Analytic Solutions for Evolving Size Distributions of Spherical Crystals or Droplets Undergoing Diffusional Growth in Different Regimes

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ABSTRACT
Motivated by simulations of slow-growing contrail cirrus, the solution of the diffusional growth equations for a population of spherical ice crystals or water droplets is reexamined. For forcing specified by the evolution of the total water content above saturation within a parcel (whether driven by vertical motions, radiative heating, turbulent mixing, etc.) three behavior regimes are identified: “very fast growth” that cannot equilibrate, “fast growth” with a narrowing size spectrum, and “slow growth” with a broadening spectrum. The boundaries between regimes, time scales involved, and evolution of the condensate mass, number, and supersaturation are determined. The slow-growth regime represents an example of “spectral ripening,” with crystal or droplet numbers falling in time because of surface tension effects. Surprisingly the diffusional growth equations for the size spectrum evolution can be solved exactly in this case: in appropriate coordinates the spectral shape becomes steady, crystal or droplet numbers fall as a forcing-dependent power law, and the mean particle mass grows linearly with time. Dependence on different physical variables, fluctuating forcing, and modifications due to kinetic theory corrections are all considered. In the limit of zero external forcing on the parcel the size-spectrum solution is mathematically equivalent to a classic result in the theory of Ostwald ripening of solid solutions. It is argued that the slow-growth regime may be important in the evolution of contrail cirrus and perhaps in setting upper limits on droplet number densities in stratiform boundary layer clouds. The theoretical results are compared with parcel model simulations for illustration and to study numerical issues in binned microphysics models.

1. Introduction

The particle size distribution (PSD) of a cloud is needed for many applications such as determining radiative properties or precipitation dynamics. Many processes are involved in its evolution including nucleation, diffusional growth, turbulent mixing, sedimentation, and aggregation. In the present work we derive analytic solutions for spectra within a parcel evolving primarily via diffusional growth for some broad regimes of imposed forcing independent of specific origin, whether vertical motions, radiative heating, or turbulent growth of the parcel. Of particular interest are regimes in which there is a steady loss of crystals or droplets but where the spectral shape, defined in appropriate coordinates, remains steady.

This study was motivated by results from large-eddy simulation (LES) of contrail-induced cirrus clouds for different conditions (D. Lewellen et al. 2011, unpublished manuscript). These typically involve localized clouds in an ice-supersaturated environment that grow in volume and ice mass only relatively slowly over times of hours through turbulent mixing and that have relatively large ice number densities in their cores. The simulations often show gradual ice crystal number loss with a simple power-law behavior in time that disappears if the dependency of the equilibrium vapor pressure on the ice surface tension (the “Kelvin effect”) is removed from the microphysics model. This occurs despite the mean crystal size being large enough that one would, a priori, consider the influence of the Kelvin effect to be negligible. This observation stimulated a series of simpler parcel simulations with the LES code and the analytic treatment of a basic set of microphysics equations.

In section 2 of this paper the equations defining the microphysics at the chosen level of approximation are presented and relevant time scales are identified. The boundaries of different behavior regimes are determined and, in the slow-growth regime, an exact solution is derived for the PSD development as a function of the
imposed forcing on the parcel. Some algebraic details are relegated to appendix A. Sample results from numerical simulations with a binned microphysics model are given for illustration and as a check of the analytic solutions. Exact but nontrivial analytic solutions provide a convenient testing ground for numerical treatments; an examination of numerical choices within binned microphysics schemes from this perspective is given in appendix B.

Section 3 discusses further the slow-growth regime, including the sensitivity to different physical variables, effects of forcing on faster or slower time scales, and effects of kinetic theory corrections to the diffusion equation. The presentation and simulation examples are given for ice clouds, but, as is well known, the underlying equations of diffusional growth are applicable (with suitably modified terms) to water droplets as well; the implications of the results for water clouds are also discussed briefly in section 3.

The surface tension–dependent crystal (or droplet) loss in the slow-growth regime is due to scavenging of moisture by larger crystals from smaller crystals and is an example of the more general phenomenon of Ostwald ripening.1 This phenomenon has received extensive study in applications to solid solutions, emulsions, and alloys. It turns out that in the limit of zero imposed forcing on the parcel the slow-growth solution presented here reproduces a classic result for Ostwald ripening of solid solutions in Lifshitz and Slyozov (1961) and Wagner (1961), dubbed “LSW theory” (Kahlweit 1975).2 There has been comparatively very little consideration of this phenomenon in the atmospheric sciences. To our knowledge, the solution of the spectral development, identification of the boundaries of the behavior regimes, and application to ice clouds considered here are all new. There has been some earlier discussion related to the ripening process within water clouds, however. Srivastava (1991) derived an expression for the increase in time of the difference of the squared radii of two droplets due to surface tension effects under the influence of an updraft. He noted that the surface tension effects can be important even at late times and sometimes give rise to droplet loss, though without solving for the rates. Korolev (1995) and Celik and Marwitz (1999) used parcel simulations to demonstrate broadening of droplet size spectra due to the ripening process. Korolev attributed this to the (irreversible) influence of fluctuations in the supersaturation level due to turbulent motions, but Celik and Marwitz (1999) concluded that the process was independent of supersaturation fluctuations for a closed parcel. Wood et al. (2002) assessed the potential importance of spectral ripening for stratiform boundary layer clouds, identifying the importance of the mean parcel in-cloud residence time. Some of the present results provide further insight on this issue.

2. Diffusional growth in different regimes

a. Motivating examples

Figure 1 shows results from a simulation of an aircraft contrail evolving into contrail cirrus over 12 h. The 3D LES includes aircraft wake motions, resolved ambient atmospheric turbulence, and binned ice microphysics adapted from the National Aeronautics and Space Administration (NASA) Ames Community Aerosol and Radiation Model for Atmospheres (CARMA; e.g., Jensen et al. 1994; Ackerman et al. 1995). The LES model and

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1 We thank J. Ross for first pointing this out to us.
2 We thank an anonymous reviewer for suggesting this reference and hence leading our attention to LSW theory.
Simulation procedures are extensions of those described in Lewellen and Lewellen (2001) and Huebsch and Lewellen (2006). The ice mass grows as the contrail plume expands in highly supersaturated ambient conditions, driven at different times by aircraft wake motions, ambient turbulence, and precipitation of ice crystals (which eventually sublimate in a subsaturated layer below). The contrail core has higher crystal number densities (of order a few per cubic centimeter even at an age of several hours, with radii of a few microns) and slower total ice mass growth than typical natural cirrus because of the origin of the ice crystals (in cooling aircraft exhausts in a low-turbulence ambient environment versus nucleation in an ambient updraft). Most notable for present purposes is a steady late-time power-law loss of ice crystal number despite a steady growth in mean crystal size, and a quasi-steady shape of the PSD (displayed logarithmically). The figure displays the crystal number integrated over the contrail volume (and averaged per meter of flight path) so the reduction seen is not a dilution effect. Nor is the crystal loss in such cases a precipitation or aggregation effect: while the ice mass is strongly influenced by growth at the plume edges and precipitation below, the crystal number (and PSD) is heavily concentrated in a slower-growing core volume of smaller crystals.

There are numerous complexities in the contrail–cirrus development including large spatial inhomogeneities. Figure 2 shows sample results run with the same code in a simpler venue, a fast-running box model/parcel mode. An initial population of ice crystals is prescribed uniformly in a parcel. The parcel is defined subsequently to follow the population of ice crystals, with a mixing of ambient air into the parcel imposed at a chosen rate, expanding the parcel’s volume over time. Note that this differs from the more common convention of defining a parcel so that its volume is fixed over time. Particle sedimentation and aggregation are not included. The results are shown integrated over the expanding parcel; in particular, the total crystal number is plotted (rather than a number density per unit volume) so that the losses in Fig. 2b are entirely due to complete evaporation of crystals, not to dilution of the parcel. Asymptotically three different behavior modes can be identified: a “very-fast-growth” regime in which the mixing of ambient supersaturated air into the parcel is so rapid that the ice mass growth cannot keep up and the supersaturation \( \delta S_i \) grows in time; a “fast-growth” regime for more moderate imposed mixing in which the total ice mass growth scales with the growth in parcel volume, \( \delta S_i \) falls in time, all crystals survive and grow, and the PSD narrows; and a “slow-growth” regime in which the crystal number falls as a mixing rate–dependent power of time, the asymptotic fall of \( \delta S_i \) is near universal, and a broader, shape-preserving spectrum is found. For both the box model and contrail simulations, the observed crystal losses are prevented if the surface tension dependence of the equilibrium saturation vapor pressure over the crystal (the “Kelvin effect”) is removed from the microphysics model.

