Parameterization of cloud droplet formation in large-scale models: Including effects of entrainment
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[1] This work offers for the first time a comprehensive parameterization suitable for large-scale models which is robust, computationally efficient, and from first principles links chemical effects, aerosol heterogeneity and entrainment with cloud droplet formation. The parameterization is based on the entraining ascending parcel model framework; mixing of outside air is parameterized in terms of a per-length entrainment rate. The integration of the droplet growth is done using the “population splitting” concept of Nenes and Seinfeld (2003). Formulations for lognormal and sectional aerosol representations are given, as well as simplifications that allow the treatment of entrainment with high computational efficiency without loss of accuracy. The concept of “critical entrainment,” a value beyond which droplet activation is not favored, is introduced and shown that it is important for defining (1) whether or not entrainment effects have an impact on droplet formation and (2) the characteristic temperature and pressure for cloud droplet formation. The performance of the parameterization was evaluated against a detailed numerical parcel model over a comprehensive range of droplet formation conditions. The agreement is always very good (mean relative error 2.3% ± 21%); errors tend to increase as entrainment approaches the critical value, but are never above 40%.


1. Introduction

[2] Atmospheric aerosols strongly impact the radiative balance of the earth. They can either scatter and absorb thermal and solar radiation (“direct effect”) or alter the radiative properties and global distribution of clouds (“indirect effect”) [Twomey, 1977; Albrecht, 1989; Ackerman et al., 2000; Lohmann and Feichter, 2005]. The increase in cloud albedo from increases in cloud condensation nuclei, CCN, is commonly termed the “first indirect” or “Twomey” effect [Twomey, 1977]. Assessment of the first indirect effect requires the detailed description of the droplet formation process by activation of precursor aerosols (CCN). The ability of an aerosol to act as a CCN is controlled by its physicochemical properties [e.g., Pruppacher and Klett, 1997]; thus the aerosol size distribution and mixing state, the solubility and hygroscopicity of the aerosol components, the presence of soluble gases, the formation of surfactant layers that can change surface tension and water vapor uptake rate, as well as other mass transfer limitations (such as dissolution kinetics [Asa-Awuku and Nenes, 2007]) play an important role in the formation of new droplets [McFiggans et al., 2005]. Besides these factors, the dynamical (e.g., updraft velocity, mixing and cooling by entrainment) and thermodynamical state (temperature, pressure, relative humidity) of the cloudy air mass profoundly impact the droplet formation process [Pruppacher and Klett, 1997].

[3] Cloud droplet formation is simulated by solving a system of ordinary differential equations that represent the condensational growth of CCN coupled with conservation of heat and water vapor mass in an ascending air parcel [e.g., Pruppacher and Klett, 1997; Nenes et al., 2001; Snider et al., 2003]. Although this theory is well established, the computational burden associated with its solution precludes its application in large-scale general circulation models (GCMs). Instead, droplet activation is calculated from computationally efficient parameterizations whose sophistication ranges from empirical correlations (relating aerosol mass or number concentration and cloud droplet number concentration) [e.g., Boucher and Lohmann, 1995; Gultepe and Isaac, 1996] to physically based prognostic representations of the activation physics [Feingold and Heymsfield, 1992; Abdul-Razzak et al., 1998; Abdul-Razzak and Ghan, 2000; Cohard et al., 2000; Nenes and Seinfeld, 2003; Fountoukis and Nenes, 2005; Ming et al., 2006].

[4] All physically based parameterizations developed to date rely on the assumption that the droplet formation is an adiabatic process (with the exception of Feingold and Heymsfield [1992] who discuss how to include entrainment); thus conditions at cloud base uniquely determine the number of CCN that activate. Adiabaticity considerably

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simplifies the equations for cloud droplet formation so they can be solved analytically (or approximated by semiempirical correlations). However, entrainment during the parcel ascent may influence its temperature and cloud liquid water content; entrainment decreases cloud droplet number relative to adiabatic conditions [Pruppacher and Lee, 1976; Pontikis et al., 1987; Jonas, 1996]. After activation, the droplet spectra can be further modified by evaporation and mixing, with important implications for the cloud characteristics [Paluch, 1979; Rogers et al., 1985; Pontikis and Hicks, 1993; Jonas, 1996; Pruppacher and Klett, 1997; Su et al., 1998]. The importance of entrainment is corroborated by in situ observations which mostly show that the liquid water content measured is lower than expected by adiabaticity [e.g., Paluch and Baumgardner, 1989; Brenguier and Chauumat, 2001; Snider et al., 2003; Conant et al., 2004; Gerber et al., 2005; Meskhidze et al., 2005; Peng et al., 2005]. Neglecting entrainment may therefore lead to an overestimation of in-cloud droplet number and an underestimation of droplet spectral width, which if used in large-scale models, may bias indirect effect assessments. Despite its complexity, developing a physically based prognostic parameterization that can explicitly treat entrainment would be a powerful tool to study aerosol-cloud interactions within large-scale models, and is the objective of this study. The new parameterization is based on the frameworks of Nenes and Seinfeld [2003] and Fountoukis and Nenes [2005], where kinetic limitations, slow water vapor accommodation, and “chemical effects” are explicitly treated. Two formulations are presented, one for sectional and one for lognormal aerosol representation.

2. Description of the Parameterization

Calculated cloud droplet number concentration is accomplished in three steps. First, the critical entrainment rate, $e_v$ (the entrainment required to completely prevent activation), and the characteristic temperature and pressure for droplet formation are computed. Afterward, aerosol size distribution/chemical composition information is used to compute the “CCN spectrum” (the concentration of CCN as a function of cloud maximum supersaturation, $s_{\text{max}}$). Then $s_{\text{max}}$ and droplet number, $N_d$, are computed.

