Timescale analysis of aerosol sensitivity during homogeneous freezing and implications for upper tropospheric water vapor budgets

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Abstract:
Using timescales for the generation and depletion of water vapor, we predict aerosol sensitivity in clouds formed by homogeneous freezing. Our timescale analysis explains why aerosol sensitivity increases dramatically with ice deposition coefficients ($α_i << 0.1$), and also why aerosol sensitivity increases as vertical velocity increases, temperature decreases, $N_a$ decreases, and aerosol size decreases. We combine existing in-situ observations with adiabatic parcel modeling to constrain $α_i \sim 0.1$ for small ice crystals forming at high ice supersaturations. Two important implications for understanding and modeling upper tropospheric water vapor budgets emerge from our results: 1) aerosol sensitivity can be appreciable at low temperatures and slow updrafts found in the upper tropical troposphere, 2) reconciling our results with recent laboratory measurements supports theory that $α_i$ increases with ice supersaturation and/or decreases with ice crystal size.

1. Introduction

The sensitivity of clouds to aerosol properties is an important area of climate research. Twomey (1974) described the first indirect effect of increasing aerosol concentrations ($N_a [m^{-3}]$) on clouds: For a fixed water content, the drop number concentration and brightness of warm clouds increases. In contrast, modeling studies have shown that the number of ice crystals ($N_i [m^{-3}]$) resulting from homogeneous freezing is relatively insensitive to upper tropospheric $N_a$ (e.g., Jensen and Toon (1994), DeMott et al. (1997), Kärcher and Lohmann (2002a, 2002b), Kärcher and Ström (2003)). In other words, these studies imply weak aerosol sensitivity, or that increasing $N_a$ has a negligible effect on $N_i$ or $η_a << 1$ where $η_a$ is an aerosol sensitivity parameter defined as:

$$η_a \equiv \frac{d( \ln N_i )}{d \ln(N_a)}$$ (1)
Observations show a positive but weak correlation between $N_i$ and $N_a$ during cold cloud formation (Siefert et al., 2004). If $\eta_a$ is larger than these studies suggest, an increase in anthropogenic $N_a$ will increase cold cloud $N_i$. For a fixed water content, increasing $N_i$ will increase cold cloud albedos and alter radiative fluxes. In addition, increasing $N_i$ will increase the drawdown of supersaturation in the upper tropical troposphere and therefore alter the water vapor budget of the stratosphere. Given the influence of cold cloud microphysical properties on radiative fluxes and water vapor budgets, it is important to understand the atmospheric conditions under which cold clouds are sensitive to changes in $N_a$.

In this paper, we investigate the physical factors that determine $\eta_a$ in cold clouds formed by homogeneous freezing. Although aerosols that serve as heterogeneous ice nuclei can alter cold cloud properties, we do not consider the impact of heterogeneous freezing on cold cloud aerosol indirect effects. Heterogeneous freezing is not well constrained by observations or theory. In addition, observations of large ice supersaturations (e.g., Ovarlez et al. (2002)) and low ice nuclei concentrations (e.g., DeMott et al., 2003) suggest that homogeneous nucleation is an important ice formation mechanism in the upper troposphere. In Section 2, we use an adiabatic parcel model with binned ice microphysics (Kay et al., 2006) to demonstrate that a simple timescale ratio explains the dependence of $\eta_a$ upon thermodynamic factors including vertical velocity ($w$ [m s$^{-1}$]) and co-varying temperature ($T$ [°C]) and pressure ($P$ [mb]), and microphysical factors including $N_a$, hydrated aerosol radius ($r_a$ [m]), and the ice deposition coefficient or mass accommodation coefficient ($\alpha_i$). In Section 3, we discuss the implications of our results for the atmosphere. In Section 4, we summarize our results and provide suggestions for future work.

Our work builds on the analytical results of Kärcher and Lohmann (2002a,b) (hereafter KL). KL found that $N_i$ is primarily controlled thermodynamically rather than microphysically, with a strong dependence upon $w$. KL compared ice crystal growth timescales to the timescale of the freezing event, and found two homogeneous freezing regimes: a “fast-growth” and a “slow-growth” regime. Yet, KL did not consider two important microphysical factors: 1) KL did not address the current uncertainty in $\alpha_i$, i.e., the fraction of impinging water vapor molecules that are incorporated into an ice crystal lattice (Pruppacher and Klett, 1997). KL assumed $\alpha_i=0.5$ in their analysis, but laboratory measurements of $\alpha_i$ vary from 0.006 to 1 (e.g., Haynes et al. (1992), Magee et al. (2006)). 2) KL did not explicitly treat the depletion of $N_a$ by freezing, a crucial factor when $N_i$ are limited by $N_a$.

