Dynamical states of low temperature cirrus

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Abstract. Low ice crystal concentration and sustained in-cloud supersaturation, commonly found in cloud observations at low temperature, challenge our understanding of cirrus formation. Heterogeneous freezing from effloresced ammonium sulfate, glassy aerosol, dust and black carbon are proposed to cause these phenomena; this requires low updrafts for cirrus characteristics to agree with observations and is at odds with the gravity wave spectrum in the upper troposphere. Background temperature fluctuations however can establish a “dynamical equilibrium” between ice production and sedimentation loss (as opposed to ice crystal formation during the first stages of cloud evolution and subsequent slow cloud decay) that explains low temperature cirrus properties. This newly-discovered state is favored at low temperatures and does not require heterogeneous nucleation to occur (the presence of ice nuclei can however facilitate its onset). Our understanding of cirrus clouds and their role in anthropogenic climate change is reshaped, as the type of dynamical forcing will set these clouds in one of two “preferred” microphysical regimes with very different susceptibility to aerosol.

1 Introduction

Cirrus clouds are composed of ice crystals that form at high altitudes and temperatures typically below 235 K (Pruppacher and Klett, 1997). They play a key role in climate by modulating the planetary radiative balance (Liou, 1986) and heat transport in the upper troposphere (Ramanathan and Collins, 1991). They strongly impact water vapor transport across the tropopause level (Jensen and Pfister, 2004) and play an important role in lower stratospheric chemistry (Peter, 1997). Cirrus may be affected by aircraft emissions (Seinfeld, 1998) and long range transport of pollutants and play an important (but highly uncertain) role in anthropogenic climate change.

A key microphysical parameter required for understanding the climate impact of cirrus is their concentration of ice crystals, \( N_c \). At temperatures between 200 and 235 K cirrus ice crystals form primarily by homogeneous freezing of supercooled deliquesced aerosol (DeMott et al., 2003; Heymsfield and Sabin, 1989), which occurs if the saturation ratio with respect to ice, \( S \), (i.e., the ratio of water vapor partial pressure to its equilibrium value over ice) reaches a characteristic threshold value, \( S_{\text{hom}} \) (Koop et al., 2000). Heterogeneous freezing of water upon existing aerosol particles (termed “ice nuclei”, IN) can also occur (at \( S \) lower than \( S_{\text{hom}} \)) and contribute to ice crystal concentrations (DeMott et al., 2003; Froyd et al., 2009), especially in polluted and dust-rich regions (Barahona et al., 2010a; Haag et al., 2003). The level of water vapor supersaturation (i.e., \( S - 1 \)) is the thermodynamic driver for ice formation, and is generated by expansion of air parcels forced by large scale dynamics, gravity waves, and small scale turbulence (Kim et al., 2003).

At temperatures below 200 K (typically near the tropical tropopause layer, TTL) the simple conceptual model for cirrus formation presented above is at odds with observations (Jensen et al., 2010; Krämer et al., 2009; Peter et al., 2006). Temperature fluctuations from mesoscale gravity waves are common at high altitudes and can produce localized vertical motion with updraft velocity as large as 1 m s\(^{-1}\) (Bacmeister et al., 1999; Herzog and Vial, 2001; Jensen and Pfister, 2004; Sato, 1990). Such motion can increase the rate of expansion cooling at the point of freezing so that a large number of ice crystals is nucleated before the local supersaturation is depleted by ice crystal growth. Homogeneous freezing driven by gravity wave motion would produce high ice crystal number concentration, \( N_c \), between 1 and 10 cm\(^{-3}\).
near the TTL (Barahona et al., 2010a). Such high concentrations however are not observed; \(N_c\) remains low, sometimes even lower than concentrations observed in weak updraught zones at cold temperatures (Krämer et al., 2009; Lawson et al., 2008). This “low \(N_c\)” paradox is accompanied by other phenomena, such as low supersaturation relaxation times (Krämer et al., 2009), which in turn leads to sustained supersaturation levels inside clouds (i.e., “the supersaturation puzzle”) (Gao et al., 2004; Krämer et al., 2009; Peter et al., 2006), high clear-sky supersaturation (Jensen et al., 2005), and broad ice crystal size distributions (i.e., large crystal sizes, Jensen et al., 2008). These phenomena occur despite the strong dynamical forcing and the ample amounts of deliquesced aerosol available for homogeneous freezing. Suppressed freezing by organics (Murray, 2008), slow water vapor transfer to the ice phase (Gao et al., 2004; Magee et al., 2006), and freezing to cubic instead of hexagonal ice (Murray et al., 2005), have been proposed to explain these features. These mechanisms are however not capable of explaining the low \(N_c\) in low temperature cirrus clouds (Peter et al., 2006). Lacking the predictive understanding of such phenomena hinders the ability of climate models to capture the climate effects of cirrus clouds and their response to anthropogenic perturbations.

Heterogeneous freezing of IN as the main path of cirrus formation has been proposed to explain the features of cirrus clouds at low temperature (Abbatt et al., 2006; Jensen et al., 2010; Murray et al., 2010). Owing to their ability to freeze ice at much lower supersaturation than homogeneous freezing requires, IN can deplete water vapor, reduce supersaturation and inhibit homogeneous freezing; this can drastically reduce the number of ice crystals that forms in the cirrus (Barahona and Nenes, 2009b; DeMott et al., 1994; Kärcher et al., 2006). Much of the anthropogenic impact on cirrus clouds and climate is thought to occur through this IN-\(N_c\) feedback mechanism (Lohmann and Feichter, 2005). Dust (Khvorostyanov et al., 2006), effloresced ammonium sulfate (Abbatt et al., 2006; Jensen et al., 2010; Wise et al., 2010), and glassy aerosol (Murray et al., 2010) have been identified as potential natural IN at the TTL.

The evolution of cirrus clouds at low \(T\) has been addressed in several studies. Using a one dimensional (1-D) cloud model, Jensen and Pfister (2004) found that the superposition of temperature fluctuations along Lagrangian trajectories near the TTL resulted in rapid cooling cycles that increased the rate of crystal production by homogeneous nucleation leading to \(N_c\) above \(1\, \text{cm}^{-3}\). Khvorostyanov et al. (2006) used a 1-D cirrus model to investigate the evolution of a cirrus layer initialized at \(200\, \text{K}\) and found that regardless of the predominant nucleation mechanism, temperature fluctuations increased the maximum \(N_c\) in the cloud (up to \(0.6\, \text{cm}^{-3}\)). \(N_c\) however rapidly decreased after the initial freezing pulse due to the vertical advection of ice crystals precluding new nucleation events and diluting \(N_c\) down to about \(0.05\, \text{cm}^{-3}\). Gensch et al. (2008) used box model simulations along Lagrangian trajectories to test homogeneous and heterogeneous nucleation scenarios in the formation of cirrus at low \(T\). It was found that only heterogeneous nucleation scenarios (with prescribed IN number concentration around \(0.1\, \text{cm}^{-3}\)) would result in \(N_c\) close to observations. This conclusion was echoed by Froyd et al. (2009) and Jensen et al. (2010) using 1-D models along prescribed \(T\) trajectories.

