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

Jennifer E. Kay[1] and Robert Wood[2]
[1] National Center for Atmospheric Research, Boulder, CO, jenkay@ucar.edu, ph: 303-497-1730
[2] Department of Atmospheric Sciences, University of Washington, Seattle, WA

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 $\alpha_i \sim 0.1$ 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:

$$\eta_a \equiv \frac{d(\ln N_i)}{d\ln(N_a)} \qquad (1)$$

Observations show a positive but weak correlation between N_i and N_a during cold cloud formation (Siefert et al., 2004). If η_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 η_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 η_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 (α_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 α_i, i.e., the fraction of impinging water vapor molecules that are incorporated into an ice crystal lattice (Pruppacher and Klett, 1997). KL assumed α_i=0.5 in their analysis, but laboratory measurements of α_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 η_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_{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_{event} [s]) when vapor deposition on newly formed ice crystals causes S_i to decrease and J_{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:

$$N_i = \int_0^{t_{event}} J_{\text{hom}}(S_i(t))\frac{4}{3}\pi r_a(t)^3 N_a(t)dt \quad (2)$$

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:

$$\frac{dS_i}{dt} = \frac{S_i}{\tau_{lift}} - \frac{S_i}{\tau_{growth}} \quad (3a)$$

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

We hypothesize that η_a can be predicted by the timescale ratio, R.

$$R \equiv \frac{\tau_{growth}}{\tau_{lift}} \quad (3b)$$

In other words, η_a is entirely determined by the competition between the rates of lifting (cooling) and ice crystal growth. When $R >> 1$ 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 (η_a approaches 1). When $R << 1$, 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 (η_a approaches 0). Note that $R>>1$ is roughly equivalent to KL's "fast growth" regime while $R << 1$ is roughly equivalent to KL's "slow growth regime".

To evaluate if R can quantitatively predict the aerosol sensitivity parameter η_a, analytical expressions for τ_{lift} and τ_{growth} are required. In an analytical analysis of a rising adiabatic parcel based on Eq. (2) and KL, the time constants τ_{lift} and τ_{growth} arise naturally and are defined as follows:

$$\tau_{lift} = [Q_1 w]^{-1} \tag{4}$$

$$\text{with } Q_1 = \frac{\Gamma}{T}\left(\left(\frac{L_s(S_i+1)}{R_v T}\right) - \frac{5}{2}\right)$$

where Q_1 is a thermodynamic constant, $\Gamma = 0.0098$ K m^{-1} is the dry adiabatic lapse rate (appropriate for the low temperatures being considered), $L_s = 2.834\times10^6$ J kg^{-1} is the latent heat of sublimation, and $R_v = 461$ J K^{-1} kg^{-1} is the gas constant for water vapor.

$$\tau_{growth} = [(KN_a)^{2/3}(S_i D^*_{v_v})]^{-1} \tag{5}$$

$$\text{with } K = \sqrt{2(\rho_{sat-i}\rho_i)}$$

$$\text{and } D^*_v = \frac{D_v}{\frac{r_a}{r_a+\lambda} + \frac{D_v}{r_a \alpha_i}\sqrt{\frac{2\pi M_w}{R_{ideal}T}}} = \frac{D_v}{\frac{r_a}{r_a+\lambda} + \frac{\lambda}{\alpha_i r_a}}$$

where K is a constant, ρ_{sat-i} is the saturation vapor density with respect to ice (kg m^{-3}), $\rho_i = 900$ 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)), λ is the molecular mean free path (m) (Eq. 16.20 in Jacobson (1999)), $M_w = 0.018015$ kg mole^{-1} is the molecular weight of water, and $R_{ideal} = 8.3145$ J K^{-1}mole^{-1} is the ideal gas constant.

By calculating τ_{lift} and τ_{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 timestep before freezing begins, herein defined as when the ice particle production rate (dN_i/dt) exceeds 1 m^{-3} s^{-1}.