2. What determines $\eta_a$?

For simplicity, we consider cold cloud formation during adiabatic ascent at constant $w$. The supersaturation with respect to ice ($S_i$) increases during ascent and, at the beginning of freezing
(time \( t = 0 \)), the homogeneous freezing rate \( (J_{\text{hom}} \, [m^{-3} \, s^{-1}]) \), an exponentially increasing function of \( S_i \) reaches a threshold value \( (J_o \, [m^{-3} \, s^{-1}]) \). Freezing stops at a later time (\( t_{\text{event}} \, [s] \)) when vapor deposition on newly formed ice crystals causes \( S_i \) to decrease and \( J_{\text{hom}} \) to decrease below \( J_o \). Given this physical picture of cloud formation, the \( N_i \) generated in a homogeneous freezing event can be approximated as:

\[
N_i = \int_0^{t_{\text{event}}} J_{\text{hom}}(S_i(t)) \frac{4}{3} \pi a_o(t)^3 N_o(t) \, dt
\]  

(2)

With this physical model of cold cloud formation, the partitioning of water between the ice and vapor phase depends on a competition between lifting (cooling), which increases \( S_i \), and ice crystal growth, which depletes \( S_i \). Using timescale notation, the dependence of \( S_i \) on the competition between lifting and growth can be expressed as:

\[
\frac{dS_i}{dt} = \frac{S_i}{\tau_{\text{lift}}} - \frac{S_i}{\tau_{\text{growth}}}
\]  

(3a)

where \( \tau_{\text{lift}} \) is the timescale for increase of \( S_i \) via cooling through ascent, and \( \tau_{\text{growth}} \) is a timescale for growth of freshly nucleated ice crystals by vapor deposition. Because ice crystal growth results from vapor deposition, \( \tau_{\text{growth}} \) is also a timescale for the drawdown of \( S_i \).

We hypothesize that \( \eta_a \) can be predicted by the timescale ratio, \( R \).

\[
R \equiv \frac{\tau_{\text{growth}}}{\tau_{\text{lift}}}
\]  

(3b)

In other words, \( \eta_a \) is entirely determined by the competition between the rates of lifting (cooling) and ice crystal growth. When \( R \gg 1 \) ice crystal growth is relatively slow when compared with the cooling that increases \( S_i \). Consequently, large \( S_i \) values occur for long periods, and almost all of the available aerosol can freeze (\( \eta_a \) approaches 1). When \( R \ll 1 \), ice crystal growth is relatively fast when compared to cooling. As a result, large \( S_i \) are quickly reduced, and only a small number of the available aerosol can freeze (\( \eta_a \) approaches 0). Note that \( R \gg 1 \) is roughly equivalent to KL’s “fast growth” regime while \( R \ll 1 \) is roughly equivalent to KL’s “slow growth regime”.

To evaluate if \( R \) can quantitatively predict the aerosol sensitivity parameter \( \eta_a \), analytical expressions for \( \tau_{\text{lift}} \) and \( \tau_{\text{growth}} \) are required. In an analytical analysis of a rising adiabatic parcel based on Eq. (2) and KL, the time constants \( \tau_{\text{lift}} \) and \( \tau_{\text{growth}} \) arise naturally and are defined as follows:
\[ \tau_{\text{lift}} = [Q_1 w]^{-1} \]  

(4)

with \[ Q_1 = \frac{\Gamma}{T} \left( \frac{L_s (S_i + 1)}{R_s T} - \frac{5}{2} \right) \]

where \( Q_1 \) is a thermodynamic constant, \( \Gamma = 0.0098 \text{ K m}^{-1} \) is the dry adiabatic lapse rate (appropriate for the low temperatures being considered), \( L_s = 2.834 \times 10^6 \text{ J kg}^{-1} \) is the latent heat of sublimation, and \( R_v = 461 \text{ J K}^{-1} \text{ kg}^{-1} \) is the gas constant for water vapor.

