Parameterization of Cloud Drop Activation Based on Analytical Asymptotic Solutions to the Supersaturation Equation

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ABSTRACT

Toward improving parameterization of cloud droplet activation in cloud and climate models, the integro–differential equation for supersaturation is solved analytically for the algebraic size spectrum of the cloud condensation nuclei (CCN) that is equivalent to the lognormal spectrum. The analytical solutions are obtained for four limiting cases that are combinations of two different values of the updraft vertical velocity (small and large) and two different values of the condensation coefficient that correspond to pure and polluted cloud drops. The characteristics of the CCN can vary within each limit. Thus, these four limits and interpolation among them cover the vast majority of cloudy conditions. Analytical expressions are obtained for the time of CCN activation, maximum supersaturation, and the concentration of activated droplets.

For small updraft vertical velocities, these quantities are the products of the power laws by six variables: CCN concentration, mean radius, soluble fraction, vertical velocities, surface tension, and condensation coefficient. At large updraft vertical velocities, the activation time and maximum supersaturation are the products of the power laws of only two variables—CCN concentration and vertical velocity—and are independent of the CCN physicochemical properties. The first limit is a generalization of the Twomey power laws, with Twomey's coefficient $C_T$ and index $k$ expressed via CCN physicochemical properties; the other three limits are new. The accuracy and regions of validity of these limits are determined by comparison with the exact numerical solution to the supersaturation equation. These solutions can be used for parameterization of drop activation in cloud and climate models and for control of numerical solutions. An advantage of this method is that it does not require running parcel models, and the drop concentrations can be obtained from lookup tables or as simple interpolation among the limiting solutions for the instantaneous model parameters.

1. Introduction

The indirect aerosol effects on radiative forcing (e.g., Twomey 1977; Albrecht 1989) remain an outstanding uncertainty in simulations of global climate change (Foster et al. 2007). The importance of these effects has motivated development of improved schemes of drop activation for climate models. Drop activation schemes used prior to the 1990s were based mostly on the Twomey (1959) power law for concentration of activated cloud condensation nuclei (CCN), $N_{dr} = C_T s_m^k$, where $C_T$ and $k$ are the empirical coefficient and index and $s_m$ is the maximum supersaturation in a cooling air parcel. The drawbacks of Twomey’s parameterization are that $N_{dr}$ is unlimited and can exceed CCN concentration $N_a$ and parameters $C_T$ and $k$ are not related to CCN physicochemical properties.

A substantial revision and intensive development of new cloud drop activation parameterizations began in the 1990s (see reviews in Charlson et al. 2001; Lohmann and Feichter 2005; McFiggans et al. 2006; Svenningsson et al. 2006; Foster et al. 2007). In particular, Kulmala et al. (1993), Shulman et al. (1996), and Laaksonen et al. (1998) generalized Köhler’s theory for the presence of soluble trace gases and slightly soluble substances. Khvorostyanov and Curry (2007, hereafter KC07) generalized Köhler’s theory for the case of small soluble...
fraction and found analytical solutions to the Köhler equation and for the CCN wet size spectra at aerosol hygroscopic growth, as well as for activity spectra at drop activation without assumption of high dilution. New methods of calculating CCN activation were developed based on lognormal (e.g., Ghan et al. 1993, 1995; Feingold et al. 1994; Fountoukis and Nenes 2005) and algebraic (Cohard et al. 1998, 2000) CCN size and activity spectra, which limited the number of activated drops so that \( N_{\text{dr}} \ll N_a \). Khvorostyanov and Curry (2006, hereafter KC06) and KC07 suggested another algebraic size and activity spectra of dry and wet CCN equivalent to the corresponding lognormal spectra, providing a generalization of the power laws that reconciled the lognormal and algebraic approaches; this approach was used in both bin and bulk cloud models (Morrison and Grabowski 2007, 2008). Parameterizations of drop nucleation have been developed using results from parcel models that are fit to various empirical functions of a few nondimensional parameters (Abdul-Razzak et al. 1998, hereafter AGR98; Abdul-Razzak and Ghan 2000, 2004; Fountoukis and Nenes 2005; Ming et al. 2006). Saleeby and Cotton (2004) developed another approach consisting of lookup tables compiled from many runs of a parcel model.

The necessity of running parcel models many times for compiling lookup tables or tuning the parameterizations makes the procedure time consuming and does not easily reveal the relevant physical relationships. An alternative approach was suggested in Khvorostyanov and Curry (2008, hereafter KC08), who derived the integro–differential equation for supersaturation using the algebraic size and activity spectra from KC06 and KC07 as a starting point. KC08 provided a numerical solution of this equation for the time of CCN activation \( t_m \), maximum supersaturation \( s_m \), and concentration of activated droplets \( N_{\text{dr}} \) as functions of the physicochemical properties and size spectrum of the CCN and the vertical velocity \( w \).

A deficiency of all numerical solutions is that they do not demonstrate the general analytical dependencies of the drop activation on various parameters. The search for analytical solutions for \( t_m \), \( s_m \), and \( N_{\text{dr}} \) is desirable because they would allow the following applications: (i) control for the accuracy of various numerical solutions, especially in the cases where the general dependencies are unclear and spread of the parcel model runs can be significant (e.g., at strong variations of the condensation coefficient or surface tension); (ii) development of newer parameterizations based on the characteristic parameters occurring in these solutions; (iii) construction of the general solutions by interpolation among the particular analytical solutions; and (iv) design of laboratory and field experiments.

It has been known since the pioneering work by Twomey (1959) that the supersaturation equation does not allow general analytical solutions, even with the simplest power-law activity spectrum, unless certain simplifications are made. The situation becomes even more complicated when more complex activity spectra are used (e.g., lognormal or algebraic) because of the impossibility of exact analytical evaluation of the characteristic integrals of this problem. Twomey considered droplet growth with the condensation coefficient \( \alpha_c \sim 1 \), that is, in the diffusion regime of droplet growth. Subsequent laboratory and field experiments and theoretical works showed that \( \alpha_c \) can be as low as \( 10^{-5} \); that is, droplets grow in the kinetic regime (Pruppacher and Klett 1997, hereafter PK97; Seinfeld and Pandis 1998; Feingold and Khvorostyanov 1997; Feingold and Chuang 2002; Chuang 2003). However, parameterizations of drop activation considered mostly the cases with \( \alpha_c \sim 1 \) with some numerical estimates of the effects of \( \alpha_c \) (Nenes et al. 2002; Ming et al. 2006), but, to our knowledge, the general analytical dependencies of \( t_m \), \( s_m \), and \( N_{\text{dr}} \) on \( \alpha_c \) have not been derived.

Here, the analytical solutions to the general integral supersaturation equation from KC08 are obtained for four limiting cases: small and large updraft vertical velocities and two values of the condensation coefficient, corresponding to the diffusion and kinetic regimes of droplet growth. These solutions express the activation time, maximum supersaturation, and concentration of activated droplets as functions of the vertical velocity and physicochemical characteristics of the CCN including explicit dependencies on the condensation coefficient and surface tension.

### 2. Basic equations

The basic equations include the size spectrum of dry aerosol, the Köhler equation for dilute solutions, an equation for droplet radius growth rate accounting for kinetic effects, and the integro–differential equation for supersaturation. The size spectrum of dry aerosol \( f_{\text{ad}}(r_{\text{ad}}) \) by the dry radii \( r_{\text{ad}} \) is described by the lognormal distribution with the aerosol number concentration \( N_a \), dispersion \( \sigma_d \), and mean geometric radius \( r_{\text{ad}} \) related to the modal radius \( r_m = r_{\text{ad}} \exp(-\ln^2\sigma_d) \). It was found in KC06 and KC07 that this lognormal size spectrum is equivalent to the algebraic size spectrum. CCN hygroscopic growth is described by the Köhler equation for supersaturation \( s = \frac{\rho_v - \rho_{\text{water}}}{\rho_{\text{water}}} \) in dilute solutions (PK97):

\[
s = \frac{A_k}{r} - \frac{B}{r^2},
\]  

(2.1)
where $r$ is the radius of an aerosol particle, $A_k$ is the Kelvin curvature parameter, and $B$ describes the Raoult effect of soluble fraction of CCN:

$$A_k = \frac{2M_k \xi_{sa}}{RT\rho_w}; \quad B = \frac{3\Phi_m M_w}{4\pi M_g}\left[1 + \frac{\xi_{sa}}{r_{ad}^{2(1+\beta)}}\right].$$

(2.2)

Here $\rho_v$, $\rho_{vo}$, and $\rho_w$ are the densities of vapor, saturated vapor, and water; a list of notations is given in appendix A. The parameterization of $B$ as the power law by the dry radius $r_{ad}$ [second equation for $B$ in (2.2)] is described in Khvorostyanov and Curry (1999), KC06, and KC07. The parameters $b$ and $\beta$ in (2.2) depend on the physicochemical properties of the soluble part of an aerosol particle and are given in appendix A. The values $\beta = 1/2$ and $\beta = 0$ correspond to the soluble fraction proportional to the volume (internally mixed aerosol) and to the surface area (e.g., a dust particle covered by a soluble film).

It was found in KC06 and KC07 that the algebraic and lognormal size spectrum correspond to the algebraic and to the surface area (e.g., a dust particle covered by a soluble film).

$$\phi_s(s) = k_{so} C_0 s^{k_{so} - 1}(1 + \eta_0 s^{k_{so}})^{-2}.$$  

(2.3)

Here $s_0$ is the mean geometric supersaturation and $\sigma_s$ is the supersaturation dispersion:

$$s_0 = r_{do}^{-(1+\beta)} \left(\frac{4A_k}{27b}\right)^{1/2}; \quad \sigma_s = s_d^{(1+\beta)}.$$  

(2.4)

The index $k_{so}$ is an analog of the Twomey (1959) power index $k_f$ and is a generalization for $s$ space that accounts for the soluble fraction of the index introduced in Ghan et al. (1993) for the size spectrum, and parameters $C_0$ and $\eta_0$ are expressed via CCN microphysical properties:

$$k_{so} = \frac{4}{\sqrt{2\pi} \ln \sigma_s} = \frac{4}{\sqrt{2\pi}(1+\beta) \ln \sigma_d};$$

(2.5)

$$C_0 = N_a s_0^{-k_{so}} = N_a \left(\frac{27b}{4A_k}\right)^{k_{so}/2} (r_{do})^{k_s(1+\beta)}; \quad \eta_0 = s_0^{-k_{so}}.$$  

(2.6)

The lognormal spectrum $f_{sa}(r_{ad})$ can also be approximated with good accuracy by the algebraic spectrum (2.3) with the replacements $s \rightarrow r_{ad}$, $s_0 \rightarrow r_{do}$, $k_{so} \rightarrow k_{ad} = 4/(\sqrt{2\pi} \sigma_s)$ (KC06, KC07).

The equation for droplet radius growth rate $\dot{r}_d$ accounts for the kinetic correction $\xi$:

$$\dot{r}_d = \frac{c_3 s}{r_d + \xi}$$

where

$$c_3 = \frac{D_v \rho_w}{\rho_g \Gamma_1}; \quad \xi = \frac{4D_v}{\alpha_c V_w}.$$  

(2.7)

in which $D_v$ is the water vapor diffusion coefficient, $\Gamma_1$ is the psychrometric correction to the growth rate due to latent heat release, $V_w$ is the thermal speed of water vapor molecules ($\Gamma_1$ and $V_w$ are defined in appendix A), and $\alpha_c$ is the condensation coefficient. The estimates for the typical conditions and detailed calculations in KC08 show that, if $\alpha_c \sim 1$, then the kinetic correction $\xi \ll r_d$, can be neglected in (2.7), and the growth of newly activated droplet proceeds in a diffusion regime. If $\alpha_c \approx 0.01$, then $\xi \gg r_d$ in (2.7), and droplet growth is kinetic (free molecular).

