heterogeneous freezing assuming an abundance of effective ice nuclei (IN) in the upper troposphere. With the aid of recent aircraft measurements and satellite observation, Jensen et al. (2010) showed that, in contrast to homogeneous nucleation, the lower ice supersaturations at which heterogeneous freezing becomes active and the limiting ice concentrations produce broader ice size distributions, which reconcile with the measurements. On the other hand, sustained high supersaturation levels inside clouds can be explained by suppressed vapor growth. Suppressed growth causes a higher cloud-base supersaturation maximum and longer supersaturation relaxation time, which results in more nucleation events and sustained supersaturation (Spichtinger et al. 2004). A low deposition coefficient can explain suppression in growth (Gao et al. 2004).

The deposition coefficient \( \alpha_d \) is one of the most uncertain, but critical, parameters in modeling crystal vapor depositional growth. This coefficient accounts for the fraction of the water molecules that impinge on the crystal surface and are ultimately incorporated into the lattice on crystal facets (prism or basal) during the depositional growth. It can be thought of as a growth efficiency. A value of \( \alpha_d = 0 \) (unity) means that no (all) molecules striking the surface are incorporated. Hence, \( \alpha_d \) describes an effective surface-based resistance (often called surface kinetic resistance) to vapor diffusion. The deposition coefficient is known to depend on the supersaturation immediately above the crystal surface (local supersaturation \( S_{\text{local}} \)), and there are various formulations of that dependence (e.g., Nelson and Baker 1996; Libbrecht 2003b). All formulations are based on the observation that growth depends on a critical supersaturation \( S_{\text{crit}} \), which itself is a function of surface properties and temperature. High values of \( S_{\text{crit}} \) mean that \( S_{\text{local}} \) needs to be high before growth of the crystal surface will commence. When \( S_{\text{local}} \) is smaller than \( S_{\text{crit}} \), it indicates that the growth has the potential to be strongly limited by surface incorporation.

The deposition coefficient not only controls the growth rate of the crystal, but it also determines the ultimate shape of the crystal. For instance, hexagonal prism crystals can grow into either columnar or platelike forms depending upon which crystal facet has a higher \( \alpha_d \) and, hence, a lower \( S_{\text{crit}} \) (e.g., Lamb and Scott 1974; Libbrecht 2003b). Because \( \alpha_d \) is important for determining the primary habit, Chen and Lamb (1994) based their “adaptive habit” approach to aspect ratio evolution on what they called the inherent growth ratio \( \Gamma \), which is defined as the ratio of the deposition coefficient along the \( c \) axis to that along the \( a \) axis (\( \alpha_c/\alpha_a \)). In Chen and Lamb’s (1994) method, spheroidal ice with two primary growth axes (\( a \) and \( c \)) is assumed: The axes are defined in relation to the hexagonal ice, with the \( c \)-axis growth direction perpendicular to the basal (or hexagonal) face and the \( a \) axis perpendicular to the prism (or rectangular) face. Chen and Lamb (1994) assumed that \( \Gamma \) depends only on the temperature. A number of articles have shown that the method can reproduce the basic evolution of aspect ratio, crystal mass, and crystal fall speed in clouds at liquid saturation (Chen and Lamb 1994; Hashino and Tripoli 2007; Sulia and Harrington 2011; Harrington et al. 2013a,b). Nevertheless, Chen and Lamb’s (1994) method did not couple \( \alpha_c \) and \( \alpha_a \) to the vapor diffusion equation; therefore, surface kinetic resistance was not included.

Surface kinetic resistance in cirrus clouds has been studied with various models, but usually \( \alpha_d \) is prescribed as a constant. Parcel model simulations (Lin et al. 2002; Gierens 2003) show that the ice number concentration produced by homogeneous freezing \( N_i \) strongly depends on the value that is assumed for \( \alpha_d \); when \( \alpha_d \) is small, slower vapor depletion near cloud base results in higher supersaturation that consequently triggers the freezing of more haze particles; therefore, larger number of smaller ice particles are produced. Low values of \( \alpha_d \) increase the supersaturation relaxation time (Spichtinger et al. 2004), which possibly explains elevated ice supersaturations observed inside cirrus clouds (Comstock 2004; Spichtinger and Gierens 2009). Kay and Wood (2008) used time-scale analysis in a parcel model study to show that aerosol sensitivity (the change of \( N_i \), resulting from homogeneous nucleation in response to the change of aerosol number concentration) increases dramatically with deposition coefficient \( \alpha_d \ll 1.0 \). By constraining model results with observations, they suggested that \( \alpha_d \) increases with ice supersaturation and/or decreases with ice crystal size. They also showed that using a constant, low value (0.006 ± 0.0015) suggested by laboratory experiments (Magee et al. 2006) is inconsistent with observations.
Harrington et al. (2009) conducted cirrus simulations with the Regional Atmospheric Modeling System (RAMS) and showed that $\alpha_d$ has a strong influence on cirrus dynamics, which then feeds back into the cloud structure and cloud lifetime. A smaller value of deposition coefficient ($\alpha_d = 0.005$), in the range of suggested values by (Magee et al. 2006), leads to inefficient growth. Ice particles in this case remain small, and precipitation rates are subsequently low, leading to cirrocumulus-like structures over time. When a large value of $\alpha_d$ (unity) is used, broken cirrus uncinus form and persist. Lohmann et al. (2008) used a global model, European Centre Hamburg Model 5 (ECHAM5), with a two-moment microphysics scheme and allowing for both homogeneous and heterogeneous nucleation to simulate cirrus evolution. By changing $\alpha_d$ from a typically used value of 0.5 to 0.006, the number of ice crystals increases significantly, and the particle sizes are smaller as a result of the weaker crystal growth. The shortwave and longwave significantly, and the particle sizes are smaller as a result of the weaker crystal growth.