The formation of cirrus clouds however exhibits complex non-linear behavior that may not be captured by box and 1-D models. Ice falling through active freezing zones (typically located at the top of the cirrus layer, Spichtinger and Gierens, 2009b) in clouds consume water vapor and can inhibit homogeneous freezing much like IN do (Kay et al., 2007; Spichtinger and Gierens, 2009b). Their effectiveness depends on their residence time in freezing zones and hence depends on their size. Large ice crystals tend to quickly fall out of freezing zones and have limited effect on new ice formation events; small crystals (typically those with terminal velocity, \(u_{\text{term}}\), less or equal to the mean updraft \(\bar{u}\) of the cirrus layer) fall slowly and can remain long enough in the upper part of the cloud to affect new freezing events. This suggests that at low temperatures, preexisting (and typically small, Krämer et al., 2009) ice crystals may locally dehydrate the freezing zone sufficiently to inhibit the formation of new ice. The rate of crystal production is not uniform through the freezing zone, as the “local” saturation ratio, \(S\), and updraft velocity, \(u\) (defined at the scale of individual cloud “parcels” \(\sim10^0–10^2\, \text{m}\), Pruppacher and Klett, 1997) may be affected by fluctuations in wind speed and temperature induced by gravity waves (Jensen et al., 2010; Kärcher and Haag, 2004; Kim et al., 2003). These internal \(S\) variations are usually neglected in cirrus cloud studies on the basis that the long-term evolution of the cloud is determined by the mean values of \(S\) and \(u\).

In this work we analyze the range of conditions for which heterogeneous freezing may explain the features of cirrus clouds at low temperature, and propose an alternative view (based on a statistical description of cirrus formation and evolution) in which the interplay of temperature fluctuations, and ice crystal production and sedimentation leads to previously unidentified natural cirrus states of low ice crystal concentration and sustained high supersaturation.

2 Heterogeneous freezing at low temperature

The impact of IN on \(N_c\) depends on their concentration, \(N_{\text{IN}}\). If too low (\(N_{\text{IN}} < 1 \times 10^{-4}\, \text{cm}^{-3}\)), a negligible impact is seen on \(N_c\), as too few (heterogeneously-frozen) ice crystals form to quench supersaturation below the homogeneous/heterogeneous freezing threshold (Barahona and Nenes, 2009b). Low \(N_c\) favors large crystal size and therefore heterogeneously frozen ice crystals may sediment out of the cloud layer before significantly modifying \(S\) (Spichtinger
and Gierens, 2009a). When \( N_{\text{IN}} \) approaches a characteristic “limiting” concentration (which depends on updraft velocity, the IN freezing threshold and size), \( N_{\text{lim}} \), supersaturation is quenched, homogeneous freezing is suppressed, and \( N_c \) decreases steeply (Barahona and Nenes, 2009b). For \( N_{\text{IN}} \geq N_{\text{lim}} \), homogeneous nucleation is inhibited and \( N_c = N_{\text{IN}} \). Thus, \( N_{\text{lim}} \) is the minimum \( N_c \) that can form in an active nucleation zone in a freshly-formed cirrus cloud (Barahona and Nenes, 2009b) and presents the maximum reduction in \( N_c \) possible from IN.

Figure 1 shows \( N_c \) accounting for the competition between homogeneous and heterogeneous freezing during cloud formation (Barahona and Nenes, 2009b) (details of the calculation are described Sect. 3.3). As \( N_c \) is strongly influenced by the vertical velocity at the point of freezing and much less sensitive to small \( T \) variations, the curves in Fig. 1 represent the peak \( N_c \) that would be obtained in box models after a single freezing event (Barahona and Nenes, 2008; Hoyle et al., 2005; Kärcher and Lohmann, 2002; Spichtinger and Gierens, 2009a). If \( N_{\text{IN}} \) is always very close to \( N_{\text{lim}} \), competition between homogeneous and heterogeneous freezing could yield \( N_c \) close to observations. This requires \( N_{\text{IN}} \sim 0.1 \text{ cm}^{-3} \), which is 20-fold higher than measured dust concentrations (\( \sim 0.005 \text{ cm}^{-3} \)) at the tropopause level (Froyd et al., 2009). Ammonium sulfate aerosol is present at much higher concentrations than dust, and can serve as IN (Abbatt et al., 2006; Fountoukis and Nenes, 2007; Shilling et al., 2009). If \( N_{\text{IN}} \) is always very close to \( N_{\text{lim}} \), competition between homogeneous and heterogeneous freezing could yield \( N_c \) close to observations. This requires \( N_{\text{IN}} \sim 0.1 \text{ cm}^{-3} \), which is 20-fold higher than measured dust concentrations (\( \sim 0.005 \text{ cm}^{-3} \)) at the tropopause level (Froyd et al., 2009). Ammonium sulfate aerosol is present at much higher concentrations than dust, and can serve as IN (Abbatt et al., 2006; Fountoukis and Nenes, 2007; Shilling et al., 2006).

To inhibit homogeneous freezing and reproduce observations of \( N_c \), the concentration of ammonium sulfate IN needs to be within 10% of \( N_{\text{lim}} \); if concentrations fall below \( 0.9N_{\text{lim}} \), homogeneous freezing is triggered and predicted \( N_c \) is significantly above observations (Fig. 1). If higher concentration than \( N_{\text{lim}} \) is present, homogeneous freezing is completely suppressed, but too many crystals still form (Barahona and Nenes, 2008). In fact, if all ammonium sulfate is available as IN, \( N_c \) from heterogeneous freezing and pure homogeneous freezing are always comparable (Fig. 2), because crystals formed from ammonium sulfate IN are very small (with size close to the dry aerosol; \( 0.02-0.05 \mu \text{m} \), Froyd et al., 2009) and grow too slowly to quench supersaturation before a large fraction of the aerosol freezes heterogeneously. \( N_c \) is within observed values only if the average size of crystals at the point of freezing is 2 \( \mu \text{m} \) or larger (Fig. 2), which is too large for upper tropospheric aerosol (Froyd et al., 2009). Experimental studies suggest that heterogeneous freezing of ammonium sulfate IN at \( T \sim 240 \text{ K} \) can be very selective (about 1 in \( 10^3 \) particles nucleate ice (Shilling et al., 2006). If the same selectivity maintains at lower \( T \), too few IN would be available to prevent homogeneous freezing (therefore resulting in high \( N_c \)). Higher nucleation selectivity (e.g., about 1 in \( 10^2 \) particles actively nucleating ice) would result in complete inhibition of homogeneous freezing and still maintain \( N_c \) close to observations (not shown). A pure heterogeneous scenario of ice nucleation on ammonium sulfate however implies a maximum supersaturation below 20% (Fig. 3b), i.e., \( S \) greater than 1.2 would be rarely observed as newly formed crystals would rapidly remove supersaturation. This is at odds with observations of relative humidity that suggest that clear-sky supersaturation in the vicinity of cirrus up to 70% (and occasionally above) is very common at low temperature (Krämer et al., 2009). Hence the low \( N_c \) and high \( S \) observed at high-level cirrus can be reconciled with box-model results only if the concentration of ammonium sulfate IN is remarkable constant (0.1 \( \pm 0.01 \text{ cm}^{-3} \)), the concentration of dust is exceptionally large, or, the fluctuations in vertical velocity from gravity wave motion are neglected.