If an air parcel is rising at vertical velocity $w$ and crosses the condensation level, drops begin to activate, supersaturation increases and reaches its maximum value $s_m$ at the time $t_m$ when activation ceases, and the concentration of activated drops is $N_{act}(s_m)$. Using the above assumptions for dry aerosol size spectra, particle activation, and droplet growth, KC08 derived the nonlinear integral–differential equation for the integral supersaturation

$$y(t) = \int_{t_0}^t [y(t)] d\tau.$$  

(2.9)

where $y(t)$ describes droplet growth after activation:

$$[1 + y(t)] y''(t) = c_1 w - y'(t) [y_{act}(t)]^{-1}.$$  

(2.8)

The parameter $c_1$ is given in appendix A, $y_{act}(t) = [4\pi D_v N_{act}(t)]^{-1}$ is introduced in KC08 as the effective supersaturation relaxation time at the initial stage of condensation, $r_{act}(t) = J_0(t)$ is an activation radius, and $J_0(t)$ is the integral:

$$J_0(t) = r_{act}(t) = \frac{k_{so}}{s_0^{k_{so}}} \int r_{ef}(t, t_0) \left[\frac{[y(t_0)]^{k_{so}} - y''(t_0)}{1 + \eta_0[y(t_0)]^{k_{so}}}\right]^2 dt_0,$$  

(2.9)

where $r_{ef}(t, t_0)$ describes droplet growth after activation time $t_0$ to the time $t_m$

$$r_{el}(t, t_0) = \frac{\left\{ [r_d(t_0) + \xi]^2 + 2c_3 [y(t) - y(t_0)] \right\}^{1/2} - \xi^2}{\left\{ [r_d(t_0) + \xi]^2 + 2c_3 [y(t) - y(t_0)] \right\}^{1/2}}.$$  

(2.10)

A solution to (2.8) can yield the values of $s_m$, time $t_m$, and $N_{act}(s_m)$. As noted by Twomey, an analytical solution of the complicated nonlinear integral–differential equation (2.8) is a formidable task. Numerical solutions of the supersaturation equation in the parcel models may lead to substantial differences among various models due to different approximations in the description of physical processes (e.g., various choices of $\alpha_c$), finite
difference schemes, and numerical techniques (e.g., Lin et al. 2002).

Therefore, instead of numerical calculation of \( s_m \) and \( N_{dr}(s_m) \), we apply the approach developed by Twomey (1959) and Sedunov (1967, 1974). It is based on approximation of the rhs of (2.8) to obtain analytically lower and upper bounds for solutions to (2.8). It will be shown that these bounds appear to be sufficiently close (differing by less than 10\%–20\%), and any solution between these bounds gives \( t_m, s_m, N_{dr} \) with sufficiently small errors (~5\%–10\%) that do not exceed the typical errors in other parameterizations. The advantage of the analytical approach is that it provides the general analytical dependences of solutions on the input parameters that are often hidden in numerical solutions.

The simplest solution to (2.8) can be obtained by substituting the first term \( c_1w \) on the rhs into the second term—that is, to search for a solution of the form \( y(t) = s(t) = c_1wt \). Linear functions of this form were used by Twomey (1959) and Sedunov (1967, 1974) to obtain the analytical bounds. We transform the second term on the rhs in (2.8) using the linear approximation for \( y(t) = s(t) \); then

\[
y'(t) = s(t) = a_m t, \quad y''(t) = a_m, \quad y(t) = \frac{(a_m/2)t^2}{2}. \tag{2.11}
\]

The form of \( a_m \) will be specified below; the choice (2.11) is sufficiently general and allows one to obtain the upper and lower bounds of \( t_m \) and \( s_m \). Substitution of (2.11) into (2.8) yields

\[
[1 + y'(t)]^{-1}y''(t) = c_1w - \Lambda(t)k^{s_m^{1/2}}. \tag{2.12}
\]

where

\[
\Lambda(t) = c_3N_{dr}s_0^{-k_{a_m}} \frac{k_{a_m^{3/2}}}{a_m^{k_{a_m^{3/2}}}} J_3(t), \tag{2.13a}
\]

\[
= c_3N_{dr}s_0^{-k_{a_m}} \frac{k_{a_m^{3/2}}}{a_m^{k_{a_m^{3/2}}}} [2J_2(t)], \tag{2.13b}
\]

with \( c_5 \) defined in appendix A and \( J_2 \) and \( J_3 \) the dimensionless integrals:

\[
J_2(t) = \int_0^1 \left[ \frac{\left[ U_s + (x - x^2) \right]^{1/2}}{\left[ U_s + (1 - x^2) \right]^{1/2}} \right]^2 \frac{x^{k_{a_m - 1}}}{(1 + \lambda_x^{x^{k_{a_m^{3/2}}}})} dx \tag{2.14a}
\]

and

\[
J_3(t) = 2J_2(t) \tag{2.14b}
\]

where \( x = t/t_0, z = x^{1/2} \), \( \lambda_x \) is the nondimensional parameter

\[
\lambda_x = \left( \frac{a_m t^{k_{a_m}}}{s_0} \right)^{k_{a_m}} = \left( \frac{3\sqrt{2} a_m t^{b/a_0} \lambda_x^{3/2}}{N_{a_m}^2} \right)^{k_{a_m}}. \tag{2.15a}
\]

and

\[
U_s = \frac{[r_{a_0}(t_0) + \xi]^2}{c_5 a_m^2 t^2}; \quad V_s = \frac{\xi}{(c_5 a_m)^{1/2}}. \tag{2.15b}
\]

The time \( t_m \) when supersaturation in the parcel reaches a maximum \( s_m \) is determined from the condition \( s'(t_m) = 0 \) or \( y'(t_m) = 0 \); then the lhs of (2.12) is zero and we obtain an algebraic expression

\[
t_m = \left[ c_1w/\Lambda(t_m) \right]^{1/(k_{a_m^{3/2}})}. \tag{2.16}
\]

After \( t_m \) is determined, it can be substituted into \( \Lambda(t_m) \) in (2.12), which can be integrated neglecting \( y' \ll 1 \) on the left-hand side, and we obtain a time-dependent solution for \( s(t) \):

\[
y'(t) = s(t) = c_1w \left[ 1 - \frac{1}{k_{a_m} + 3} \left( \frac{t}{t_m} \right)^{k_{a_m^{3/2}}} \right]. \tag{2.17}
\]

where we used (2.16) to express \( \Lambda(t_m) \) via \( t_m \). Equation (2.17) describes, with good accuracy, the time evolution of supersaturation to the time \( t_m \) and slightly beyond and gives \( s_m \) at the time \( t_m \):

\[
y'(t_m) = s_m(t_m) = c_1w \left[ 1 - \frac{1}{k_{a_m} + 3} \right] = \alpha_{k1} c_1w, \tag{2.18}
\]

where

\[
\alpha_{k1} = \frac{k_{a_m} + 2}{k_{a_m} + 3}. \tag{2.19}
\]

Equations (2.16) and (2.18) form a system of equations that allows for evaluation of \( t_m \) and \( s_m \) in various particular cases when the integrals \( J_2 \) and \( J_3 \) are specified. The asymptotic expressions for the integrals \( J_2 \) and \( J_3 \) will be obtained below and will allow us to derive four limits of the activation process, depending on \( \lambda_x \) and \( \alpha_c \).

The lower and upper bounds for \( t_m, s_m, N_{dr} \) can be evaluated using (2.11), \( y'(t) = s(t) = a_m t \), and (2.18) to relate \( s_m \) and \( t_m \). This is illustrated in Fig. 1, showing upper and lower bounds for \( s(t) \). It is usually presented as a schematic diagram [e.g., Fig. 1 in Twomey (1959) or Fig. 13–3 in PK97], but it is plotted here based on calculations of all curves using (2.17), (2.20), and (2.21).
Twomey (1959) approximation
gles); and the Sedunov (1967, 1974)
circles), yielding a lower bound for the integrals and an upper
simpler] on the right-hand side of the supersaturation
bound for ammonium sulfate),
s
area under the curve DCB equal to
overestimates the integral
J
parabolic analytical solution (2.17) (solid circles); linear
approximation \( s_m(t) = c_1 w t \) (2.20) yielding the upper bound
for integrals \( J_2 \) and \( J_3 \) and lower bound for \( s_m \) (diamonds); the
Twomey (1959) approximation \( s_m(t) = c_1 w (t - t_m) + s_m \) (triangles); and the Sedunov (1967, 1974) \( s_m(t) = s_m(t_m) \) (2.21) (open
circles), yielding a lower bound for the integrals and an upper
bound for \( s_m, t_m \). The last two straight lines intersect parabola
(2.17) at \( s_m \). Calculations are performed with the input parameters
\( N_a = 500 \text{ cm}^{-3}, r_{d0} = 0.05 \mu \text{m}, \alpha_d = 1.8, \beta = 0.5 \) (soluble fraction is
proportional to the volume), \( b = 0.25 \) (50% of soluble fraction of
ammonium sulfate), \( T = 10^\circ \text{C}, \) and \( p = 800 \text{ hPa} \).

It was shown by Twomey (1959) and Sedunov (1967, 1974) that a choice
\[
a_m = c_1 w; \quad s_m(t) = c_1 w t \tag{2.20}
\]
overestimates the integral \( J_0 \) [of the type (2.9) but simpler] on the right-hand side of the supersaturation
equation of the type (2.8) because the straight line \( s_m(t) \)
(DE in Fig. 1) lies above the exact curve \( s(t) \) and the area
below \( s(t) \) is less than the area under the curve DCB equal to \( J_0 \). Therefore, (2.20)
underestimates \( s_m \) and yields Twomey's lower bound
for \( s_m \). Twomey showed that another approximation
\( s_{un,T}(t) = c_1 w (t - t_m) + s_m \) underestimates the \( J_0 \) be-
cause the straight line \( s_{un,T}(t) \) (AC in Fig. 1) lies below
\( s(t) \) and the area of the triangle ABC is less than the
area below \( s(t) \) equal to \( J_0 \). Therefore, using \( s_{un,T}(t) \)
overestimates \( s_m(t_m) \) and gives the upper bound for \( s_m \).
Sedunov (1967, 1974) proved that another choice shown in
Fig. 1,
\[
a_m = s_m/t_m; \quad s_{un,s}(t) = s_m(t/t_m), \tag{2.21}
\]
also underestimates the rhs of (2.8) and gives an upper
bound for \( t_m \) and \( s_m \); this bound is closer to the exact
solution because the line \( s_{un,s}(t) \) (DC) lies above \( s_{un,T}(t) \)
(AC) and better approximates \( J_0 \). Owing to the coeffi-
cient \( \alpha_k < 1 \), \( s_m \) calculated with (2.18) is lower than
point E and much closer to point C; thus (2.18) will be
used for evaluation of the bounds.

The integral \( J_0 \) in (2.9) here and \( J_2 \) and \( J_3 \) are much
more complicated than \( J_0 \) estimated by Twomey and Sedunov, which corresponds to the first limit considered
in section 3, but the same is valid for these integrals in
the other limits: (2.20) overestimates \( s(t) \) and \( J_0 \) and
yields a lower bound for \( s_m \), whereas (2.21) underesti-
mates \( s(t) \) and \( J_0 \) and yields an upper bound for \( s_m \). This
is illustrated in Fig. 1 and will be proven for all four
limits in subsequent sections.

The drop concentration can be calculated from (2.3)
with \( s = s_m \), and can be written as
\[
N_{dr}(s_m) = \int_0^{s_m} \phi(s') ds' = \frac{C_{w_m} k_m}{(1 + \eta_0 k_m)} \tag{2.22}
= C(s_m)^{k_m}. \tag{2.23}
\]
The first form (2.22) is similar to Twomey’s power law
with correction at large \( s_m \), as derived by Ghan et al.
(1993, 1995) and modified in KC06 and KC08, ac-
counting for the drop growth after activation and vari-
sable fractions. The second form (2.23) is the gener-
ized power law; the coefficient \( C(s_m) \) and index
\( k(s_m) \) are continuous functions of the maximum super-
saturation or of the vertical velocity. The equations for
\( C(s_m) \) and \( k(s_m) \) are given in KC06 and KC08:
\[
C(s_m) = C_{w_0}^{s_0} \left[ 1 + \left( \frac{s_m}{s_0} \right)^{k_0} \right]^{-1}, \tag{2.24}
\]
\[
k(s_m) = k_{s_0} \left[ 1 + \left( \frac{s_m}{s_0} \right)^{k_0} \right]^{-1}, \tag{2.25}
\]
and
\[
\chi(s_m) = k_{s_0} \left( \frac{s_m}{s_0} \right)^{k_0} \left[ 1 + \left( \frac{s_m}{s_0} \right)^{k_0} \right]^{-1}. \tag{2.26}
\]
Equation (2.16) for \( t_m \) depends on \( \Lambda \) and contains the
integrals \( J_2(t_m) \) or \( J_3(t_m) \), which themselves depend on
\( t_m \) and, in general, cannot be evaluated analytically
without some simplifications. However, these equations
are easily solved numerically and lookup tables are
easily calculated, as illustrated in KC08. Then \( N_{dr}(s_m) \)
can be calculated in a cloud or climate model using
the modified power law described in KC08 and these
lookup tables.