2. Model description

a. Lagrangian microphysical parcel model

We modified the Lagrangian microphysical parcel model of Sulia and Harrington (2011) to explore the influence of surface kinetics on homogeneous freezing in cirrus. Differential equations for environmental variables (temperature, pressure, altitude, and supersaturation), along with differential equations for the growth of drops and ice crystals compose the model. The liquid and ice spectra are also Lagrangian, and each spectrum is divided into $n_l$ and $n_i$ bins for liquid and ice, respectively. The number of ice bins varies with the nucleation rate but is set to a maximum of 1000 (see below). Vapor diffusion equations for the growth of the $n_l$ drop and $n_i$ ice bins, and the differential equations for the environmental variables are solved at each time step using the variable ordinary differential equation (VODE; Brown et al. 1989) package. Sedimentation and entrainment–mixing processes are not included.

b. Homogeneous freezing of solution particles

We consider homogeneous freezing only because it is better constrained by observations and theory (Koop et al. 2000) than heterogeneous ice nucleation mechanisms, and prior studies have revealed sensitivity of homogeneous freezing to growth (Lin et al. 2002; Gierens 2003). How the onset of heterogeneous freezing responds to the surface kinetic effects will be explored in future work.

The background haze particles are assumed to be aqueous ammonium sulfate with a number concentration of 150 cm$^{-3}$ lognormally distributed into 10 size bins. The geometric mean radius is 0.015 $\mu$m, and geometric standard deviation $\sigma = 1.48$, similar to the haze particle distribution used in prior parcel model simulations (Jensen et al. 1994; Lin et al. 2002). The homogeneous nucleation rate is parameterized following Koop et al. (2000):

$$\log[I_{\text{hom}}(a_w, T)] = -906.7 + 8502\Delta a_w - 26924\Delta a_w^2 + 29180\Delta a_w^3,$$

$$\Delta a_w = a_w - a_{\text{ice}}(T), \quad \text{and}$$

$$a_{\text{ice}}(T) = \exp \left[ \frac{210.368 + 131.438T - 3.2373 \times 10^{-6}/T - 41.7291 \times \ln(T)}{8.31441T} \right],$$

where $I_{\text{hom}}$ is the homogeneous nucleation rate; $a_w$ is the aerosol water activity, which is defined by the ratio between the water vapor pressures of the solution and of pure water under the same conditions; and $a_{\text{ice}}$ is the
water activity of a solution in equilibrium with ice. Following Kay and Wood (2008), we assume that $a_w$ approximately equals the relative humidity. We note here that in tests where $a_w$ is calculated based on haze particle properties (not shown), the results follow closely to those that assume $a_w$ equals the relative humidity.

During freezing, ice bins are populated following the probabilistic approach of Ervens et al. (2011): A new ice particle class forms if at least 1% of the solution droplets in a given size bin are predicted to freeze. This binning method limits the number of ice class bins that are formed from nucleation to a maximum of 1000. The method also appropriately sorts the frozen drops in size because freezing occurs first for the largest drops.

c. Ice crystal depositional growth model: KLAH

Modeling vapor depositional growth follows the Kinetically Limited Adaptive Habit (KLAH) model (Zhang and Harrington 2014). This approach combines the adaptive habit model of Chen and Lamb (1994) with predicted $\alpha_d$ for each axis direction based on laboratory-derived $S_{crit}$. KLAH follows the traditional flux-matching method for including surface kinetics (Pruppacher and Klett 1997, p. 420) in mass growth but extends the method to the major and minor axes of any single crystal. Zhang and Harrington (2014) show that these may be included through modified vapor diffusivity $D'_v$ and thermal conductivity $k'_T$ in the capacitance growth equation:

\[
\frac{dm}{dt} = 4\pi C(c, a)S_i \left( \frac{RT}{M_w D'_v \ell_v(T)} + \frac{\ell_v}{M_w k'_T} \left( \frac{\ell_v}{RT} - 1 \right) \right)^{-1},
\]

where $C$ is the capacitance of a spheroid with two primary semiaxis lengths $c$ and $a$, $S_i$ is the ice supersaturation, $\ell_v$ is the ice equilibrium vapor pressure, $R$ is the universal gas constant, $T$ is the temperature, $M_w$ is the molecular mass of water, and $\ell_v$ is the enthalpy of sublimation. As shown by Zhang and Harrington (2014), $D'_v$ and $k'_T$ are modified for explicit dependence on $\alpha_d$ along the $a$ and $c$ axes:

\[
D'_v = \frac{2}{3} \frac{D_v}{\ell_v(\alpha_d)C + C/3^a C_\Delta} + \frac{1}{3} \frac{D_v}{\ell_v(\alpha_d)C + C/3^c C_\Delta}
\]

and

\[
k'_T = \frac{2}{3} \frac{k_T}{\ell_T(\alpha_T,a)C + C/3^a C_\Delta} + \frac{1}{3} \frac{k_T}{\ell_T(\alpha_T,c)C + C/3^c C_\Delta},
\]

where $\alpha_d$ and $\alpha_c$ are deposition coefficients for the $a$ and $c$ axis, respectively; $\ell_v(\alpha) = 4D_v({\alpha}/(\alpha_T))$ is the kinetic length scale for vapor diffusion; $\ell_T(\alpha_T) = 4k_T((\alpha_T)/\alpha_T C_\Delta)$ is the kinetic length scale for thermal diffusion (e.g., Mordy 1959; Harrington et al. 2009); and $C_\Delta = C(a + \Delta, c + \Delta)$ is the capacitance evaluated at the mean “jump” distance $\Delta$ (approximately the mean free path) away from the spheroidal particle surface. The mean molecular speeds are $\bar{v}_r$ and $\bar{v}_e$ for vapor and dry air, respectively, and $\mu_a$ and $c_p$ are the air density and specific heat capacity at constant pressure of dry air. Because the thermal accommodation coefficient $\alpha_T$ is thought to be near 1 (Shaw and Lamb 1999), its impact on growth is relatively minor.

