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

Jennifer E. Kay¹ and Robert Wood²

Received 13 November 2007; revised 18 February 2008; accepted 17 April 2008; published 23 May 2008.

[1] 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 ($\alpha_i$) $\ll 0.1$, and also why aerosol sensitivity increases as vertical velocity increases, temperature decreases, aerosol number decreases, and aerosol size decreases. We combine existing in-situ observations with adiabatic parcel modeling to constrain $\alpha_i \geq 0.1$ 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 moderate updrafts ($\sim 5 \text{ cm/s}$) in the upper tropical troposphere, 2) reconciling our results with recent laboratory measurements supports theory that $\alpha_i$ increases with ice supersaturation and/or decreases with ice crystal size.


1. Introduction

[2] 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 ($N_a \text{ [m}^{-3} \text{]}$) on clouds: For a fixed water content, the drop number concentration and brightness of warm clouds increases.

[3] In contrast, modeling studies have shown that the number of ice crystals ($N_i \text{ [m}^{-3} \text{]}$) 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 $\eta_a \ll 1$ where $\eta_a$ is an aerosol sensitivity parameter defined as:

$$\eta_a \equiv \frac{d(\ln N_i)}{d \ln (N_a)}$$

Observations show a positive but weak correlation between $N_i$ and $N_a$ during cold cloud formation [Seifert 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 $N_a$.

[4] 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; Jensen et al., 2001] and low ice nuclei concentrations [e.g., DeMott et al., 2003] suggest that homogeneous freezing of aqueous aerosols 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 ($w \text{ [m s}^{-1} \text{]}$) and co-varying temperature ($T \text{ [}^\circ\text{C}]$) and pressure ($P \text{ [mb]}$), and microphysical factors including $N_m$, dry aerosol radius ($r_{a, dry} \text{ [m]}$), 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.

[5] Our work builds on the analytical results of Kärcher and Lohmann (2002a, 2002b) (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 \( N_f \) are limited by \( N_a \).

2. What Determines \( \eta_a \)?

[6] For simplicity, we consider cold cloud formation during adiabatic ascent at constant \( \omega \). The supersaturation with respect to ice \( (S_i) \) increases during ascent and, at the beginning of freezing \( (t = 0) \), the homogeneous freezing rate \( (J_{\text{hom}} \left[ \text{m}^3 \text{s}^{-1} \right]) \), an exponentially increasing function of \( S_i \) reaches a threshold value \( (J_o \left[ \text{m}^3 \text{s}^{-1} \right]) \). Freezing stops at a later time \( (t_{\text{event}} [\text{s}]) \) 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 \( N_f \) generated in a homogeneous freezing event can be approximated as:

\[
N_f = \int_0^{t_{\text{event}}} J_{\text{hom}}(S_i(t)) \frac{4}{3} \pi r_a(t)^3 N_a(t) dt
\]

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:

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

where \( \tau_{\text{lift}} \) is the timescale for increase of \( S_i \) via cooling, 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 \( S_i \).

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

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

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 \( (\eta_a \text{ approaches } 1) \). 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 \( (\eta_a \text{ approaches } 0) \). 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”.

[8] 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 equation (2) and KL, the time constants \( \tau_{\text{lift}} \) and \( \tau_{\text{growth}} \) arise naturally and are defined as follows:

\[
\tau_{\text{lift}} = [Q_1 \omega]^{-1}
\]

with

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

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.

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

with

\[
K = \sqrt{2 \rho_{\text{sat}} a (\rho)}
\]

and

\[
D_v^* = \frac{D_v}{r_a + \lambda_{\text{ff}} \sqrt{\frac{2 \pi M_w}{R_{\text{ideal}} T}}} = \frac{D_v}{r_a + \lambda_{\text{ff}} \sqrt{\frac{2 \pi M_w}{R_{\text{ideal}} T}}}
\]

where \( K \) is a constant, \( \rho_{\text{sat}} \) is the saturation vapor density with respect to ice \( (\text{kg m}^{-3}) \), \( \rho_i = 900 \text{ kg m}^{-3} \) is the density of ice, \( D_v^* \) is the modified vapor diffusivity \( (\text{m}^2 \text{s}^{-1}) \) [Pruppacher and Klett, 1997, equation (13-14)] 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 \( (\text{m}^2 \text{s}^{-1}) \) [Pruppacher and Klett, 1997, equation (13-3)], \( \lambda \) is the molecular mean free path \( (\text{m}) \) [Jacobson, 1999, equation (16.20)], \( M_w = 0.018015 \text{ kg mole}^{-1} \) is the molecular weight of water, and \( R_{\text{ideal}} = 8.3145 \text{ J K}^{-1} \text{ mole}^{-1} \) is the ideal gas constant.

[9] 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
η_a can be predicted by R alone. In all cases, we calculated R using the parcel model output at the time step before freezing begins, herein defined as when the ice particle production rate (dN_i/dt) exceeds 1 m^{-3}s^{-1}. When α_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 α_i = 0.01 than when α_i = 1.