\[ \tau_{\text{growth}} = [(K N_a)^{2/3} (S_i D_v^*)]^{-1} \]  

(5)

with \[ K = \sqrt{2 \left( \rho_{\text{sat-i}} / \rho_i \right)} \]

and \( D_v^* = \frac{D_v}{r_a + \lambda} = \sqrt{\frac{2 \pi M_w}{r_a \alpha_i}} \frac{R_{\text{ideal}} T}{R_v} \frac{r_a + \lambda}{r_a + \lambda} \frac{\alpha_i}{r_a} \]

where \( K \) is a constant, \( \rho_{\text{sat-i}} \) is the saturation vapor density with respect to ice (kg m\(^{-3}\)), \( \rho_i = 900 \text{ kg m}^{-3} \) is the density of ice, \( D_v^* \) is the modified vapor diffusivity (m\(^2\) s\(^{-1}\)) (Eq. 13-14 in Pruppacher and Klett (1997)) which includes impedances to growth due to vapor diffusivity and surface processes but neglects the relatively small thermal impedance to growth, \( D_v \) is the vapor diffusivity (m\(^2\) s\(^{-1}\)) (Eq. 13-3 in Pruppacher and Klett (1997)), \( \lambda \) is the molecular mean free path (m) (Eq. 16.20 in Jacobson (1999)), \( M_w = 0.018015 \text{ kg mole}^{-1} \) is the molecular weight of water, and \( R_{\text{ideal}} = 8.3145 \text{ J K}^{-1} \text{ mole}^{-1} \) is the ideal gas constant.

By calculating \( \tau_{\text{lift}} \) and \( \tau_{\text{growth}} \) in a number of model experiments (adiabatic parcel model with binned microphysics, configuration described in Table 1), we evaluate if \( \eta_a \) can be predicted by \( R \) alone. In all cases, we calculated \( R \) using the parcel model output at the timestep before freezing begins, herein defined as when the ice particle production rate \((\text{d}N_i/\text{d}t)\) exceeds 1 m\(^3\) s\(^{-1}\).

To introduce our parcel model experiments, we first show an experiment in which we only vary \( \alpha_i \) (Figure 1). Decreasing \( \alpha_i \) increases \( R \) by increasing \( \tau_{\text{growth}} \) without affecting \( \tau_{\text{lift}} \). Indeed, the increase in \( S_i \) and \( J_{\text{hom}} \) resulting from long \( \tau_{\text{growth}} \) is why small \( \alpha_i \) lead to large \( N_i \) (Gierens et al., 2003). The \( \alpha_i \) lifting experiment reveals the dramatic effect of \( R \) on the sensitivity of \( N_i \) to \( N_a \) and on the
drawdown of $S_i$. When efficient growth is assumed ($R<<1$, $\alpha_i > 0.1$, blue curves Figure 1), $J_{hom}$ and $S_i$ are quickly reduced by ice crystal growth, and $N_i$ is not sensitive to $N_a$. In contrast, with inefficient growth ($\alpha_i <<0.1$, $R>>1$, red curves Figure 1), $J_{hom}$ and $S_i$ reach large values and the sensitivity of $N_i$ to $N_a$ increases. It is interesting to note that only the lowest $\alpha_i$ ($\alpha_i =0.001$) result in persistent high $S_i$ ($S_i > 40\%$) after the freezing event ends. Surprisingly, $S_i$ is depleted faster when $\alpha_i=0.01$ than when $\alpha_i = 1$. This counter-intuitive result is explained as follows: When $\alpha_i$ decreases, individual particles grow inefficiently allowing both the peak $S_i$ and $J_{hom}$ to increase. When the peak $J_{hom}$ increases, $N_i$ and the total surface area dramatically increase. Thus, even though individual particles are growing inefficiently, the increase in total ice surface area allows the $S_i$ drawdown to be faster when $\alpha_i=0.01$ than when $\alpha_i = 1$.