The freezing fraction of organic glassy aerosol can be much lower than that of ammonium sulfate and maintain \( N_{\text{IN}} \) close to \( N_{\text{lim}} \) (hence yield low \( N_c \), Fig. 3a) if the vertical velocity is below 15 cm s\(^{-1}\) (Murray et al., 2010). At larger updrafts however, homogeneous nucleation is triggered, producing high \( N_c \) (Fig. 3a). When integrated over a normal distribution of updrafts with standard deviation \( \sigma_u \) (Fig. 3c), \( N_c \) remains within observed values for \( \sigma_u \) up to 40 cm s\(^{-1}\) at \( T = 195 \text{ K} \). The onset of homogeneous nucleation occurs at even lower \( u \) for colder temperatures and \( N_c \) deviates from observations for \( \sigma_u \) as low as 10 cm s\(^{-1}\) at \( T = 185 \text{ K} \). Predominance of heterogeneous nucleation from glassy IN would also imply maximum supersaturation around 30% if the new formed ice crystals efficiently remove
Fig. 2. Simulations of ice crystal concentration by pure heterogeneous freezing. $N_c$ is presented as a function of the initial size of the ice nuclei. Conditions (Lawson et al., 2008) used were $T = 185\, \text{K}$, $p = 100\, \text{hPa}$, and vapor-to-ice deposition coefficient, $\alpha_d$, of 0.07 (dashed line), and 1.0 (solid line). The IN population was assumed to be monodisperse with total number concentration of $100\, \text{cm}^{-3}$ (Lawson et al., 2008). Freezing of solid ammonium sulfate was assumed to occur in a “burst” around the heterogeneous freezing threshold described by sigmoidal freezing spectrum with inflection point $S_{\text{het}} = 15\%$ (Abbatt et al., 2006), where 99% of the aerosol freeze within a 2% supersaturation interval about $S_{\text{het}}$ (inset plot).

supersaturation (Barahona et al., 2010a; Murray et al., 2010). However as the freezing fraction of glassy IN is small, $S$ can increase even after heterogeneous nucleation has occurred. Still, $S$ would have to remain below $S_{\text{hom}}$ for $N_c$ to remain low unless homogeneous freezing is suppressed. The later scenario is however not supported by observations. Both, in-cloud and clear-sky RH are generally limited by the homogeneous freezing threshold indicating efficient supersaturation removal by homogeneously-frozen ice crystals (Krämer et al., 2009; Selkirk et al., 2010). Although uncertainty in RH can be typically up to 20% (Krämer et al., 2009), it is still smaller than the difference between the homogeneous and heterogeneous freezing thresholds, typically between 30% and 40%, giving this support to the idea that homogeneous freezing occurs at low temperatures. All together, this implies that in the presence of (ubiquitous) $T$ fluctuations, the presence of glassy IN may contribute, but not fully account for the observed characteristics of low $T$ cirrus.

3 Parcel statistical ensemble model

The main processes affecting the evolution of $N_c$ and mean saturation ratio, $S_o$, within a cirrus layer are the freezing of new ice, the sedimentation of existing ice crystals, the lifting of air masses (which generates supersaturation), and the relaxation (i.e., mass transfer) of water vapor to/from the ice phase. The magnitude of each process can be expressed in terms of a characteristic timescale, i.e., $\tau_{\text{fr}}$, $\tau_{\text{sed}}$, $\tau_{\text{lift}}$, and $\tau_{\text{rel}}$ for freezing, sedimentation, lifting, and relaxation, respectively. Fluctuations in $S$ and $u$ can have a strong impact on all cloud processes; we therefore represent them in terms of a...
probability distribution centered about the cirrus-average saturation ratio, \( S_o \), and vertical velocity, \( \bar{u} \). The width of these probability distributions is largely determined by the mean amplitude of temperature fluctuations, \( \delta T \) (Bacmeister et al., 1999; Hoyle et al., 2005; Kärcher and Burkhardt, 2008). The rate of ice production is given by the frequency with which \( S \) exceeds the homogeneous freezing threshold (Kärcher and Burkhardt, 2008) times the length and intensity of each freezing event (hence \( \tau_{fr} \)) (Barahona and Nenes, 2008; Pruppacher and Klett, 1997). The same fluctuations also affect the local mass transfer rate between the ice and vapor phases, so that when averaged over the cloud, water deposition/sublimation occurs at an “effective” saturation ratio, \( S_{eff} \), that may differ from \( S_o \).

Supersaturation and crystal number in the cirrus cloud are determined using a “Lagrangian statistical ensemble” approach. This involves determining the time-dependant state of \( i \) homogeneous Lagrangian “parcels” that move with a (time-dependant) vertical velocity, \( u_i \); ensemble averaging of the parcel solutions (outlined below) weighted by the appropriate probability distribution give approximate equations that describe the time-dependant properties for the whole cirrus. From these considerations, simple equations can be derived that represent the evolution of \( N_c \) and \( S_o \) in the cirrus (Sects. 3.1, 3.2).

### 3.1 Evolution of saturation ratio

In the absence of ice nucleation, the rate of change of saturation ratio, \( S \), within the \( i^{th} \) Lagrangian parcel is given by (Barahona and Nenes, 2009a; Seinfeld and Pandis, 1998)

\[
\frac{dS_i}{dt} = \alpha u_i S_i - \gamma \int_{D_{min}}^{\infty} \frac{dD}{D_{c,i}} \frac{D_{c,i} n_{c,i}(D_c) dD_c}{D_{c,i}}
\]

where \( \alpha = \frac{g \Delta H_{M_a}}{c_p R \bar{u}} - g M_a \rho_a \Delta H_e \) and \( \gamma = \frac{\Delta H_{M_a}}{\rho_i \bar{M}_w \rho_a \Delta H_e} \), \( \Delta H_e \) is the latent heat of sublimation of water, \( g \) is the acceleration of gravity, \( c_p \) is the heat capacity of air, \( p_i^{th} \) is the ice saturation vapor pressure at \( T \) (Murphy and Koop, 2005), \( p \) is the ambient pressure, \( M_w \) and \( M_a \) are the molar masses of water and air, respectively, \( R \) is the universal gas constant, \( \rho_i \) and \( \rho_a \) are the ice and air densities, respectively, and \( D_{c,i} \) is the volume equivalent diameter of an ice particle (assuming spherical shape). \( n_{c,i}(D_c) \) is the ice crystal size distribution in the \( i^{th} \) parcel, and

\[
\frac{dD_{c,i}}{dt} = \frac{G(S_i - 1)}{D_{c,i}}
\]

where \( G \approx \left[ \frac{\rho_i R T}{4 \rho_d D_{c,i} M_w} + \frac{\Delta H_{M_a}}{4 k_a T} \left( \frac{\Delta H_{M_a}}{RT} - 1 \right) \right]^{-1} \), \( k_a \) is the thermal conductivity of air, \( D_c = D_c(T, p, \alpha_d) \) is the water vapor diffusion coefficient from the gas to ice phase corrected for non-continuum effects, and \( \alpha_d \) is the water vapor deposition coefficient. Substituting Eq. (2) into Eq. (1) provides after evaluation of the integral,

\[
\frac{dS_i}{dt} = \alpha u_i S_i - \frac{(S_i - 1)}{\tau_{rel,i}}
\]

where \( \tau_{rel,i} = (\beta N_{c,i} \bar{D}_{c,i})^{-1} \) is the relaxation time scale in the \( i^{th} \) parcel, \( \beta = \gamma G \), and, \( N_{c,i}, \bar{D}_{c,i} \) are the concentration and mean size of ice crystals in the \( i^{th} \) parcel, respectively.

Equation (3) provides the supersaturation “state” for every Lagrangian parcel considered in the ensemble. Knowledge of the distribution of \( u_i \) (from the spectrum of gravity waves in the cirrus) can then be used to “drive” the parcels in the ensemble to find the resulting distribution of \( S_i \). Averaging is carried out first over all parcels reaching a given cloud level with vertical velocity \( u_j \) (referred to as the “\( j^{th} \) cloud velocity state”), and then averaging over all cloud states. Based on this, the average saturation ratio, \( S_o \), of the cloud over a time interval \( \Delta t \) is

\[
S_o(t) = \int_{-\infty}^{+\infty} \int_{-\infty}^{1} \int_{-\infty}^{1} S_i(\mu, \bar{x}, \tau) P(\mu, \bar{x}, \tau) d\tau d\tilde{x} d\mu
\]

where \( \mu = \frac{u}{\bar{u}}, u \) and \( \bar{u} \) are the instantaneous and average vertical velocity, respectively, \( \tilde{x} \) denotes the position in the cloud, \( \tau = \frac{t}{\bar{t}} \), where \( \bar{t} \) is the averaging time, and \( X(t) \) is the domain of \( \bar{x} \). \( P(\mu, \bar{x}, \tau) \) is the normalized probability at time \( \bar{t} \) of finding a parcel between position \( \bar{x} \) and \( \bar{x} + d\bar{x} \) (where \( d\bar{x} = \frac{dxdydz}{V_{load}} \)), with vertical velocity within \( u \) and \( u + du \).