KLAH parameterizes the dependence of $\alpha_d$ on temperature and supersaturation following Nelson and Baker (1996):

\[
\alpha_d = \left( \frac{S_{local}}{S_{crit}} \right)^m \tanh \left( \frac{S_{crit}}{S_{local}} \right)^m,
\]

along both $a$ and $c$ axes. Figure 1a shows $\alpha_d$ variation with normalized local supersaturation ($S_{local}/S_{crit}$) for various values of $m$ in Eq. (3). The parameter $m$ controls the steepness of the black curves (Fig. 1a) and can be
thought of as accounting for different surface types (Nelson and Baker 1996). For instance, when \( m = 1 \) the surface consists of permanent spiral dislocations, and the hyperbolic tangent form of \( \alpha_d \) is strictly for this type of growth (Burton et al. 1951). When \( m = 30 \), molecular attachment requires the nucleation of 2D islands; therefore, a sharp increase of \( \alpha_d \) occurs at \( S_{\text{crit}} \). Although spiral dislocation and 2D nucleation growth types are clearly distinguished in the literature, it is not clear how the surface growth type is related to the ambient conditions. In addition, other surface growth types exist, such as ice surfaces that consist of numerous stacked islands, surfaces that are completely molecularly rough so that \( \alpha_d \) is one, or surfaces that are covered with a quasi liquid layer at higher temperatures (Kuroda and Lacmann 1982; Ming et al. 1988; Ewing 2004). Modifying the hyperbolic tangent form with \( m \) was devised by Nelson and Baker (1996) as a way to use one basic formulation to cover other surface growth types. In our parcel model simulation, we assume that \( m \) value decreases with rising saturation ratio \((e/e_s)\) from 30 at low saturations \((e/e_s < 0.15)\) to \( m = 1 \) at liquid saturation \((e/e_s = 1)\). The implicit assumption is that the number of nucleated ledges should rise with saturation ratio.

The critical supersaturation is determined from laboratory measurements (i.e., Nelson and Knight 1998; Libbrecht 2003b), while \( S_{\text{local}} \) is derived from the diffusion model [see derivation and formula in Zhang and Harrington (2014)]. It is the local supersaturation that controls the growth of the particle, since it is the ratio of \( S_{\text{local}}/S_{\text{crit}} \) that determines \( \alpha_d \). However, the ambient supersaturation is often far larger than \( S_{\text{local}} \) or \( S_{\text{crit}} \). For example, Fig. 1b shows the variation of \( \alpha_d \) with ambient supersaturation at a temperature of \(-35^\circ\text{C}, 500\text{-hPa}\) pressure, particle size of 10 \( \mu \text{m} \), and \( S_{\text{crit}} \) of 6%. In these cases, \( S_{\text{local}} \) is often near \( S_{\text{crit}} \); however, note that, regardless of the \( m \) value, \( \alpha_d \) is nearly always low. For relatively large \( m \) values \((m = 15-30)\), when surface requires 2D island nucleation, \( \alpha_d \) is less than 0.1 for nearly all ambient supersaturations that are below 20%.

Values of \( \alpha_d \) below 0.1 typically produce inhibited growth (Gierens 2003). Hence, 2D nucleation can lead to kinetically limited growth over a broad range of ambient supersaturations. When many ledges exist on the crystal surface \((m = 1-5)\), ambient supersaturations can still be relatively large \((5\%-15\%)\) when growth is inhibited by surface kinetics.

Because the growth efficiencies for two axis lengths are predicted, the KLAH method predicts the aspect ratio and, hence, the shape of the ice particles as a function of temperature and supersaturation. Aspect ratio \( \phi \) evolves following the mass distribution hypothesis of Chen and Lamb (1994):

\[
\frac{dc}{da} = \frac{\alpha_f(T,s_i) c}{\alpha_d(T,s_i) a} = \Gamma(T,s_i) \phi, \tag{4}
\]

where we have explicitly indicated both the temperature and supersaturation dependence of \( \alpha_d \). This has been done to distinguish the KLAH from Chen and Lamb (1994), in which it is assumed that \( \Gamma \) depends only on the temperature. The KLAH model was tested against the numerical hexagonal model of Wood et al. (2001) and produces reasonable predictions of the particle mass, \( \alpha_d \), \( \phi \), and size (Zhang and Harrington 2014).

3. Parcel model simulation

Our previous studies with the KLAH model show that the effects of surface kinetic resistance significantly reduce mass growth and alter axis-length evolution in situations with low saturation states and temperatures (Zhang and Harrington 2014). From that work, it appears as if surface kinetic resistance is most critical for reduced mass and axis-length growth rates when \( T < -20^\circ\text{C} \) and the saturation state is below that of liquid. We therefore focus our attention on simulated cirrus, as they can have low saturation states. Cloud depth is set to approximately 700 m to ensure that complete homogeneous freezing occurs and that the parcel reaches an approximate steady-state ice supersaturation. Cases that require a longer time to reach steady-state supersaturation occur when nucleated ice number concentration is low and the total vapor uptake by ice is inefficient.

We did two sets of parcel model simulations. One set of simulations forms ice instantaneously at a given concentration and ice water content (IWC), assuming that the formed ice particles are monodisperse (nucleated at one size). These simulations effectively sever the link between ice nucleation and depositional growth and form a baseline with which the nucleation simulations can be compared. The second simulation set takes into account the homogeneous freezing process and assumes an initial lognormal distribution of aqueous ammonium sulfate particles (Sulia and Harrington 2011). Homogeneous freezing and ice crystal growth are driven by a constant updraft \( w \) ranging from 5 to 120 cm s\(^{-1}\). For ice particle growth, we examined two situations: one in which surface kinetics are fixed and a second where surface kinetics evolve through predicted deposition coefficients. Three cases are examined for fixed surface kinetics, and spherical ice is assumed for simplicity. The first case uses standard capacitance theory (No Kinetics); the second and third cases assume a constant \( \alpha_d \) of 1 (Sphrl.0) and 0.01 (Sphrl.01). These simulations are similar to prior studies that have fixed \( \alpha_d \) (e.g., Gierens et al. 2003; Harrington et al. 2009). It is worth noting that the no-surface-kinetics case does not mean that \( \alpha_d \) equals
to 1. Even when $\alpha_d = 1$, growth rates can be reduced [see Fig. 1 in Xue et al. (2005)] if the particle size is near the mean free path $\Delta$, which at cirrus pressures (300 hPa) is close to the size of newly nucleated ice particles ($\Delta \sim 0.2 \mu m$ vs a nucleated size of about 1 $\mu m$). This growth reduction depends primarily on the ratio of the mean free path to particle size (Knudsen number); hence, we refer to it as the “mean free path” or “small particle” effect. In the case when ice forms instantaneously with a prescribed radius of 5 $\mu m$, ice particles are already significantly larger than the mean free path, so the no-surface-kinetic results resemble $\alpha_d = 1$ results. Last, we examine the cases when $\alpha_d$ is predicted for spherical growth (reduced KLAH model) and nonspherical growth with $\alpha_d$ predicted on both the $a$ and $c$ axes (KLAH model). Table 1 gives the input parameters for the different crystal growth models.