Although the α_i lifting experiment revealed that there are complex relationships between η_a and atmospheric variables, our parcel model runs suggest that the aerosol sensitivity parameter η_a can be predicted by R alone (Figure 2a). When multiple parcel model experiments are plotted on one R versus η_a graph (Figure 2a), they collapse onto a single curve. In other words, one can predict changes in η_a by evaluating the effect of changing external atmospheric conditions on R (equation (3)).

Using a sensitivity test approach, we explored the influence of plausible variations in thermodynamic (w, T, P) and microphysical (α_i, r_{a-dry}) variables on η_a and R (Figures 2b–2d). Through R, we can understand the physical basis for the influence of these parameters on η_a. Aerosol sensitivity increases with w through τ_{lift}. With α_i > 0.1, η_a is small; however when α_i < 0.1, τ_{growth} increases and η_a increases dramatically. Aerosol sensitivity increases when T ≲ -70°C because low T increases τ_{growth}. Finally, variations in r_{a-dry} have only a limited influence on η_a. As r_{a-dry} decreases, η_a slightly increases because for a fixed N_a, reducing r_{a-dry} increases τ_{growth}.

3. Implications for the Atmosphere

When α_i < 0.1, plausible variations in α_i can dramatically change η_a and alter the sensitivity of η_a to variations in other microphysical and thermodynamic variables. The influence of α_i on τ_{growth} is more important at
low $\alpha_i$ because $D_i^e$ does not depend directly on $\alpha_i$, but on $r_i^e (r_i^a + \lambda + \lambda/\alpha_i r_i^a) \approx 1 + \lambda/\alpha_i r_i^a$ (see equation (5)). When $\lambda/\alpha_i r_i^a < 1$, 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 of magnitude larger than INCA-observed $N_i$. When present, shattering of ice crystals by aircraft probes [e.g., Field et al., 2003; McFarquhar et al., 2007] and “aging” of ice crystal size distributions (e.g., via dispersion) would reduce observed $N_i$ and increase the value of $\alpha_i$ required to match INCA observations with parcel modeling experiments. Indeed, matching modeled and observed $N_i$ during a wave cloud case from SUCCESS suggests $\alpha_i > 0.5$ [Jensen et al., 1998]. Uncertainty in the INCA-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 $< \alpha_i < 1$) is consistent with the mean observed $N_i$.

In summary, atmospheric observations suggest that $\eta_a$ rarely approaches 1 and $\alpha_i$ are $\geq 0.1$ 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], 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). 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 $T = -50^\circ$C and $\alpha_i = 0.1$). Second, there is a significant increase in $\eta_a$ at low temperature. These results contrast with those of Hoyle et al. [2005], who suggested that $T$ and $P$ effects on $\tau_{growth}$ are in balance. Finally, $\eta_a$ increases as $N_{ai}$ 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 aerosol sensitivity during homogeneous freezing ($\eta_a$, equation (1)). We found the following.

1. The $\eta_a$ can be explained and predicted using a single timescale ratio, $R$ (equation (3)).

2. The $\eta_a$ increases dramatically when $\alpha_i < 0.1$, but we constrain $\alpha_i \geq 0.1$ for small ice crystals forming at high $S_i$. With $\alpha_i = 0.1$, our model shows that $\eta_a$ is small under most atmospheric conditions, but quickly increases to appreciable values with large $w$ ($w > 100$ cm s$^{-1}$ when $T = -50^\circ$C) or at low $T$ ($w < 5$ cm s$^{-1}$ when $T = -80^\circ$C).

Simultaneous observations of $N_{ai}$, $N_i$, and $w$ 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. Observations of low $N_{ai}$ in the TTL [e.g., Peter et al., 2003] may seem confounding to our modeling results, but could be consistent if homogeneous freezing occurs at very low $w$ (e.g.,

![Figure 3. Maximum $N_i$ contoured as a function of vertical velocity ($w$) and aerosol number concentration ($N_{ai}$) from the parcel model lifting experiments with $\alpha_i = 0.1$, $T_0 = -50^\circ$C, $P_0 = 250$ mb, and $r_{a-dry} = 0.2 \mu m$. Colors indicate the aerosol sensitivity parameter $\eta_a$ (equation (1)) and range from the thermodynamically limited freezing regime ($\eta_a = 0$) to the aerosol-limited freezing regime ($\eta_a = 1$). The green line shows where $\eta_a = 0.05$ line for $T_0 = -80^\circ$C and $P_0 = 100$ 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]).]
mm/s proposed by Luo et al. [2003] or if measurements fail to detect small ice crystals. Finally, our work demonstrates the importance of constraining $\alpha_i$ variations as a function of $S_i$ and ice crystal size. Understanding $\alpha_i$ variations 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].

[21] Acknowledgments. J.E.K. was supported by NSF-ATM-02-1147. R.W. was supported by startup funds from the University of Washington. Both authors thank Dr. Marcia Baker for her encouragement and her contributions to this work.

References


J. E. Kay, Climate and Global Dynamics, National Center for Atmospheric Research, P.O. Box 3000, Boulder, CO 80307-3000, USA.

R. Wood, Department of Atmospheric Sciences, University of Washington, Seattle, WA 98195, USA.