Equation (1) can be simplified, by considering that fluctuations generated by gravity waves are random in nature (i.e., follow a Gaussian distribution, Fig. 4d). Thus, under the assumption that \( P(\mu, \bar{x}, \tau) \) does not vary with space and time over \( \Delta t \), \( P(\mu, \bar{x}, \tau) \propto P(\mu) \) and Eq. (4) simplifies to

\[
S_o(t) = \int_{-\infty}^{+\infty} \int_{-\infty}^{1} \int_{-\infty}^{1} S_i(\mu, \bar{x}, \tau) P(\mu) d\tau d\tilde{x} d\mu
\]

Equation (5) assumes that \( S_o \) is affected by processes that act throughout the volume of the cirrus cloud. Other processes, like entrainment and radiative cooling, are neglected. Although this will not affect the conclusions of our study, they could be included in future studies e.g. indirectly through appropriate modification of the vertical velocity distribution (Barahona and Nenes, 2007).

Defining \( \tilde{S}_j = \int_{X(t) \bar{x}(t)}^{1} S_j(\mu, \bar{x}, \tau) d\tau d\tilde{x} \) as the average supersaturation of parcels in the “\( j^{th} \)” velocity state over the time interval \( \Delta t \), Eq. (5) can be rewritten as

\[
S_o(t) = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} \tilde{S}_j(\mu_j) P(\mu_j) d\mu_j - \infty
\]

the time derivative of which gives,
Substitution of Eq. (3) into above provides

\[
\frac{d\tilde{S}_j}{dt} = \int_{\chi(t)}^{1} \left\{ \alpha u_j S_i \left( \frac{S_i - 1}{\tau_{rel,i}} \right) d\tau d\tilde{x} = \alpha u_j \int_{\chi(t)}^{1} S_i d\tau d\tilde{x} \right. \\
- \int_{\chi(t)}^{1} \left( \frac{S_i - 1}{\tau_{rel,i}} \right) d\tau d\tilde{x}
\]

(10)

which can be rewritten as,

\[
\frac{d\tilde{S}_j}{dt} = \alpha u_j \tilde{S}_j - \int_{\chi(t)}^{1} \left( \frac{S_i - 1}{\tau_{rel,i}} \right) d\tau d\tilde{x}
\]

(11)

Introducing \( \tilde{S}_{eff,j} \) so that,

\[
\int_{\chi(t)}^{1} \int_{\tau_{rel,i}}^{\infty} \left( \frac{S_i - 1}{\tau_{rel,i}} \right) d\tau d\tilde{x} = \left( \tilde{S}_{eff,j} - 1 \right) \int_{\chi(t)}^{1} \int_{\tau_{rel,i}}^{\infty} d\tau d\tilde{x}
\]

(12)

From Eq. (3),

\[
\frac{1}{\tau_{rel,j}} = \int_{\tau_{rel,i}}^{\infty} \frac{1}{\tau_{rel,i}} d\tau d\tilde{x} = [\beta N_c D_c]^j_{\mu=\mu_j}
\]

(13)

Combining Eqs. (12) and (13), Eq. (11) can be written as

\[
\frac{d\tilde{S}_j}{dt} = \alpha u_j \tilde{S}_j - \frac{\tilde{S}_{eff,j} - 1}{\tau_{rel,j}}
\]

(14)

where \( \tau_{rel,j} \) is the relaxation time scale associated with the \( j \)th state. \( \tilde{S}_{eff,j} \) is an “effective” saturation ratio for deposition/sublimation processes, defined below. Introducing Eq. (14) into Eq. (8),

\[
\frac{dS_0}{dt} = \int_{-\infty}^{+\infty} \left( \alpha u_j \tilde{S}_j - \frac{\tilde{S}_{eff,j} - 1}{\tau_{rel,j}} \right) P(\mu_j)d\mu_j
\]

(15)

or,

\[
\frac{dS_0}{dt} = \int_{-\infty}^{+\infty} \alpha u_j \tilde{S}_j P(\mu_j)d\mu_j - \int_{-\infty}^{+\infty} \frac{\tilde{S}_{eff,j} - 1}{\tau_{rel,j}} P(\mu_j)d\mu_j
\]

(16)

The first term in the right hand side of Eq. (16) must be equal to \( \alpha \tilde{S}_0 \), as in the absence of deposition/sublimation, \( S_0 \) in the layer increases exponentially with time (Pruppacher and Klett, 1997). With this, Eq. (16) becomes,

\[
\frac{dS_0}{dt} = \frac{dS_0}{\tau_{lift}} - \int_{-\infty}^{+\infty} \left( \frac{\tilde{S}_{eff,j} - 1}{\tau_{rel,j}} \right) P(\mu_j)d\mu_j
\]

(17)

where \( \tau_{lift} = (\alpha \tilde{u})^{-1} \). Equation (17) must be solved for each time step specifying \( P(\mu_j) \) and then evaluating \( \tilde{S}_{eff,j} \) and the integral on the right hand side. Since \( P(\mu_j) \) is determined
by the random overlapping of gravity waves of different frequency and amplitude (e.g., \(u_j\) is given by a Fourier series in time, Sect. 3.3), then for a time step of integration much smaller than \(\tau_{\text{lift}}\) (\(\sim 10^2\) s) Eq. (17) can be approximated by

\[
\frac{dS_0}{dt} = \alpha \bar{u} S_0 - \frac{\tilde{S}_{\text{eff},i} - 1}{\tau_{\text{rel},i}} \bigg|_{\mu = \mu_j} \tag{18}
\]

where \(\tilde{S}_{\text{eff},i}\) and \(\tau_{\text{rel},i}\) are calculated at the instantaneous vertical velocity.

Equation (18) gives the evolution of the \(S_0\) in the cirrus cloud; its solution however requires the knowledge of \(\tilde{S}_{\text{eff},i}\). This is accomplished by considering the properties of the different parcels reaching the cloud layer at \(t\). For example, if \(S_i\) in the \(i\)th parcel is a pseudo-steady state, \(\frac{dS_i}{dt} \sim 0\) (Korolev and Mazin, 2003) and from Eq. (3),

\[
S_{i,ss} = \frac{\tau_{\text{lift},i}}{\tau_{\text{lift},i} - \tau_{\text{rel},i}} \tag{19}
\]

where \(\tau_{\text{lift},i} = (\alpha \bar{u} \tau_t)^{-1}\) and \(S_{i,ss}\) is the steady state saturation ratio in the \(i\)th parcel. If \(\tau_{\text{lift},i} < 0\) then \(S_{i,ss} < 1\), and vice-versa. Thus, if \(u_j < 0\), the layer would likely be subsaturated over \(\Delta t\) (e.g., Eq. 6), and vice-versa when \(u_j > 0\). Thus, depending on the sign of \(u_j\) there is net deposition/sublimation of water vapor in the cloud layer. Not all parcels however reach steady state; therefore the degree of saturation/subsaturation associated with the \(j\)th state depends on the probability distribution of saturation within the cloudy layer, \(P_S(S, S_0, \Delta T)\), which is a function of \(S_0\) and the average amplitude of temperature fluctuations, \(\Delta T\). Thus, \(S_{\text{eff}}\) for \(u_j < 0\) is found by averaging over all states that would lead to subsaturation, i.e., \(P_S(S, \Delta T, S_0)\) for which \(S < 1\). Similarly, when \(u_j > 0\), the supersaturated \((S > 1)\) region of \(P_S(S, \Delta T, S_0)\) is used,

\[
\tilde{S}_{\text{eff},j} = \frac{\int S dP_S(S, \Delta T, S_0) dS}{\int dP_S(S, \Delta T, S_0) dS} \tag{20}
\]

where \(a = \left\{ \begin{array}{ll} 1 & \text{ if } u_j > 0 \\ 0 & \text{ if } u_j \leq 0 \end{array} \right\} \) and \(b = \left\{ \begin{array}{ll} S_{\text{hom}} & \text{ if } u_j > 0 \\ 1 & \text{ if } u_j \leq 0 \end{array} \right\} \)

The homogeneous freezing threshold, \(S_{\text{hom}}\), is set as the upper limit of \(P_S(\Delta T, S_0)\) as ice crystal production quickly removes supersaturation above \(S_{\text{hom}}\) (Kärcher and Burkhardt, 2008; Kärcher and Haag, 2004).