The crystal loss in the slow-growth regime is due to larger crystals in effect scavenging moisture from smaller ones because the ambient water vapor is slightly above the equilibrium saturation level for the large crystals but slightly below for the small ones. A matrix of parcel cases was simulated varying initial crystal number, ambient temperature and humidity, mixing rates into the parcel, and the ambient supersaturation.

**Fig. 2.** Results of box model simulations with full-binned microphysics for parcels of ice crystals expanding in an ambient atmosphere with \( T = 205 \text{ K}, P = 250 \text{ hPa}, \) and relative humidity with respect to ice of 110%. (a) Total ice mass in the expanding parcel (kg); (b) total crystal number in the expanding parcel; (c) supersaturation; (d) crystal size spectrum at \( t = 3 \times 10^4 \text{ s} \). In each case there are \( 10^{10} \) crystals in a 1 m\(^3\) volume initially and a power-law growth of the total water mass above saturation \([M_{eq} \text{ in Eq. (13)}]\) is imposed; \( M_{eq} = \tau^\alpha \), with \( \alpha = -0.25, 0, 0.25, 0.5, \) and 0.75 for the “slow-growth” cases (solid lines, in order of increasing line thickness); 1, 1.25, and 1.5 for the “fast-growth” cases (long dashed); and 2 for “very-fast-growth” (short dashed). A mass ratio of 1.1 between neighboring ice bins was used for the simulations, with an initial distribution uniform across five adjacent bins centered around 0.4-\( \mu \text{m} \) radius.
constant velocity or oscillatory vertical motions, and physical and numerical parameters relevant to the microphysics. Sample results will be presented below to illustrate the behavior in the different growth regimes.

\[ M_p(t) = m_0 \int_\xi^\infty d\xi e^f(\xi, t). \]  

(2)

Here \( \xi \) represents a minimum size threshold below which ice crystals are assumed to evaporate completely away. Imposing the size cutoff in this fashion, leaving \( f \) a smooth function even at \( \xi = \), proves convenient in the following analysis. Considering only diffusional growth/sublimation, the distribution function satisfies

\[ \frac{\partial f(\xi, t)}{\partial t} = - \frac{\partial}{\partial \xi} \left( \frac{d\xi}{dt} f \right). \]  

(3)

This is simply a statement that diffusional growth conserves crystal number, merely shifting it in \( \xi \) space; the right-hand side is the divergence of the local flux in this space. In this approximation any new nucleation of crystals, aggregation, or differential sorting from sedimentation is omitted. The only source of change in \( N \) is that is included is loss of crystal number through total evaporation, incorporated via the cutoff \( \xi = \). It will be shown posteriori in appendix A that the crystal loss rates are independent of the choice of \( \xi \) provided \( \xi \) is sufficiently small relative to the mean crystal size.

The diffusional growth of a crystal of mass \( m \) and radius \( r \) can be approximated by (see, e.g., Rogers 1976; Pruppacher and Klett 1997; Straka 2009)

\[ \frac{dm}{dt} = \frac{4\pi r (1 + \delta S_i - e^{\sigma_{iv}/r})}{L_s/KR_sT^3 + R_s/T^3 A}. \]  

(4)

with

\[ a_k = \frac{2\sigma_{iv}}{\rho R_s T^3}. \]  

(5)

Here \( \delta S_i = (q_i - q_t)q_i - 1 \) is the supersaturation, \( e \) the vapor pressure with respect to ice, \( L_s \) the latent heat of sublimation, \( R_s \) and \( D \) the gas constant and diffusivity for water vapor, and \( K \) the thermal diffusivity. The \( a_k \) term is taken by analogy to the Kelvin equation for droplets but using the surface tension for a water vapor–ice interface \( \sigma_{iv} \). Quantitatively this is an uncertain approximation given the complexities of ice growth, although a Kelvin effect is almost certainly present in some form. The value of \( \sigma_{iv} \) is also uncertain; in the simulations we use the temperature dependence from Hale and Plummer (1974). For cirrus-type conditions this gives \( a_k = 2 \times 10^{-3} \) \( \mu m \), much smaller than the mean crystal sizes we will consider, so that we can safely approximate \( e^{\sigma_{iv}/r} \approx 1 + a_k/r \). We cannot, however, drop the Kelvin correction entirely without greatly altering the solution in some cases, as is clear from the simulation results. In many regimes \( \delta S_i \) is

\[ N_p(t) = \int_\xi^\infty d\xi f(\xi, t). \]  

(1)

b. Equation set

In each of the three behavior regimes qualitatively identified in Fig. 2 the behavior exhibits enough simplicity (e.g., power-law scaling and self-similar PSD shapes) that it suggests that an analytic treatment might be fruitful. With this motivation we introduce a simplified equation set for the microphysics. We will find, despite the simplifications, that the analytic solutions predict the simulation results (which include more accurate microphysics) quite well. To proceed we consider the evolution of a parcel defined by following a specified population of ice crystals, governed by a reduced set of dynamics. Let us denote the total ice crystal number, ice mass, and air mass of the parcel as \( N_p, M_p \), and \( M_t \), respectively, and its temperature, density, total water mixing ratio, ice-mass mixing ratio, and saturation mixing ratio (with respect to ice) as \( T, \rho, q, q_t (= M_p/M_t) \), and \( q_o \), respectively. We consider the external forcing on the parcel (e.g., its vertical motion, radiative heating, or turbulent mixing of air into the parcel) as prescribed. Vertical motion or radiative heating drive changes in \( T \) (and hence in \( q_t \)); turbulent mixing into the parcel directly increases \( M_p \) (which by itself tends to decrease \( q_t \)) and can alter \( T \) or \( q_t \) if the temperature or mixing ratio outside the parcel differs from that within. We will show that the forcing enters the analysis below essentially only through its effect on the total water content of the parcel above saturation [cf. Eq. (13)] so we will not differentiate between the different sources of forcing in the following. Note that since we have defined the parcel to follow a chosen crystal population rather than a fixed air volume, the effect of the external forcing on \( N_p \) and \( M_p \) is solely through the ice microphysics; dilution alone leaves them unchanged. We assume that the parcel is chosen to be small enough that it can be approximated as well mixed throughout its evolution. For simplicity we assume that the ice crystals are spherical. Since it is applications with very small ice crystals (or water droplets) that are intended, this is not entirely unreasonable.

It proves convenient to treat the PSD with a logarithmic coordinate \( \xi = \ln(m/m_0) \), where \( m \) is the mass of an individual ice crystal and \( m_0 \) is a mass scale specified below. The number of crystals in the parcel with \( \xi \) lying between \( \xi \) and \( \xi + d\xi \) will be denoted \( f(\xi, t) \). Then, by definition,

\[ M_p(t) = m_0 \int_\xi^\infty d\xi e^f(\xi, t). \]  

Here \( \xi \) represents a minimum size threshold below which ice crystals are assumed to evaporate completely away. Imposing the size cutoff in this fashion, leaving \( f \) a smooth function even at \( \xi = \), proves convenient in the following analysis. Considering only diffusional growth/sublimation, the distribution function satisfies

\[ \frac{\partial f(\xi, t)}{\partial t} = - \frac{\partial}{\partial \xi} \left( \frac{d\xi}{dt} f \right). \]  

(3)

This is simply a statement that diffusional growth conserves crystal number, merely shifting it in \( \xi \) space; the right-hand side is the divergence of the local flux in this space. In this approximation any new nucleation of crystals, aggregation, or differential sorting from sedimentation is omitted. The only source of change in \( N \) is that is included is loss of crystal number through total evaporation, incorporated via the cutoff \( \xi = \). It will be shown posteriori in appendix A that the crystal loss rates are independent of the choice of \( \xi \) provided \( \xi \) is sufficiently small relative to the mean crystal size.