2.1. Representation of CCN Spectrum

Two representations of the aerosol size distribution and chemical composition are used to represent the CCN spectrum: the “sectional” and “lognormal” representations. The first is a “sectional representation,” in which the aerosol size distribution is divided into an arbitrary number of size classes (sections) each with a uniform profile of aerosol number concentration and chemical composition between its boundaries [Nenes and Seinfeld, 2003]. Using Köhler theory, the aerosol size distribution is mapped onto supersaturation space to obtain the critical supersaturation distribution function, $n'(s)$,

$$ n'(s) = \frac{dN}{ds} = \frac{N_i}{s_{c,i} - s_{c,i-1}} \quad s_{c,i} < s < s_{c,i-1} $$

where $N_i$ is the aerosol number concentration of section $i$, $s$ is supersaturation, and $s_{c,i}$ and $s_{c,i-1}$ represent the lower and upper bounds of each supersaturation bin, respectively. The “CCN spectrum,” $F'(s)$, is obtained by integration of equation (1) from $s' = 0$ to $s' = s$.

$$ F'(s) = \int_{0}^{s} n'(s')ds' = \sum_{i=1}^{n_m} \frac{N_i}{s_{c,i} - s_{c,i-1}} \left[ \frac{s - s_{c,i-1}}{s_{c,i} - s_{c,i-1}} \right] $$

where $s_{c,i}$ and $s_{c,i-1}$ are the critical supersaturations for the boundaries of section $i$, and population $i$. The sectional representation provides the most general description of the aerosol size distribution, as it can describe the presence of externally or internally mixed aerosol populations [Nenes and Seinfeld, 2003].

In the “lognormal representation,” we assume each lognormal population mode has a uniform chemical composition and the aerosol follows a lognormal size distribution. Following Fountoukis and Nenes [2005], $n'(s)$ and $F'(s)$, are given by,

$$ n'(s) = \frac{dN}{ds} = \sum_{i=1}^{n_m} \frac{2N_i}{3s_v\sqrt{2\pi} \ln \sigma_i} \exp\left[ -\frac{\ln^2(s_{c,i}/s)}{2\ln^2 \sigma_i} \right] $$

$$ F'(s) = \int_{0}^{s} n'(s')ds' = \sum_{i=1}^{n_m} \frac{N_i}{2} \operatorname{erfc}\left[ -\frac{\ln(s_{c,i}/s)}{3\sqrt{2} \ln \sigma_i} \right] $$

where $\sigma_i$ is the geometric standard deviation for lognormal mode $i$, $s_{c,i}$ is the critical supersaturation of a particle with a diameter equal to the geometric mean diameter of the mode $i$, and $n_m$ is the number of lognormal modes in the population.

If the maximum supersaturation, $s_{\text{max}}$, in the cloud parcel is known, then the maximum activated droplet number $N_d$ is given by

$$ N_d = F'(s_{\text{max}}) $$

2.2. Calculation of $s_{\text{max}}$

The rate of change of water vapor supersaturation, $s$, in a homogeneously mixed entraining parcel ascending with constant velocity $V$, can be written as [Seinfeld and Pandis, 1998]

$$ \frac{ds}{dt} = \alpha V + \gamma \frac{dw}{dt} + eV \left( \frac{\Delta H_w M_w}{RT^2} \right) \left( \frac{\Delta H_l}{c_p} \left( w - w' \right) + \left( T - T' \right) \right) $$

where, $\alpha = \frac{eM_e \Delta H_l}{c_p RT^2}$, $\gamma = \frac{pM_e}{\rho(T) M_w}$, and $e$ is the entrainment rate (m$^{-1}$), $w$ and $w'$ are the water vapor mixing ratios in the parcel and in the ambient, respectively,
and \( R \) is the universal gas constant. The water mass balance in the entraining parcel is [Prudduy and Klett, 1977; Seinfeld and Pandis, 1998]

\[
\frac{dw}{dt} = -\frac{dW}{dt} - eV(w + W - w')
\]

(7)

where \( W \) is the liquid water mixing ratio in the parcel, and \( \frac{dW}{dt} \) is the rate of condensation of liquid water onto the drops. Using (7) and assuming \( 1 + s \approx 1 \), equation (6) can be written as

\[
\frac{ds}{dt} = \alpha V - \gamma \left[ \frac{dW}{dt} + eVW \right] + eV \left[ \frac{\Delta H_s M_s}{RT^2} (T - T') - (1 - RH) \right]
\]

(8)

where \( RH \) is the ambient fractional relative humidity. The first term in the right hand side of equation (8) represents the tendency of supersaturation to increase from expansion (cooling); the second term expresses the decrease of supersaturation from depletion of water vapor by condensational growth of water droplets and by entrainment of liquid water during the parcel ascent. The third term is the combined effect of entraining dry air, where cooling from mixing at \( T' \) tends to increase the supersaturation, and mixing with subsaturated dry air decreases \( s \).

[10] The use of the entrainment rate to parameterize mixing implies a constant aerosol mixing ratio during the parcel ascent where the entrained aerosol have the same properties of the precloud aerosol; the short timescale of cloud formation precludes significant transformation or ageing of the aerosol during the activation event [Su et al., 1998]. Although a simplification, it is more realistic than adiabatic activation (which is what all current physically based parameterizations employ). The extension of our formulation to include such chemistry and vertically heterogeneous aerosol mixing is certainly possible, but beyond the scope of this manuscript.