Another way of studying the kinetics of drop nuclea-
tion is based on simplifying \( J_2 \) or \( J_3 \), finding the asymp-
totic limits of the solutions, and then constructing inter-
polations among them. There are four particular cases, or
four limits, when the general solution described above
can be simplified and approximate analytical solutions
for $r_{sd}$ and $N_{dc}$ can be found. These cases are naturally separated by the values of the vertical velocities (or maximum supersaturations $s_m$), determined by the values of $\lambda_s$ in (2.15a), and of the condensation coefficients $\alpha_c$ that determine diffusion growth of the activated droplets (sufficiently pure water drops) or kinetic growth regime (polluted water drops). Thus, these four limits are

1. $\lambda_s \ll 1$ (small vertical velocities $w$), $\alpha_c \sim 1$ (diffusion droplet growth limit);
2. $\lambda_s \ll 1$ (small vertical velocities $w$), $\alpha_c \ll 1$ (kinetic droplet growth limit);
3. $\lambda_s \gg 1$ (large vertical velocities $w$), $\alpha_c \sim 1$ (diffusion droplet growth limit); and
4. $\lambda_s \gg 1$ (large vertical velocities $w$), $\alpha_c \ll 1$ (kinetic droplet growth limit).

Equations (2.22) and (2.23) for $N_{dc}(s_m)$ and solutions for $s_m$ represent a generalization of the corresponding expressions from Twomey (1959) and Sedunov (1967, 1974), which were based on the power law for $\phi_a(s)$ and correspond to the first limit here, as described in section 3. These four limits, along with the situations intermediate among them, cover the vast majority of the situations that can be met in atmospheric clouds. Thus, if we find these limits and construct interpolations among them, the problem of parameterization can be solved. However, these limits have physical meaning without interpolation since each of them corresponds to some particular type of cloud with a certain range of vertical velocities. For example, limits 1 and 2 may correspond to stratiform clouds or fog with low vertical velocities $w$, and limits 3 and 4 correspond to convective clouds with higher $w$. Thus, for simulation of these cloud types, it can be sufficient to use these limits without interpolation over the range of vertical velocities.

The applicability of these limits is determined by the scaling parameters $s_m/s_0$ and $\lambda_s = (s_m/s_0)^{5/3}$. Figure 2 shows that the values $\alpha_c = 10^{10}$ (the physical value of $\alpha_c \leq 1$, but $\alpha_c = 10^{10}$ means absence of kinetic correction, $\xi = 0$) and $\alpha_c = 1$ yields close results; that is, the value $\alpha_c = 1$, which may be characteristic of pure water drops (e.g., AGR98), is close to Twomey’s case with $\xi = 0$. The regions of the four limits are depicted in Fig. 2b on the $\lambda_s$–$w$ diagram. One can see that the first limit, $\lambda_s \ll 1$, is valid with $\alpha_c \sim 1$ and small $w$. The second limit with $\alpha_c \ll 1$ is marked in Fig. 1 at $w \leq 1$ around the ordinate axis. The third and fourth limits are reached at $w > 1$–2 m s$^{-1}$. This representation is somewhat schematic and these regions may extend to larger areas with varying parameters. The applicability of these limits will be illustrated in more detail in the next sections by comparison of these limits with the exact solutions.

3. Limit 1: Small vertical velocity, diffusional growth regime

This limit corresponds to $\lambda_s \ll 1$ (low $w$ or low $s$) and $\alpha_c \sim 1$ (diffusion growth regime of activated drops); thus, the denominator of the second fraction in the integrand of $J_1$ [(2.14b)] tends to unity. This is equivalent to the absence of denominator in the differential CCN activity spectrum (2.3), which is then similar to the Twomey (1959) power law

$$
\phi_a(s) = k_{a0}C_0 s^{5/3}.
$$

(3.1)

When $\alpha_c \sim 1$, we can neglect small $\xi$ and also assume small $r_{sd}(t_0)$. With these assumptions and (3.1), limit 1 is
equivalent to the model of activation considered by Twomey, Sedunov, and all subsequent similar models, but their empirical coefficient $C_0$ and index $k_0$ were expressed in (2.5) and (2.6) via aerosol parameters. As shown in appendix B, $J_s$ in this case is reduced to Twomey's integral with the Euler beta function $B(x, z)$:

$$J^1_3 = B\left(\frac{k_0}{2}, \frac{3}{2}\right). \quad (3.2)$$

a. Lower bound

Substitution of (3.2) into (2.16) for $l_m$, using $A$ from (2.13a) with $a_m = c_i w_m$ from (2.20), solving for $l_m$, and using $c_5$ from appendix A yields the lower bound $l^{(1)}_m$ in the first limit:

$$l^{(1)}_m = K^{(1)}_{lm} + \frac{1}{2}\left\{w\left[(k_0^{1/2} - (k_0^{1/2})^2) \right]^{1/2}\right. \quad (3.3)$$

$$K^{(1)}_{lm}(T, p) = \frac{c_i^{1/2}}{2\pi D_c^{1/2}} k_0 B\left(\frac{k_0}{2}, \frac{3}{2}\right)^{1/2} \quad (3.4)$$

(The superscript $i$ hereafter denotes the $i$th limit with $i = 1–4$, and the lower subscript $l$ means hereafter lower bound). The maximum supersaturation is from (2.18) in this limit

$$s^{(1)}_m = \alpha_{k_1} K^{(1)}_{sm} N_a^{(1/2)} (k_0^{1/2})^{k_0} W^{2/(2(k_0^{1/2})^2)} \quad (3.5)$$

$$K^{(1)}_{sm}(T, p) = \frac{2\pi D_c^{1/2}}{c_i^{1/2}} k_0 B\left(\frac{k_0}{2}, \frac{3}{2}\right)^{1/2} \quad (3.6)$$

and $\alpha_{k_1}$ was defined in (2.19). Note that $s^{(1)}_m$ and $l^{(1)}_m$ are related as $s^{(1)}_m = \alpha_{k_1} c_i w^{(1)}_m$, that is, almost as in Twomey's upper approximation (2.20) $s_{ov}(t)$ but with the coefficient $\alpha_{k_1}$. Table 1 shows that $s^{(4)}_m$ is only 11%–24% lower than $s_{ov}(t)$. The droplet concentration from (2.22) is

$$N^{(1)}_{dr}(s_m) = N_a K^{(1)}_{sm} s_0^{(1)} k_0$$

$$= \alpha_{k_1} K^{(1)}_{sm} N_a^{(1/2)} (k_0^{1/2})^{k_0} W^{2/(2(k_0^{1/2})^2)} \quad (3.7)$$

The powers of $w$ in the expressions for $s_m$ and $N_{dr}$ are the same as derived by Twomey (1959), but his empirical indices and coefficients are expressed now via aerosol microphysical parameters. The coefficient $\alpha_{k_1}$ is similar to the first major term in the Twomey power series expansion for the lower bound of $s_m$ [Twomey's

$$E_{(7)}$. The more detailed expressions can be obtained if $\sigma_d$ is expressed via $r_{d0}$ and $b$ from (2.4) and $A_k$ is expressed via $\zeta_{sa}$ and $T$ using (2.2):

$$l^{(1)}_m = K^{(1)}_{lm} \frac{Q_w^{1/2}}{w^{(1/2)}} N_a^{(1/2)} (k_0^{1/2})^{k_0} W^{2/(2(k_0^{1/2})^2)} \times \frac{s_{3/4}}{s_{sa}}/\left(2(k_0^{1/2})^2\right) \quad (3.9)$$

$Q_w$ is given in appendix A. Equation (3.8) shows that $l_m$ decreases (i.e., activation ceases faster) with the increase of $N_a$, $r_{d0}$, solubility $b$ (i.e., $e_v$), and updraft $w$ and with the decrease of surface tension in the presence of surfactants (e.g., organics). In particular, with $k_0 \sim 2$ ($\sigma_d = 1.7, \beta = 0.5$), (3.8) yields

$$l^{(1)}_m = K^{(1)}_{lm} (T) N_a^{(1/4)} r_{d0}^{(3/4)} b^{(1/4)} \frac{r_{d0}^{(3/4)}}{\zeta_{sa}} W^{(5/8)} \quad (3.9)$$

The analytical dependencies for another $\sigma_d$ can be found using the $k_0 - \sigma_d$ relation from Table 1. For $s_m$, we obtain from (3.5) and (2.4)

$$s^{(1)}_m = \alpha_{k_1} K^{(1)}_{sm}(T) Q_w^{1/2} N_a^{(1/2)} (k_0^{1/2})^{k_0} W^{2/(2(k_0^{1/2})^2)} \times b^{(1/4)} \frac{s_{3/4}}{s_{sa}}/\left(2(k_0^{1/2})^2\right) \quad (3.10)$$

Thus, $s_m$ increases with increasing $w$ and $\zeta_{sa}$ and decreases with an increasing supersaturation absorption rate, that is, increasing $N_a$, $r_{d0}$, and $b$ (or $e_v$). In particular, with $k_0 \sim 2$, we have

$$s^{(1)}_m = \alpha_{k_1} K^{(1)}_{sm}(T) N_a^{(1/4)} r_{d0}^{(3/4)} b^{(1/4)} \frac{r_{d0}^{(3/4)}}{\zeta_{sa}} W^{(5/8)} \quad (3.10a)$$

For $N_{dr}$, we have from (3.7) using (2.4) and (3.10)
\[ N_{dr,l}^{(1)} = \alpha_{k1} k_{w} \left( N_{sm}^{(1)}(T) \right)^{k_{a}} Q_{w}^{-k_{a}/(k_{a}+2)} N_{a}^{2(k_{a}+2)} r_{db}^{2k_{c}(1+\beta)/(k_{a}+2)} \times \beta^{k_{a}/(k_{a}+2)} b_{sa}^{-3k_{a}/[2(k_{a}+2)]} T^{3k_{a}/[2(k_{a}+2)]} W^{3k_{a}/[2(k_{a}+2)]}. \]  

(3.11)

The index of \( \xi_{sa} \) is negative—that is, the maximum drop concentration grows with decreasing surface tension [as with increasing concentration of organics; e.g., Facchini et al. (1999, 2000) and Abdul-Razzak and Ghan (2004)]—and the other five power indices are positive; thus, \( N_{dr} \) increases with the increase of any of the four factors: \( N_{a}, r_{db}, b \) (or \( \epsilon_{b} \)), and \( w \). The power by \( T \) is overwhelmed by decreasing \( K_{sm}^{(1)}(T) \), and \( N_{dr} \) decreases with increasing \( T \) as obtained in KCS08 in agreement with Saleebey and Cotton (2004). In particular, with \( k_{a} \sim 2 \), we have

\[ N_{dr,l} \sim [K_{sm}^{(1)}(T)]^{k_{a}} N_{a}^{3/2} r_{db}^{1/2} b_{sa}^{-3/4} T^{3/4} W^{3/4}. \]  

(3.11a)

The variations \( \delta N_{dr} \) predicted by (3.11) at small variations of the surface tension \( \delta \xi_{sa} \) are as follows:

\[ \frac{\delta N_{dr,l}^{(1)}}{N_{dr,l}^{(1)}} = -C_{N\xi} \frac{\delta \xi_{sa}}{\xi_{sa}}; \quad C_{N\xi} = \frac{3k_{a} b_{sa}}{2(k_{a}+2)}. \]  

(3.11b)