The diffusional growth of a crystal of mass \( m \) and radius \( r \) can be approximated by (see, e.g., Rogers 1976; Pruppacher and Klett 1997; Straka 2009)

\[ \frac{dm}{dt} = \frac{4\pi r (1 + \delta S_i - e^{\sigma_{iv}/r})}{L_s/KR_sT^3 + R_s/T^3 A}. \]  

(4)

with

\[ a_k = \frac{2\sigma_{iv}}{\rho R_s T^3}. \]  

(5)

Here \( \delta S_i = (q_i - q_t)q_i - 1 \) is the supersaturation, \( e \) the vapor pressure with respect to ice, \( L_s \) the latent heat of sublimation, \( R_s \) and \( D \) the gas constant and diffusivity for water vapor, and \( K \) the thermal diffusivity. The \( a_k \) term is taken by analogy to the Kelvin equation for droplets but using the surface tension for a water vapor–ice interface \( \sigma_{iv} \). Quantitatively this is an uncertain approximation given the complexities of ice growth, although a Kelvin effect is almost certainly present in some form. The value of \( \sigma_{iv} \) is also uncertain; in the simulations we use the temperature dependence from Hale and Plummer (1974). For cirrus-type conditions this gives \( a_k = 2 \times 10^{-3} \) \( \mu m \), much smaller than the mean crystal sizes we will consider, so that we can safely approximate \( e^{\sigma_{iv}/r} \approx 1 + a_k/r \). We cannot, however, drop the Kelvin correction entirely without greatly altering the solution in some cases, as is clear from the simulation results. In many regimes \( \delta S_i \) is
driven toward zero, making \( a_k \) a critical dimensional scale in Eq. (4). For this reason, perverse as it may seem a priori, we nondimensionalize by choosing (for ice density \( \rho_i \))

\[
m_0 = \frac{4}{3} \pi \rho_i a_k^3. \tag{6}
\]

With these choices and writing \( m \) and \( r \) in terms of \( \zeta \), Eq. (4) can be reexpressed as

\[
\frac{d \zeta}{dt} = G(e^{-2\zeta/3} \delta S_i - e^{-\zeta}), \tag{7}
\]

where \( G \) is a known thermodynamic function:

\[
G = \frac{4\pi a_k}{m_0} \left[ \frac{L_i^2}{K_{R_i} T^2} + \frac{R_i T}{e_i(T) D} \right]^{-1} \approx \frac{4\pi a_k}{m_0} \rho_i q_s D. \tag{8}
\]

The last approximation in Eq. (8), effected by dropping the \( L_i \)-dependent term, is good to a few percent for typical cirrus conditions but is not required for most of the analysis here. For now we neglect kinetic theory corrections to the diffusion (which are, however, present in the simulations) so \( G \) is independent of crystal radius. This will be revisited later.

c. Time scales and regime boundaries

Applying the simplified equation set (1)–(3) and (7) to the externally forced parcels of crystal populations, we now aim, as a function of that forcing, to determine the boundaries between the three behavior regimes identified qualitatively in the simulations in Fig. 2. We will also identify the time scales involved to reach those behavior regimes.

For the ice mass evolution in the parcel we have

\[
\frac{dM_p}{dt} = m_0 G \int_{\xi_c}^\infty d\zeta (e^{\xi/3} \delta S_i - 1)f \tag{9}
\]

\[
= m_0 G N_p \left( \delta S_i \frac{\bar{\tau}}{a_k} - 1 \right) \tag{10}
\]

\[
= -\frac{1}{\tau_c} \left\{ M_p - M_p \left[ q_i - q_s \left( 1 + \frac{a_k}{\bar{\tau}} \right) \right] \right\}. \tag{11}
\]

Expression (9) follows by differentiating Eq. (2) and using Eqs. (3) and (7), and an integration by parts.\(^3\) Then Eq. (10) follows using the mean crystal radius \( \bar{\tau} = a_k N_p^{-1} \int d\zeta e^{\xi/3}f \)

and finally Eq. (11) by using the definition of \( \delta S_i \) and defining the time scale

\[
\tau_c = \frac{M_p q_s a_k}{m_0 G N_p r} \approx (4\pi D \tau N_p)^{-1}, \tag{12}
\]

where \( N_p = \rho N_p / M_p \) is the local ice crystal number density. The approximation in Eq. (12) follows from that in Eq. (8) and identifies \( \tau_c \) as (for spherical crystals) the relaxation time scale given by Khvorostyanov and Sassen (1998).

The terms \( M_p(t), q_i(t), \) and \( q_s(t) \) are considered as prescribed forcing driving the parcel dynamics. If changes in these are not too rapid, then on time scales long compared to \( \tau_c \), Eq. (11) shows that the ice mass will be driven to an equilibrium value set directly by the forcing

\[
M_p \approx M_p(q_i - q_s) = M_{eq}. \tag{13}
\]

In this case (the fast and slow growth regimes introduced earlier) \( M_p(t) \) can be considered prescribed as well, and \( \delta S_i \) is driven small compared to supersaturation levels outside of the parcel. This is not true in the very-fast-growth regime; for instance, if ambient air is mixed into the parcel fast enough, the crystal growth will not be rapid enough to compensate and \( \delta S_i \) will instead approach its level in the ambient.

To establish the boundaries of the three regimes we solve Eq. (10) for \( \delta S_i \),

\[
\delta S_i = c_s^{-1} \left( \frac{m_0 N_p}{M_p} \right)^{1/3} \frac{1}{\left( \frac{1}{m_0 G N_p} \frac{dM_p}{dt} + 1 \right)}, \tag{14}
\]

where we have expressed \( \bar{\tau} \) in terms of the mean crystal mass \( \bar{m} \) and a constant \( c_s \), depending on the PSD:

\[
\bar{\tau} = c_s a_k \left( \frac{\bar{m}}{m_0} \right)^{1/3} \text{ with } \bar{m} = \frac{M_p}{N_p}. \tag{15}
\]

In what we identify as the fast-growth regime we expect Eq. (13) to apply, \( \delta S_i \) to be driven small in time; \( N_p \) to remain constant, and the second term on the right-hand side of Eq. (14) (which represents the Kelvin-effect piece) to remain negligible compared to the first. The time dependence on the right-hand side of Eq. (14) in this case is carried solely through \( M_p \) and possibly \( G \) [which from Eq. (8) varies with \( q_s \)], which are effectively externally prescribed. These expectations of the fast-growth regime will be violated and the very-fast-growth regime entered if the forcing is prescribed such that \( \delta S_i \) ends up growing in time. From Eq. (14) this will occur if (and only if) \( G^{-1} dM_{eq}^2 / dt \) grows in time. On the other hand, if

\(^3\) The boundary terms from the integration by parts are dropped because \( f(\infty) \) vanishes and \( f(\xi_c) \) is assumed to be negligibly small, as will again be confirmed a posteriori in appendix A.
$G^{-1} dM_{\text{eq}}/dt$ decreases with time, then on a time scale of order

$$\tau_s(t) = \frac{m(t)}{m_0 G} = \frac{q_i q_s}{q_s q_i} e^{-\frac{q_s (4\pi D N_p)}{q_i}}.$$  \hspace{1cm} (16)

the second term on the right-hand side of Eq. (14) will become comparable with the first, violating the conditions of the fast-growth regime so as to cross over into the slow-growth regime. If, for example, the forcing is as in Fig. 2, driven by mixing into the parcel so that $G$ may be considered constant and $M_{\text{eq}}$ increases as a power law, then the fast-growth regime is predicted for

$$1 \leq \alpha \leq 3/2 \quad \text{for} \quad M_{\text{eq}}(t) \sim t^{\alpha},$$  \hspace{1cm} (17)

with the slow-growth regime for $\alpha < 1$ and very-fast-growth regime for $\alpha > \frac{3}{2}$. The simulations are found to confirm this prediction (e.g., Fig. 2).