3. Implications for the atmosphere

When $\alpha_i <<0.1$, plausible variations in $\alpha_i$ can dramatically change $\eta_a$ and alter the sensitivity of $\eta_a$ to variations in other microphysical and thermodynamic variables. The influence of $\alpha_i$ on $\tau_{growth}$ is more important at low $\alpha_i$ because $D_i^*$ does not depend directly on $\alpha_i$, but on $r_a/(r_a + \lambda) + \lambda/\alpha_i r_a \approx 1 + \lambda/\alpha_i r_a$ (Eq. 5). When $\lambda/\alpha_i r_a <<1$, the precise value of $\alpha_i$ is unimportant because diffusive impediments to growth are more important than surface impediments to growth. Reviews of laboratory measurements at cold cloud temperatures suggest that $\alpha_i$ for ice crystals could be as low as 0.001 and as high as 1 (Haynes et al., 1992); recent laboratory measurements found $\alpha_i=0.006$.
(Magee et al., 2006). Given the sensitivity of $\eta_a$ to $\alpha_i$ when $\alpha_i < 0.1$, discrepancies between $\alpha_i$ measurements must be resolved.

Fortunately, existing observations can be used to constrain $\alpha_i$ for small ice crystals forming at high $S_i$ in the atmosphere. In general, observed $N_i$ (0.001-10 cm$^{-3}$ (e.g., Mace et al. 2001, Kärcher and Ström, 2003) rarely approach observed $N_a$ (10-500 cm$^{-3}$ (e.g., Rogers et al, 1998, Minikin et al. 2003)). The INCA field campaign (Kärcher and Ström, 2003) provides a unique opportunity to constrain $\alpha_i$. Using INCA measurements, we require $\alpha_i \approx 0.1$ to simultaneously match the mean $N_i$, $N_a$, $T$, and $w$ in lifting parcel model experiments (Table 2). With $\alpha_i = 0.006$ (Magee et al., 2006), modeled $N_i$ are orders of magnitude larger than INCA-observed $N_i$. If present, shattering of ice crystals by aircraft probes (e.g., Field et al, 2003, 2006) would reduce observed $N_i$ and increase the value of $\alpha_i$ required to match INCA-observed values with parcel modeling experiments. Uncertainty in the observed $w$ also has an important effect on the constrained $\alpha_i$. If a large range of $w$ are considered (3 cm/s < $w$ < 50 cm/s), a much larger range of $\alpha_i$ (0.01 < $\alpha_i$ < 1) are consistent with the mean observed $N_i$.

In summary, atmospheric observations suggest that $\eta_a$ rarely approaches 1 and $\alpha_i$ are ~ 0.1 for small ice crystals forming at high $S_i$. Therefore, laboratory observations of $\alpha_i = 0.006$ (Magee et al., 2006), which were made at $S_i < 20\%$, may only be appropriate for large ice crystals or at low $S_i$. There is a theoretical basis for the latter possibility (Nelson and Baker (1996), Wood et al. (2001)), but further measurements are required to constrain the behavior of $\alpha_i$ as a function of $S_i$ and ice crystal size.

Assuming $\alpha_i = 0.1$ for small ice crystals forming at large $S_i$, cold clouds in the atmosphere primarily form in a regime where $\eta_a << 1$ (Figure 3). In other words, the $N_i$ resulting from homogeneous freezing is generally thermodynamically-limited, not aerosol-limited. This outcome agrees with KL, Hoyle et al. (2005), Kärcher and Ström, (2003), and Kay et al. (2006), who all found that atmospheric $N_i$ are primarily controlled by $w$. With $\alpha_i = 0.1$, our modeling results do suggest there are conditions under which $N_i$ does depend on $N_a$. First, $\eta_a$ increases at very large $w$ (approximately $w > 100$ cm s$^{-1}$ when $T = -50$ C and $\alpha_i = 0.1$). Second, there is a significant increase in $\eta_a$ at temperatures such as those where cirrus clouds form in the tropical upper troposphere. Finally, $\eta_a$ increases as $N_a$ decreases, which can be important at high $w$ or low $T$. 
4. Summary and Discussion

In this study, we used analytical analysis, parcel modeling, and observations to understand the sensitivity of ice crystal concentration produced by homogeneous freezing to changes in aerosols ($\eta_a$ - Eq. 1). Our primary findings were:

- The dependence of $\eta_a$ on a large number of microphysical and thermodynamic variables can be explained and predicted using a single timescale ratio, $R$ (Eq. 3).
- In modeling sensitivity experiments, $\eta_a$ increases dramatically when $\alpha_i < 0.1$, but also when $w$ increases, $T$ decreases, $N_a$ decreases, or $r_a$ decreases.
- Using existing atmospheric observations and simple modeling, we suggest $\alpha_i \geq 0.1$ for small ice crystals forming at high $S_i$. As a consequence $\eta_a$ is small under most atmospheric conditions, but may increase at large $w$ ($w > 100$ cm s$^{-1}$) or at low $T$ ($T < -70 \, ^\circ C$).
- In order to reconcile laboratory and atmospheric observations, we suggest that $\alpha_i$ likely decreases with ice crystal size or increases with $S_i$.