In particular, with \( k_{a} \sim 2 \), then \( \delta N_{dr,l}/N_{dr,l} \sim (-3/4) \delta \xi_{sa}/\xi_{sa} \), which exactly coincides with the expression derived in Facchini et al. (1999). For a broader size spectrum with \( \sigma_{d} \sim 2.5 \) and \( \beta = 0.5 \), we have \( k_{a} \sim 1.16 \) (Table 1) and \( C_{N\xi} \approx 0.55 \); that is, the dependence of \( N_{dr,l} \) on surface tension becomes weaker for broader CCN spectra, but the general qualitative dependence of \( N_{dr,l} \) on \( \xi_{sa} \) in the first limit is similar to that in Facchini et al. (1999) and Abdul-Razzak and Ghan (2004). Note that these conclusions are drawn since we here consider \( \xi_{sa} \) as a variable and do not consider effects of its variations due to redistribution of organics between the surface and volume in CCN, as in Li et al. (1998) and Abdul-Razzak and Ghan (2004). According to Facchini et al., a 30% decrease in \( \xi_{sa} \) should cause a \( \sim 20\% \) increase in \( N_{dr} \), which should lead to an increase in top of the atmosphere albedo locally by \( \sim 1\% \) and could cause a global mean forcing with an upper limit of \( \sim 1 \) W m\(^{-2}\). The opposite effects of decreasing \( N_{dr} \) with increasing organic coating of CCN and decrease of \( \xi_{sa} \) were predicted by Feingold and Chuang (2002) and other studies reviewed in Foster et al. (2007). The equations here allow estimations of the effects of surface tension with various other CCN properties. These effects can be noticeable for small \( w \), but it will be shown in sections 5 and 6 that \( N_{aq} \) is highly insensitive to surface tension at high \( w \) (>1–2 \) m \( \text{s}^{-1} \).

b. Upper bound

Evaluation of the upper bound in this and other limits is a little more complicated and is done using the system of (2.16) and (2.18) to determine the two unknown, \( t_{m} \) and \( s_{m} \). Substitution of \( \xi_{sa}^{(1)} \) from (3.2) and \( a_{m} = s_{m}/t_{m} \) from (2.21) into (2.13a) yields \( \Lambda(t_{m}) \). Substituting it into (2.16) and (2.18) and solving for \( t_{m} \), we obtain for \( \mu_{k}^{(1)} \), \( t_{mu}^{(1)} \), \( s_{mu}^{(1)} \), and \( N_{dr,u}^{(1)} \) in the upper bound (subscript \( u \)) of the first limit almost the same expressions as (3.8), (3.10), and (3.11) but multiplied by the coefficient \( \alpha_{low,up}^{(1)} \) for \( t_{mu}^{(1)} \) in (3.8), by \( \alpha_{up}^{(1)} \) instead of \( \alpha_{k1} \), for \( s_{ml}^{(1)} \) in (3.10), and \( \alpha_{up}^{(1)} k_{a} \) instead of \( \alpha_{k1} k_{a} \) for \( N_{dr,u}^{(1)} \) in (3.11). The relation \( s_{mu}^{(1)} = \alpha_{up}^{(1)} c_{1} w_{mu}^{(1)} [\text{similar to Twomey's upper approximation (2.20)}] \) arises again, but with the coefficient \( \alpha_{up}^{(1)} \sim 1 \). The relation of the lower and upper bounds in the first limit is determined by the coefficient \( \alpha_{up}^{(1)} \):

\[ t_{mu}^{(1)} = \alpha_{low,up}^{(1)} t_{ml}^{(1)}; \quad s_{mu}^{(1)} = \alpha_{low,up}^{(1)} s_{ml}^{(1)}; \quad N_{dr,u}^{(1)} = (\alpha_{up}^{(1)} k_{a}) N_{dr,l}^{(1)}. \]  

(3.12)

where

\[ \alpha_{up}^{(1)} = \alpha_{k}^{(1)} 1/[2(k_{a}+2)] = [(k_{a}+2)/(k_{a}+3)]^{1/[2(k_{a}+2)]}; \]  

(3.13a)

\[ \alpha_{low,up}^{(1)} = \alpha_{up}^{(1)} / \alpha_{k}^{(1)} = [(k_{a}+3)/(k_{a}+2)]^{(2k_{a}+3)}/[2(k_{a}+2)]. \]  

(3.13b)

The coefficients \( \alpha_{k}^{(1)}, \alpha_{up}^{(1)}, \) and \( \alpha_{low,up}^{(1)} \) along with the index \( k_{a} \) are given in Table 1 for various size dispersions \( \sigma_{d}, \) One can see that \( k_{a} \) decreases from 5.84 at \( \sigma_{d} = 1.2 \) (narrow spectra) to 1.16 at \( \sigma_{d} = 2.5 \) (wide spectra). All the coefficients are close to 1, and their variations are much smaller: \( \alpha_{k} \) decreases from 0.89 to 0.76 and \( \alpha_{up} \) decreases from 0.993 to 0.957. Thus, the lower bound for \( s_{m} \) obtained with (2.18) is 11%–26% lower than would be obtained if applied the linear relation (2.20) for the maximum values as \( s_{m} = c_{1} w_{sm} \), and the upper bound is only 1%–4% lower. Calculated variations of the coefficients with \( \beta = 0 \) (not shown here) are even smaller. For all \( \sigma_{d}, \alpha_{up}^{(1)} > \alpha_{k}^{(1)} \); that is, the condition is satisfied that the upper bound is higher than the lower bound. However, the difference between the bounds is rather small. The coefficient \( \alpha_{low,up}^{(1)} \), which characterizes the ratio of the upper to the lower bounds, increases from 1.116 to 1.260 in this range; that is, the difference does not exceed 12% and 26% for the narrow and wide spectra, respectively. If one assumes that the true value is close to the mean of the upper and lower bounds, the error would be halved: \( \sim 6\% \) and 13\%. 
A comparison was performed of these approximate analytical limits for $t_m$, $s_m$, and $N_{dr}$ with a more precise numerical solution of the complete supersaturation Eq. (2.8) with (2.22)–(2.26) as described in KC08, referred to hereafter as the “exact solution.” Figure 3 shows that the agreement between the first limit and exact solution for $t_m$ and $s_m$ is good for both bounds at $w \leq 30$–50 cm s$^{-1}$. A comparison of $s_m$ with the parameterization from AGR98 also shows satisfactory agreement; the curve calculated using AGR98 algorithm is closer to our lower bound for $s_m$ at small $w$ and coincides with the exact solution at $w > 0.5$ m s$^{-1}$.

Figure 3c shows a comparison of $N_{dr}$ calculated using three methods. The first limit for $N_{dr}$ (3.11), that is, the traditional power law (solid squares), is close to the exact solution only at $w = 1$–4 cm s$^{-1}$ (diamonds). At greater $w$ using the power law may significantly overestimate droplet concentrations. This curve and conclusion are in close agreement with those from Ghan et al. (1993, Fig. 7): This was the major reason for revisions of the Twomey theory. The upper bound of the “modified power law” (2.23) is close to the exact solution to $w \sim 0.5$ m s$^{-1}$; the error does not exceed 10%–15%. Thus, the modified power law for $N_{dr}$ allows one to decrease the errors. This comparison justifies the validity of the first limit and shows the region of its applicability.

c. Comparison with the Twomey power law

A more detailed comparison with the Twomey power law allows expression of Twomey’s empirical coefficients $C_T$ and $k$ via aerosol microphysical parameters. The expression for maximum supersaturation, derived in more general form in PK97, their Eq. (13-40), can be rewritten as

\[
\begin{align*}
    s_m &= C_T^{-1/(k+2)} C_{ST} w^{3/2(k+2)}; \quad (3.14a) \\
    C_{ST} &= \left[ \frac{A_1^{3/2} A_2^{3/5} k B(k/2, 3/2)}{2 \pi \rho_w A_3} \right]^{1/(k+2)}, \quad (3.14b)
\end{align*}
\]

where $C_T$ is Twomey’s empirical constant of the activation spectrum and we introduced a new coefficient $C_{ST}$ for convenience of comparison (a misprint in PK97 in the sign of the power index of $C_T$ is corrected). The coefficients $A_1$ and $A_2$ describe supersaturation generation and absorption, respectively, in the supersaturation equation [PK97, Eq. (13–30)]

\[
\frac{ds}{dt} = A_1 w - A_2 \frac{d\rho_L}{dt}, \quad (3.15)
\]

where $\rho_L$ is the liquid water content and $A_3$ in (3.14b) is the coefficient in drop radius growth rate [Eq. (13–32) in

---

**Fig. 3.** Comparison of the lower and upper bounds of the first limit (solid and open circles) at $\alpha_c = 1$ with the exact solutions (diamonds): (a) maximum time $t_m$; (b) maximum supersaturation $s_m$ compared with parameterization from AGR98; (c) concentration of activated droplets $N_{dr}$ calculated as the power law (3.11) (solid squares), the modified power law (2.22) with the lower and upper bounds for $s_m$ from (b), and exact solution (2.22) with the exact $s_m$ and $\alpha_c = 1$. Calculations are performed with the same parameters as in Fig. 2.
PK97). In our notation, \( A_1 = c_1 \), \( A_3 = c_3 \). The expression for \( A_2 \) in PK97, their Eq. (13–31),

\[
A_2 = \frac{R_u T}{(M_w/M_a)\epsilon_{sw}} + \frac{(M_w/M_a)I_c^2}{p T c_p}
\]

with \( \epsilon_{sw} \) being the saturated vapor pressure, can be simplified using the equations of state for water vapor and air as \( A_2 = \Gamma_1 / \rho_{sw} \), where \( \Gamma_1 \) is the psychrometric correction defined in appendix A. Substitution of \( A_1, A_2, \) and \( A_3 \) into (3.14b) yields

\[
C_{ST} = \left[ \frac{c_{1/2}^2 \rho_{sw}}{2 \pi \rho_{w} \Gamma_1 c_{1/2}^2 2B(k/2, 3/2)} \right]^{1/(k+2)} \quad (3.16)
\]

It follows from (2.3) or (3.1) that, for \( k = k_{d0} \), in this limit \((s_m/s_0)^{k_s} \ll 1\). Then, writing \( c_{1/2}^2 = c_3^2 \) and using (2.7) for \( c_3 \), we see that \( C_{ST} \) exactly coincides with our \( K^{(1)}_{mm} \) in (3.6). Equating our expression for the upper bound of \( s_{mm} \) (3.10) with correction (3.12) and Twomey’s (3.14a), dividing by \( C_{ST} = K^{(1)}_{mm} \), and using the definitions of \( b \) and \( Q_w \), we obtain

\[
C_T = 10^{-2k_{d0}}\alpha_{k1}^{-1/2} \left( \frac{2^{5/3} M_w}{3 R\rho_w} \right)^{-3k_{d0}/2} N_a
\]

\[
\times \frac{c_{1/2}^2\sigma_{d0}}{T^{3k_{d0}/2} T_{d0}^{(1+\beta)} (\rho_{d0} M_w / \rho_{w} M_d)^{k_{d0}/2}} \quad (3.17)
\]

Equation (3.17) is valid for both \( \beta = 1/2 \) and \( \beta = 0 \) and \( s_{mm} \) measured in percent. Thus, Twomey’s empirical coefficient \( C_T \) is expressed via \( N_a, r_{d0}, \sigma_d \) (in \( k_{d0} \)), \( T, \xi_a, \beta, b \) (or \( e_s \)), \( \rho_w \), and other properties of CCN. In addition, these dependencies are nonlinear—governed by the powers of \( k_{d0} \). It is not surprising that measurements made over a period of decades have found such different values for \( C_T \), varying by several orders of magnitude in various air masses and clouds (Hegg and Hobbs 1992; Table 9.1 in PK97; Seinfeld and Pandis 1998). Twomey’s empirical index \( k \), equal to \( k_{d0} \) at \( s \ll 1\% \), is also expressed with (2.5) via \( \sigma_d \) and \( \beta \). Equations (3.17) and (2.5) explain many features of the observed values of \( C_T \) and \( k \). In particular, since \( C_T \sim N_a, C_T \) has higher values in continental than in maritime air masses, as well as greater values in polluted areas. Further, \( C_T \) also increases with increasing soluble fraction \( e_s \), mean radius of CCN \( r_{d0} \), and with decreasing surface tension in the presence of surfactants.