[11] By definition, the liquid water mixing ratio, \( W \), is [Prudduy and Klett, 1977]

\[
W = \frac{\pi}{6} \frac{\rho_w}{\rho_d} \int_0^{s_{\text{max}}} D_p^5 n'(s')ds'
\]

(9)

where \( \rho_w \) and \( \rho_d \) are the liquid water and air densities, respectively, and \( D_p \) is the droplet wet diameter. From (9), the rate of water condensation on the droplet population is obtained,

\[
\frac{dW}{dt} = \frac{\pi}{2} \frac{\rho_w}{\rho_d} \int_0^{s_{\text{max}}} D_p^5 \frac{dD_p}{dt} n'(s')ds'
\]

(10)

neglecting curvature effects, equation (10) can be written as [Twomey, 1959; Nenes and Seinfeld, 2003]

\[
\frac{dW}{dt} = \frac{\pi}{2} \frac{\rho_w}{\rho_d} G_{\text{smax}} \int_0^{s_{\text{max}}} \left( D_p^5 \tau + 2G \int_{s_{\text{max}}}^{s_{\text{max}}} s(t)dt \right)^{1/2} n'(s')ds'
\]

(11)

with

\[
G = \frac{4}{\rho_v \rho_d} G_{\text{FP}} + \frac{\Delta H_v \rho_v}{k_v T} \left( \frac{\Delta H_s M_s}{RT} - 1 \right)
\]

(12)

where \( k_v \) is the thermal conductivity of air, \( D'_p \) is the water vapor mass transfer coefficient from the gas to droplet phase corrected for noncontinuum effects, computed as suggested by Fountoukis and Nenes [2005], \( \tau \) represents the time at which the parcel supersaturation exceeds the CCN critical supersaturation; for CCN that instantaneously activate, \( D_p(\tau) \) is equal to the CCN critical diameter, \( D_c = D_p(\tau) \) (where \( A = \frac{4\pi M_s}{\rho_s} \) and \( \sigma \) is the surface tension of the droplet at the point of activation [Nenes et al., 2001]). The maximum supersaturation, \( s_{\text{max}} \), controls the total number of droplets that can form and is computed from equation (8) by setting \( \frac{dW}{dt} = 0 \). Integrating (11) and substituting in (8) one obtains at \( s_{\text{max}} \),

\[
\alpha V + eV \left[ \frac{\Delta H_s M_s}{RT^2} (T - T') - (1 - RH) \right] = I_e(0,s_{\text{max}}) = 0
\]

(13)

where the second term in equation (13) is known as the “condensation integral” [Nenes and Seinfeld, 2003],

\[
I_e(0,s_{\text{max}}) = G_{s_{\text{max}}} \int_0^{s_{\text{max}}} \left( D_p^5 \tau + 2G \int_{s_{\text{max}}}^{s_{\text{max}}} s(t)dt \right)^{1/2} n'(s')ds' + eV \frac{\Delta H_s M_s}{RT^2} (T - T') - (1 - RH)
\]

(14a)

If \( I_e(0,s_{\text{max}}) \) is known, equation (13) can be solved for \( s_{\text{max}} \).

[12] The condensation integral (equation (14a)) can be simplified, as the term \( eVW \) in equation (8) in many cases can be neglected. Simulations suggest (not shown) that this simplification can be applied when in-cloud \( s_{\text{max}} < 0.8\% \) and \( e < 0.1c_v \); equation (8) then reduces to

\[
\frac{ds}{dt} = \left\{ \alpha + e \left[ \frac{\Delta H_s M_s}{RT^2} (T - T') - (1 - RH) \right] \right\} V - \gamma \left[ \frac{dW}{dt} \right] \]

(14b)

and \( I_e(0,s_{\text{max}}) \) would simplify to:

\[
I_e(0,s_{\text{max}}) = G_{s_{\text{max}}} \int_0^{s_{\text{max}}} \left( D_p^5 \tau + 2G \int_{s_{\text{max}}}^{s_{\text{max}}} s(t)dt \right)^{1/2} n'(s')ds'
\]

(14c)

[13] This simplified representation of entrainment is equivalent to using the adiabatic formulation of the parameterization [Nenes and Seinfeld, 2003; Fountoukis and Nenes, 2005] after modifying the factor \( \alpha \) in the expansion cooling term to include entrainment effects, i.e., \( \alpha = \frac{\rho M_s \Delta H_s}{c_p RT} - \frac{\rho M_v}{RT} + e \left[ \frac{\Delta H_s M_s}{RT^2} (T - T') - (1 - RH) \right] \).

2.3. Computing the Condensation Integral \( I_e(0,s_{\text{max}}) \)

[14] The “population splitting” concept of Nenes and Seinfeld [2003] can be used to approximate \( I_e(0,s_{\text{max}}) \) as the sum of two terms,

\[
I_e(0,s_{\text{max}}) = I_{e,1}(0,s_{\text{part}}) + I_{e,2}(s_{\text{part}}, s_{\text{max}})
\]

(15)
where \( I_{e,1}(0, s_{\text{part}}) \) represents the growth of CCN that either do not strictly activate or experience significant growth beyond their critical diameter [Nenes and Seinfeld, 2003], so that \( D_p^*(\tau) \ll 2G_{\text{max}} \int_{s(t)dt}^\infty s(t)dt \); \( 2G_{\text{max}} \int_{s(t)dt}^\infty s(t)dt \) can be evaluated using Twomey’s lower bound [Twomey, 1959] which after substitution gives for \( I_{e,1}(0, s_{\text{part}}) \),

\[
I_{e,1}(0, s_{\text{part}}) = \left( \frac{G}{\alpha F} \right)^{1/2} \int_0^{s_{\text{max}}} \left( s_{\text{max}}^2 - s^2 \right)^{1/2} \left( G s_{\text{max}} + eG \left( s_{\text{max}}^2 - s^2 \right) \right) n'(s') ds'
\]

(16)

\[ \text{where} \]

\[ u_{\text{part}} = \frac{\ln(\frac{s_{\text{part}}}{s_{\text{max}}})^2}{3\sqrt{2} \ln \sigma_i}; \quad u_{\text{max}} = \frac{\ln(\frac{s_{\text{part}}}{s_{\text{max}}})^2}{3\sqrt{2} \ln \sigma_i} \]

(20)