Predicting $\alpha_d$ requires values of $S_{crit}$, which are known only for temperatures above $-40^\circ C$ (e.g., Nelson and Knight 1998; Libbrecht 2003b; Zhang and Harrington 2014). For lower temperatures, we use lower and upper bounds on $S_{crit}$ that are plausible and take into account that some evidence suggests $S_{crit}$ increases with decreasing temperature (Libbrecht 2003b). The lower bound to $S_{crit}$ assumes the value at $-40^\circ C$ ($S_{crit,a} = 6\%$ and $S_{crit,c} = 3\%$; Zhang and Harrington 2014), producing columnar growth. To simulate plate growth, we simply switch the $S_{crit}$ axis values, and spherical growth uses the average $S_{crit}$. The upper bound to $S_{crit}$ assumes an average value of 13\% based on in situ measurements (Gao et al. 2004); $S_{crit,e}$ and $S_{crit,a}$ are determined by assuming the ratio of $S_{crit,a}/S_{crit,e} = 2$ at $-40^\circ C$.

4. Parcel model: No nucleation feedbacks

The parcel model allows us to study the possible feedbacks between ice formation and growth processes in a well-controlled, albeit idealized, manner. As discussed above, we first conduct simulations with instantaneous nucleation of ice so that feedbacks between nucleation and growth do not occur. We prescribe a monodisperse population of ice particles with a number density equal to $1 cm^{-3}$. The particles are initially spherical, consistent with homogeneously frozen liquid, with a radius of 5 $\mu m$. We note that using a gamma size distribution produces similar results as when the initial population is monodisperse (not shown). The reason that no difference occurs here is because of an effect described by Sheridan et al. (2009): initially smaller ice particles can grow more rapidly, overtaking the initially larger ice particles. Therefore, even if we started the simulations with an initial spectral spread in size, the ice distribution narrowed first before broadening, as in Sheridan et al. (2009). The overall effect is that there was no difference between the gamma and monodisperse results. A low and a high initial ice relative humidity value, RH$_i$ = 125\% and RH$_i$ = 105\%, are used along with $T_{init} = -46^\circ C$ and $P_{init} = 250 hPa$. For these simulations, the parcel has a constant updraft of $w = 20 cm s^{-1}$.

a. Case 1: High RH$_i$

Vertical profiles of $S_i$ and IWC, using the different ice crystal growth models elaborated in Table 1, are shown in Fig. 2 for the high-RH$_i$ case. The supersaturation decreases with height for all simulations, except for the spherical model with constant $\alpha_d = 0.01$ (Sphr.01). In this case, vapor uptake by ice growth is strongly limited by surface resistance. Because $\alpha_d$ is constant, the supersaturation produced by adiabatic cooling is much larger than that consumed by vapor deposition as the parcel is initially lifted. The ice supersaturation eventually reaches steady state, but only near the end of the simulation, when the supersaturation production by the updraft balances the vapor consumption by ice growth.

For the models predicting $\alpha_d$, the steady-state supersaturation asymptotically approaches a value in the
The range of \( S_{\text{crit}} \) and \( S_{\text{crit}} \), similar to the results of Zhang and Harrington (2014). When particle initial size is prescribed to be 5 \( \mu m \), the ratio \( C/C_0 \) that appears in Eq. (2) is approximately equal to 1; therefore \( a_d = 1.0 \) (Sphr1.0) and the no-surface-kinetic cases have nearly identical results (only Sphr1.0 is shown).

The low, constant \( a_d \) (Sphr.01) also leads to much smaller IWC (Fig. 2b), in comparison to the other growth models in the lower 300 m of the cloud. As \( S_{\text{local}} \) approaches steady state, the IWC differences between the models become small. Models that assume a smaller \( S_{\text{crit}} \) predict larger IWC than models with a larger value, since more water remains as vapor when \( S_{\text{crit}} \) is high. This makes physical sense: A lower \( S_{\text{crit}} \) value means that the ice surfaces begin growing at a lower value of the supersaturation over the crystal axes.

The manner in which particle habit evolves does not impact the predicted supersaturation and IWC by more than a few percent. In general, \( S_{\text{local}} \) over the crystal surface is usually near \( S_{\text{crit}} \), since \( S_{\text{crit}} \) controls surface incorporation. This provides a control on \( S_{\text{local}} \), which is somewhat higher than \( S_{\text{local}} \) because of vapor and thermal diffusion effects. This indicates that \( S_{\text{crit}} \) is the primary control on \( S_{\text{local}} \), which then determines the growth rate of the IWC. Though habit evolution does not impact \( S_{\text{local}} \), the axis evolution is impacted (Fig. 3), and this will be critical for other physical processes, such as sedimentation, collection, and radiative properties. For the low- and high-\( S_{\text{crit}} \) cases, the radius profiles for the spherical models are similar except for the first 200–300 m of lifting, and this matches the supersaturation evolution in Fig. 2. The predicted \( a_d \) values in the small-\( S_{\text{crit}} \) case are larger than the large-\( S_{\text{crit}} \) case for all \( a_d \), as expected: lower \( S_{\text{crit}} \) means molecular incorporation begins at a lower supersaturation; hence, the growth efficiency is higher. However, it is counterintuitive that columns could become longer when \( S_{\text{crit}} \) is larger rather than when it is smaller (cf. Figs. 3a and 3c). This interesting result shows that it is not only the \( S_{\text{crit}} \) on each axis that controls \( \phi \) change, it is also controlled by how the vapor field responds to changes in \( \phi \).

The effect of \( S_{\text{crit}} \) on \( \phi \) evolution (Fig. 4) shows that a large \( S_{\text{crit}} \) produces particles with more extreme shapes (larger \( \phi \) deviations from unity). The larger \( S_{\text{crit}} \) causes the minor axis to nearly cease growing earlier, therefore leading to more extreme aspect ratios. For column growth, \( \alpha \)-axis cessation begins (growth rate drops below 0.01 \( \mu m \) s\(^{-1} \)) at 142 and 136 s into the simulations for low- and high-\( S_{\text{crit}} \) cases, respectively. While for plate growth, \( c \)-axis cessation begins around 112 and 96 s for both low and high \( S_{\text{crit}} \) (cf. Figs. 3a and 3c). After the minor axis stops growing, the available vapor is distributed on the growing (major) axis, which accelerates growth in that dimension. This effect occurs for both column and plate growth; however, the resulting aspect ratio difference of plates is less pronounced. This is because the mass is deposited roughly two-dimensionally for plate growth, as compared with one-dimensionally for column growth.