The drop concentration is evaluated, according to Twomey, as \( N_{dr} = C_T s_{mm} \) using (3.14a) and (3.14b):

\[
N_{dr} = C_T^{2/(2+k)} C_{ST} W^{3/(2(k+2))} \quad (3.18)
\]

Substituting any values of CCN parameters into (3.17) and (3.18) yields concrete values of \( C_T \) and \( N_{dr} \) for a given air mass. It should be noted that the typical calculated values of \( C_T \) and \( k \) in the limit \( s \ll 1\% \) are usually much greater than often measured, \( k \sim 0.2–0.6 \) at \( s = 0.2\%–1\% \), and then used in parameterizations. This reflects a substantial difference between Twomey’s model using typical \( k \sim 0.2–0.6 \) (PK97) corresponding to some fixed and sufficiently high \( s \) and our model with calculated \( k(s) \), which is a generalization of Twomey’s model for arbitrary continuous \( s \). This is illustrated in Fig. 4, which depicts \( C(s_m) \) and \( k(s_m) \) calculated with (2.24)–(2.26) and presented as functions of \( s_m \) and \( w \) simultaneously: \( C(s_m) \approx 8000 \, \text{cm}^{-3} \) at \( w = 1 \, \text{cm s}^{-1} \) \((s_m = 0.043\%)\) and decreases to the limit \(-500 \, \text{cm}^{-3} \) at
\[ w \approx 1-2 \text{ m s}^{-1} \ (s_m = 0.5\%-1\%). \] Experimental data on \( C(s_m) \) are not readily available. However, the calculated index \( k(s_m) \), which also decreases from 1.67 to below 0.2 in this range of \( s_m \) or \( w \) (Fig. 4b), is in a good agreement with data both from cloud chamber and field experiments (e.g., Juisto and Lala 1981; Yum and Hudson 2001). This indicates that this model generalizes the Twomey model owing to the kinetic drop growth regime that contained only the beta function, now \( J_3 \) is substantially different than that in the Twomey model owing to the kinetic drop growth rate. The integral \( J_3 \) for this case is evaluated in appendix B:

\[ J_3(t) = 4^{-3} D_v^{-3} V_w^{-1} \alpha_c^{-3} (c_s a_m)^{3/2} \frac{k_0}{2} \cdot 3. \] (4.1)

In contrast to the first limit (3.2) for \( J_3^{(1)} \), the diffusion regime that contained only the beta function, now \( J_3^{(2)} \) contains also several coefficients in dimensionless combination.

a. Lower bound

Substituting (4.1) into (2.16) for \( t_m \) with \( a_m = c_1 w \) from (2.20) we obtain the lower bound (superscript 2 means the second limit; subscript \( f \) means lower bound):

\[ t_m^{(2)} = K_m^{(2)} N_a^{-1/3(k_0+5)} k_0/(k_0+5) \cdot W^{-1(k_0+2)/(k_0+5)} \cdot \alpha_c^{-3(k_0+5)}, \] (4.2)

\[ K_m^{(2)}(T, p) = \left[ \frac{\pi k_0}{128} c_1^{-3} c_2 D_v^{-2} V_w^{-3} B \left( \frac{k_0}{2} \cdot 3 \right) \right]^{-1(k_0+5)}. \] (4.3)

The lower bound for \( s_m \) is calculated using (2.18):

\[ s_m^{(2)} = \alpha_k K_m^{(2)} N_a^{-1/3(k_0+5)} k_0/(k_0+5) \cdot W^{-3(k_0+5)} \cdot \alpha_c^{-3(k_0+5)}, \] (4.4)

\[ K_m^{(2)}(T, p) = \left[ \frac{\pi k_0}{128} c_1^{-3} c_2 D_v^{-2} V_w^{-3} B \left( \frac{k_0}{2} \cdot 3 \right) \right]^{-1(k_0+5)}. \] (4.5)

The lower bound for \( N_a \) is calculated from (2.22) taking into account that \( s_m \ll s_0 \) (lower s limit):

\[ N_{dr}^{(2)}(s_m) = (\alpha_k) K_N^{(2)} N_a^{5(k_0+5)} \times s_0^{-5k_0/(k_0+5)} W^{k_0/(k_0+5)} \cdot \alpha_c^{-3(k_0+5)}. \] (4.6)

\[ K_N^{(2)}(T, p) = \left[ \frac{\pi k_0}{128} c_1^{-3} c_2 D_v^{-2} V_w^{-3} B \left( \frac{k_0}{2} \cdot 3 \right) \right]^{-1(k_0+5)}. \] (4.7)

As in the previous limit, these equations can be expressed via \( r_{d0} \) and \( b \) instead of \( s_0 \) using (2.4) for \( s_0 \), and via \( s_\alpha \) and \( T \) instead of \( A_k \) using (2.2); thus, we obtain for \( t_m \):

\[ t_m^{(2)} = K_m^{(2)} C_w^{5/3} N_a^{-1/3(k_0+5)} r_{d0}^{-1(1+b)/(k_0+5)} b^{-k_0/(2(k_0+5))} \times s_\alpha^{-3k_0/(2(k_0+5))} W^{-3(k_0+5)} \cdot \alpha_c^{-3(k_0+5)}. \] (4.8)

For \( k_0 = 2 \), it follows from (4.8) in the kinetic regime:

\[ s_m^{(2)} = \alpha_k^{1/3} K_m^{(2)} N_a^{-1/7} r_{d0}^{-3/7} b^{-1/7} s_\alpha^{3/7} T^{-3/7} W^{-4/7} \cdot \alpha_c^{-3/7}. \] (4.9a)

Comparing with the diffusion limit (3.9), we see that all six dependencies (except for \( \alpha_c \)) are weaker in the kinetic regime. The maximum supersaturation from (4.4), (4.5), and (2.4) becomes

\[ s_m^{(2)} = \alpha_k^{1/3} K_m^{(2)} C_w^{5/3} N_a^{-1/3(k_0+5)} r_{d0}^{-1(1+b)/(k_0+5)} b^{-k_0/(2(k_0+5))} \times s_\alpha^{-3k_0/(2(k_0+5))} W^{-3(k_0+5)} \cdot \alpha_c^{-3(k_0+5)}. \] (4.9)

For \( k_0 = 2 \), (4.9) gives in this kinetic regime

\[ s_m^{(2)} = \alpha_k^{1/3} K_m^{(2)} N_a^{-1/7} r_{d0}^{-3/7} b^{-1/7} s_\alpha^{3/7} T^{-3/7} W^{-4/7} \cdot \alpha_c^{-3/7}. \] (4.9a)

Comparing with the diffusion limit (3.10a), we see that the dependencies on \( N_a \), \( r_{d0} \), \( s_\alpha \), \( T \), and \( b \) are substantially weaker in the kinetic regime, and the dependence on \( w \) is slightly stronger. The droplet concentration from (4.6) and (4.7) converts into

\[ N_{dr}^{(2)}(s_m) = (\alpha_k) K_N^{(2)} C_w^{5/3} N_a^{-1/7} r_{d0}^{-3/7} b^{-1/7} s_\alpha^{3/7} T^{-3/7} W^{-4/7} \cdot \alpha_c^{-3/7}. \] (4.10)

In particular, for \( k_0 = 2 \), (4.10) gives in the kinetic regime

\[ N_{dr}^{(2)} = K_{nd}^{(2)} N_a^{5/7} b^{15/7} s_\alpha^{15/7} T^{-15/7} W^{-6/7} \cdot \alpha_c^{-6/7}. \] (4.10a)
Comparing this to the diffusional growth limit (3.11a) from section 3, we see that the dependencies on the six parameters \((N_{sa}, r_{d0}, b, \zeta, T, w)\) become somewhat stronger in the kinetic limit. In addition, this limit includes explicit dependence on the condensation coefficient \(\alpha_c\). An advantage of this method of asymptotic limits is that it allows for simple estimates of the effects of variations of the condensation coefficients on \(t_{m}, s_{m},\) and \(N_{dr}\). It follows from (4.8)–(4.10) that

\[
\frac{t_{m}^{(2)}}{s_{m}^{(2)}} \sim \alpha_{c}^{-3(k_{m}+5)},
\]

\[
N_{dr}^{(2)} \sim N_{dr}^{(1)} \alpha_{c}^{-3k_{m}/(k_{m}+5)}.
\]

**b. Upper bound**

Derivation of the upper bound, as for the first limit, is based on the use of the system of equations, (2.16) and (2.18). Substituting (4.1) for \(J^{(2)}_{s}\) with \(a_{m} = s_{m}/t_{m}\) from (2.21) into this system and solving for \(t_{m}, s_{m}\), we obtain for \(t_{m}^{(2)}, s_{m}^{(2)}\) and \(N_{dr}^{(2)}\) in the upper bound (subscript \(u\)) of the second limit (superscript 2) almost the same expressions as (4.8)–(4.10) but multiplied by the coefficient \(\alpha_{up}^{(2)}\) for \(t_{m}^{(2)}\) in (4.8), by \(\alpha_{up}^{(2)}\) instead of \(\alpha_{k1}\) for \(s_{m}^{(2)}\) in (4.9), and by \((\alpha_{up}^{(2)})^{k_{m}}\) instead of \(\alpha_{k0}\) for \(N_{dr}^{(2)}\) in (4.10). That is, the relation of the limits is

\[
t_{m}^{(2)} = \alpha_{up}^{(2)} t_{m}^{(1)}, \quad s_{m}^{(2)} = \alpha_{up}^{(2)} s_{m}^{(1)},
\]

\[
N_{dr}^{(2)} = (\alpha_{up}^{(2)})^{k_{m}} N_{dr}^{(1)},
\]

where

\[
\alpha_{up}^{(2)} = \alpha_{k1}^{2(k_{m}+5)} = [(k_{m} + 2)(k_{m} + 3)]^{2(k_{m}+5)},
\]

\[
\alpha_{low}^{(2)} = \alpha_{up}^{(2)} / \alpha_{k1} = [(k_{m} + 3)(k_{m} + 2)]^{(k_{m}+3)/(k_{m}+5)}.
\]

The coefficients \(\alpha_{k1}, \alpha_{up}^{(2)},\) and \(\alpha_{low}^{(2)}\) are given in Table 1. As in the first limit, all the coefficients are close to 1, and \(\alpha_{up}^{(2)}\) is 2%–8% lower than 1. In this range of \(\sigma_{d}\), \(\alpha_{up}^{(2)}\) increases from 1.099 to 1.20 and characterizes the ratio of \(t_{m}\) and \(s_{m}\) in the upper to lower bounds. Shown in Fig. 5 is a comparison of the upper bound of the second limit with the exact solution to the supersaturation Eq. (2.8). The agreement becomes sufficiently good at \(\alpha_{c} = 0.04\), and \(w = 2–3 \text{ cm s}^{-1}\). A surprising finding is that the value \(\alpha_{c} = 0.04\), used in many cloud models, is close to the boundary of the kinetic regime, and the values of \(t_{m}, s_{m},\) and \(N_{dr}\) are substantially greater in this regime than in the first limit with \(\alpha_{c} = 1\) and a diffusion regime (cf. Figs. 3 and 5).