[15] \( I_{e,2}(s_{\text{part}}, s_{\text{max}}) \) represents the growth of CCN that are recently activated and have not grown significantly beyond their critical diameter, i.e., \( D_p^*(\tau) \gg 2G_{\text{max}} \int_{s(t)dt}^\infty s(t)dt \). With this, and assuming \( D_p^*(\tau) = D_e \), \( I_{e,2}(s_{\text{part}}, s_{\text{max}}) \) can be written as

\[
I_{e,2}(s_{\text{part}}, s_{\text{max}}) = \frac{N_{A_f}}{3\sigma_{g,t}} \left( G s_{\text{max}} \exp \left( \frac{9 \ln^2 \sigma_i}{8} \right) \right. \\
\cdot \left. \left( \frac{u_{\text{part}} - 3 \ln \sigma_i}{\sqrt{2}} \right) \right. \\
\cdot \left. \left( \frac{u_{\text{max}} - 3 \ln \sigma_i}{\sqrt{2}} \right) \right. \\
\cdot \left. \left( \frac{eV}{3} \right) \left( \frac{24 \ln^2 \sigma_i}{3 \sigma_{g,t}} \right) \right. \\
\cdot \left. \left( \frac{8 \ln^2 \sigma_i}{8} \right) \right. \\
\cdot \left. \left( \frac{u_{\text{part}} - 9 \ln \sigma_i}{\sqrt{2}} \right) \right. \\
\cdot \left. \left( \frac{u_{\text{max}} - 9 \ln \sigma_i}{\sqrt{2}} \right) \right) 
\]

(19)

[16] The “partitioning critical supersaturation,” \( s_{\text{part}} \), separates the two CCN populations expressed by equation (15), and is calculated using the “discriminant criterion” [Nenes and Seinfeld, 2003], i.e., the sign of \( \Delta = (s_{\text{max}}^2 - \frac{164\sigma_i V}{9\sigma_{\text{max}}})^{1/2} \); \( \Delta > 0 \) indicates a droplet growth regime primarily free from kinetic limitations, and \( \Delta < 0 \) a regime where kinetic limitations are dominant [Nenes and Seinfeld, 2003]. When \( \Delta \geq 0 \), \( s_{\text{part}} = s_{\text{max}} \left\{ \frac{1}{2} \left[ 1 + (1 - \frac{164\sigma_i V}{9\sigma_{\text{max}}})^{1/2} \right] \right\}^{1/2} \); when \( \Delta < 0 \), \( s_{\text{part}} \) is calculated using \( s_{\text{part}} = s_{\text{max}} \min \{ (1 - \frac{1074}{5} s_{\text{max}}^{-0.3824}) 1.0 \} \) [Nenes and Seinfeld, 2003].

### 2.3.1. \( I_e(0, s_{\text{max}}) \) for Lognormal Aerosol

[17] For lognormal aerosol, substitution of equation (3) into equations (16) and (17), and approximating \( (1 - \frac{s_{\text{part}}}{s_{\text{max}}})^{1/2} \approx (1 - \frac{1}{2} \frac{s_{\text{part}}}{s_{\text{max}}}) \) gives,

\[
I_{e,1}(0, s_{\text{part}}) = \left( \frac{G}{\alpha F} \right)^{1/2} \frac{N_f}{2} \left( G s_{\text{max}} \right. \\
\cdot \frac{1}{2} \left. \left( \frac{s_{g,t}}{s_{\text{max}}} \right) ^2 \exp \left( \frac{9 \ln^2 \sigma_i}{2} \right) \right. \\
\cdot \left. \left( \frac{3}{\sqrt{2}} \ln \sigma_i + u_{\text{part}} \right) \right. \\
\cdot \left. \left( \frac{27 \ln^2 \sigma_i}{2} \right) \right. \\
\cdot \left. \left( \frac{3}{\sqrt{2}} \ln \sigma_i + u_{\text{part}} \right) \right. \\
\cdot \left. \left( \frac{1}{2} \frac{s_{g,t}}{s_{\text{max}}} \right) \right. \\
\cdot \left. \left( \frac{27 \ln^2 \sigma_i}{2} \right) \right. \\
\cdot \left. \left( \frac{3}{\sqrt{2}} \ln \sigma_i + u_{\text{part}} \right) \right. \\
\cdot \left. \left( G s_{\text{max}} + eG \left( s_{\text{max}}^2 - s^2 \right) \right) \right. \\
\cdot \left. \left( \frac{1}{2} \right) \right) 
\]

(18)

\[
I_{e,2}(s_{\text{part}}, s_{\text{max}}) = \sum_{j=1}^{s_{\text{max}}+1} \left\{ \frac{2G_{\text{max}}}{3} \ln \frac{s_{j}}{s_{j-1}} \right\} \\
\cdot \left\{ \frac{N_{A_j}}{s_{j} - s_{j-1}} \right\} \\
\cdot \left\{ \frac{1}{s_{j}} \left( \frac{N_{A_{j}}}{s_{j} - s_{j+1}} \right) \right\} \\
\cdot \left\{ \frac{2G_{\text{max}}}{3} \ln \frac{s_{\text{max}}}{s_{\text{j}}} \right\} \\
\cdot \left\{ \frac{N_{A_{j}}}{s_{\text{j}} - s_{j-1}} \right\} \\
\cdot \left\{ \frac{1}{s_{j}} \left( \frac{N_{A_{j}}}{s_{j} - s_{j+1}} \right) \right\} \\
\cdot \left\{ \frac{2G_{\text{max}}}{3} \ln \frac{s_{\text{max}}}{s_{j}} \right\} \\
\cdot \left\{ \frac{N_{A_{j}}}{s_{\text{j}} - s_{j-1}} \right\}
\]

(22)
i is the sections that bound $s_{\text{part}}$ and $s_{\text{max}}$, respectively. When no entrainment is considered, equations (21) and (22) reduce to the solution of Nenes and Seinfeld [2003] for adiabatic conditions. Depending on the aerosol formulation used, $s_{\text{max}}$ is calculated by substituting (18) and (19), or (21) and (22), in (15) and then into (13), which is then solved using the bisection method. Using $s_{\text{max}}, N_d$ is computed from equation (5).