A few other points about the time scales: first, in the very-fast-growth regime $t \gg \tau_s$, is not generally reached because $\tau_s$ itself in Eq. (12) is increasing sufficiently fast in time; second, from Eq. (16) the time scale $\tau_s$ for equilibrating to the slow-growth behavior is generally much longer than the time scale $\tau_s$ for approaching the ice mass equilibrium in Eq. (13); and finally, given the strong inverse dependence of $\tau_s$ on $N_p$, the slow-growth regime is only encountered in practice given high enough number densities and/or long enough integration times. To give representative estimates of time scales and crystal radii involved, consider nominal cirrus conditions of $T = 220$ K and $3 \text{ mg m}^{-3}$ ice water content. For number densities $N_p \approx 1000 \text{ cm}^{-3}$ such as one might find in a young ($\approx 1 \text{ min old}$) contrail, this leads to mean volume radius $r_{\text{mv}} \approx 1 \mu m$, $\tau_r \approx 3 \text{ s}$, and $\tau_s \approx 1 \text{ min}$; for $N_p \approx 5 \text{ cm}^{-3}$ such as one might find in a contrail core a few hours old, $r_{\text{mv}} \approx 5 \mu m$, $\tau_r \approx 2 \text{ min}$, and $\tau_s \approx 4 \text{ h}$; and for $N_p \approx 0.5 \text{ cm}^{-3}$ such as might be more representative of aged natural cirrus, $r_{\text{mv}} \approx 12 \mu m$, $\tau_r \approx 10 \text{ min}$, and $\tau_s \approx 1.5 \text{ days}$. Note that $\tau_s$ tends to increase steadily in time as crystal number densities fall; in practice it is enough for $t$ to be comparable with $\tau_s(t)$ for the slow growth regime to be reached (since over the preceding development $\tau_s$ was smaller).

The behavior in the fast-growth regime is relatively simple: $N_p$ remains constant while $M_{\phi}(t)$ is given by Eq. (13) and $dS_\phi$ by Eq. (14). When a population of crystals or droplets all grow in time via vapor diffusion the relative width of the size spectrum shrinks in time (e.g., Rogers 1976). The asymptotic behavior is simply a monodisperse spectrum with $m = M_{\phi}(t)/N_p$. In the simulation results of Fig. 2 this expected behavior is seen approximated by a spectral peak with width limited by the bin resolution.

d. The slow-growth regime

We now consider the slow-growth regime in more detail, in particular solving for the evolving PSD on time scales of order $\tau_s$. The full derivation is somewhat involved but is useful to document because the same techniques can be used to find related solutions for modified forcing or growth kernel [Eq. (7)]. Some readers may prefer to jump directly to the solution in Eq. (37); confirming that this solves the desired Eqs. (1)–(3) and (7) is a comparatively straightforward exercise in substitution.

Figure 3 shows the development of the PSD from a box model parcel simulation with the full-binned microphysics that is expected to be in the slow-growth regime. Plotted logarithmically, the spectral shape, after broadening from its initial conditions (essentially on time scale $\tau_s$), becomes steady in time, shifting downward as crystals are lost and rightward as the mean crystal mass increases. This implies that the distribution function can be expressed in the self-similar form

$$f(\xi, t) = e^{\phi(x) + Q(t)} \quad \text{with} \quad x = \xi - \lambda(t).$$  \hspace{1cm} (18)

We assume this form and will check a posteriori its consistency with different forms of the external forcing. We have the freedom to shift a constant between $\phi$ and $Q$ and

Note that this corresponds to a steady broadening in time when plotted linearly with radius.
to define \( \lambda \) in terms of any fixed feature of the spectrum, allowing us to set the normalizations:

\[
\int_{-\infty}^{\infty} dx \, e^{\phi(x)} = 1; \quad \int_{-\infty}^{\infty} dx \, e^{\phi(x)+x} = 1. \tag{19}
\]

The latter corresponds to the choice

\[
e^\lambda = \frac{m}{m_0}. \tag{20}\]

With these conventions Eqs. (1) and (2) become simply

\[
N_p(t) = e^{Q(t)}, \tag{21}
\]

\[
M_p(t) = m_0 e^{Q(t)+\lambda(t)}. \tag{22}
\]

Here we have assumed that the spectrum falls sufficiently rapidly that the lower limits of the integrals in Eqs. (1) and (2) can be extended to \(-\infty\) with negligible change (as is confirmed in appendix A). Substituting Eq. (18) into Eq. (2) can be extended to

\[
\text{all } \lambda \text{ and } x. \tag{23}
\]

Equivalently, for the crystal number,

\[
N_p(t) = \frac{M_p(t)}{m(t)} = \frac{M_p(t)}{m(t)} + \Lambda m_0 G(t - t_0) \tag{28}
\]

\[
\approx \frac{M_p(t)}{\Lambda m_0 G t} \text{ for } t \gg \tau_s. \tag{29}
\]

The mean crystal mass increases linearly\(^5\) changing appreciably on time scale \(\tau_s\), and the crystal number falls at a rate to make this consistent with the given \(M_p(t)\) [from Eq. (13)]. The asymptotic result for \(N_p(t)\) becomes independent of the initial state. Equation (25) together with Eq. (27) provides the evolution of the supersaturation:

\[
\delta S_i = \tilde{S} \left( \frac{m_0}{m(t)} \right)^{1/3} \tag{30}
\]

\[
\approx \tilde{S}(\Lambda G t)^{-1/3} \text{ for } t \gg \tau_s. \tag{31}
\]

Differentiating Eq. (22) and using Eqs. (13), (21), (24), and (26) gives a constraint relating \(\Lambda\) and \(\tilde{Q}\) to the imposed forcing:

\[
\Lambda + \tilde{Q} = \frac{1}{m_0 G N_p} \frac{dM_{eq}}{dt}. \tag{32}
\]

Using Eq. (28) for \(N_p\) in Eq. (32), this provides a constraint on the form of the “imposed” \(M_{eq}\) evolution for it to be consistent with the ansatz (18):

\[
M_{eq}(t) = M_{eq}(t_0)[1 + \beta(t - t_0)]^\alpha, \tag{33}
\]

with

\[
\alpha = 1 + \tilde{Q}/\Lambda \tag{34}
\]

and

\[
\beta = m_0 G A / m(t_0). \tag{35}
\]

\(^5\) More generally, for time-varying \(G\), \(m(t) = m(t_0) + \Lambda m_0 \int_{t_0}^{t_0} G dt\) with corresponding changes to \(N_p(t)\).
In practice, it is sufficient if $M_{eq}(t)$ is approximately of the form of Eq. (33) for development on time scales of order $t_s$. As discussed later, variations on longer time scales are followed in a quasi-steady sense and on shorter time scales in a low-pass-filtered sense. Turbulent mixing of moist air into a parcel often naturally drives an approximately exponential growth in $M_{eq}(t)$. Note that the $a$, $1$ requirement in Eq. (17) for the slow-growth regime is consistent with the requirement (given the absence of nucleation) that $N_p(t)$, as given in Eq. (29), never grows.

A constraint between $L$, $\sim Q$, and the applied forcing is provided by Eq. (34), with the simplest case being that of $\sim Q = 2 L$ for constant $M_p$ (or growing only very slowly over time scales of order $t_y$). An additional constraint involving $\hat{S}$ can be obtained from Eq. (14) by using Eqs. (20) and (32) and multiplying through by $e^{\lambda/3}$.

$$\hat{S} = \frac{1}{2b^3} \Lambda + \frac{3}{2b^3} \hat{Q} + 1. \quad (36)$$

For $M_{eq} \sim r^a$ with $a < 1$ the simulation results are found to be consistent with the asymptotic behaviors predicted in Eqs. (27), (29), and (31) for $\eta$, $N_p$, and $\delta S$, respectively (e.g., Figs. 2 and 4) and with Eqs. (34) and (36) as well. The linear growth of $\eta$ is found as predicted even when $M_{eq}$ is falling (e.g., Fig. 4 for $a = -0.25$). The change in slopes in Fig. 4 is due to $\Lambda$ changing with $a$ as discussed below.

In appendix A the PSD is solved for explicitly. Written in terms of radius it is

$$f(\zeta, t) d\zeta = d(lnr) \frac{3 \gamma e^{\gamma N_p(r/r_{mx})} e^{-\gamma(1-r/r_{mx})}}{2(r/2r_{mx} + 1)^{1+4\gamma/3}(1-r/r_{mx})^{2+5\gamma/3}}. \quad (37)$$

where $\gamma = 1 - a$ is determined by the forcing Eq. (33) and $r_{mx}$ is a maximum radius related to the mean volume radius $r_{mv}$ or mean mass $\overline{m}$ by

$$r_{mx} = b(\gamma) r_{mv} = b(\gamma) \left[ \frac{3m(t)}{4\pi \rho_i} \right]^{1/3}. \quad (38)$$

An integral for $b(\gamma)$ is given in appendix A and is plotted in Fig. 5. The other constants introduced earlier are all determined in terms of $b$ in appendix A. For reference,

$$\Lambda = \frac{1}{2b^3}; \quad \hat{S} = \frac{3}{2b^3} \hat{Q} = -\frac{\gamma}{2b^3}; \quad c_s = \frac{1}{3b^2} (1 - \gamma + 2b^3). \quad (39)$$

The PSD [Eq. (37)] is plotted for sample values of $\gamma$ in Fig. 6. It falls off exponentially as $r \to r_{mx}$, only modestly above $r_{mv}$. Modestly below $r_{mv}$ there is a “crossover” radius at $r = \hat{S}^{-1} r_{mv}$: above this size the crystals grow while below they evaporate. The time dependence of Eq. (37) is carried through $N_p(t)$ and $r_{mx}(t)$ and given by Eqs. (27), (28), and (38). The PSD [Eq. (37)] and parameter values [Eq. (39)] are reproduced by simulation results up to numerical limitations as discussed in appendix B.