**FIG. 5.** Comparison of the upper bound of the second limit (solid symbols and lines) with the corresponding exact solutions to the supersaturation equation (corresponding open symbols, dashed lines): (a) maximum time \(t_{max}\); (b) maximum supersaturation \(s_{max}\); (c) concentration of activated droplets \(N_{dr}\). Calculations are performed with the parameters \(N_p = 500 \text{ cm}^{-3}, r_{d0} = 0.05 \mu m, \sigma_{d} = 1.8, \beta = 0.5, b = 0.25, T = 10^\circ C,\) and \(p = 800 \text{ hPa}\). Agreement (accuracy of the second limit) becomes better at \(\alpha_{c} < 0.04\) — that is, \(\alpha_{c} = 0.04\) is close to the boundary of the kinetic regime.
5. Limit 3: Large vertical velocity, diffusional growth regime

At large \( w \)—that is, in the limit \( s_m \gg s_0 (\lambda_s \gg 1) \)—the integral \( J_3 \) in (2.14b) in general cannot be reduced to the beta function and it is more convenient to use (2.16) for evaluation of \( \tilde{J}_2 \), with (2.13b) for \( \Lambda \) and (2.14a) for \( J_2 \). The integral \( J_2^{(3)}(t) \) in this limit is evaluated in appendix B as

\[
J_2^{(3)}(t) \approx \frac{1}{k_{c0}^{(3)}} \left( a_{m}^{(3)} \right)^{k_{a}}. \tag{5.1}
\]

The error of this analytical \( J_2^{(3)}(t) \) relative to the exact numerical calculation is less than 4\% for \( \lambda_s \approx 30 \). Substituting (5.1) into (2.13b) for \( \Lambda \) and then into (2.16) yields an expression for \( t_m^{(3)} \):

\[
t_m^{(3)} = (4\pi D_v c_1^{-1/3} c_3^{2/3})^{-1/2} N_a^{-1/2} w^{1/2}. \tag{5.2}
\]

a. Lower bound

Substituting here \( a_m = c_{3w} \) from (2.20) we obtain a simple asymptotic for the lower bound (subscript \( l \)) of the third limit (superscript 3) at large \( w \):

\[
l_m^{(3)} = K_{ml}^{(3)} N_a^{-1/2} w^{-1/4}, \tag{5.3}
\]

\[
K_{ml}^{(3)} = [4\pi D_v c_1^{1/2} c_3^{1/2}]^{-1/2}. \tag{5.4}
\]

This gives for \( s_m \) from (2.18),

\[
s_m^{(3)} = \alpha_{kl} K_{sm}^{(3)} N_a^{-1/2} w^{3/4}, \tag{5.5}
\]

\[
K_{sm}^{(3)} = [4\pi D_v c_1^{-3/2} c_3^{1/2}]^{-1/2}. \tag{5.6}
\]

b. Upper bound

The upper bound, as in the previous sections, is obtained by substituting \( a_m = s_m / t_m \) from (2.21) into (5.2), then substituting the resulting relation \( t_m(s_m) \) into the system of equations, (2.16) and (2.18), and solving for \( t_m, s_m \). Then we obtain \( t_m \), similar to (5.3) but multiplied by the coefficient \( \alpha_{\text{upp}}^{(3)} \) and \( s_m \), similar to (5.5) but with coefficient \( \alpha_{\text{upp}}^{(3)} \) instead of \( \alpha_{kl} \) so that

\[
t_m^{(3)} = \alpha_{\text{low,upp}}^{(3)} t_m^{(3)}, \quad s_m^{(3)} = \alpha_{\text{low,upp}}^{(3)} s_m^{(3)} \tag{5.7}
\]

where

\[
\alpha_{\text{up}}^{(3)} = \alpha_{1/4}^{(3)} = [(k_{s0} + 2)(k_{s0} + 3)]^{1/4}, \tag{5.8}
\]

\[
\alpha_{\text{low,upp}}^{(3)} = \alpha_{\text{up}}^{(3)} / \alpha_{kl} = \alpha_{k1}^{(3)} = [(k_{s0} + 3)/(k_{s0} + 2)]^{3/4}. \tag{5.9}
\]

The coefficients \( \alpha_{k1}, \alpha_{\text{up}}^{(3)}, \) and \( \alpha_{\text{low,upp}}^{(3)} \) are given in Table 1. They are also close to 1. The coefficient \( \alpha_{k1} \) for the lower bound is the same for all four limits as the upper bound \( \alpha_{\text{upp}}^{(3)} \) is lower than 1 by 3–7\% and the ratio of the upper to lower limits, \( \alpha_{\text{low,upp}}^{(3)} \), is 1.09–1.23. That is, the error of any solution average between these bounds does not exceed 5\%–11\%.

Equations (5.3)–(5.9) show that \( t_m \) and \( s_m \) in the limit of high \( w \) are also related by the linear relation similar to (2.18) with some coefficients \( \leq 1 \) and also have the form of power laws but only by \( N_a \) and \( w \). The dependencies of \( t_m \) and \( s_m \) on \( r_{d0}, \sigma_{d0}, b, \beta \), and on curvature parameter \( \Lambda \) or surface tension \( \xi_{sa} \), which were very substantial in the first and second limits at small \( w \), disappear from the coefficients and power indices in the limit of high \( w \). Thus, in the third limit with large \( w \) and \( \alpha_c \sim 1 \) (sufficiently pure drops) we come to an unexpected result: the maximum supersaturation \( s_m \) and activation time \( t_m \) are independent of the CCN size spectrum and chemical composition. Thus, \( t_m \) and \( s_m \) depend only on CCN concentration \( N_a \) and vertical velocities \( w \), and the indices of these dependencies are constant and universal for various CCN; that is, they are the same for different values of \( k_{d0} \). This somewhat paradoxical result can be explained by the fact that the third limit deals with the region \( \lambda_s \gg 1 \) where \( s_m \) is much greater than the mean geometric \( s_0 \). Thus, the vast majority of CCN will be activated, no matter what their individual properties are. This independence of the physical and chemical properties of CCN indicates that the effects of surface tension and chemical composition on the global albedo and radiative balance should be evident in stratiform clouds and become much weaker in convective clouds.

Figure 6 shows that the \( t_m \) and \( s_m \) in the upper bound of the third limit approach the corresponding exact solutions at \( w > 1–2 \text{ m s}^{-1} \), but this conclusion is valid only for \( \alpha_c \sim 1 \) and becomes invalid at \( \alpha_c = 0.1 \). At \( w < 0.7–1 \text{ m s}^{-1} \), the third limit is invalid. This independence of \( r_{d0}, \sigma_{d0}, b \) (or \( s_0 \)) at high \( w \) was illustrated in Figs. 3, 5, and 7 in KC08, where \( t_m(w) \) and \( s_m(w) \) in double-log coordinates represented almost straight lines with slopes slightly increasing toward large \( w \). The corresponding curves \( t_m(w) \) and \( s_m(w) \) converged at \( w \geq 0.5–2 \text{ m s}^{-1} \) toward the limits with fixed slopes. The simple equations of the third limit here provide an explanation for this as well as exact values of the slopes. The expression (2.22) for \( N_{dr} \) shows that \( N_{dr} \) tends to \( N_a \) at \( \lambda_s \gg 1 \) \( (s_m \gg s_0) \); we can approximate it by expanding the denominator in (2.22) into a power series by \( s_0 / s_m \sim 1 / \lambda_s \) and retaining two terms—that is, \( N_{dr} \) or fraction activated \( N_{dr} / N_a \) is a parabolic function of \( (s_m / s_0)^{-k_{sa}} \):

\[
N_{dr} = N_a [1 - (s_m / s_0)^{-k_{sa}} + (1/2)(s_m / s_0)^{-2k_{sa}}]. \tag{5.10}
\]
and $s_m$ is defined in (5.5) and (5.7). Figure 6 shows that $N_{dr}$ in the third limit at $\alpha_c \sim 1$ also approaches the exact solution at $w > 1-2$ m s$^{-1}$ but, at $w \leq 0.5$ m s$^{-1}$, $N_{dr}$ increases with decreasing $w$; that is, the number of terms in expansion (5.10) becomes insufficient and (5.10) becomes invalid. In contrast to $t_m$ and $s_m$, the value of $N_{dr}$ depends of the CCN size spectrum and chemical composition via $\alpha_c$, but this dependence is substantially weaker than in the first and second limits.

6. Limit 4: Large vertical velocity, kinetic growth regime

When $\alpha_c \ll 1$, then $\xi \gg r_d(t_0)$, and $r_d(t_0)$ can be neglected in $U_s$, so $U_s = V_s^2$, furthermore, $(1 - x^2) \ll U_s^{-1/2}$ in $J_2$, and $(1 - x^2) \ll V_s$. The integral $J_2$ in the fourth limit is evaluated in appendix B:

$$J_2^{(4)}(t_m) = \frac{1}{128} \frac{a_m^{3/2} - k_m^{3/2} \alpha_c^{3/5} v_w^3 k_0}{k_0 D_v^{3/2}}. \quad (6.1)$$

a. Lower bound

Substituting (6.1) into (2.13b) and (2.16) and using (2.20) for $a_m$, we obtain an algebraic equation for $t_m$, which gives the lower bound

$$t_m^{(4)} = K_m^{(4)} N_{dr}^{-1/5} w^{-2/5} \alpha_c^{-3/5}, \quad (6.2)$$

$$K_m^{(4)} = \left[ (\pi/64) c_1^2 c_3^2 V_0^3 D_v^{-2} \right]^{-1/5}. \quad (6.3)$$

The lower bound for maximum supersaturation $s_m$ is calculated from (6.2) and (2.18):

$$s_m^{(4)} = \alpha_{k1} K_m^{(4)} N_{dr}^{-1/5} w^{3/5} \alpha_c^{-3/5}, \quad (6.4)$$

$$K_m^{(4)} = \left[ (\pi/64) c_1^2 c_3^2 V_0^3 D_v^{-2} \right]^{-1/5}. \quad (6.5)$$

b. Upper bound

The upper bound for $t_{mu}$ and $s_{mu}$ is obtained as in the previous sections using (2.21) for $a_m$, substituting $J_2$ from (6.1) into (2.16), (2.18), and solving the system of two equations. Then we obtain $t_{mu}$ similar to (6.2) but multiplied by the coefficient $\alpha_{up}$ and $s_{mu}$ similar to (6.4) but with coefficient $\alpha_{up}$ instead of $\alpha_{k1}$, so that

$$t_{mu} = \alpha_{low,up}^{(4)} t_{mu}^{(4)}, \quad s_{mu} = \alpha_{low,up}^{(4)} s_{mu}^{(4)}, \quad (6.6)$$

where

$$\alpha_{up} = \alpha_{k1}^{2/5} = [(k_0 + 2)/(k_0 + 3)]^{2/5}, \quad (6.7)$$

$$\alpha_{low,up}^{(4)} = \alpha_{up}^{(4)} \alpha_{k1} = \alpha_{k3}^{2/5} = [(k_0 + 3)/(k_0 + 2)]^{2/5}. \quad (6.8)$$

The coefficients $\alpha_{k1}$, $\alpha_{up}$, and $\alpha_{low,up}$ are given in Table 1. They also are close to 1. The coefficient $\alpha_{k1}$ for
the lower limit is the same, \( \alpha_{\text{up}}^{(4)} \) for the upper limit is lower than 1 by 5\%--10\%, and \( \alpha_{\text{low}}^{(4)} \), the ratio of the upper to lower limits, is 1.07–1.18. That is, the error of any solution mean between these limits does not exceed 3.5\%--9\%.

Thus, the activation time \( t_m \) and maximum supersaturation \( s_m \) in both bounds of the fourth limit do not depend on the CCN size spectrum \( \{r_d, \alpha_d\} \), surface tension, soluble fraction \( \beta, b, \) or \( \varepsilon_m \) or chemical composition; they depend only on CCN concentration \( N_d \), vertical velocities \( w \), and condensation coefficient \( \alpha_c \).

These dependencies have the form of the power laws, their indices are constant and universal for various CCN similar to the third limit, and—in contrast to the first and second limits at small \( w \), where the indices depend on the size dispersions—the drop concentration \( N_d \) can be calculated from (5.10) as a parabolic function of \( \lambda_b \) below \( N_f \). It depends on physicochemical properties of CCN as in the third limit but much weaker than in the first and second limits.

The characteristics of the fourth limit are compared with the exact solution in Fig. 7 for various \( \alpha_c \). The fourth limit calculated with \( \alpha_c = 1 \) is far from the exact solution at all \( w \), which is quite natural since \( \alpha_c = 1 \) is characteristic of diffusion regime. With \( \alpha_c = 0.04 \), the \( t_m \) and \( s_m \) in the fourth limit are already close to the exact solution. This means that \( \alpha_c = 0.04 \), often used in the models, is closer to the kinetic than to diffusion regime. For \( \alpha_c = 0.01 \), characteristic of polluted clouds, the parameters \( t_m, s_m \), and \( N_d \) almost merge with exact solutions at \( w \approx 10 \) cm s\(^{-1} \).