2.4. Critical Entrainment Rate

[20] Entrainment reduces $s_{\text{max}}$ (hence $N_d$) achieved during cloud formation. Therefore a “critical entrainment rate,” $e_c$, must exist, above which mixing of outside air prevents supersaturation and cloud droplet formation. If $e$ is below $e_c$, supersaturation develops and CCN activate into cloud droplets. $e_c$ is therefore a bound between two regions: one that forms a cloud, (where $s > 0$ and there is a maximum in the supersaturation profile for finite time), and another where $s < 0$ and no maximum supersaturation is reached. For $e > e_c$, $s$ is always negative; at $e_c$, $s_{\text{max}} = 0$ and $\frac{ds}{dt} = 0$ are reached at a finite time (Figure 1).

[21] The critical entrainment rate can be calculated from equation (8). Since at $e_c, s_{\text{max}} = 0$, then neglecting any water vapor condensation from the aerosol particles before supersaturation gives $\frac{\partial w}{\partial t} \approx 0$. At these conditions, $\frac{ds}{dt} = 0$ is reached when water availability from cooling via expansion and mixing at $T'$ balance the depletion from mixing with subsaturated air. With this, equation (8) can be solved for $e_c$ to give

$$e_c = \frac{\alpha}{(1 - RH) - \frac{\Delta H M_e}{RT^2}(T - T') + \gamma W}$$

(23a)

[22] Consistent with existing studies [i.e., Paluch, 1979; Paluch and Baumgardner, 1989; Raga et al., 1990; Grabowski and Pawlowka, 1993], $e_c$ is a strong function of the parcel thermodynamic state and its environment, (i.e., $T, RH, T'$). $W$ in equation (23a) is the mass mixing ratio of water just before supersaturation ($RH = 1.0$). An order of magnitude analysis of the terms in the denominator of equation (23a) (not shown) suggests that $\gamma W$ is of the order of $10^{-3}$ to $10^{-2}$ and is in most of the cases much smaller than the other terms (of order $10^{-5}$), so can be neglected

$$e_c = \frac{\alpha}{(1 - RH) - \frac{\Delta H M_e}{RT^2}(T - T') + \gamma W}$$

(23b)

equation (23b) is an excellent approximation of equation (23a) except in very polluted environment (aerosol mass > $10^5 \mu g m^{-3}$) and ambient conditions close to saturation ($RH > 0.95$ and $T - T' < 0.3^\circ C$); in such cases neglecting the term $\gamma W$ would introduce some bias in the calculation of $e_c$ and equation (23a) should be used.

2.5. Characteristic $T$ and $p$ for Droplet Formation

[23] To calculate $s_{\text{max}}$ and $N_d$, it is necessary to define “characteristic” conditions for droplet formation. Cloud base conditions (those at the lifting condensation level) are typically used since most of the droplets activate during the first stages of cloud formation [Pruppacher and Klett, 1997; Nenes et al., 2001]. Entrainment affects cloud base conditions by mixing of ambient (dry) air, so they become a function of the entrainment rate. It would however be desirable (and computationally efficient) to define characteristic $T$ and $p$ for droplet formation that does not depend on $e$. We choose $T$ and $p$ at $e = 0.98 e_c$, (since by definition, cloud base is not defined at $e = e_c$) as small temperature variations exert a large influence on $s_{\text{max}}$ as $e$ becomes closer to $e_c$ (shown below), whereas their influence on $s_{\text{max}}$ at adiabatic conditions is much lower. Selecting “characteristic” cloud formation conditions at $e = 0.98 e_c$ closely approximates the actual cloud base $p$ and $T$ for high $e$, where its influence on $s_{\text{max}}$ is more important.

[24] Cloud base conditions are computed from conservation of energy and water vapor mass for an unsaturated entraining parcel [Seinfeld and Pandis, 1998],

$$\frac{dT}{dz} = \frac{\delta T}{\delta z} + e(T - T')$$

(24)

$$\frac{dw}{dz} = e(\frac{M_p}{M} \frac{p(T')}{RH - w})$$

(25)

[25] The water mass and energy balances should be solved simultaneously with equation (23a) or (23b) (depending on the liquid water content and ambient conditions, section 2.4), to obtain $e_c$ and the characteristic $T$ and $p$ for droplet formation. If the difference between the parcel and the ambient temperatures, $T - T'$, is constant during the parcel ascent (constant lapse rate) then the ambient air will remain subsaturated assuring that other effects i.e., secondary activation due to isobaric mixing [e.g., Korolev and Isaac, 2000], or mixing with supersaturated environments [e.g., Pontikis et al., 1987] are second order.

2.6. Application of the Parameterization

[26] Application of the parameterization is presented in Figure 2. The input consists of the aerosol distribution
characteristics, the physicochemical properties of the aerosol population (i.e., density, composition, solubility, surfactant characteristics), the initial temperature, \(T_0\), and pressure, \(p_0\), entrainment rate, \(e\), the ambient temperature, \(T_0\), and relative humidity, \(\text{RH}\), and the water vapor uptake coefficient, \(\alpha_c\). Initial conditions \(T_0\) and \(p_0\) are used to calculate the characteristic \(T\) and \(p\) of the cloud and the critical entrainment rate, \(e_c\), by solving simultaneously equations (23) to (25). After \(e_c\) is determined, the CCN spectrum is computed using either equation (2) or (4).