In the limit of zero external forcing on the parcel (i.e., for $\gamma = 1$), Eq. (37) reduces to the classic solution for the shape of the particle size spectrum produced by Ostwald ripening as given by LSW theory (Lifshitz and Slyozov 1961; Wagner 1961). A more pedagogical derivation of this limit (in the context of solid solutions) can be found in Lifshitz and Pitaevskii (1981). Interestingly, the full solution of Eq. (37) turns out to be mathematically closely related to the generalization of the LSW result to an arbitrary number of spatial dimensions as given by Yao.
et al. (1993) if we replace $\gamma$ in Eq. (37) by one-third the spatial dimension.

3. Further discussion of the slow-growth regime

a. Dependence on different physical variables

From the analytic solution we can identify the effects of different physical parameters on slow-growth regime behavior. When it applies, the solution is determined solely through $M_{eq}$, $G$, and an initial condition $N_p(t_0)$. For illustration Fig. 7 shows simulation results (with the full-binned microphysics model) where $P$, $T$, $q_i/q_v$, and $N_p(t_0)$ have been specified in each case and $M_{eq}$ has been held constant in time ($\gamma = 1$). Varying only $q_i/q_v$ leaves $G$ and $N_p(t_0)$ unchanged but scales $M_{eq}$. This leaves $\bar{m}$ and $\delta S_i$ development unaffected [cf. Eqs. (27) and (30)] but scales the asymptotic value of $N_p(t)$ [cf. Eq. (29)] and hence the overall scale of the size spectrum (cf. Figs. 7a–d). Varying $T$ (or $P$) for fixed $q_i/q_v$ and parcel volume scales $G$ and $M_{eq}$ by approximately the same factor [through the change in $\rho g \tilde{q}_i$; cf. Eqs. (8) and (13)]. This leaves $N_p(t)$ unchanged asymptotically but changes the $\bar{m}$ evolution and shifts the center of the size spectrum and level of $\delta S_i$ accordingly (cf. Figs. 7e–h).

As noted following Eq. (29), and seen in Figs. 7i–l, the asymptotic solution becomes independent of $N_p(0)$, but varying $N_p(0)$ alters the approach to that solution by changing $\bar{m}(0)$ and also the time scale $\tau_s$ to reach the slow-growth solution. The case illustrated with smallest $N_p(0)$ in Figs. 7i,j takes of order $\sim 10^3$ s to reach the slow-growth solution (as judged from the linear growth of $\bar{m}$) but has not yet reached the asymptotic state by simulation’s end as in the other cases. For fixed conditions the slow-growth solution provides a time-varying upper bound [Eq. (29)] on $N_p$ or, by using Eq. (13), on number density $N_p$.

\begin{equation}
N_p \leq \frac{\rho(q_i - q_v)}{\Lambda n_0 G t}
\end{equation}

The ice/water-vapor surface tension $\sigma_{iw}$ enters the equations only through the length scale $a_k$. In turn $n_0$ scales as $a_k^2$ and $G$ as $a_k^{-2}$. The analytic treatment thus predicts that $\bar{m}$ will asymptotically scale linearly with $\sigma_{iw}$ [from Eq. (27)], $N_p$ will scale with $\sigma_{iw}^{-1}$ [from Eq. (29)], and $\delta S_i$ with $\sigma_{iw}^{2/3}$ [from Eq. (31)]. The simulation results with the more complete microphysics are again found to be consistent with these predictions (Figs. 7m–p). The quicker approach of $N_p(t)$ to power-law scaling with doubled $\sigma_{iw}$ (and slower approach with halved $\sigma_{iw}$) seen in Fig. 7m arises from the scaling of $\tau_s$ with $\sigma_{iw}^{-1}$ [from Eq. (16)].

Figures 2 and 4 provide sample simulation results when $M_{eq}$ varies in time ($\gamma < 1$) through plume mixing. They are again in excellent agreement with the theoretical predictions. In these cases $T$ and $q_i$ were taken the same inside and outside of the parcel, so the expansion of $M_p$ varies only $M_{eq}$ not $G$. The asymptotic crystal number varies as $N_p \sim t^{\alpha-1}$ and the change in $\bar{m}$ in different cases is through the change in $\Lambda$ produced by changing $\alpha$. The slow-growth solution depends on $M_{eq}$ and $G$ but not otherwise on how the changes are produced (i.e., whether driven by vertical motions, diabatic temperature change, mixing when $T$ and $q_i$ differ inside and outside the parcel, etc.).

b. Forcing on different time scales

The slow-growth solution is more generally encountered than the specific form of the forcing Eq. (33) might suggest. Slow variations (compared to time scale $\tau_s$) of nearly arbitrary form will to good approximation follow the $\alpha = 0$ solution for the current value of $M_{eq}(t)$. Equation (35) provides little additional constraint: if the forcing is of the form of Eq. (33) but with $\beta$ too large to satisfy Eq. (35), it can be brought into compliance simply by shifting the time origin $t_0$ and if $\beta$ is initially too small to satisfy Eq. (35), then the parcel will initially evolve according to the $\alpha = 0$ solution, reducing $\bar{m}(t)$ until Eq. (35) can be satisfied and the asymptotic behavior reached.

From Eq. (16) it is clear that forcing on time scales short compared to $\tau_s$ but still long compared to $\tau_s$ are of practical importance. Examples in contrail evolution include both gravity wave oscillations at late times and circulations about the aircraft trailing vortex cores at early times. Figure 8 compares parcel cases with and without a superposed “high-frequency” forcing. The “low-pass filtered” development matches that of the slow-growth solution with no oscillation (given sufficient particle size bin resolution; see appendix B), even when the oscillatory deviations become quite large $[\delta S_i$ oscillating in sign with an amplitude an order of magnitude larger than the low-pass
mean; $N_p(t)$ dropping in staircase fashion; and the PSD alternately narrowing and broadening).

The survival of the slow-growth solution under these conditions can be understood as follows. Decompose variables into low-pass filtered and fluctuating components, e.g., $\delta S_i = \langle \delta S_i \rangle + \delta S_i'$. We ignore the variation $G'$ relative to $\langle G \rangle$ since it is generally small, but $\delta S_i'$ and $f'$ may become large relative to $\langle \delta S_i \rangle$ and $\langle f \rangle$, respectively.
are (a) ice mass \((m_g)\); (b) crystal number; (c) size spectrum around 3 \(\times 10^3\) s, with the solid lines given every 60 s over a 600-s oscillation period.

From Eq. (10) an imposed oscillation in \(M_p\) leads to an oscillation in \(\delta S_i\). It is \(\delta S'_i\) that drives the spectrum fluctuations \(f'\) and from Eq. (3) the two are \(\pi/2\) out of phase \((df'/dt - \delta S'_i)\). Low-pass filtering Eqs. (1)–(3) and using Eq. (7), the resulting equations for the low-pass filtered variables are identical to their counterparts with no high-frequency component with the exception of an additional term, \(-\partial/\partial t' (G e^{-2/3} \delta S'_i f'))\). If \(\delta S'_i\) and \(f'\) are \(\pi/2\) out of phase, however, \(\delta S'_i f'\) vanishes, so that the behavior of the low-pass filtered variables is just the solution considered previously.