b. Case 2: Low \( R_{\text{Hi}} \)

Surface kinetic resistance is sensitive to the ambient saturation (Zhang and Harrington 2014), so we also show results using a low initial ice relative humidity, \( R_{\text{Hi}} = 105\% \). Figure 5a shows that all of the ice growth models produce a maximum \( S_{\text{local}} \) with a subsequent relaxation toward steady state. It takes longer for \( S_{\text{local}} \) to relax when growth is inefficient (\( a_d = 0.01 \)). IWC at the end of the simulations is about 1/4 less than in the
high-RH_i simulations. While the relative differences among the growth models are qualitatively similar to the high-RH_i simulations, there are some notable differences. Unlike the high-RH_i simulation, there is a more pronounced separation in the supersaturations among the simulated cases. In particular, the spherical cases produce higher Si (about 1%–2%) than the nonspherical cases, both in low- and high-RH_i cases. This is because of larger growth rates that are produced by nonspherical particles, as discussed in Zhang and Harrington (2014).

In both high- and low-RH_i cases, when predicting \( \alpha_d \), the resulting supersaturation relaxes to approximately the \( S_{\text{crit}} \) value. This provides modeling evidence that low deposition coefficients are a potential candidate to explain the large-scale persistence of supersaturation inside cirrus clouds, as hypothesized by Peter et al. (2006).

The most striking difference among the simulations is illustrated in Fig. 6, which shows the \( \phi \) evolution. The aspect ratio can reach a value higher than 60 for columns and lower than 0.2 for plates, which is more extreme than predicted in the high-RH_i case. As discussed in the last section, at low \( S_i \), the minor axis growth slows and nearly ceases earlier than at higher \( S_i \). At lower \( S_i \), the \( S_{\text{local}} \) over the minor axis drops below the critical value more quickly, so most of the mass increase goes to extending the major axis, leading to more extreme \( \phi \).

5. Parcel model: Homogeneous nucleation

Homogeneous freezing depends strongly on the environmental saturation that, in turn, is sensitive to the value of \( \alpha_d \), as our results above and others have shown (e.g., Lohmann et al. 2008). That ice growth feeds back with ice nucleation is relatively well known (Lin et al. 2002); however, it remains an open question as to how evolving \( \alpha_d \) and \( \phi \) may alter the homogeneous freezing.
rates. In the simulations below, the initial temperature is $-46^\circ$C, and the initial RH is 125%, which is below liquid saturation. Homogeneous nucleation occurs based on the water activity, but generally the freezing happens at about $-47.7^\circ$C, which we define as cloud base. Cloud base has an ice relative humidity of around 147%, or near liquid saturation, which is consistent with other studies (DeMott et al. 1994; Gierens 2003). For these simulations, the parcel also has a constant updraft of $w = 20$ cm s$^{-1}$.

a. Ice number concentration, IWC, and particle size

As Fig. 7 shows, the nucleation rates are sensitive to the treatment of $\alpha_d$. For spherical growth, small $\alpha_d$ results in inefficient depositional growth, leading to a high $S_i$ maximum and high nucleation rates. As soon as a large number of smaller particles are nucleated in the small $\alpha_d$ case ($\alpha_d = 0.01$), $S_i$ drops significantly as the large number of small particles deplete the vapor (Fig. 7). Spherical growth with a constant $\alpha_d = 0.01$ (Sphr0.01) produces $N_i$ that are about three orders of magnitude greater, with much smaller particles, than when no surface kinetics is included, similar to prior studies (Gierens 2003). Because of the large difference in $N_i$ between the two cases, the supersaturation profile changes drastically in comparison to the instantaneous-nucleation simulations. With feedbacks to homogeneous freezing included, $S_i$ drops sharply to a very small value for $\alpha_d = 0.01$, because the large number of nucleated ice particles causes rapid vapor consumption. Note that the nucleated concentration is distinctly different between the Sphr1.0 and No Kinetics cases. This is because of the mean-free-path effect described earlier: the newly formed ice particles are small enough that growth rates are reduced. In the case with $\alpha_d = 1$ (Sphr1.0), the predicted $N_i$ has a similar value at steady state to the cases with predicted $\alpha_d$. Additionally, the steady-state $S_i$ reaches a value higher than Sphr0.01 but lower than the predicted $\alpha_d$ cases.

When $\alpha_d$ is predicted, $N_i$ and $S_i$ fall between the no-surface-kinetics and constant $\alpha_d = 0.01$ simulations. Spherical and nonspherical growth have about the same average value of $S_{crit}$, so all of the models that predict $\alpha_d$ tend to have $S_i$ values that reach steady state at about the same value. As a consequence, the overall nucleated concentration, and therefore IWC, is similar and does not depend on particle shape. The mean-free-path effect explains why there is no dependence of ice nucleation rate on the particle habit that is assumed: during initial droplet freezing and ice crystal growth, the particles are

![Fig. 4](image4.png)

**Fig. 4.** Aspect ratio evolution with height for the high-RH$_i$ (125%) case. Shown are results for columns and plates from the KLAH model at small (black) and large (red) $S_{crit}$.

![Fig. 5](image5.png)

**Fig. 5.** Parcel model results for the low-RH$_i$ (105%) case with instantaneous ice nucleation. Shown are the evolutions of (a) $S_i$ and (b) IWC. Different crystal growth models are indicated on the figure and simulation identifiers are provided in Table 1.
small and quasi spherical. If $\alpha_d$ is high, the vapor diffusion rate is limited by the ratio of large mean free path to particle size (or the Knudsen number). Near cloud base, when freezing occurs, the supersaturation is very high, so the predicted $\alpha_d$ is always near 1 (Figs. 8b,d). Therefore, the nucleated concentrations should match those with a constant $\alpha_d = 1$, which indeed they do. However, once nucleation ceases (Fig. 7; Ni, at 180–200 m), as $S_i$ decreases because of ice growth, $\alpha_d$ begins to decrease, and the value can drop to as low as about 0.001 (Fig. 8d), when $S_i$ reaches steady state (Fig. 7; $S_i$). These results suggest that $\alpha_d = 1$ can be assumed during homogeneous freezing; however, it cannot be assumed once nucleation ceases, since $\alpha_d$ depends on the supersaturation.