### 3.2 Evolution of ice crystal number concentration

The evolution of the number concentration within a cloudy layer is given by

\[
\frac{dN_c}{dt} = \frac{dN_c}{dt} \bigg|_{\text{fr}} + \frac{dN_c}{dt} \bigg|_{\text{sed}} \tag{21}
\]

where \(\frac{dN_c}{dt} \bigg|_{\text{fr}}\) is the rate production of ice crystals within the layer, and \(\frac{dN_c}{dt} \bigg|_{\text{sed}}\) is their sedimentation rate. If homogeneous and heterogeneous nucleation are active, \(\frac{dN_c}{dt} \bigg|_{\text{fr}}\) is given by,

\[
\frac{dN_c}{dt} \bigg|_{\text{fr}} = \frac{dN_c}{dt} \bigg|_{\text{fr,hom}} + \frac{dN_c}{dt} \bigg|_{\text{fr,het}} \tag{22}
\]

where \(\frac{dN_c}{dt} \bigg|_{\text{fr,hom}}\) and \(\frac{dN_c}{dt} \bigg|_{\text{fr,het}}\) are the ice crystal production rates from homogeneous and heterogeneous nucleation, respectively.

#### 3.2.1 Homogeneous nucleation

Ice crystal production by homogeneous nucleation is driven by local motions and occurs within single parcels when \(S_i > S_{\text{hom}}\). The maximum ice crystal concentration produced by homogeneous freezing within the \(i\)th parcel is given by (Barahona and Nenes, 2008; Pruppacher and Klett, 1997)

\[
N_{c,i} = N_0 \left\{ 1 - \exp \left( - \int_{0}^{t_{\text{max},i}} \bar{v}_0 J(S_i) dt \right) \right\} \tag{23}
\]

which can be approximated by (Barahona and Nenes 2008)

\[
\frac{dN_{c,i}}{dt} \bigg|_{\text{fr,hom}} \approx N_0 \bar{v}_0 J_{\text{max,i}} \exp \left( - \frac{S_{\text{max,i}}}{\bar{v}_0} \right) \tag{24}
\]

where \(J_{\text{max,i}} = J(S_{\text{max},i})\). \(S_{\text{max},i}\) is the maximum saturation ratio reached in the \(i\)th parcel, calculated by setting \(\frac{dS_i}{dt} = 0\) in Eq. (1).

\[
S_{\text{max},i} = \frac{\gamma}{\bar{u} \alpha_i} \int_{D_{\text{c,i}}}^{\infty} \int_D D_{\text{c},i} \frac{dD_{\text{c},i}}{dt} n_{i,nuc}(D_c) dD_c \tag{25}
\]

where \(n_{i,nuc}(D_c)\) is the size distribution of the recently nucleated ice crystals, and \(D_0\) is the mean size of the deliquesced aerosol. Equation (25) assumes that only recently nucleated ice crystals are contained within the parcel. In reality, a fraction of preexisting crystals remain in nucleation zones (typically located near the cloud top, Spichtinger and Gierens, 2009b) inhibiting the homogeneous freezing of ice. Ice crystals experience gravitational settling, hence only those crystals with terminal velocity, \(\bar{u}_{\text{term}}\), below \(\bar{u}\) would be found at
the cloud top. Adding the consumption of water vapor from preexisting crystals to the right hand side of Eq. (25) gives

\[
S_{\text{max}, i} = \frac{\gamma}{\alpha u_i} \left[ \int_{D_{\text{min}}}^{D_{\text{term}}} D^n c_i n_{\text{i, nuc}}(D_c) dD_c + \int_{D_{\text{term}}}^{\infty} D^n c_i n_c(D_c) dD_c \right]
\]

(26)

where \( n_c(D_c) \) is the cloud ice crystal size distribution, \( D_{\text{term}} \) is the size of the crystal for which \( u_{\text{term}} = \bar{u} \), and \( D_{\text{min}} \) is the minimum size of the preexisting crystals in the cloud. Equation (26) can be combined with Eq. (2) to obtain

\[
S_{\text{max}, i} = \frac{\gamma}{\alpha u_i} \left[ \int_{D_{\text{min}}}^{D_{\text{term}}} D^n c_i n_{\text{i, nuc}}(D_c, S_{\text{max}, i}) dD_c + GN_c \bar{D}_{\text{c}} f_p(S_{\text{max}, i} - 1) \right]
\]

(27)

where, \( f_p = \frac{1}{N_c D_{\text{term}}} \int_{D_{\text{min}}}^{D_{\text{term}}} D_c n_c(D_c) dD_c \), is the fraction of preexisting ice crystals remaining in nucleation zones. As ice crystals remaining in the cloud layer were produced by preexisting freezing events, Eq. (27) provides a link between the history of different parcels and the nucleation of new crystals. The analytical solution of Eq. (27) is presented elsewhere (Barahona and Nenes, 2009b; Barahona et al., 2010b).

The rate of ice crystal production by homogeneous nucleation in the \( j \)th cloud velocity state is given by the concentration of nucleated crystals over the freezing timescale,

\[
\left. \frac{dN_c}{dt} \right|_{\text{fr, hom}} = \frac{P_i(S > S_{\text{hom}})N_0}{\tau_{\text{hom}, j}} H_\text{c}(u_j)
\]

(28)

where \( \tau_{\text{hom}, j}^{-1} = \bar{v}_0 J_{\text{max}, j} \exp \left( -\frac{\bar{v}_0}{\alpha u_j} \int_0^{S_{\text{max}}} J(S) dS \right) \).

\( H_\text{c}(u_j) \) is the Heaviside function and is introduced because homogeneous nucleation is very unlikely in parcels with negative vertical velocity (i.e., updraft must be maintained for some time before \( S_{\text{hom}} \) is reached after which it is quickly depleted by crystal nucleation and growth, Barahona and Nenes, 2008; Kärcher and Lohmann, 2002). \( P_i(S > S_{\text{hom}}) \) represents the fraction of parcels for which \( S > S_{\text{hom}} \). Using the same averaging procedure as for the supersaturation equation, we obtain

\[
\left. \frac{dN_c}{dt} \right|_{\text{fr, hom}} = N_0 P_i(S > S_{\text{hom}}) H_\text{c}(\mu_j) \left. \right|_{\mu = \mu_j}
\]

(29)

3.2.2 Heterogeneous nucleation

The formulation of the ice crystal production rate by heterogeneous freezing is simplified by using the ice nucleation spectrum, \( N_\text{IN}(S, T) \) (Barahona and Nenes, 2009a).