7. Interpolation equations and comparison with exact solutions

General solutions for \( t_m \) and \( s_m \) can be constructed by interpolating between the limiting solutions with some interpolation function \( \Omega_w(w, w_{sc}) \) or \( \Omega_w(\alpha, \alpha_{\text{sc}}) \). For example, interpolations between the first and third limits can be constructed as

\[
s_m^{(3)}(w) = s_m^{(1)}(\Omega_w(w, w_{sc}) + s_m^{(3)}[1 - \Omega_w(w, w_{sc})])
+ \Delta s_{m13} \Omega_w(w, w_{sc})[1 - \Omega_w(w, w_{sc})],
\]

(7.1)

where \( w_{sc} \) is a scaling parameter. The function \( \Omega_w(w, w_{sc}) \) should satisfy the conditions \( \Omega_w(w = 0) \to 1 \), \( \Omega_w(w \gg w_{sc}) \to 0 \), which ensure corresponding limits at small and large \( w \). The third term on the rhs of this equation is added to make these fitting functions closer to the exact solutions at the intermediate \( w \), and the product \( \Omega_w(1 - \Omega_w) \) vanishes at \( w \to 0 \) and \( w \gg w_{sc} \).

Figure 8a shows \( s_m \) calculated with Eq. (7.1) and \( \Omega_w(w, w_{sc}) = \exp(-w/w_{sc}) \) with \( w_{sc} = 1.8 \) m s\(^{-1} \) and

\[
\Delta s_{m13} = s_m^{(3)}(5 \text{ m s}^{-1}) - s_m^{(3)}(1 \text{ cm s}^{-1}).
\]

The fitted interpolated curve (solid circles) tends to the corresponding limits at \( w \to 0 \) and \( w \gg w_{sc} \), lies above both limits owing to addition of the term with \( \Delta s_{m13} \), and is very close to the exact curve. The \( N_d(w) \) calculated with this interpolated \( s_m(w) \) and the modified power laws

![Fig. 7. Comparison of the upper bound of the fourth limit (solid symbols) with the corresponding exact solutions at various values of condensation coefficient indicated in the legend (open symbols): (a) activation time \( t_m \); (b) maximum supersaturation \( s_m \); (c) concentration of activated droplets \( N_d \). Calculations are performed with the same parameters as in Fig. 2.](image-url)
values of a nondimensional parameter \( \lambda \) (2.15a) and of the condensation coefficient \( \alpha_w \). The parameter \( \lambda \) is proportional to \( w, r_{d0}, \) and \( e_{w} \), and for the typical for CCN values the \( \lambda \) criterion is equivalent to the two limits of small and large updrafts \( w \). Thus, there are four combinations of two limits of \( w \) (small and large) and two limits of the condensation coefficient: \( \alpha_w \sim 1 \) (pure cloud drops; diffusional regime of droplet growth) and \( \alpha_w \ll 1 \) (polluted cloud drops; kinetic growth regime). The other characteristics of CCN \( (N_{a}, r_{d0}, \) and \( \sigma_{dr} \), composition, soluble fraction \( e_{w} \), and its distribution in the CCN, temperature, and pressure) can vary within each of these limits. Thus, these four limits and interpolation among them cover the vast majority of cloudy conditions.

Analytical expressions are obtained for the activation time \( t_{m} \), maximum supersaturation \( s_{m} \), and the concentration of activated droplets \( N_{dr} \). Since the exact analytical solution of the integro–differential supersaturation equation is not feasible, the solutions are found using and generalizing the methods from Twomey (1959) and Sedunov (1967, 1974) as the lower and upper bounds for each quantity. The solutions for the bounds are almost identical and differ only by the coefficients. These coefficients are close to 1 and to each other and differ not more than 10%–20%; that is, any superposition between these bounds has a smaller error.

There is significant difference between the regimes of small updrafts \( w \) and large \( w \). In the limit of small \( w \) both in the diffusion and kinetic regimes (first and second limits), the expressions for \( t_{m}, s_{m}, \) and \( N_{dr} \) are the products of six power laws of \( N_{a}, r_{d0}, b, w, \xi_{sa}, \) and \( \alpha_{w} \) with coefficients depending on pressure and indices depending on the size dispersion and soluble fraction. The modified power laws (2.22) and (2.23) substantially better approximate \( N_{dr} \). The temperature dependence is negative \( (N_{dr} \) decreases with increasing \( T \). The first limit of low \( w \) and diffusional growth is a generalization of the Twomey (1959) power laws, allowing Twomey’s empirical coefficient \( C_{T} \) and power index \( k \) to be expressed directly as functions of the CCN microphysical parameters and chemical composition and extended from small supersaturations to any values. The limit of low \( w \) and kinetic growth provides for the first time an analytical dependence on the condensation coefficient and surface tension that can be verified both experimentally and using other theoretical methods, for example, parcel modeling. The often-used value \( \alpha_{w} = 0.04 \) is close to the boundary of kinetic regime. A comparison of these two limits with the exact numerical solutions from KC08 shows that the regime of “small \( w \)” can be valid up to \( w \leq 0.2–0.5 \) m s\(^{-1}\), that is, for synoptic-scale updrafts and turbulent fluctuations of vertical velocity.

8. Summary and conclusions

The integro–differential equation for the integral water supersaturation in clouds, derived and solved numerically previously by the authors (KC08), is solved here analytically for some limiting cases. The size spectrum of dry CCN is assumed to be algebraic equivalent to lognormal with the concentration \( N_{a} \), mean geometric radius \( r_{d0} \), dispersion \( \sigma_{dr} \), and solubility parameter \( b \) proportional to the soluble fraction \( e_{w} \). CCN may have variable chemical composition and surface tension \( \xi_{sa} \). The analytical solutions are obtained for four limiting cases that are determined by the

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Fig. 8. Illustration of the method of interpolation between the first and third limits and comparison with exact solutions: (a) maximum supersaturation \( s_{m} \); (b) droplet concentration \( N_{dr} \) calculated as described in the text. The parameters are as in Fig. 2.

(2.22) or (2.23) is also close to the exact curve. This example indicates that more precise interpolation expressions for \( N_{dr} \) can be obtained using (2.22) or (2.23) and interpolation among various analytical limits for \( s_{m} \). A detailed elaboration of this interpolation procedure among all four limits requires a special careful choice of several functions \( \Omega_{w} \), which is beyond the scope of this paper and will be described elsewhere.
The solutions for large vertical velocities ($w > 1$--$2$ m s$^{-1}$) are substantially different from the small vertical velocity solutions. The quantities $t_m$ and $s_m$ are also the products of power laws, but only by $w$ and $N_w$, and are independent of CCN properties. This fairly surprising finding can be explained by the fact that $s_m$ at large $w$ is much greater than the mean geometric supersaturation $s_0$, the majority of CCN have been activated, and therefore the dependence on CCN properties weakens and vanishes. The limits of $w$ for onset of this regime increase with decreasing $r_d$ and soluble fraction. These expressions allow significant simplification of calculating drop nucleation in sufficiently strong updrafts in convective clouds.

In contrast to some previous parameterizations predicting power laws by $s_m$ and unlimited $N_{dr}$, the expression for $N_{dr}$ found here for large $w$ is described by the modified power laws (2.22) and (2.23) and is bounded by $N_d$, as in Ghan et al. 1993, 1995; AGR98; Cohard et al. 1998, 2000; Fountoukis and Nenes 2005; KC06; KC08). A comparison of these two “large $w$” limits with exact numerical solutions from KC08 showed good agreement with accuracy of a few percent, indicating that the regime of large $w$ can be valid for $N_{dr}$ at $w \geq 1$--$2$ m s$^{-1}$.

These asymptotic analytical solutions can be used as controls for the accuracy and optimization of numerical solutions, estimating cloud drop spectral dispersions and the indirect dispersion effect on climate (e.g., Liu et al. 2006), design of field and laboratory experiments, parameterization of droplet activation by interpolation among the four limits for cloud and climate models, physically interpreting and understanding variations of the activation process with changing atmospheric composition, and investigations of aerosol indirect radiative forcing.

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APPENDIX A

List of Notations

- $A_k = \frac{2M}{k_BT_{ps}}$ Kelvin curvature parameter
- $a_m$ slope in approximation $s(t) = a_m (t)$, (2.11)
- $B = \frac{3\varphi M_s M_w}{4\pi M \rho_w}$ parameter of Raoults effect of CCN soluble fraction
- $b = (\nu \Phi) e_0 \rho_w \frac{M_s}{M_w}$ for $\beta = 1/2$

$$b = r_{ad1} e_0 (\nu \Phi) e_0 \frac{M_s}{M_w}$$ for $\beta = 0$

- $C(s_m)$ coefficient of activity spectrum, (2.23)
- $C_0$ parameter of activity spectrum, (2.6)
- $C_T$ Twomey’s empirical constant, (3.17)
- $c_1(T) = \left( \frac{L_w M_s}{c_p M_w} - 1 \right) \frac{g}{R_T}$, in (2.8), (2.12)
- $c_3$ parameter of droplet growth rate, (2.7)
- $c_5 = 2\pi D_v \sqrt{3} k_s$ parameter in (2.13a,b)
- $c_p$ specific heat capacity of air
- $D_v$ water vapor diffusion coefficient
- $g$ gravitational constant
- $J_2(t), J_3(t)$ integrals (2.14a), (2.14b)
- $k(s_m)$ index of activity spectrum, (2.25)
- $k_{d0} = 4/(\sqrt{2 \pi \sigma_d})$ power index of the size spectrum
- $k_{d0}$ index of activity spectrum at $s < 1$, (2.5)
- $L_{cc}$ latent heat of condensation
- $M_w, M_s$ molecular weights of water and air
- $M_s$ molecular weight of the soluble fraction
- $m_s$ mass of the soluble fraction
- $N_{a, dr}$ aerosol and drop number concentration
- $p$ pressure
- $Q_w = (2.53 M_w / 3 R_p w)^{3/2}$, (3.8)
- $R$ universal gas constant
- $R_m, R_v$ gas constants for air and water vapor
- $r_{ad}, r_d$ radius of a dry CCN and a droplet
- $r_{ad1}$ scaling radius in b with $\beta = 0$
- $r_{d0}$ and $r_m$ the mean geometric and the modal radii of CCN size spectrum
- $s = (\rho_w - \rho_m)/\rho_m$ supersaturation
- $s_0$ mean geometric supersaturation, (2.4)
- $s_m$ maximum supersaturation in a parcel
- $s_{ml}, s_{mu}$ lower and upper bounds of $s_m$
- $T$ temperature (K)
- $t$ time
- $t_m$ time when maximum $s_m$ is reached
- $t_{ml}, t_{mu}$ lower and upper bounds of $t_m$
- $U_s, V_s$ nondimensional parameters (2.15b)
- $V_w = \left( \frac{M_s}{M_w} \right)^{1/2}$ thermal speed of water vapor molecules, (2.7)
- $w$ vertical velocity
- $y(t) = \int_{t_0}^{t} s(t') dt'$ the integral supersaturation
- $\alpha_c$ condensation coefficient
- $\alpha_{k1} = (k_{d0} + 2)/(k_{d0} + 3)$, (2.19)
- $\alpha_{k1}^{(1)} = \alpha_{k1}^{(1/2)}$, (3.13a)
- $\alpha_{k1}^{(2)} = \alpha_{k1}^{(2k_{d0} + 5)}/(2k_{d0} + 5)$, (3.13b)
- $\alpha_{k1}^{(2)} = \alpha_{k1}^{(2k_{d0} + 5)}$, (4.14)
- $\alpha_{k1}^{(2)} = \alpha_{k1}^{(2k_{d0} + 5)}$, (4.15)
- $\alpha_{k1}^{(3)} = \alpha_{k1}^{(3)}$, (5.8)
- $\alpha_{k1}^{(3)} = \alpha_{k1}^{(3)}$, (5.9)
\[ \alpha_{up}^{(4)} = \alpha_{ki}^{2/3} \]  

(6.7)

\[ \alpha_{low,up}^{(4)} = \alpha_{up}^{(4)} / \alpha_{ki} = \alpha_{ki}^{-3/5}. \]  

(6.8)

\[ B(x, z) \]  