The parcel model may provide an explanation for the apparent contradiction between cirrus parcel model (Kay and Wood 2008) results and laboratory studies (Magee et al. 2006; Skrotzki et al. 2013). Kay and Wood (2008) suggested that an $\alpha_d$ value of 1 is needed in the model to match real-world cirrus $N_i$ observations, and some laboratory studies suggest high values of $\alpha_d$ as well (0.2–1.0) (Skrotzki et al. 2013). On the other hand, low values of $\alpha_d$ (~0.006) have also been obtained from laboratory measurements at low $S_i$ (Magee et al. 2006), results which tend to match those from ice crystal growth studies over ranges of supersaturation (Libbrecht 2003b).

Our parcel model results suggest that these findings might be reconciled in the following way. Near cloud base, where $S_i$ is high, $\alpha_d$ is predicted to be near unity, matching Kay and Wood’s (2008) result. However, above cloud base, as $S_i$ decreases, $\alpha_d$ becomes small, which is consistent with Magee et al.’s (2006) data and the supersaturation dependence exhibited in Libbrecht’s (2003b) data.

Though predicting particle shape produces only relatively small differences in mass, $S_i$, and $N_i$ in general, the properties of the simulated crystals and their size spectra do depend on habit evolution. For instance, the predicted particle axis lengths in the KLAH model depend critically upon the value of $S_{crit}$, with columns becoming longer and plates becoming wider at larger $S_{crit}$ (Fig. 8), leading to aspect ratios that are more extreme (Fig. 9). The physical reasons are the same as we pointed out earlier: larger $S_{crit}$ means that the minor axis ceases growing earlier, leading to more growth along the major axis and more extreme aspect ratios (Fig. 9).

Physically, the changes in shape are driven by the predicted $\alpha_d$ shown in Fig. 8. For column growth at a low value of $S_{crit}$, the predicted $\alpha_d$ is about one order of magnitude larger for the $c$, as compared to the $a$, axis. Note that the difference between the deposition coefficients for the $c$ and $a$ axes increases, for columns, by about two orders of magnitude when $S_{crit}$ is large. The deposition coefficient along the $a$ axis is so small that the growth of that axis effectively ceases. Note that the difference between the deposition coefficient along $c$ is nearly the same (about 0.07) for either the low- or high-$S_{crit}$ cases (Figs. 8b,d): It is the earlier slowing of $a$-axis growth that leads to the greater $c$-axis growth when $S_{crit}$ is large. Hence, each particle has essentially the same overall growth efficiency, and this is the reason for the similar IWC between the cases. It is interesting to note that observations of crystals with extreme aspect ratios at high altitudes and low supersaturations have been made (i.e., Jensen et al. 2010). However, laboratory measurements sometimes show crystals with low aspect ratios at high temperatures (Bailey and Hallett 2009) and at low pressures but high supersaturations (Gonda 1980). Reconciling the various measurements with theory will require significant future work.

Finally, it is worth pointing out that, whether $S_{crit}$ is large or small, the values of $S_i$, $N_i$ and IWC from predicted $\alpha_d$ cases in Fig. 7 are very similar, which suggests that the nucleated $N_i$ does not depend upon whether spherical particles or nonspherical particles are used. This result indicates that, if one wishes to capture the impacts of surface kinetics on nucleated ice concentration and mass only, a spherical model may well provide an accurate estimate.

The fact that there are large differences in the evolution of the average axis lengths suggests that there should be commensurate impacts on the size distributions of axis length and $\phi$. Indeed, as Fig. 10 shows, the evolution of the ice size spectrum is strongly impacted by the value of $S_{crit}$. When $S_{crit}$ is small, column-
Plate-like crystals are "blocky," having \( \phi \) close to unity and size spectra that are relatively narrow. However, when \( S_{\text{crit}} \) is large, aspect ratios of both crystal types become much more extreme (up to 12.4 for columns and 0.17 for plates) and the size spectra broaden significantly. The more extreme aspect ratios and broader spectra are, again, because of the near cessation of growth along a single-axis direction early on in the simulations. The increase in spectrum breadth is due primarily to the timing of axis growth cessation. An ice particle must become large enough in size before the local supersaturation over the minor crystal axis is low enough for growth to cease. Particles that nucleate at an earlier time experience the near cessation of growth of the minor axis earlier, leading to a broader size spectrum by the end of the simulation.

Ice size spectra at the end of the simulation for all growth models are presented in Fig. 11, with the results shown for small values of \( S_{\text{crit}} \). Regardless of whether \( \alpha_d \) is predicted, the size spectra for spherical particles are nearly always narrow and with smaller sizes than those for nonspherical growth. The only exception is that, when surface kinetics are not considered, ice particle maximum lengths are slightly larger than plate growth at low \( S_{\text{crit}} \). The predicted size distribution for Sphr1.0 and Sphr resemble each other, because the mean-free-path/small-particle effect acts similarly at cloud base for both cases.

b. Effect of updraft velocity

The strength of the updrafts controls the generation rate of supersaturation and, therefore, impacts the ice nucleation rates. Updraft control on nucleation is well known and was demonstrated early on by Heymsfield and Sabin (1989), among others. Because the results presented above used a single updraft speed, we examine situations where the imposed updraft speed is set to values of 5, 20, 50, 100, and 120 cm s\(^{-1}\), when the \( S_{\text{crit}} \) is

![Fig. 7. Parcel model results with nucleation process: the evolution of (left) supersaturation with respect to ice, (center) nucleated ice number concentration, and (right) ice water content for (a) a parcel lifted from an initial height to 720 m and (b) a close-up for the nucleation zone. The asterisk indicates where nucleation starts.](image-url)
small. Note that Sphr1.0 is not shown, because the results closely resemble those for Sphr. Figure 12a shows that stronger updrafts produce higher ice concentrations, which is consistent with previous studies (Lin et al. 2002). At each updraft speed, spherical growth with constant $\alpha_d = 0.01$ (Sphr.01) predicts the highest $N_i$, while spherical growth with no kinetic effects (No Kinetics) predicts the lowest $N_i$. Note that the predicted $N_i$ asymptotically approaches a constant value for $w > 50$ cm s$^{-1}$ in the case Sphr.01, because all of the available drops are frozen at these updraft speeds. Regardless of updraft speed, the growth methods that predict $\alpha_d$ produce ice concentrations between No Kinetics and the small $\alpha_d$ (Sphr.01) cases that depend weakly on the predicted shape of the ice crystals. These results are consistent with those of the prior sections. The nucleated concentration does not depend on shape, because $\alpha_d$ is close to 1 near cloud base, where ice is nucleated. In addition, near cloud base, all of the particles are nearly isometric. So one would expect that the nucleated concentration would be about the same, regardless of the particle shape. This result provides further evidence that particle shape and $\alpha_d$ may not play a role in determining overall nucleated numbers and mass. Instead, it appears to be the mean-free-path effect that impacts the nucleated concentration.