Assuming that \( N_\text{IN}(S, T) \) is weakly dependent on \( T \) and \( N_\text{IN}(S, T) = N_{\text{o, het}} f_{\text{het}}(S) \), then following Eq. (29) we write,

\[
\left. \frac{dN_c}{dt} \right|_{\text{fr, het}} = N_{\text{o, het}} \left. \frac{dP_i}{dS} \right|_{\text{fr, het}} \frac{dS}{dt} P_i(S > S_{\text{het}}) H_\text{c}(\mu_j) \left. \right|_{\mu = \mu_j}
\]

(30)

which can be written as

\[
\left. \frac{dN_c}{dt} \right|_{\text{fr, het}} = N_{\text{o, het}} P_i(S > S_{\text{het}}) \frac{H_\text{c}(\mu_j)}{\tau_{\text{het}, j}} \left. \right|_{\mu = \mu_j}
\]

(32)

where \( \tau_{\text{het}, j} = \left( \frac{dP_i}{dS} \bar{a}_{\text{het}, j} S \right)^{-1} \).

3.2.3 Sedimentation of ice crystals

Sedimentation processes out of the cloud layer depend primarily on the bulk properties of the cloud, i.e., the mean ice crystal size distribution and number concentration (interaction of individual parcels with falling crystals within the layer is accounted for in Eq. 27). The ice crystal loss rate by sedimentation is then given by,

\[
\frac{dN_c}{dt} \mid_{\text{sed}} = \frac{1}{H} \int_{D_{\text{min}}}^{D_{\text{term}}} \bar{u}_{\text{term}}(D_c)n(D_c)dD_c
\]

(33)

where \( H \) is the cloud layer thickness. As \( u_{\text{term}} \sim D_c \) (Heymsfield and Iaquinta, 2000), Eq. (33) can be further simplified to

\[
\frac{dN_c}{dt} \mid_{\text{sed}} = \frac{N_c \bar{u}_{\text{term}}}{H} = \frac{N_c}{\tau_{\text{sed}}}
\]

(34)

where \( \bar{u}_{\text{term}} = u_{\text{term}}(\bar{D}_c) \).

3.3 Numerical solution

3.3.1 Competition between homogeneous and heterogeneous freezing

Calculation of ice crystal number concentration, \( N_c \) in situ cirrus from combined homogeneous and heterogeneous nucleation in Figs. 1 and 3 is done using an analytical parameterization developed for in situ formed cirrus clouds and freezing fractions below 0.6 (Barahona and Nenes, 2009b). When the calculated freezing fraction exceeds 0.6, a sigmoidal increase in \( N_c \) is assumed (Barahona et al., 2010a),

Atmos. Chem. Phys., 11, 3757–3771, 2011
in agreement with parcel model simulations and field observations (Barahona and Nenes, 2008; DeMott et al., 2003).

For combined homogeneous and heterogeneous nucleation, it was assumed that the IN freeze instantaneously at a super-saturation freezing threshold, \( q_{\text{het}} \), of 15%, typical of deposition mode IN (Abbatt et al., 2006), with a 0.1 µm diameter at freezing (Froyd et al., 2009). Glassy aerosol was assumed to have a total concentration of 50 cm\(^{-3}\) and a freezing fraction given by the nucleation spectrum of Murray et al. (2010).

### 3.3.2 Vertical velocity spectrum

Observations suggest that the spectrum of gravity wave perturbations near the tropopause is pseudorandom in nature, a result of the superposition of waves from different sources, and therefore varies temporally and spatially (Bacmeister et al., 1999; Jensen and Pfister, 2004; Kim et al., 2003). A representative spectrum of vertical velocity fluctuations can be generated using a Fourier series (Bacmeister et al., 1999; Jensen and Pfister, 2004) of the form

\[
u = \bar{\nu} + \sum_j A(\omega_j) \cos(\omega_j t + m H + \varphi)
\]

where \( m \) is the vertical wave number, \( H \) is the cloud thickness, and \( \omega_j, A(\omega_j), \) and \( \varphi \), are the wave frequency, phase, and amplitude, respectively. We have adopted this representation as follows.

For each cirrus simulation, a time series of \( u \) was generated over the frequency interval \( \omega = [3.35 \times 10^{-7}, 9.44 \times 10^{-4}] \) Hz (Jensen and Pfister, 2004), using randomly generated \( \varphi \) and \( m \). \( A(\omega_j) \) was calculated using a power spectrum scaling law of \(-1.85\) for \( \omega_j \geq 1 \times 10^{-5} \) Hz and of \(-0.25\) for \( \omega_j \leq 1 \times 10^{-5} \) Hz (Herzog and Vial, 2001; Jensen and Pfister, 2004). This procedure resulted in a normal distribution of \( u \) (Fig. 4d) centered around \( \bar{u} \). The maximum amplitude was assumed to occur at \( \omega_j = 1 \times 10^{-3} \) Hz (Jensen and Pfister, 2004) as it reproduces the results of Gayet et al. (2004) (Fig. 4 green line) which give positive \( u \) around 0.23 m s\(^{-1}\) for \( \delta T = 1 \) K (i.e., \( A(1 \times 10^{-3}) \approx 2.1 \delta T / \bar{u} \)). Representative time series for \( u(t) \) are presented in Fig. 5a.

### 3.3.3 Ice crystal production

The homogeneous freezing timescale, \( \tau_{\text{hom}, j} \), was calculated using the parameterization of Barahona and Nenes (2008, 2009b, a). Precursor aerosol was assumed to be composed of deliquesced ammonium sulfate, lognormally distributed with dry mean geometric diameter of 40 nm, geometric dispersion of 2.3, and number concentration of 100 cm\(^{-3}\) (Lawson et al., 2008). To account for possible compositional impacts on crystal growth kinetics, the water-vapor deposition coefficient was varied between 0.006 (Magee et al., 2006) and 1.0. Homogeneous freezing is described using the parameterization of Koop et al. (2000). The term \( P_j \) (\( S > S_{\text{hom}} \)) in Eq. (28) is the probability of finding \( S \) above \( S_{\text{hom}} \), and is introduced to account for the threshold behavior of homogeneous freezing (Kärcher and Burkhardt, 2008; Koop et al., 2000). The effect of preexisting ice crystals on freezing was accounted for by allowing a fraction of \( N_c \) to deplete water vapor and increase \( \tau_{\text{hom}, j} \) (Barahona and Nenes, 2009b; Barahona et al., 2010b). The fraction of preexisting crystals remaining in freezing zones was calculated as

\[
f_{\text{f,ps}} = \left. \frac{1}{N_c \bar{D}_c} \int_{D_{\text{min}}}^{D_{\text{term}}} D_c n_c(D_c) dD_c \right| \frac{\text{total water content}}{D_{\text{term}}}
\]

where \( n(D_c) \) is the ice crystal size distribution, \( D_{\text{min}} \) is the minimum pre-existing crystal size, and \( D_{\text{term}} \) is the crystal size for which its terminal velocity, \( u_{\text{term}} \), is equal to the uplift velocity of the cirrus later, \( \bar{u} \). \( u_{\text{term}} \) was calculated assuming ice crystals have columnar shape with maximum dimension equal to \( \bar{D}_c \) (Heymsfield and Iaquinta, 2000). Following Heymsfield and Platt (1984) it was assumed \( n(D_c) = AD_c^{-3.15} \); the parameters \( A \) and \( D_{\text{min}} \) were calculated from the moments of \( n(D_c) \):

\[
N_c = \int_{D_{\text{min}}}^{\infty} n(D_c) dD_c = \frac{1}{N_c} \int_{D_{\text{min}}}^{\infty} D_c n(D_c) dD_c.
\]

The calculation of \( \bar{D}_c \) is described below. Integration of equations of Eqs. (18) and (21) was accomplished using a fixed time step of 2 s. Initial values for \( N_{c,0} = 0.01 \) cm\(^{-3}\) and \( S_0 = 1.0 \) were set. Sensitivity to using different initial values affected only the time required to establish dynamic equilibrium (by a few hours) and is assessed in Fig. 8.