For small-amplitude fluctuations, \(N_p(t)\) is found to have just the decay predicted by the slow-growth solution with a small oscillation superposed. Since no crystal sources have been included this cannot remain the case for larger-amplitude oscillatory forcing that would imply \(N_p(t)\) increasing at times. Instead, a periodic width modulation of the spectrum occurs (Fig. 8), leading to a staircase structure for \(N_p(t)\). This does not change the mean crystal loss rate, but it can increase the numerical requirements for an accurate solution (appendix B).

c. Kinetic theory effects

Equation (4) is based on the continuum diffusion equation. When crystals are small enough relative to the molecular mean free path, corrections arise that can be estimated from kinetic theory and absorbed into a modified diffusion constant (see, e.g., Rogers 1976; Pruppacher and Klett 1997):

\[
D^* = D/(1 + \lambda_p/r) \quad \text{with} \quad \lambda_p = D \left( \frac{2\pi}{R_b T} \right)^{1/2}.
\]  

The form for \(\lambda_p\) is that given by Fukuta and Walter (1970). That used in CARMA has additional Knudsen number dependence, but only the basic behavior, rather than the precise form, concerns us here. The ice deposition (or accommodation) coefficient \(\beta_i\) is (along with its counterpart the condensation coefficient on liquid water) not well determined experimentally [e.g., see Tables 5.4 and 5.5 of Pruppacher and Klett (1997)]. Values chosen by different cirrus modelers also vary widely and significantly impact the crystal number densities produced by homogeneous freezing nucleation in cirrus simulations (e.g., Lin et al. 2002). Kinetic theory corrections are often considered important only for crystal/droplet formation, dropping out as radii grow. As we have seen with the Kelvin term, however, sometimes such effects can remain important even for later evolution. Figure 9 showing binned microphysics simulations varying \(\beta_i\), confirms that to be the case here as well.

For larger crystals and/or \(\beta_i\), the corrections in Eq. (41) can already be incorporated in the previous analysis as a slow time variation of \(G\) [e.g., with \(D^*\) evaluated at \(r = r_{mv}\) replacing \(D\) in Eq. (8)]. For small enough \(r\) and \(\beta_i\), the \(\lambda_p\) term dominates in Eq. (41) and \(G\) becomes approximately proportional to \(r, G = \tilde{G}\). The previous analysis can be repeated but with different scaling exponents appearing in some expressions and \(\tilde{G}\) replacing \(G\). Again there are slow-, fast-, and very-fast-growth regimes but with shifted boundaries between them: for

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6 The linearly increasing amplitude of the oscillation in \(\tilde{S}(t)\) in Fig. 8 is also predicted by Eq. (10) given the \(r^{-1}\) dependence of \(N_p\).

7 This may account for the differing conclusions on the effects of oscillations between Korolev (1995) and Celik and Marwitz (1999).
1.1, \(N_p(0) = 10^{10}, \alpha = 0,\) and bin mass ratio = 2.

\(M_p \sim t^\alpha\) slow growth requires \(\alpha < \frac{3}{2}\) and very fast growth \(\alpha > 3.\)

In the slow-growth regime for small \(\beta_i,\) Eq. (23) is replaced by

\[
\frac{d\phi}{dx} = \frac{\dot{Q}}{\Lambda} - \frac{1}{3} e^{-x/3} \dot{S} + e^{-2x/3},
\]

where

\[
\Lambda = \dot{G}^{-1} e^{2x/3} \frac{d\lambda}{dt}, \quad \dot{S} = \alpha/3 \delta S_t, \quad \text{and}
\]

\[
\dot{Q} = \dot{G}^{-1} e^{2x/3} \frac{dQ}{dt}
\]

are constant in time. The analog of Eq. (27) becomes

\[
\bar{m}(t) = \bar{m}(t_0) \left\{ 1 + \frac{2}{3} \left[ \frac{m_0}{\bar{m}(t_0)} \right]^{2/3} \bar{G} \lambda(t - t_0) \right\}^{3/2}
\]

and asymptotically in time

\[
N_p(t) \sim t^{-3/2}.
\]

The \(t^{-3/2}\) asymptotic decay for small \(\beta_i\) is apparent in Fig. 9.

Equation (42) may be integrated for the spectral shape leading to

\[
f(\xi, t) d\xi = 3d(\ln r) \gamma e^{3\gamma} N_p \frac{(r/r_m)^2}{(1 - r/r_m)^2 + 3\gamma} e^{-3\gamma(1 - r/r_m)}. \tag{46}
\]

As before, \(r_m = b(\gamma)r_m\), but with a different integral expression for \(b,\) and

\[
\Lambda = \frac{1}{b^2}, \quad \dot{S} = \frac{2}{b}, \quad \text{and} \quad \dot{Q} = \frac{-\gamma}{b^2}. \tag{47}
\]

d. Water clouds

The forms of the equations governing diffusional growth are the same for water droplets as for ice crystals. This makes the present analysis relevant to water clouds if the corresponding values for surface tension and latent heat and the full expression for \(G\) in Eq. (8) are used.

The treatment of the Kelvin effect is on much firmer footing for water droplets, although the omission of collection growth likely limits the regime of applicability of the analysis sooner than it does for ice. For droplets one must also consider solute effects. Given that the Kelvin effect has been shown to play a role even for droplets much larger than the characteristic radius for that effect \(a_k,\) one must consider whether solute effects might behave similarly and have to be included even for larger droplets. Including both Kelvin and solute effects to lowest order in \(1/r,\) the saturation term in the numerator of the growth Eq. (4) becomes (see, e.g., Rogers 1976)

\[
\delta S_t - \frac{a_k}{r} \left[ 1 - \frac{1}{3} \left( \frac{r^*}{r} \right)^2 \right], \tag{48}
\]

where \(r^*\) is the critical radius corresponding to the peak of the Köhler curve at which condensation nuclei become “activated.” Thus for droplets even modestly larger than \(r^*\) the leading solute effect [i.e., the \(r^*\) term in Eq. (48)] becomes small compared to the leading Kelvin effect term, as noted earlier by Srivastava (1991). Thus the treatment given here without solute effects should be applicable to water clouds provided \(r_m \gg r^*\) (i.e., given a clear separation between cloud droplets and haze particles). This may not be satisfied in cases with very large condensation nuclei; such complications remain an open question for the analysis.

If the slow-growth regime appears in water clouds it will arise first in conditions with higher number densities and lower velocity scales (e.g., boundary layer stratiform clouds). We are in agreement with Wood et al. (2002) on the importance of the mean parcel in-cloud residence time in determining whether the “ripening” process is significant and further have here identified \(\tau_r\) as the appropriate time scale for comparison and determined its dependence

\[8\] The approximate form given in Eq. (8) is not appropriate for the higher saturation pressures generally found in water clouds.
on other physical parameters. Wood et al. (2002) note that the ripening process might sometimes significantly affect autoconversion rates. We raise the possibility here that it may provide an upper bound on droplet number densities [e.g., Eq. (40)] that might sometimes govern, potentially affecting the radiative properties of the cloud as well.

Applying Eq. (40) in a way that can be compared with in situ measurements of cloud droplet number densities is problematic because the theory assumes a homogeneous parcel with known history while the measurements necessarily average over many parcels with potentially quite different histories. The limit [Eq. (40)] depends on the residence time of the parcel within the cloud, the local liquid water content \( q_L \), and to some extent on the prior vertical motion or dilution of that parcel on time scales comparable to \( \tau_\lambda \), since these can affect \( \Lambda \). Despite this difficulty we can at least make an order of magnitude estimate of whether spectral ripening may be playing a role in limiting observed number densities or whether the theoretical limit is far removed from observations. Assuming nominal stratocumulus values \( (T = 286 \text{ K}, \text{ height} = 400 \text{ m}, \text{ liquid water mixing ratio of } q_L = 0.2 \text{ g kg}^{-1}) \), taking \( \Lambda = 0.167 \) (as predicted for \( \alpha = 0 \)), and neglecting kinetic theory corrections, the number density limit applied to droplets becomes

\[
N_p \leq \frac{9.9 \times 10^5}{t},
\]

in units of per cubic centimeter for in-cloud residence time \( t \) in seconds. The estimate in Eq. (49) changes proportionally with \( q_L \) variations with temperature or altitude are secondary. The in-cloud residence time should scale with typical circulation turnover times in the boundary layer but might differ appreciably in magnitude since most parcels will spend much of their time outside of the core updraft or downdraft. Wood et al. (2002) used a parcel trajectory analysis and a simple turbulence representation to estimate probability distribution functions (PDFs) of in-cloud residence time for two cases: low or “fairly vigorous” turbulence (vertical wind standard deviations of 0.2 or 0.6 m s\(^{-1}\), respectively). They found mean in-cloud residence times for these cases of order 100 and 10 min, respectively, with standard deviations similar to the means in each case. Inserted into Eq. (49) these times provide number density estimates of 165 or 1650 cm\(^{-3}\). These are at least in the correct ballpark for observations of marine stratocumulus (e.g., Wang et al. 2009), suggesting that the limits on the number densities given by the present analysis may sometimes play a role. It is well known that as CCN concentrations increase, the fraction activated tends to decrease because of competition for available moisture during the activation process. The present work suggests that spectral ripening effects after activation may push this limit further so that increases in CCN concentration past a certain point will no longer lead to significant increases in cloud droplet number density. The relative role of these two mechanisms in setting limits on droplet concentrations in stratocumulus or stratus layers in different regimes might best be assessed quantitatively via LES with detailed microphysics, where the effects might be disentangled by suitable variation of microphysics parameters.