That particle shape has little influence on surface kinetic feedbacks to mass and nucleated number is further demonstrated by the lack of any variation in the cloud-base supersaturation maximum $S_\Delta$ (Fig. 12b) among the growth methods that predict $\alpha_d$. As in prior studies, when ice growth is extremely inefficient (the constant $\alpha_d = 0.01$ case), $S_\Delta$ is the greatest because of the low vapor uptake, leading to the nucleation of large numbers of particles. This result, again, underscores that using a constant, low value of $\alpha_d$, or ignoring surface kinetics entirely, leads to either over- or underestimates of particle numbers.
As one might expect, particle size tends to decrease in general (Figs. 12c,d) as updraft speed rises because of the shorter growth times. If we scale the axis lengths by growth time to expose the average growth speed, then there is a general increase with vertical velocity (Figs. 12e,f). The radius predicted by Sphr falls within the range of the No Kinetics and Sphr.01 cases discussed above.

Interestingly, the nonspherical models produce crystals with the longest axis lengths at the lowest updraft speeds (Fig. 12d). For instance, column growth produces long columns at an updraft speed of only 5 cm s$^{-1}$, while more isometric, blocky shapes occur at higher updraft speeds. Again, this is because of the slowing, or complete cessation, of growth along the minor axis as the supersaturation production decreases. One must be careful when interpreting these results, however, since each parcel is lifted over the same total distance. Hence, at 5 cm s$^{-1}$, the ice particles have a longer growth time but also less immediately available vapor, since vertical motion production of saturation is weaker. Figures 13a and 13b show how $\phi$ changes with height, and Figs. 13c and 13d show how $\phi$ changes with growth time. These four plots confirm that ice particles with more extreme aspect ratios are produced as updraft speed is reduced, and it is the earlier cessation of minor axis growth causing the more extreme $\phi$ at lower updrafts.

6. Summary, discussion, and conclusions

Modeling the evolution of atmospheric cold clouds is challenging because of the wide variety of crystal shapes and because surface molecular incorporation becomes critical at the temperatures and saturation states of the upper troposphere. Though all surfaces are rough to some degree, at low temperatures, crystal facets become smoother molecularly (Markov 2003); therefore, the formation of surface sites capable of efficient molecular incorporation from the vapor becomes less energetically favorable. Consequently, higher supersaturations, typically above some critical value $S_{\text{crit}}$ are needed for promoting accommodating steps on the surface of a crystal before its face grows substantially. While this is demonstrably true for monocrystals like hexagonal plates and columns (Libbrecht 2003a), it is likely also true for the facets of more complex crystal shapes, such as rosettes. Recent studies have suggested that to better retrieve cirrus micro- and macrophysical properties with satellite-measured radiances, crystal surfaces need to be artificially roughened in the scattering calculations [see Baran (2012) and references therein]. However, Magee et al. (2014) indicate that even crystals with substantial surface roughness can have inhibited growth. The growth efficiency, expressed quantitatively as the deposition coefficient $a_{\text{cd}}$, decreases to values substantially...

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**Fig. 9.** Aspect ratio evolution with height when homogeneous nucleation is computed. Shown are results for columns and plates from the KLAH model at small (black) and large (red) $S_{\text{crit}}$.

**Fig. 10.** The ice particle size distribution in binned classes (10 bins) at the end of parcel model simulation is shown for (a) prolate spheroidal (columnlike) and (b) oblate spheroidal (platelike) particles. Here, the $x$ axis indicates the size of the maximum dimension of a spheroidal particle [$R_{\text{max}} = \text{maximum}\ (a, c)$]. The color scale shows the aspect ratio of the particles at each $R_{\text{max}}$. Small- and large-$S_{\text{crit}}$ case results are labeled in black and red, respectively.
below unity for growth at low temperatures and supersaturations (Magee et al. 2006). Though including the influences of $a_d$ on the growth of ice is known to be important, most studies ignore it, while some use a constant value (Harrington et al. 2009), at times using the value determined for liquid drops (Lin et al. 2002).

Furthermore, though the explicit temperature and supersaturation dependence on $a_d$ is recognized, to our knowledge, no prior studies have attempted to include consistent dependency of $a_d$ on these environmental parameters, though some ad hoc methods have been used (Gierens 2003).

**FIG. 11.** The ice particle size distribution at the end of parcel model simulations from all ice growth models is shown in binned classes similar to curves in Fig. 10. Different symbols indicate different ice growth methods.

**FIG. 12.** The effect of updraft speeds on (a) $N_i$; (b) $\Delta S$ (peak $S_i$ minus $S_i, S_j$ at cloud base); (c),(d) the axis lengths at the end of the simulation; and (e),(f) the axis lengths scaled by growth time (averaged axis growth speed).
In this paper, we used both spherical and nonspherical ice crystal growth models with different treatments of $\alpha_d$ to examine the impacts of ice growth and deposition coefficient prediction on feedbacks to homogeneous freezing in parcel model simulations of cirrus. The key findings from our parcel model simulations are as follows.

When spherical particles are assumed:
- When $\alpha_d = 0.01$, the model predicts high $N_i$, consistent with prior studies.
- Assuming $\alpha_d = 1$ is a good approximation for modeling the feedbacks between growth and homogeneous freezing as long as surface mean-free-path effects are included; however, this assumption leads to short supersaturation relaxation time and low $S_i$ at steady state.
- Predicted $\alpha_d$ is near unity at cloud base because of the high supersaturation required for homogeneous freezing, but $\alpha_d$ then decreases to values as low as 0.001 above cloud base. Predicting $\alpha_d$ produces $N_i$ intermediate to the constant $\alpha_d$ cases and relatively high steady-state $S_i$.