We suggest that future studies investigate the implications of this study for water vapor budgets at low $T$. Hoyle et al. (2005) proposed that co-varying $T$ and $P$ decreases have competing effects on ice crystal growth rates, and as a result, the $N_i$ generated by homogeneous freezing does not depend on $T$. In contrast, our results suggest the balancing of $T$ and $P$ effects on $\tau_{growth}$ is not universal and that $\tau_{growth}$, $\eta_a$, and $N_i$ do increase at low $T$. Simultaneous observations of $N_a$ and $N_i$ at low $T$ could be used to estimate atmospheric $\alpha_i$ and to evaluate the dependence of $\eta_a$ on $T$ presented in this study, but it will be critical to make accurate estimates of $w$ in addition to microphysical parameters in order to determine these constraints. Finally, future work should constrain variations in $\alpha_i$ as a function of $S_i$ and ice crystal size. Constraining variations in $\alpha_i$ will be especially important when evaluating if $\alpha_i$ can explain atmospheric observations of large persistent $S_i$ in the upper troposphere (Peter et al., 2006).

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Figure Captions

Figure 1. Time series of parcel model lifting experiments (Table 1) with $w = 10 \text{ cm s}^{-1}$, $T_0 = -50 \degree \text{C}$, $P_0 = 250 \text{ mb}$, $N_a=100 \text{ cm}^{-3}$, and $r_a=0.2 \mu \text{m}$.

Figure 2. Aerosol sensitivity vs. R for all parcel model runs (panel A) and for a sensitivity test with a range of $\alpha$ (panel B), $T_0$ (panel C) and $r_{a\text{-dry}}$ (panel D). For panels B-D: 1) circles $w=2 \text{ cm s}^{-1}$, diamonds $w=10 \text{ cm s}^{-1}$, and squares $=100 \text{ cm s}^{-1}$. 2) Aerosol sensitivity was calculated using the change in $N_i$ from model runs with $N_a=100 \text{ cm}^{-3}$ and $N_a=500 \text{ cm}^{-3}$ (see Eq. 1). 3) Unless otherwise indicated, values are base values: $T_0 = -50 \degree \text{C}$, $P_0 = 250 \text{ mb}$, and $r_a=0.2 \mu \text{m}$.

Figure 3. Maximum $N_i$ contoured as a function of vertical velocity ($w$) and aerosol number concentration ($N_a$) from the parcel model lifting experiments with $\alpha=0.1$, $T_0 = -50 \degree \text{C}$, $P_0 = 250 \text{ mb}$, and $r_a=0.2 \mu \text{m}$. Colors indicate the aerosol sensitivity parameter $\eta_a$ (Eq. 1) and range from the thermodynamically limited nucleation regime ($\eta_a=0$) to the aerosol-limited nucleation regime ($\eta_a=1$). The green line shows where $\eta_a=0.5$ line for $T=-80 \degree \text{C}$ and $P_0=100 \text{ mb}$. The INCA field campaign observations are indicated in the transparent white circle (10-90% percentile values taken from Kärcher and Ström (2003) and Minikin et al. (2003)).
Table 1. Parcel model description and configuration used for this study. In all model runs, $\alpha_i$ does not depend on ice crystal size or on ice supersaturation.