The heterogeneous freezing timescale, \( \tau_{\text{het}, j} \), (e.g., Eq. 32) was calculated at each time step using the instantaneous vertical velocity, \( u_j \), and the effective supersaturation \( S_{\text{eff}, j} \). For the simulations presented in Sect. 4, the aerosol freezing fraction, \( f_{\text{het}} \), was calculated from the spectrum of Murray et al. (2010). \( N_{0,\text{het}} \) and \( S_{\text{het}} \) were set to 1 cm\(^{-3}\) and 1.35, respectively (Murray et al., 2010).

---

Fig. 5. Time series of (a) updraft velocity, \( u \), (b) total water content, \( q_{\text{tot}} \), and (c) mean ice crystal diameter, \( \bar{D}_c \), for the conditions presented in Fig. 4; (d) time series of \( \bar{D}_c \) for initial \( S \) and \( N_c \) of \(-0.2\) and 0, respectively, and homogeneous and heterogeneous nucleation active (conditions similar to Fig. 8c).
3.3.4 Ice crystal sedimentation

The rate of ice crystal sedimentation over the cloud scale, $H$, was assumed proportional to the terminal velocity of the mean crystal size $\bar{D}_c$ (Eq. 33). Other removal processes (ice crystal sublimation and detrainment) are neglected; $H$ however was varied over a wide interval (100 to 5000 m) to account for the uncertainty associated with neglecting these processes. $\bar{D}_c$ was calculated so that the total water vapor in the layer was partitioned between ice and vapor phases, i.e.,

$$\bar{D}_c = \left( \frac{6q_{\text{ice}}}{\tau N_c} \right)^{1/3},$$

where $q_{\text{ice}} = q_{\text{tot}} - \frac{p^0 S_0 M_w}{RT}$, $\rho_i$ is the ice density (Pruppacher and Klett, 1997), $R$ is the universal gas constant, $M_w$ is the molecular mass of water, and $p^0$ is the saturation water vapor pressure over ice (Murphy and Koop, 2005); the minimum ice crystal size was set to 4 $\mu$m in agreement with theoretical studies and experimental observations (Barahona and Nenes, 2008; Durran et al., 2009; Krämer et al., 2009). Loss of total water content, $q_{\text{tot}}$, from the cloudy layer is also accounted for by solution of

$$\frac{dN_{\text{tot}}}{dt} = \frac{-\pi}{6} \rho_i \bar{D}_c^3 \frac{dN_c}{dt} \text{,}$$

Representative time profiles of $\bar{D}_c$ and $q_{\text{tot}}$ are presented in Fig. 5. The timescale of relaxation at $u = u_j$, $\tau_{\text{rel},j}$, was calculated using $N_c$ and $\bar{D}_c$ of the cloud layer (e.g., Eq. 13). Mass transfer limitation from non-continuum effects are taken into account in the calculation of $\tau_{\text{hom},j}$ but neglected in the calculation of $\tau_{\text{rel},j}$. The latter is justified as $\bar{D}_c > 10 \mu$m.

4 Cirrus in dynamical equilibrium

The model developed in Sect. 3 is used to describe the evolution of a cirrus layer at low temperature taking into account the effect of internal $S$ variations on $N_c$. Forward integration of Eqs. (18) and (29) and (34) is carried out using the procedure described in Sect. 3.3. A wide range of initial conditions and model parameters are selected to describe the cirrus evolution under different scenarios.

Figure 4 shows the evolution of a cirrus layer subject to gravity-wave fluctuations with an initial average temperature of 195 K and lifting at $\bar{u} = 1$ cm s$^{-1}$. Only homogeneous freezing is considered (heterogeneous freezing is “switched off”, i.e., $N_{\text{0,het}} = 0$). For values of $\delta T > 1$ K, the cloud initially experiences a strong homogeneous nucleation pulse, so that $N_c$ initially increases steeply (Fig. 4a); the consumption of water vapor by crystal growth decreases $S_0$ (Fig. 4b) which prevents any new freezing events. $N_c$ slowly decreases from sedimentation loss; only after enough ice crystals sediment out of the cloud layer, $S_0$ increases (e.g., $\delta T = 1$ K, green lines) and new freezing events occur. For $\delta T > 1.4$ K (purple lines) this is possible even if the layer remains on average slightly subsaturated ($S_0 \approx 1$) because the probability distribution of $S$ is broad enough for a non-negligible probability with $S > S_{\text{hom}}$. However it is likely that recently formed crystals will sublimate within a few hours in the subsaturated environment returning the moisture to the layer (ice crystal removal by sublimation is not considered). The cirrus is then maintained by new, independent freezing events. This “pulse-decay” behavior is characterized by $\tau_{\text{sed}} \gg \tau_{\text{rel}}$ so ice crystals reside long enough in the cloud to relax supersaturation (Fig. 4c); this behavior is also consistent with the parcel model concept of cirrus (where high $N_c$ and low $S_0$ coexist within the parcel). The subsaturation levels (Fig. 4b) achieved in this state are in agreement with in situ observations of relative humidity in dissipating clouds (Gao et al., 2004; Krämer et al., 2009).

The cirrus evolution is however quite different when $\delta T$ is small; the distribution of $S$ is narrow, and substantial ice production is only possible after enough supersaturation (i.e., $S_0$) builds up in the cloudy layer to allow a non-negligible probability where $S > S_{\text{hom}}$. Thus, freezing events producing large $N_c$ (associated with large $u$ fluctuations; Fig. 4d) are less frequent. Low $N_c$ allows the formation of large ice crystals (Fig. 5c, d) which sediment out of the layer before substantially depleting supersaturation, leading to new freezing events. This “dynamic equilibrium” between ice production and loss is a previously unidentified microphysical regime of cirrus, characterized by $\tau_{\text{sed}} \sim \tau_{\text{rel},j}$ (Fig. 4c): it maintains low $N_c$ and high $S_0$ in the cloudy layer (Fig. 4a, b) and is consistent with observations of low-temperature cirrus. Clouds in “dynamic equilibrium” also exhibit broad crystal size distribution, because large ice crystals coexist with freshly-formed (small) crystals in the cloud. The average ice crystal size in this case converges to values around 15 to 20 $\mu$m, in agreement with observations (Krämer et al., 2009). Before equilibrium is reached, $D_c$ exhibits larger values due to the model’s sensitivity to initial conditions (Fig. 5c, d).

When simulations (such as those of Fig. 4) are placed on a “state diagram” of $N_c$ vs. $S_0$, the two microphysical regimes described above clearly emerge. Examples are presented in Figs. 6 and 7 for a range model parameters and a variety of $\delta T$ (lines of distinct color). For example, decreasing the cloud thickness to 100 m (Fig. 4b) increases the sedimentation rate (Eq. 34) allowing $S$ to replenish quickly and facilitating the onset of equilibrium. Similarly, the rate of ice crystal growth increases with $T$ (Fig. 4d) increasing the average ice crystal size (hence decreasing $\tau_{\text{rel}}$ and $\tau_{\text{sed}}$) and facilitating the onset of equilibrium. Dynamical equilibrium would however lead to lower $N_c$ than observed at $T \sim 225$ K (Gayet et al., 2004; Krämer et al., 2009) and it is likely that clouds at these conditions would evolve following a pulse-decay behavior. Figure 7 shows state diagrams for different values of $\bar{u}$, $T$ and $H$ for the similar initial conditions. In general, progression towards a “dynamic equilibrium” is favored when supersaturation replenishes quickly (i.e., at high $\bar{u}$) as ice crystal growth and sedimentation are favored (leading to low $\tau_{\text{sed}}$), and vice-versa for “pulse-decay” behavior. Rapid consumption of supersaturation by the growing ice crystals also decreases the time between freezing pulses and replenishment of $S$. Thus the period of the $S$ and $N_c$ oscillations in the dynamic equilibrium state is mainly controlled by $\tau_{\text{rel}}$. 