When nonspherical particle shapes are allowed to evolve:
- Predicted $N_i$, IWC, and $S_i$ resemble the results from the spherical models when $\alpha_d$ is predicted.
- Under the same conditions, the large $S_{\text{crit}}$ case produces more extreme aspect ratios than the small $S_{\text{crit}}$ case because of larger critical supersaturations on the minor axis causing the earlier cessation of minor-axis growth.
- Updraft strength impacts the evolution of particle shapes, with weaker updrafts leading to more extreme aspect ratios.

Our results suggest that, when surface kinetic resistance is included for spherical ice growth, and $\alpha_d$ is kept constant as in most prior methods, the number of nucleated ice particles can increase by more than two orders of magnitude, and size of the particles decreases by one order of magnitude (when $\alpha_d = 0.01$), as compared to simulations that effectively ignore surface kinetic effects. However, assuming a constant value for $\alpha_d$ that is similar to that used by others, $\alpha_d = 0.01$, the ice number concentration is overestimated compared with...
available observations (Lawson et al. 2008; Krämer et al. 2009). Predicting $a_d$ leads to ice concentrations that are about an order of magnitude higher than when surface kinetics are ignored, one order of magnitude lower than when $a_d = 0.01$, but similar to $a_d = 1$. However, compared to the case where $a_d$ is predicted, using a constant value of $a_d = 1$ results in a much smaller steady-state ice supersaturation than the ice supersaturation found persisting in cirrus clouds in the upper troposphere (Peter et al. 2006; Krämer et al. 2009). The predicted $a_d$ values from cloud base to steady state range from about 1.0 to very low values of around 0.001. Our results suggest $a_d = 1$ is likely a good approximation for modeling homogeneous freezing if the mean-free-path effect is included, but assuming $a_d = 1$ may not be appropriate after nucleation ceases.

Our results also show that the spherical growth model with predicted $a_d$ produces similar evolution of $N_i$ and IWC to the nonspherical KLAH model because of the mean-free-path effect during nucleation and initial growth, and simulations with varying updraft speeds illustrate that this result is robust. Consequently, if one wishes to predict the primary influence of varying $a_d$ on nucleated ice concentrations and IWC, using a spherical growth model in which $a_d$ is predicted based on the mean $S_{crit}$ appears to be robust. This result also indicates that averaging the $S_{crit}$ values for each axis length produces input values for single-axis growth methods, such as spherical growth, that are relatively accurate. Therefore, it appears possible to average input data from laboratory experiments that determine growth along two axes [such as Libbrecht (2003b)] for single-axis growth methods.

While the evolution of nonspherical particles does not appear, at least in the cases simulated here, to have an impact on the nucleated number or IWC evolution of the simulated cirrus parcel, it does have an impact on the resulting shapes of the particles. For instance, if $S_{crit}$ is relatively low along each direction, each axis will grow more efficiently because lower ice supersaturation is required to initiate growth along each axis. This leads to crystals with aspect ratios close to unity because each axis grows relatively efficiently and because the small amount of available vapor at cirrus temperatures must be distributed over each axis. If $S_{crit}$ is larger, then the disparity in $S_{crit}$ between the axes leads to the near cessation of growth along the minor axis. The result is more extreme aspect ratios and particle major-axis lengths that become much larger than when a smaller $S_{crit}$ is assumed. This effect becomes most pronounced at low updraft speeds where supersaturation production is weak. Because $S_{crit}$ is critical to ice shape evolution, determining the $S_{crit}$ values in the laboratory is critical for predicting the ice growth in cold clouds. Future work should focus on determining $S_{crit}$ and, while axis-dependent values would be best, averages also have value, as our results illustrate.

Since cirrus clouds are often composed of crystals that are irregular in shape or are rosettes formed through polycrystalline nucleation (Bailey and Hallett 2009; Baran 2012), our spherical particle results probably bear most strongly on the growth that would occur in real cirrus. Hexagonal plates and columns occur far less often but were included to indicate the possible effects that evolving habits may have on the simulated cirrus. The range of the aspect ratio predicted by our model falls within the range of some results from Bailey and Hallett (2004, 2009). However, there is also an apparent contradiction with the work of Bailey and Hallett (2009). Our model produces thinner plates and columns at lower RHc, as opposed to high RHc, as Bailey and Hallett (2009) measured. This contradiction could be due to a number of factors. First, our model begins habit growth at the instant a water drop freezes. Since droplets are small when they freeze (5–10 µm), minor-axis growth cessation will start earlier when the relative humidity is low, thus leading to extreme aspect ratios for some crystals. It is possible that the sizes of the nucleated ice particles in Bailey and Hallett (2009) were larger or that our model begins habit evolution too early. Second, at high relative humidity, ice crystals undergo facet instabilities that lead to hollowing and can amplify growth rates and lead to thinner particles with more extreme shapes. We cannot treat facet instabilities in our model at this stage, and this could be a reason for the differences between our model and Bailey and Hallett (2009) at high relative humidity. Third, Bailey and Hallett’s ice was grown on a substrate that could have produced permanent defects in the ice crystals, therefore leading to dislocation growth. If growth occurs by dislocations, then one would expect more efficient growth on both axes even at low ice supersaturations. In our model, we assume growth occurs primarily by 2D nucleation, which has a very strong supersaturation dependence compared to dislocation growth (Fig. 1). Thus, it is possible that the surface mechanism controlling growth is different between Bailey and Hallett’s work and our modeling study. Finally, it is worth noting that not all measurements of ice growth at higher supersaturations show that crystals become thinner with more extreme aspect ratios. The measurements of Gonda (1980), taken near liquid saturation, produced crystals with aspect ratios near 1 at low atmospheric pressures.

Our parcel model study is an exploratory study of whether surface kinetics impact cirrus ice concentrations during homogeneous freezing. The parcel model simulations are most representative of cirrus driven by
buoyant processes (cloud-top radiative cooling) in isolation from any synoptic drivers, with homogeneous nucleation as the only ice source. Situations like the one we have modeled here are ideal: the dynamics are prescribed, sedimentation and parcel mixing cannot occur, and no radiative processes exist. However, the advantage of our approach is that it allows the examination of detailed microphysical processes that cannot be examined in an Eulerian framework. The parcel framework allows us to explicitly evolve the deposition coefficients of individual ice particles, and examine the potential impacts on both nucleation and the early evolution of simple ice particle shapes. The treatment of heterogeneous nucleation and the effect of more complex dynamic processes will be extended in further work.

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