To introduce our parcel model experiments, we first show an experiment in which we only vary α_i (Figure 1). Decreasing α_i increases R by increasing τ_{growth} without affecting τ_{lift}. Indeed, the increase in S_i and J_{hom} resulting from long τ_{growth} is why small α_i lead to large N_i (Gierens et al., 2003). The α_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 (α_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 α_i (α_i =0.001) result in persistent high S_i (S_i > 40%) after the freezing event ends. Surprisingly, S_i is depleted faster when α_i=0.01 than when α_i = 1. This counter-intuitive result is explained as follows: 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 vs. η_a graph (Figure 2, panel A), 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 (Eq. 3).

Using a sensitivity test approach, we explored the influence of plausible variations in thermodynamic (*w*, *T*, *P*) and microphysical (α_i, r_a) variables on η_a and *R* (Figure 2b-d). 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 $\alpha_i > 0.1$, η_a is small; however when $\alpha_i < 0.1$, 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 have only a limited influence on η_a. As r_a decreases, η_a slightly increases because for a fixed N_a, reducing r_a 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 α_i because D_v^* does not depend directly on α_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 α_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 α_i for ice crystals could be as low as 0.001 and as high as 1 (Haynes et al., 1992); recent laboratory measurements found α_i=0.006

(Magee et al., 2006). Given the sensitivity of η_a to α_i when α_i <0.1, discrepancies between α_i measurements must be resolved.

Fortunately, existing observations can be used to constrain α_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 α_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 α_i required to match INCA-observed values with parcel modeling experiments. Uncertainty in the observed w also has an important effect on the constrained α_i. If a large range of w are considered (3 cm/s < w < 50 cm/s), a much larger range of α_i ($0.01 < \alpha_i < 1$) are consistent with the mean observed N_i.

In summary, atmospheric observations suggest that η_a rarely approaches 1 and α_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 α_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, η_a increases at very large w (approximately $w > 100$ cm s^{-1} when T=-50 C and α_i=0.1). Second, there is a significant increase in η_a at temperatures such as those where cirrus clouds form in the tropical upper troposphere. Finally, η_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 (η_a - Eq. 1). Our primary findings were:

- The dependence of η_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, η_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 η_a is small under most atmospheric conditions, but may increase at large w ($w > 100$ cm s^{-1}) or at low T ($T < -70$ °C).
- In order to reconcile laboratory and atmospheric observations, we suggest that α_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 τ_{growth} is not universal and that τ_{growth}, η_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 α_i and to evaluate the dependence of η_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 α_i as a function of S_i and ice crystal size. Constraining variations in α_i will be especially important when evaluating if α_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 cm s^{-1}, T_0 = -50 C, P_0 = 250 mb, N_a=100 cm^{-3}, and r_a=0.2 μm.

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

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 α_i=0.1, T_0 = -50 °C, P_0 = 250 mb, and r_a=0.2 μm. Colors indicate the aerosol sensitivity parameter η_a (Eq. 1) and range from the thermodynamically limited nucleation regime (η_a=0) to the aerosol-limited nucleation regime (η_a=1). The green line shows where η_a=0.5 line for T=-80 °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)).