4. Summary and concluding remarks

We have reconsidered an old subject, the evolution of water droplet or ice crystal size distributions in the approximation that diffusional growth is the only microphysical process at work. Within a parcel defined by a fixed particle population, outside forcing (e.g., radiational heating, ascent, turbulent mixing into the parcel, etc.) affects the behavior essentially through its effect on the total water mass above saturation within the parcel \( M_{eq} \). As a function of this forcing we have identified different asymptotic behavior regimes, the boundaries between them, and the time scales involved. For slow enough growth of \( M_{eq}(t) \), a regime is eventually reached in which the PSD in logarithmic coordinates is of constant shape, particle numbers are continually lost through a surface tension–dependent spectral ripening process, and the mean particle mass grows linearly in time. At the level of the equations considered we have presented a full analytic solution for the microphysical development of a well-mixed parcel in this slow-growth regime.

In the limit of constant \( M_{eq} \) the slow-growth solution given here reproduces the classic LSW theory result for the PSD produced by Ostwald ripening of a solid solution in a diffusion-limited regime (Lifshitz and Slyozov 1961; Wagner 1961). The generalization given here to cases including different imposed external forcing may be of interest in the more heavily studied applications of Ostwald ripening to solid solutions, emulsions, and alloys. It must be noted, however, that the present analysis (like LSW) treats the diffusional growth of particles in a “mean field” approximation (i.e., particle–particle interactions are ignored). It is thus valid only in the limit of the particles occupying a small total volume fraction, a condition often violated in these other applications of

For reference, given the nominal conditions considered, these number densities lead to mean volume droplet radii of 6.6 or 3.1 \( \mu \text{m} \) and \( \tau_\lambda \sim 15 \) or 1.5 min respectively (i.e., about 15\% of the in-cloud residence time in each case).
Ostwald ripening. The inclusion of the effects of finite volume fraction has been a principal focus of research aimed at extending LSW theory (e.g., Marqusee and Ross 1984; Yao et al. 1993).

One advantage of a nontrivial exact solution is that it provides a useful venue for exploring numerical issues. In the present case the theoretical predictions have been verified with spectrally resolved parcel model simulations, and the numerical requirements for different levels of fidelity clarified in the process.

Several caveats to the physical applicability of the present analysis apply. The appearance of the slow-growth regime requires high particle number densities and/or long residence times. It is likely encountered in some contrails and contrail cirrus; simulations show the spectral ripening process playing an important role in crystal number losses at both early and late times (D. Lewellen et al. 2011, unpublished manuscript). It may be encountered in some low-turbulence, high-number density water or ice clouds as well. Estimates of the residence time and forcing-dependent number densities from the analytic solution are not inconsistent with the high end of number densities observed in cold upper-tropospheric cirrus or in marine stratocumulus (e.g., Krämer et al. 2009; Wang et al. 2009). More quantitative observational tests are complicated by the difficulties in determining mixing rates or residence times and, in the cirrus case, with problems in measuring small crystals (e.g., Jensen et al. 2009).

Since sedimentation sorting, aggregation, direct radiative heating of crystals, and nonspherical shapes are excluded from the analysis, particle sizes must be sufficiently modest (though they can still be much bigger than the Kelvin scale $a_k$). The slow-growth regime in particular relies on a simple balance involving diffusional growth and spectral ripening for the simple scaling behavior of the PSD. When other processes become dominant, completely different behavior regimes can arise [e.g., the simple scaling behavior of the PSD governed by sedimentation induced aggregation identified by Field and Heymsfeld (2003)].

At least within the high-number-density cores of contrails out to 1-h age there is evidence that ice crystals are predominantly small (a few microns in radius) and nearly spherical (Schröder et al. 2000). For much larger crystals, shapes become highly nonspherical and can be expected to quantitatively change the present analysis significantly, although the three qualitative growth regimes identified here should still be represented. A constant nonspherical shape (independent of size) could be handled simply by scaling $a_k$ with an appropriate shape factor in the analysis. The sensitivity would then be entirely analogous to the changes seen in varying $\sigma_{siv}$. In practice, however, the shapes will almost certainly be size dependent, changing the $r$ dependence of the growth kernel [Eq. (7)]. For some simple choices of dependency the analysis could still be carried through in similar fashion (as was done for example in section 3c with the change in growth kernel in the limit of very small accommodation coefficient). Not having a compelling candidate for this dependency, however, the treatment of nonspherical crystals in this analysis must remain an open question for now.

Even when diffusional growth dominates, there is a sizable degree of uncertainty in applying the analytic solutions in the slow-growth regime because of the sensitivity to the accommodation coefficient coupled with its uncertain determination for both ice and water and, in the ice case, the uncertainty in the treatment of surface energy (Kelvin) effects. If the asymptotic crystal/droplet populations in the slow growth regime could be measured in a controlled setting in the laboratory, this sensitivity might possibly be used to advantage. Comparison with the analytic solution might then provide some useful constraints on the value of the accommodation coefficients and treatment of the Kelvin effect. A measurement of integrated particle numbers over long times with a smooth asymptotic behavior has potential advantages over instantaneous measurements of particle size growth rates, although uncertainties in the measurements and from processes not included in the analysis might render the comparison ineffective.

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APPENDIX A

The Slow-Growth Regime Size Spectrum

We wish to integrate Eq. (23) and impose the constraints of Eq. (19). Defining $\gamma = 1 - \alpha = -\tilde{Q}/\Lambda$ from Eq. (34) and substituting $y = e^{\gamma z}$ these become

$$\frac{dy}{dz} = \frac{1}{y} \left[ -3\gamma^2 - 2(\tilde{S}/\Lambda)y + 3/\Lambda \right], \quad (A1)$$

$$3 \int_0^\infty dy \, y^{-1} e^\phi = 1, \quad (A2)$$

$$3 \int_0^\infty dy \, y^2 e^\phi = 1. \quad (A3)$$
For some $a$, $b$, $c$ the denominator of Eq. (A1) can be factored:
\[ f_1(y) = y^3 - (S/\Lambda)y + 1/\Lambda = (y - a)(y - b)(y - c). \]  
(A4)

The nature of the roots can be deduced as follows. In the limit $y \to -\infty$, $\frac{d\phi}{dy}$ behaves as $-3\sqrt{y}$ so that the integrand in Eq. (A3) scales as $y^2e^\phi \sim y^{2-3\gamma}$. Thus Eq. (A3) cannot be satisfied for $y \to -\infty$; the integral must be cut off at some finite $y$. Consistency requires one of the roots (say $b$) in Eq. (A4) to be real and positive, in which case $y = b$ will represent the upper end of the size spectrum. We can determine whether $y = b$ is a single, double, or triple root of $f_1$. Integrating Eq. (A1) as $y \to b$ assuming a single root one finds the spectrum $e^\phi$ nonanalytic as $y \to b$; a double or triple root insures analyticity. A double root requires
\[ f_1(b) = \frac{df_1(b)}{dy} = 0. \]  
(A5)

A triple root requires in addition that $d^2f_1(b)/dy^2 = 0$, which forces $b = 0$ and is therefore ruled out. With $b$ being a double root of $f_1$, Eqs. (A4) and (A5) then require
\[ \Lambda = 1/2b^3, \quad S = 3/2b, \]  
(A6)

and $f_1(y) = (y + 2b)(b - y)^2$. From these Eq. (A1) becomes
\[ \frac{d\phi}{dy} = \frac{3}{y} \left[ -\gamma y^3 - 2b^2y + 2b^3 \right]. \]  
(A7)

This can be integrated by the method of partial fractions and exponentiated to give
\[ e^\phi = \frac{c_0b^3}{(y + 2b)^{1+4\gamma/3}(b - y)^{2+5\gamma/3}}e^{-\gamma(y^2 - y)}. \]  
(A8)