\[ s_{\text{max}} = \frac{\alpha V + e V}{1 + \left(1 - \frac{16 \alpha V}{9 G s_{\text{max}}^3}ight)^{1/2}} \]

\[ s_{\text{max}} = \min\left\{ \frac{2 \cdot 1.04 A_{\text{BS24}}}{3}, 1.0 \right\} \]

\[ \frac{\Delta > 0}{\Delta < 0} \]

\[ s_{\text{max}} = \frac{\Delta}{9 G s_{\text{max}}^3} \]

\[ \gamma = \frac{\pi D_1^2}{2 D_0} \]

\[ N_d = F(s_{\text{max}}) \]

**3. Evaluation of the Parameterization**

**3.1. Conditions Considered**

The sectional and the lognormal formulations of the parameterization were evaluated against the numerical solution of a detailed numerical parcel model [Nenes et al., 2001] with explicit treatment of entrainment, equations (6)–(12) [Seinfeld and Pandis, 1998]. Four trimodal aerosol size distributions were selected (Table 1) that represent characteristic types of global aerosol (urban, clean continental, background and marine). The aerosol in all simulations is assumed to be composed of 50% by volume ammonium sulfate, and 50% insoluble material. In all numerical simulations, the aerosol is initially in equilibrium with the ambient relative humidity, \(\text{RH}\). \(T_0\) and \(\text{RH}\) were kept constant through the simulations. Initial parcel pressure and temperature were 1.013 kPa and 290 K, respectively. The water vapor mass uptake coefficient, \(\alpha_c\), was set to 0.06 as suggested by Meskhidze et al. [2005] and Fountoukis et al. [2007] to represent growth kinetics of ambient aerosols; runs were also done using \(\alpha_c = 1.0\).

The performance of the parameterization under adiabatic conditions has been extensively studied and evaluated against parcel model and in situ observations [Nenes and Seinfeld, 2003; Fountoukis and Nenes, 2005; Meskhidze et al., 2005].

**Table 1. Aerosol Lognormal Distributions Used in This Study [Whitby, 1978]**

<table>
<thead>
<tr>
<th>Aerosol Type</th>
<th>Nuclei Mode</th>
<th>Accumulation Mode</th>
<th>Coarse Mode</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(D_g)</td>
<td>(\sigma)</td>
<td>(N_i)</td>
</tr>
<tr>
<td>Marine</td>
<td>0.010</td>
<td>1.6</td>
<td>340</td>
</tr>
<tr>
<td>Continental</td>
<td>0.016</td>
<td>1.6</td>
<td>1000</td>
</tr>
<tr>
<td>Background</td>
<td>0.016</td>
<td>1.7</td>
<td>6400</td>
</tr>
<tr>
<td>Urban</td>
<td>0.014</td>
<td>1.8</td>
<td>106000</td>
</tr>
</tbody>
</table>

\(D_g\) is the modal diameter (\(\mu\)m), \(N_i\) is the number of dry particles (\(\text{cm}^{-3}\)), and \(\sigma\) is the geometric standard deviation. The size distributions refer to dry size with 50% insoluble fraction.
Table 2. Critical Entrainment Rate, $e_c$, and Characteristic $T$ and $p$
for Cloud Droplet Formation

<table>
<thead>
<tr>
<th>RH, %</th>
<th>$T' - T$, K</th>
<th>$e_c$, m$^{-1}$</th>
<th>$P$, Pa</th>
<th>$T$, K</th>
</tr>
</thead>
<tbody>
<tr>
<td>60</td>
<td>1.0</td>
<td>$1.69 \times 10^{-3}$</td>
<td>75,760</td>
<td>263.1</td>
</tr>
<tr>
<td>70</td>
<td>1.0</td>
<td>$2.21 \times 10^{-3}$</td>
<td>84,360</td>
<td>272.0</td>
</tr>
<tr>
<td>80</td>
<td>1.0</td>
<td>$3.55 \times 10^{-3}$</td>
<td>91,880</td>
<td>279.2</td>
</tr>
<tr>
<td>90</td>
<td>1.0</td>
<td>$11.6 \times 10^{-3}$</td>
<td>98,640</td>
<td>285.2</td>
</tr>
<tr>
<td>90</td>
<td>0.0</td>
<td>$2.45 \times 10^{-3}$</td>
<td>88,210</td>
<td>278.7</td>
</tr>
<tr>
<td>90</td>
<td>0.0</td>
<td>$4.66 \times 10^{-3}$</td>
<td>95,100</td>
<td>284.8</td>
</tr>
<tr>
<td>70</td>
<td>2.0</td>
<td>$3.05 \times 10^{-3}$</td>
<td>88,570</td>
<td>272.4</td>
</tr>
<tr>
<td>80</td>
<td>2.0</td>
<td>$6.45 \times 10^{-3}$</td>
<td>95,890</td>
<td>279.6</td>
</tr>
<tr>
<td>97</td>
<td>0.3</td>
<td>$3.62 \times 10^{-2}$</td>
<td>100,567</td>
<td>288.7</td>
</tr>
<tr>
<td>97</td>
<td>0.3</td>
<td>$1.78 \times 10^{-1}$</td>
<td>101,171</td>
<td>289.2</td>
</tr>
</tbody>
</table>

$^{a}$RH and $T' - T$ are assumed constant during parcel ascent. Initial temperature and pressure of the parcel are 290 K and 101,325 Pa, respectively.

[30] Adiabatic conditions ($e = 0$) set the upper limit for $s_{\text{max}}$ in the ascending parcel; as $e \to e_c$, $s_{\text{max}} \to 0$. The point at which $s_{\text{max}} = 0$ is calculated using equation (23b); an example of the resulting $s_{\text{max}}$ versus $e$ profiles is shown in Figure 3. It can be seen that $e_c$ does not depend explicitly on $V$ (although $e$ may [Paluch, 1979; Jonas, 1996]) or on the type of aerosol considered (equation (23b)). Figure 3 shows that the calculation of $e_c$ (equation (23b)) is in good agreement with the extrapolation of the $s_{\text{max}}$ versus $e$ curve from numerical results, to the point where $s_{\text{max}} = 0$ (hence $e = e_c$). However, obtaining a solution with the parcel model for $e \approx e_c$ becomes numerically challenging (owing to the stiffness of the system of equations (6)–(12) under such conditions); therefore comparisons are made for $e \leq 0.9e_c$. The parameterization does not exhibit such numerical issues and solutions are smooth up to 0.99$e_c$.