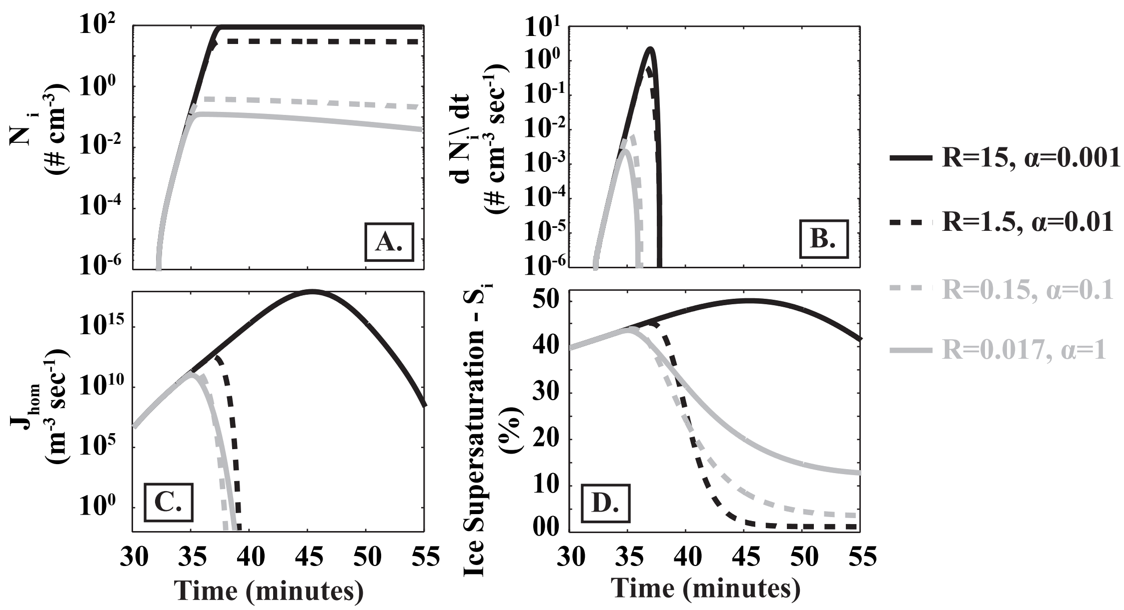
Tables:

Table 1. Parcel model description and configuration used for this study. In all model runs, α_i does not depend on ice crystal size or on ice supersaturation.

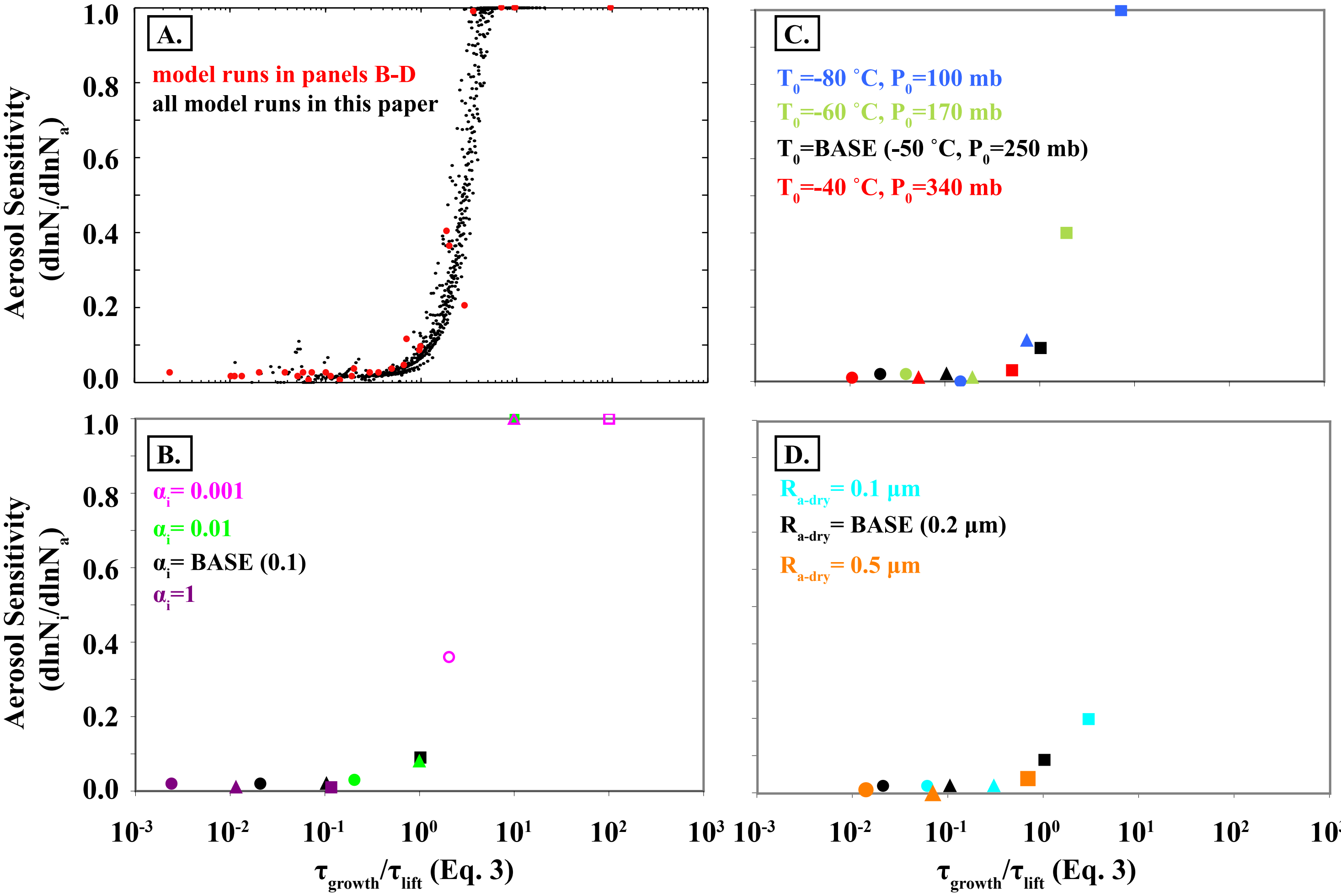
Parcel model description	Model configuration
• description and validation in Kay et al. (2006) • binned ice microphysics (300 bins) • aerosol activation using Köhler curve, monodisperse sulfuric acid aerosol with an individual dry weight of 10^{-16} kg and variable number concentration • saturation vapor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