<table>
<thead>
<tr>
<th>Parcel model description</th>
<th>Model configuration</th>
</tr>
</thead>
<tbody>
<tr>
<td>• description and validation in Kay et al. (2006)</td>
<td>• parcel lifted at a constant vertical velocity (w)</td>
</tr>
<tr>
<td>• binned ice microphysics (300 bins)</td>
<td>• homogeneous nucleation (Koop et al., 2000) only</td>
</tr>
<tr>
<td>• aerosol activation using Köhler curve, monodisperse sulfuric acid aerosol</td>
<td>• ice crystal fallout included with a</td>
</tr>
<tr>
<td>with an individual dry weight of $10^{-16}$ kg and variable number concentration</td>
<td>parcel depth=100 m</td>
</tr>
<tr>
<td>• saturation vapor pressures $e_s$ and $e_{s,ice}$ from Murphy and Koop (2005)</td>
<td></td>
</tr>
</tbody>
</table>

Table 2. INCA observations (Kärcher and Ström, 2003) and parcel model predicted $N_i$. Parcel model $N_i$ are found from constant lifting experiments (Table 1) with INCA-observed $w$, $T$, and $N_a$, and $P_0 = 250$ mb. $\alpha_i$ values of 0.057 and 0.13 were required to match the mean Scotland and Chile observations respectively.

<table>
<thead>
<tr>
<th>INCA Observations (Kärcher and Ström, 2003)</th>
<th>Parcel Model $N_i$ (# cm$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$w$ (cm s$^{-1}$) $T$ (°C) $N_a$ (# cm$^{-3}$) $N_i$ (# cm$^{-3}$) $\alpha_i =$</td>
<td>$\alpha_i =$ $\alpha_i =$ $\alpha_i =$</td>
</tr>
<tr>
<td></td>
<td>1</td>
</tr>
<tr>
<td>Scotland</td>
<td>26.2</td>
</tr>
<tr>
<td>Chile</td>
<td>23</td>
</tr>
</tbody>
</table>
\section*{Figure 1}

\begin{figure}[h]
\centering
\begin{subfigure}{0.45\textwidth}
\centering
\includegraphics[width=\textwidth]{figure1a.png}
\caption{\(N_i\) (\# cm\(^{-3}\))}
\end{subfigure}
\hfill
\begin{subfigure}{0.45\textwidth}
\centering
\includegraphics[width=\textwidth]{figure1b.png}
\caption{\(dN_i/dt\) (\# cm\(^{-3}\) sec\(^{-1}\))}
\end{subfigure}
\begin{subfigure}{0.45\textwidth}
\centering
\includegraphics[width=\textwidth]{figure1c.png}
\caption{\(J_{\text{tot}}\) (m\(^{-3}\) sec\(^{-1}\))}
\end{subfigure}
\hfill
\begin{subfigure}{0.45\textwidth}
\centering
\includegraphics[width=\textwidth]{figure1d.png}
\caption{Ice Supersaturation - \(S_i\) (%)}
\end{subfigure}
\caption{Variation of ice number concentration, ice growth rate, ice mass flux, and ice supersaturation with time for different values of \(R\) and \(\alpha\).}
\end{figure}

- \(R=0.017, \alpha=1\)
- \(R=0.15, \alpha=0.1\)
- \(R=1.5, \alpha=0.01\)
- \(R=15, \alpha=0.001\)
Aerosol Sensitivity \( \frac{d \ln N_i}{d \ln N_a} \).

\[ \begin{align*}
T_0 &= 80 \, ^\circ \text{C}, P_0 = 100 \text{ mb} \\
T_0 &= 60 \, ^\circ \text{C}, P_0 = 170 \text{ mb} \\
T_0 &= \text{BASE (-50} \, ^\circ \text{C}, P_0 = 250 \text{ mb)} \\
T_0 &= 40 \, ^\circ \text{C}, P_0 = 340 \text{ mb}
\end{align*} \]

\( \alpha_i = 0.001 \)
\( \alpha_i = 0.01 \)
\( \alpha_i = \text{BASE (0.1)} \)
\( \alpha_i = 1 \)

\( R_{a-dry} = 0.1 \mu m \)
\( R_{a-dry} = \text{BASE (0.2} \mu m) \)
\( R_{a-dry} = 0.5 \mu m \)

Model runs in panels B-D
All model runs in this paper
INCA Observations

Aerosol sensitivity - $\eta_a$ ($T=-50^\circ C$)

Vertical velocity - $w$ (cm s$^{-1}$)

Aerosol number concentration - $N_a$ (# cm$^{-3}$)

thermodynamically limited freezing

aerosol limited freezing

$\eta_a=0.5$ ($T=-80^\circ C$)