All simulations performed in this work show that most of the variation on $s_{\text{max}}$ and $N_d$ from entrainment occurs for $e > 0.1e_c$ (Figure 3).

3.4. Effects of RH and $T' - T'$

[31] It is important to evaluate the parameterization for a wide range of RH and $T'$. In all simulations (Table 3), the parameterization closely reproduced the results of the parcel model. Figure 4 shows $s_{\text{max}}$ versus $e$ (Figure 4, left) and $N_d$ versus $e$ (Figure 4, right) profiles for $T' - T'$ equal 0 and 1°C. Increasing $T' - T'$ tends to increase $e_c$ because cooling from mixing with ambient air increases $s$ (therefore a larger $e$ is required to prevent supersaturation), and offsets the effect of mixing with subsaturated air (which decreases $s$). Similarly, increasing ambient RH increases $e_c$ because a larger mixing rate is required to prevent supersaturation (not shown). Regardless of RH and $T' - T'$, Figures 3 and 4 clearly show that entrainment becomes important when $e > 0.1e_c$. The tendency of the parameterization to overpredict $s_{\text{max}}$ is discussed in section 3.5.

3.5. Evaluation of $s_{\text{max}}$ and $N_d$ Predictions

[32] Figure 5 presents $s_{\text{max}}$ calculated by the parcel model and the parameterization, for the four trimodal aerosol populations of Table 1 and conditions of Table 3. The parameterization closely reproduces the results of the parcel model (average error 26%, standard deviation 36%). The agreement is best for background aerosol at high RH (80 and 90%) whereas for marine and continental aerosols, an overprediction of above 25% can be found, which increases as RH decreases. This systematic overprediction is attributed to the expression used for representing droplet growth kinetics. Equation (16) for population 1 (in which $D_p^3(\tau) \ll 2G I_{s_{\text{max}}}(s(d)dt)$ tends to underestimate the size of droplets [Nenes and Seinfeld, 2003]. When most of the activated CCN are composed of such drops (e.g., where most CCN at the point of activation are either far above or way below their $D_c$) there is an underestimation of surface area for water vapor condensation. This means the condensation integral $I_{s_{\text{max}}}$ is underestimated; to compensate, $s_{\text{max}}$ must increase (i.e., be overestimated) to satisfy equation (13).

For marine aerosols, this occurs when most of the CCN are activated (i.e., $s_{\text{max}}$ is large) so overestimation of $s_{\text{max}}$ does not significantly impact $N_d$. However, for polluted urban
aerosol, where $s_{\text{max}}$ is low, most of the CCN are not activated, so overestimation in $s_{\text{max}}$ will lead to some overestimation in $N_d$.

[33] The parameterization closely reproduces the parcel model predictions at high $V$ and $RH$, and low to moderate entrainment (i.e., high to moderate $s_{\text{max}}$). When entrainment strongly impacts $s_{\text{max}}$ ($s_{\text{max}}$ is low, i.e., low $RH$, high $T - T'$,

Figure 3. $s_{\text{max}}$ profiles for (top left) background, (top right) marine, (bottom left) continental, and (bottom right) urban aerosols. Parcel model results are represented as solid lines, and parameterization results are given as dashed lines. $RH = 80\%$, $T - T' = 1.0$, $V = 0.1$ m s$^{-1}$ (bottom lines), 1.0 m s$^{-1}$ (middle lines) and 5.0 m s$^{-1}$ (top lines). $e_c$ for this set of runs is equal to $3.55 \times 10^{-1}$ m$^{-1}$.

Figure 4. (left) $s_{\text{max}}$ and (right) $N_d$ for marine aerosol. $T - T = 1^\circ\text{C}$, $RH = 80\%$, $V = 1.0$ m s$^{-1}$.
The parameterization tends to overpredict \( N_d \) which is more noticeable for the background and urban aerosols (Table 4). This is depicted in Figure 6; for low \( e \) values, most points fall around the 1:1 line, whereas for \( e \) values close to \( e_c \), \( N_d \) is slightly overestimated. Figures 6 and 7 show that the difference between \( N_d \) calculated by the parameterization by the parcel model is usually the largest (above 25%) when \( RH \) is low (60%) and \( T - T' \) (2°C) is high. It can be noticed as well in Figure 7 that overprediction in \( s_{max} \) found at high updraft velocities for marine aerosol (Figure 5) does not correspond to a \( N_d \) overprediction. Increasing the temperature difference \( T - T' \), will offset these effects; however the influence of \( RH \) dominates over the mixing at \( T' \).

The small value of the mean relative error (with reference to the parcel model results, Table 4) in parameterized \( N_d \) (2.3%), suggests that there are no significant systematic biases for droplet calculation. The standard deviation of the relative error is about 20% which is remarkably small given the diverse set of conditions considered in the evaluation, and the numerically challenging nature of entrainment.