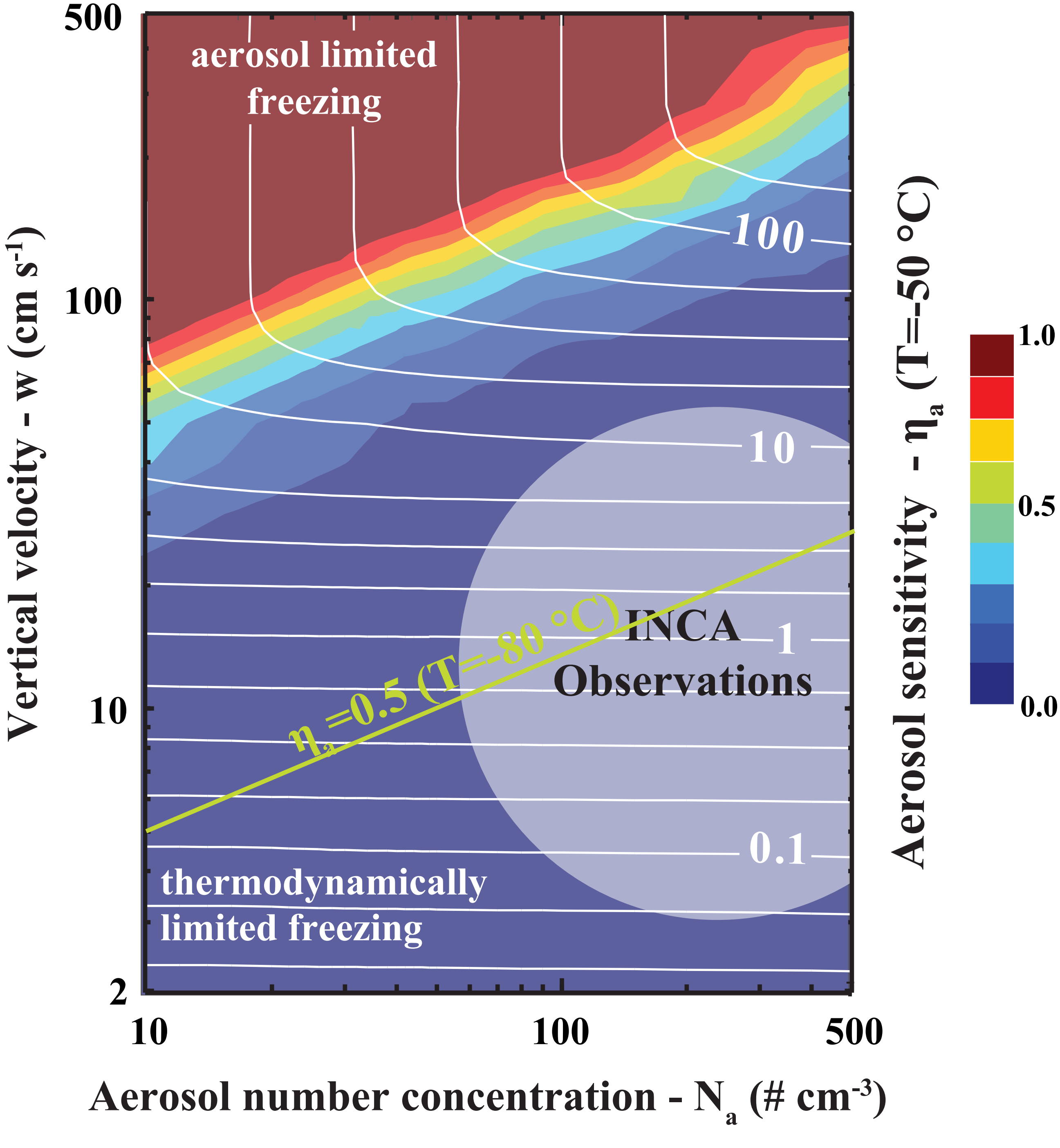
Table 2. INCA observations (Kärcher and Ström, 2003) and parcel model predicted N_i. Parcel model N_i are found from constant lifting experiments (Table 1) with INCA-observed w, T, and N_a, and P_0 = 250 mb. α_i values of 0.057 and 0.13 were required to match the mean Scotland and Chile observations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