Atmos. Chem. Phys., 11, 3757–3771, 2011
Fig. 6. Sensitivity of $N_c$ and $S_o$ evolution to cloud formation conditions for different values of $\delta T$ (color scheme same as in Fig. 4); (a) same conditions as in Fig. 4, (b) cloud thickness, $H = 100$ m (increased ice crystal removal rate), (c) deposition coefficient equal to 0.006 (Magee et al., 2006) (slow water vapor transfer), and (d) initial temperature 225 K and cloud lifting at 5 cm s$^{-1}$. The yellow star in each panel indicates initial conditions. The arrows indicate the temporal progression along each trajectory. The integration time was 40 h cases, except in (d) were it was 15 h.

Clouds in the “dynamic equilibrium” regime are also much less sensitive to slow water vapor deposition than predicted by box-model simulations. Figure 6c shows that dynamic equilibrium is possible even for $\alpha_d$ as low as 0.006 (compared to $\alpha_d = 1$ used in Figs. 4 and 6a for the same simulation conditions). Still, the high rate of production of ice crystals for $\alpha_d = 0.006$ increases $N_c$ and decreases $\bar{D}_c$, therefore increasing $\tau_{sed}$ and slowing the replenishment of supersaturation. Thus, dynamic equilibrium is only possible for $\delta T < 0.8$ as opposed to $\delta T < 1.0$ for $\alpha_d = 1$. Figure 6c however shows that the existence of strong kinetic limitations to the diffusional growth of ice crystals cannot be ruled out.

4.1 Effect of heterogeneous IN and initial conditions

It is important to study the sensitivity of the dynamical states of cirrus to initial conditions used in the simulation and to the presence of heterogeneous IN. Figure 8a shows $S$ and $N_c$ for the conditions of Figs. 4 and 6a but starting at $S_{o,ini} = -0.4$ and $N_{c,0} = 0$. For this set of initial conditions, the onset of oscillating behavior is delayed by a few hours before supersaturation is reached. This implies that the temperature of the first freezing pulse is lower than in the case with $S_{o,ini} = 1$, slightly increasing its strength; the system however dampens out these variations and eventually follows a similar trajectory as in Fig. 6a.

Figure 8b and c present simulations where both homogeneous and heterogeneous nucleation are active for different initial conditions. IN are assumed to originate from glassy aerosol with $N_{o,het} = 1$ cm$^{-3}$. By allowing ice crystal formation at low $S$, heterogeneous IN decrease the ice crystal production rate by homogeneous nucleation hence lower the maximum $N_c$. This makes the system more stable to high amplitude vertical velocity perturbations, facilitating the onset of dynamic equilibrium, which can be maintained up to $\delta T \sim 1.2$. The “stabilizing” effect of IN is exemplified in Fig. 8c where homogeneous freezing was suppressed, i.e., only heterogeneous freezing is active. This leads to a damped response to vertical velocity fluctuations so that dynamical equilibrium is possible even for $\delta T$ as high as 1.8 K. Thus, in a cirrus cloud where heterogeneous nucleation is dominant, dynamical equilibrium is very robust against external perturbations. The values of $N_c$ and $S$ at the equilibrium state are however not significantly influenced by the initial conditions nor the ice nucleation mechanism, e.g., after sometime the system oscillates about the same values as in Figs. 6 and 7. This means that the presence of IN can “help” the cirrus to achieve dynamical equilibrium without modifying the equilibrium values.
Figures 6 to 8 also show that the “dynamic equilibrium” state occurs spontaneously when $\delta T$ goes below a characteristic transition value (which depends on $\bar{u}$, $T$ and the predominant freezing mechanism). It can also be reached after a cloud initially resides in a “pulse-decay” state, if $\delta T$ is close to the characteristic value ($\delta T \sim 1$ K in Fig. 6). When maximum $N_c$ and time-averaged $S_o$ are presented on the state diagram for all simulations considered where homogeneous freezing is active, the conditions of $\delta T$ that separate “pulse-decay” and “dynamic equilibrium” regimes seem to be universal (Fig. 9). If only heterogeneous nucleation is active, the cloud resides mostly in the dynamic equilibrium state.

5 Conclusions and implications

From the discussion above, cold cirrus clouds will reside in the “dynamic equilibrium” regime if $\delta T$ is below a characteristic threshold. High-amplitude, orographically-generated gravity waves are ubiquitous (Kim et al., 2003) but often lose intensity with altitude, weakening their contribution to the background spectrum of temperature fluctuations. $\delta T$ can thus decrease enough at high altitude for cirrus to transition from a “pulse-decay” to a “dynamic equilibrium” state (Fig. 9). This would explain why low $N_c$ and high $S_o$ are observed at low temperatures near the tropopause. Dynamical equilibrium is also possible at warmer conditions (particularly for high $\bar{u}$; Fig. 6d) but require small $\delta T$; given that high amplitude fluctuations are widespread at lower altitudes (Hoyle et al., 2005), cirrus clouds are likely forced to always follow a pulse-decaying behavior. Heterogeneous IN nevertheless may help to stabilize the system so that the dynamic equilibrium manifests at higher $\delta T$ than for clouds with pure homogeneous freezing.

In summary, cirrus clouds at low temperature exhibit characteristics (e.g., low $N_c$ and sustained high saturation ratios) that cannot be explained with the simple “conventional” picture of homogeneous freezing driven by expansion cooling in the presence of ubiquitous temperature fluctuations. Even if heterogeneous nucleation is dominant, conventional models of cirrus could explain the characteristics of low-$T$ cirrus only for weak updrafts, and require neglecting the higher amplitude components of the vertical velocity spectrum. Instead, we show that small-scale fluctuations from the action of gravity waves can switch a cloud into a previously unknown “dynamic equilibrium” regime, with sustained levels of low $N_c$ and high saturation ratios consistent with “puzzling” characteristics observed in low temperature cirrus.

With this study, a new understanding for cirrus clouds emerges, where the “unperturbed” microphysical state is one of dynamical equilibrium with low crystal number and high supersaturation. Only when the mean amplitude of temperature fluctuations exceeds a threshold value ($\delta T \sim 1$ K when homogeneous freezing is active) cirrus exhibit the well-known “pulse-decay” microphysical state. Throughout much of the atmosphere, the latter state dominates, simply because $\delta T$ is larger than the characteristic threshold value. In the TTL, $\delta T$ is still remarkably large (0.6–0.8 K) (Bacmeister et al., 1999; Jensen and Pfister, 2004; Sato, 1990), but does not systematically exceed the threshold for “pulse-decay” behavior, so cirrus regress to their “unperturbed” dynamic-equilibrium state. The presence of heterogeneous IN can...
also dampen the effect of vertical velocity perturbations on ice crystal production facilitating the transition to dynamic equilibrium. However, the existence of IN is not a necessary condition to explain the characteristics of cirrus clouds at low temperature. Figure 6c shows that dynamic equilibrium states can also exist under conditions of strong kinetic limitations (very low $d_d$) and therefore their existence cannot be ruled out based on simplified models of cirrus formation.

The structure and responses of cirrus to dynamical and microphysical forcings can also be portrayed. For example, cirrus formed in the region of convective anvils might exhibit “pulse-decay” state until gravity-wave fluctuations decay to below the $\delta T$ threshold and transition to a dynamic-equilibrium state. For the same reasons, IN impacts on cirrus properties can be strong for clouds in pulse-decay state, but not for clouds in dynamic equilibrium; e.g., IN can force the cloud to fall towards equilibrium however will not modify the equilibrium state. In conclusion, the discovery of dynamic equilibrium states reshapes our understanding of cirrus clouds and their role in anthropogenic climate change, as the type of dynamical forcing and the presence of IN will set these clouds in one of two “preferred” microphysical regimes with very different susceptibility to anthropogenic aerosol.

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