Euler’s beta function

\[ \beta \]  

describes composition of CCN in (2.2)

\[ \Gamma_1 = 1 + \frac{t^2}{c_r R_s^2 \rho_s} \]  

psychometric correction to the drop growth rate, (2.7)

\[ \epsilon_m \]  

mass soluble fraction of CCN

\[ \epsilon_v \]  

volume soluble fraction of CCN

\[ \epsilon_{r,0} \]  

reference soluble fraction for \( \beta = 0 \)

\[ \xi_{sa} \]  

surface tension at the CCN surface

\[ \eta_0 \]  

parameter of activity spectrum, (2.6)

\[ \lambda_s = (a_m t/s_0)^{k_o} \]  

parameter, (2.15a)

\[ \nu \]  

the number of ions in solution

\[ \xi \]  

kinetic correction to radius growth rate, (2.7)

\[ \rho_{av}, \rho_{os}, \rho_w \]  

densities of vapor, saturated vapor, and water

\[ \sigma_d \]  

the CCN dry size distribution

\[ \sigma_s \]  

supersaturation spectrum dispersion, (2.4)

\[ \Phi_s \]  

osmotic potential, (2.2)

\[ \varphi_s(s) \]  

CCN activity spectrum, (2.3)

\[ \chi(s_m) \]  

parameter, (2.26)

APPENDIX B

Evaluation of the Integrals \( J_2 \) and \( J_3 \) for Four Limiting Cases

The \( J_2 \) and \( J_3 \) are the dimensionless integrals introduced in section 2:

\[ J_2(t) = \int_0^1 \frac{\left[U_x + (1 - x^3)^{1/2} - V_x\right]^2}{\left[U_x + (1 - x^3)^{1/2}\right]^{(1 + \lambda_s)z_{m}}^{(1 + \lambda_s)z_{m}}^{2}} \]  

(1 + \lambda_s x^{k_o})^{2} dx. \]  

(B.1)

\[ J_3(x) = 2J_2(t) = \int_0^1 \frac{\left[U_x + (1 - z)^{1/2} - V_x\right]^2}{\left[U_x + (1 - z)^{1/2}\right]^{(1 + \lambda_s)z_{m}}^{(1 + \lambda_s)z_{m}}^{2}} \]  

(1 + \lambda_s x^{k_o})^{2} z^{k_o/2 - 1} dx. \]  

(B.2)

Here \( x = n t_0, z = x^{3/2} \), and the three nondimensional parameters \( \lambda_s, U_x \), and \( V_x \) are

\[ \lambda_s = \left(\frac{a_m}{s_0}\right)^{k_o} = \left(\frac{3\sqrt{3}a_m}{2} \frac{b_{1/2} r_{d,0}^{1/2} + \rho_{0}}{A_{k}^{3/2}}\right)^{k_o}; \]  

(B.3)

\[ U_x = \frac{r_d t_0 + \xi}{c_s a_m^2}; \quad V_x = \frac{\xi}{(c_s a_m)^{1/2}}. \]  

(B.4)

The limiting values of \( J_2 \) and \( J_3 \) are evaluated below for the four limits described in sections 2–6.

a. Limit 1: Small \( w \), diffusional growth regime

In this case, \( \lambda_s \ll 1 \); thus, the denonominator of the second fraction in the integrand of \( J_3 \) tends to unity, which corresponds to the power-law activity spectrum (3.1). We make additional approximations in (B.2), consider the case \( \alpha_c \sim 1 \), neglect small \( \xi \), and assume small \( r_d(t_0) \). Then \( U_x \) and \( V_x \) vanish, and the integral \( J_3 \) (B.2) is simplified and reduced to the Euler beta function \( B(x, z) \), as in Twomey’s model:

\[ J_3^{(1)} = \int_0^1 (1 - x)^{1/2} dx = B\left(\frac{k_o}{2}, \frac{3}{2}\right). \]  

(B.5)

b. Limit 2: Small \( w \), \( \alpha_c \ll 1 \)

When \( \alpha_c \ll 1 \), then \( \xi \gg r_d(t_0) \) and \( r_d(t_0) \) can be neglected in (2.15b) or (B.4) for \( U_x \); then \( U_x \approx \xi^2 / (a_m c_s d^2) = V_x^2 \). We also have in (B.2) for \( J_3 \) the inequalities \( (1 - z) < U_x \) and \( (1 - z) < V_x^2 \). Expansion of the first fraction in the integrand of \( J_3 \) in (B.2) by the small parameter \( (1 - z)V_x^2 \) yields

\[ \frac{(1 - z)^2}{4V_x^3} = \frac{(1 - z)^2 (c_s a_m)^{3/2} \lambda_s}{4\xi^3}. \]  

(B.6)

Substituting this expression for the first fraction into \( J_3 \), we obtain

\[ J_3^{(2)}(t) = \frac{(c_s a_m)^{3/2} \lambda_s^2}{4\xi^3} \int_0^1 (1 - z)^2 (1 + \lambda_s x^{k_o})^{2} z^{k_o/2 - 1} dx. \]  

(B.7)

For \( \lambda_s \ll 1 \), we can neglect in the denominator of the integrand in (B.7) the second term; that is, the activation spectrum is also approximately by the power law (3.1). Then, using (2.7) for \( \xi \):

\[ J_3^{(2)}(t) = \frac{(c_s a_m)^{3/2} \lambda_s^2}{4\xi^3} \int_0^1 z^{k_o/2 - 1} (1 - z)^2 dx. \]  

(B.8)

\[ = 4^{-4} D_v^{-3} V_x^4 a_c^{-2} (c_s a_m)^{3/2} \lambda_s B\left(\frac{k_o}{2}, \frac{3}{2}\right). \]  

(c. Limit 3: Large \( w \), diffusional growth regime

At large \( w \)—that is, in the limit \( s_m \gg s_0 (\lambda_c \gg 1) \)—the integral \( J_3 \) in (2.14b) or (B.2) in general cannot be reduced to the beta function, and it is more convenient to use \( J_3^{(3)}(t) \). Let us consider two functions in the subintegral expression for \( J_2 \) in (B.1):
$$\phi_{nd}(x) = \frac{x^{k_{x0} - 1}}{(1 + \lambda_s x^{k_{x0}})^2};$$

$$\psi(x) = \frac{\left\{ [U_s + (1 - x^2)]^{1/2} - V_s \right\}^2}{[U_s + (1 - x^2)]^{1/2}}. \quad (B.9)$$

Then $J_2$ can be written as

$$J_2(t) = \int_{0}^{1} \psi(x) \phi_{nd}(x) dx. \quad (B.10)$$

The function $\phi_{nd}(x)$ represents the algebraic CCN activity spectrum in nondimensional form. Its maximum evaluated from the condition $d[\phi_{nd}(x)]/dx = 0$ is located at

$$x_m = \left( \frac{1}{\lambda_s k_{x0} + 1} \right)^{1/k_{x0}}. \quad (B.11)$$

For example, if $k_{x0} = 2$, then $x_m \approx 0.1$ for $\lambda_s \sim 30$ and $x_m \approx 0.18$ for $\lambda_s \sim 10$ (i.e., $x_m \ll 1$ at $\lambda_s > 10$). It was shown in KC06 that this differential CCN activity spectrum has the shape of a “smoothed” Dirac delta function, and the nondimensional function $\phi_{nd}(x)$ also has this property. This is illustrated in Fig. B1, which shows that $\phi_{nd}(x)$ has a sharp maximum at $x = x_m$, while $\psi(x)$ is a smooth function. Thus, the major contribution to $J_2$ comes from the region around $x_m$, and we can use for evaluation of $J_2$ the “quasi $\delta$ function” property of $\phi_{nd}(x)$, or the “mean value theorem” from the mathematical analysis:

$$J_2^{(3)}(t) = \int_{0}^{1} \psi(x) \phi_{nd}(x) dx = \psi(x_m) \int_{0}^{1} \phi_{nd}(x) dx. \quad (B.12)$$

The last integral is not equal to 1 and still should be evaluated since $\phi_{nd}(x)$ is proportional to the smoothed delta function but not equal. To find this integral, we use the property found in KC06 that the CCN differential spectrum is a smoothed $\delta$ function and is a derivative of the droplet concentration defined in (2.22), which is a smoothed Heaviside step function, found in a similar form by Ghan et al. (1993, 1995). The corresponding fraction activated $F_{nd}(x) = N_{nd}(x)/N_a$ can be written from (2.22) in nondimensional form as

$$F_{nd}(x) = \frac{\lambda_s x^{k_{x0}}}{1 + \lambda_s x^{k_{x0}}}. \quad (B.13)$$

One can see that

$$\phi_{nd}(x) = \frac{1}{k_{x0} \lambda_s} \frac{dF_{nd}(x)}{dx}. \quad (B.14)$$

Thus,

$$\int_{0}^{1} \phi_{nd}(x) dx = \frac{1}{k_{x0} \lambda_s} \int_{0}^{1} \frac{dF_{nd}(x)}{dx} dx = \frac{1}{k_{x0} \lambda_s} \left[ F_{nd}(1) - F_{nd}(0) \right] = \frac{1}{k_{x0} \lambda_s (1 + \lambda_s)} \approx \frac{1}{k_{x0} \lambda_s}. \quad (B.15)$$

The last approximate equation uses the fact that $\lambda_s \gg 1$. With $\alpha_s \sim 1$ and $r_d(t_0) = 0$, the values $U_s \ll 1$ and $V_s \ll 1$, and $\psi(x_m) \approx (1 - x_m)$. Figure B1 shows that at $\lambda_s \gg 1$ and $x_m \ll 1$, the value of $\psi(x_m)$ is very close to 1—for instance, $\psi(x_m) = 0.997$ in this example, and thus it can be assumed that $\psi(x_m) = 1$. Substituting (B.15) into (B.12) with $\psi(x_m) = 1$ and using (2.15a) for $\lambda_s$ yields

$$J_2^{(3)} \approx \frac{1}{k_{x0} \lambda_s} = \frac{1}{k_{x0}} \left( \frac{s_0}{a m} \right)^{k_{x0}}. \quad (B.16)$$

The exact numerical calculation of $J_2$ with the functions $\phi_{nd}(x)$ and $\psi(x)$ shown in Fig. B1 yields $J_2^{(3)} \approx 1.726 \times 10^{-2}$, and calculation with approximation (B.16) and $\lambda_s = 29.7$, corresponding to Fig. B1, yields $J_2^{(3)} \approx 1.795 \times 10^{-2}$; that is, the error is less than 4%, so (B.16) can be used with good accuracy in this limit.
d. Limit 4: Large w (λs ≫ 1), kinetic growth regime (αc ≪ 1)

Now we consider the analytical evaluation of the integral \( J_2 \) in (2.14a) or (B.1) in the fourth limit, \( s_m \gg s_0 \) (\( λ_s \gg 1 \)), and \( α_c \ll 1 \). When \( α_c \ll 1 \), then \( ξ \gg r_d(t_0) \), and \( r_d(t_0) \) can be neglected in \( U_s \); then \( U_s = V_s^2 \); besides, \( (1 - x^2) ≪ U_s \), and \( (1 - x^2) ≪ \frac{1}{U_s} \). Similar to the third limit in the previous section, we present the integrand of \( J_2 (B1) \) as \( Ψ_{nd} (x) \phi (x) \) with functions defined in (B.9). Similar to the second limit, expansion of \( ψ(x) \) by the small parameter \( (1 - x^2)/V_s^2 \) yields

\[
\psi(x) \approx \frac{(1 - x^2)^2}{4V_s^2} = \frac{(a_m c_m x_m^{3/2})}{4s^3} (1 - x_m^2)^2 \frac{1}{k_{dl} \lambda_j^3}. \tag{B.17}
\]

As for the third limit, the function \( Ψ_{nd} (x) \) has a sharp maximum at \( x = x_m ≪ 1 \) when \( λ_s \gg 1 \), and \( ψ(x) \) is a smooth function. Using again the quasi-\( δ \)-function property of \( Ψ_{nd} (x) \) and the mean value theorem and substituting (B.17) into (B.12), accounting for (B.15), yields

\[
J^{(4)}_2 (t_m) \approx \frac{(a_m c_m x_m^{3/2})}{4s^3} (1 - x_m^2)^2 \frac{1}{k_{dl} \lambda_j^3}. \tag{B.18}
\]

Using (2.15a) for \( λ_s \), (2.7) for \( \xi \), and using again the relation \( (1 - x_m^2) = 1 \), \( J^{(4)}_2 \) becomes finally

\[
J^{(4)}_2 (t_m) \approx \frac{1}{128} \frac{a_m c_m x_m^{3/2}}{k_{dl} s_0 D_0^3}. \tag{B.19}
\]

REFERENCES