	INCA Observations (Kärcher and Ström, 2003)				**Parcel Model N_i (# cm^{-3})**			
	w (cm s^{-1})	T (ºC)	N_a (# cm^{-3})	N_i (# cm^{-3})	α_i = 1	α_i = 0.1	α_i= 0.01	α_i= 0.006
Scotland	26.2	-48.3	300	5.3	0.5	2.3	102.8	212.6
Chile	23	-46.8	110	1.1	0.4	1.4	61.3	102.7



A.
B.
C.
D.
N i (# cm-3)
d Ni\ dt (# cm-3 sec-1)
Jhom (m-3 sec-1)
Ice Supersaturation - Si (%)
Time (minutes)
Time (minutes)
R=15, α=0.001
R=1.5, α=0.01
R=0.15, α=0.1
R=0.017, α=1


A.
model runs in panels B-D
all model runs in this paper
Aerosol Sensitivity
(dlnN$_i$/dlnN$_a$)
1.0
0.8
0.6
0.4
0.2
0.0
C.
T$_0$=-80 ˚C, P$_0$=100 mb
T$_0$=-60 ˚C, P$_0$=170 mb
T$_0$=BASE (-50 ˚C, P$_0$=250 mb)
T$_0$=-40 ˚C, P$_0$=340 mb
B.
α$_i$= 0.001
α$_i$= 0.01
α$_i$= BASE (0.1)
α$_i$=1
D.
R$_{a\text{-}dry}$= 0.1 µm
R$_{a\text{-}dry}$= BASE (0.2 µm)
R$_{a\text{-}dry}$= 0.5 µm
10^{-3}
10^{-2}
10^{-1}
10^{0}
10^{1}
10^{2}
10^{3}
τ$_{growth}$/τ$_{lift}$ (Eq. 3)



aerosol limited
freezing
100
10
1
0.1
INCA
Observations
η_a=0.5 (T=-80 °C)
thermodynamically
limited freezing
500
100
10
2
10
100
500
Vertical velocity - w (cm s^{-1})
Aerosol number concentration - N_a (# cm^{-3})
Aerosol sensitivity - η_a (T=-50 °C)
1.0
0.5
0.0