The integration constant $c_0$ and parameter $b$ are constrained by Eqs. (A2) and (A3), where the integrations run from 0 to $b$. Define moments of the spectrum
\[ \kappa_n = \int_0^b dy y^n e^\phi = c_0 b^{1+n-3\gamma} \xi_n(y), \]  
(A9)

where
\[ \xi_n(y) = \int_0^1 dz \frac{z^{n+3}}{(z + 2)^{1+4\gamma/3}(1 - z)^{2+5\gamma/3}}e^{-\gamma(1 - z)}. \]  
(A10)

Solving Eqs. (A2) and (A3) for $b$ and $c_0$ gives
\[ b = \left[ \frac{\xi_{-1}(\gamma)}{\xi_2(\gamma)} \right]^{1/3}, \]  
(A11)
\[ c_0 = \frac{b^{3\gamma}}{3\xi_{-1}(\gamma)}. \]  
(A12)

We can give a closed-form expression for $c_0$ while clarifying how particle loss appears in the present formulation. Equation (3) for the PSD does not explicitly include any loss term; evaporating members are simply pushed to ever smaller sizes. Physically, below some small cutoff [defined at $\xi = \xi_-$ in Eqs. (1) and (2)] the crystals or droplets evaporate away and are removed from the spectrum. Differentiating Eq. (1), using Eq. (3), and integrating by parts, we have
\[ \frac{dN_p}{dt} = \int_{\xi_-}^{\infty} d\xi \frac{\partial f}{\partial \xi} \frac{\partial \xi}{\partial t} = \int_{\xi_-}^{\infty} d\xi \frac{\partial \xi}{\partial t} \frac{d\xi}{dt} = \left[ \frac{d\xi}{dt} f \right]_{\xi=\xi_-}. \]  
(A13)

Using Eq. (7) in Eq. (A13), equating with the derivative of Eq. (21), multiplying by $e^{\lambda - \gamma/\lambda}$, and using various definitions this gives
\[ \tilde{Q} = \left[ (y^2 - 2S - y^3)e^\phi \right]_{y=\xi_-}. \]  
(A14)

Using Eq. (A8) to evaluate Eq. (A14) for $y_\gamma \ll 1$, $b$ (the physical case of interest) the result, $\tilde{Q} = -c_0^22^{-1-4\gamma/3}b^{-3-3\gamma}e^{-\gamma}$, proves independent of the specific choice of $y_\gamma$, as required to be consistent with Eq. (28) and as promised in the main text. Equating this determination of $\tilde{Q}$ with that from $\tilde{Q} = -\gamma \Lambda$ [cf. Eq. (34)] and using Eq. (A6), one can solve finally for $c_0$:
\[ c_0 = 4\gamma e^{\gamma}(b^32^{4/3})^{\gamma/3}. \]  
(A15)

Similarly using Eq. (A8) for $y_\gamma \ll 1$, $b$ to evaluate the contributions to the normalization integrals (A2) and (A3) from integrating between 0 and $y_\gamma$ one finds them to be less than $y_\gamma$ and $y_\gamma^b$, respectively (i.e., negligible contributions to the full integrals in the $y_\gamma \ll 1$ limit). This justifies the promise in the main text that extending the integration limits in Eq. (1) and (2) from $\xi_-$ to $-\infty$ [as was effectively done in writing Eqs. (21) and (22)] leads to negligible error.

We have checked numerically that Eq. (A15) is satisfied by $b$ and $c_0$ as given in Eqs. (A11) and (A12), and through differentiation that Eq. (A8) satisfies Eq. (A1). For a chosen $\gamma$ the solution for the spectrum is unique: integrating Eq. (A1) leaves four constants ($c_0$, $\tilde{Q}$, $S$, and $\Lambda$) that are fixed by the constraints from the crystal loss given above, Eqs. (34), (A2), and (A3). Thus the validity of the
argument leading to the form of $f_1$ has effectively been checked a posteriori.

Finally, one can derive a recursion relation for the moments $\kappa_n(y)$. Multiplying Eq. (A7) through by $y^{n+1} e^{\psi} (y + 2b)(b - y)^2$, integrating from 0 to $b$, and using integration by parts to transform the $d\psi/dy$ terms, one obtains after some algebra

$$(n + 3 - 3\gamma)\kappa_{n+2} = 2b^2(n + 3)\kappa_n - 2b^3(3 - n)\kappa_{n-1}. \tag{A16}$$

Equations (A2) and (A3) give $\kappa_{-1} = \kappa_2 = \frac{1}{3}$. Choosing $n = 0$ in Eq. (A16), this gives an expression for $\kappa_0$ and hence for $c_s$:

$$c_s = \frac{1}{3b^2} (1 - \gamma + 2b^3). \tag{A17}$$

This is in agreement with Eq. (36). Thus we have represented the PSD and $Q$, $S$, and $\Lambda$ entirely algebraically in terms of the imposed $\gamma$ and the single parameter $b(\gamma)$ given by Eq. (A11). The latter is plotted in Fig. 5.

As defined, $y$ is directly related to the crystal (or droplet) radius $r$:

$$y = e^{x/3} = e^{(6-\Lambda)x/3} = \left(\frac{m}{m_v}\right)^{1/3} = \left(\frac{r}{r_{mv}}\right)^{1/3}, \tag{A18}$$

where $r_{mv}$ is the mean volume radius. Using Eqs. (A8), (A15), and (21) in Eq. (A18) this then leads directly to the form of the PSD given in Eq. (37).

**APPENDIX B**

**Numerical Issues for Binned Microphysics Schemes**

A nontrivial analytic solution provides a convenient testing ground for addressing numerical requirements.

Figures B1 and B2 compare results for different ice bin resolutions with the analytic results for a case in the slow-growth regime. For this and other cases, the correct quantitative scaling exponents and qualitative behavior overall are found even for quite modest bin resolution. Predictions for other variables and the PSD smoothly approach the theoretical predictions as the bin resolution is refined. At coarser resolution the inability to resolve the steep falloff at $r_{mx}$ leads directly to the overprediction of $\Lambda$ and hence $m$ and the corresponding [given fixed $M_p(t)$] underprediction of $N_p(t)$. Not smoothly resolving...
the spectral peak is the cause of the saw teeth seen in Fig. B1b, arising as the spectral peak passes in time over successive size bins.

In the fast-growth regime the PSD progressively narrows in time, but finite bin resolution sets a width limit in simulations (cf. Fig. 2d). The effect on simulation fidelity is minor because the most important variables are otherwise constrained to the correct behavior: \( N_p \) constant, \( M_p(t) = M_{eq}(t) \), and \( \delta S \) constrained by Eq. (14). The numerical width limit can become a more serious problem when an oscillatory forcing is superposed on otherwise slow-growth conditions as in Fig. B3. Physically the PSD alternately broadens and narrows, with the crystal loss rates increasing or vanishing accordingly. For insufficient bin resolution some of the narrowing is prevented, leading to overprediction of crystal losses. The resolution needs increase with the amplitude of oscillations of \( S(t) \). From Eq. (14) this rises with increasing amplitude or frequency of the forcing oscillation and over time as \( N_p \) drops.

Figure B4 illustrates the effects of varying the time step. The most rapid fractional changes in crystal mass are for the smallest crystals, which then require the smallest time steps. As the time step is increased, the evaporation of the smallest crystals is unphysically slowed, leading to the upturning at small \( r \) of the simulated PSD from the theoretical linear falloff. This is, however, relatively harmless since the rate of crystal loss is set by other dynamics and therefore unaffected. The critical limit is \( dt < \tau_r \); otherwise, as seen in the figure, the \( M \) and \( N \) development can be severely impacted.

As noted in the discussion leading to Eq. (A15) the crystal loss rate is independent of the choice of smallest mass cutting off the spectrum, and this is confirmed in the simulations. Moreover, the dynamics can be captured quite well with only a modest window of size bins, provided it spans the spectral peak. For efficiency our microphysics implementation includes an option allowing the bin window to slide up or down in size range locally in time and space as best follows the dynamics (D. Lewellen et al. 2011, unpublished manuscript). The example in Fig. B4 shows that for bin mass ratio of 2 a local bin window of as few as 10 bins is more than adequate to accurately follow the development in the slow-growth regime over long times (and even fewer suffice in the fast-growth regime).
REFERENCES