All simulations up to this point were done for a “base case” value of \( \alpha_c = 0.06 \). We repeated the assessment

Table 4. Average Error (Standard Deviation) of Parameterized \( N_d \) Versus Parcel Model \( N_d \) for \( e < 0.9e_c \)

<table>
<thead>
<tr>
<th>Aerosol Type</th>
<th>All Data</th>
<th>( \Delta &lt; 0 )</th>
<th>( \Delta &gt; 0 )</th>
<th>All Data</th>
<th>( \Delta &lt; 0 )</th>
<th>( \Delta &gt; 0 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Background</td>
<td>4.1(23.4)</td>
<td>-8.0(9.8)</td>
<td>22.7(22.6)</td>
<td>138(148)</td>
<td>193(133)</td>
<td>50(28)</td>
</tr>
<tr>
<td>Marine</td>
<td>6.8(16.4)</td>
<td>4.7(11.7)</td>
<td>N/A</td>
<td>5(6)</td>
<td>6(5)</td>
<td>N/A</td>
</tr>
<tr>
<td>Continental</td>
<td>-4.0(15.7)</td>
<td>-3.8(12.8)</td>
<td>4.8(19.0)</td>
<td>44(49)</td>
<td>51(42)</td>
<td>17(10)</td>
</tr>
<tr>
<td>Urban</td>
<td>6.5(22.8)</td>
<td>6.1(9.08)</td>
<td>3.6(23.7)</td>
<td>688(714)</td>
<td>984(669)</td>
<td>410(560)</td>
</tr>
<tr>
<td>All aerosol</td>
<td>2.3(21.8)</td>
<td>-1.3(12.8)</td>
<td>9.4(24.4)</td>
<td>136(354)</td>
<td>81(171)</td>
<td>265(471)</td>
</tr>
</tbody>
</table>

Figure 5. \( s_{max} \) as predicted by the parameterization versus the parcel model. Simulations presented are for (top left) background, (top right) marine, (bottom left) continental, (bottom right) and urban aerosols. \( V = 0.1 \text{ m s}^{-1} \) (white), \( V = 1.0 \text{ m s}^{-1} \) (gray), and \( V = 5.0 \text{ m s}^{-1} \) (dark gray). \( T, T - T', RH, \) and \( p \) are presented in Tables 2 and 3. Dashed lines represent ±25% deviation.
for $\alpha_c = 1.0$, the results of which are shown in Figure 6. Increasing $\alpha_c$ intensifies water vapor condensation onto the drops; competition for water vapor increases, hence $s_{\text{max}}$ and $N_d$ decrease [Fountoukis and Nenes, 2005]. As expected, the general decrease in $s_{\text{max}}$ slightly increases $N_d$ error ($-16 \pm 9\%$) compared to the base case.

Further insight into the influence of kinetic limitations on the parameterization performance can be gained by analyzing the discriminant criterion, $\Delta$. Figure 8 shows that in most cases tested for marine and continental aerosols, the droplet growth is free from kinetic limitations ($\Delta > 0$) whereas for background and urban aerosols the growth regime is generally kinetically limited ($\Delta < 0$). Table 4 shows larger average error in $N_d$ for kinetic limited regimes; however, the difference in errors between the two regimes is not statistically significant (Table 4). $\Delta$ is essentially a function of $s_{\text{max}}$ (hence $e$), therefore it is important to investigate if $N_d$ error will be correlated with $s_{\text{max}}$ (Figure 9). It is noticeable that although there is no correlation between $s_{\text{max}}$ and the average relative error in $N_d$, the dispersion in the error is higher for low $s_{\text{max}}$. In all simulations shown in Figure 9, the threshold $s_{\text{max}}$ for dominantly kinetically limited CCN activation ($\Delta = 0$) ranges between 0.1% and 0.3% depending of the aerosol type, so it is clear that a larger discrepancy between the parameterization and the parcel model (although below 40%) may be expected for kinetically limited regimes. Figure 6 supports this, as most of the points outside the 25% deviation interval correspond to $e > 0.6\alpha_c$.

The parameterization was also assessed for systematic biases with respect to activation fraction (i.e., the fraction of aerosol that activate into droplets). This is an important test, as the CCN spectrum shape changes considerably from low to high activation ratios. Most of the results reside close to 1:1 line (not shown) for all aerosol types, a larger parcel model–parameterization discrepancy results at $V = 0.1 \text{ m s}^{-1}$, low RH (60 and 70%), and low $\alpha_c$ (i.e., low activation fractions). The dispersion of the data at low $V$ was about the same for the different kinds of aerosol considered, which suggested that the parameterization is not biased with respect to activation fraction.

### 3.6. Comprehensive Versus Simplified Parameterization

We assess the simplified version of the parameterization (based on using equation (14c)) by comparing its predictions of droplet number with that of the comprehen-
sive formulation (based on equation (14a)). The assessment is done for all the cloud drop formation conditions shown in Tables 1–3; in addition we also included a set of simulations for ambient $RH = 98\%$ and $T - T' = 0.3$. The results of the formulation intercomparisons are shown in Figure 10, which shows $N_d/N_{d,\text{adiabatic}}$ predicted by the comprehensive formulation versus $N_d/N_{d,\text{adiabatic}}$ from the simplified parameterization. When $N_d/N_{d,\text{adiabatic}} < 0.4$, the difference between the two formulations is negligible (not shown); for higher values of $N_d/N_{d,\text{adiabatic}}$, the simplified parameterization tends to overestimate $N_d$ when $V$ (i.e., $s_{\text{max}}$) and ambient $RH$ are high (Figure 10). This bias is not surprising, as neglecting the $eVW$ term in equation (8) eliminates the effects of entrained water, and affects $s_{\text{max}}$. The simulations suggest that the simplified parameterization can safely be used when $s_{\text{max}}$ does not exceed 0.8%.

### 3.7. Computational Requirements of Parameterization

The computational efficiency of the new parameterization is assessed by the CPU time required relative to the Fountoukis and Nenes [2005] parameterization; the latter is selected because the equations, code structure and algorithm are very similar to the new formulation. We compare the two parameterizations, for all the cases in Tables 1–3 which have $e = 0$. The inclusion of entrainment increased the CPU time by about 30%, most of which arises from the increased complexity of the equations (section 2.3), whereas the calculation of $e_c$ and characteristic $T$ and $p$ (section 2.5) has a minor impact. Compared to the parcel model, the parameterization is orders of magnitude faster; the difference becomes even larger as $e$ approaches $e_c$ (where a numerical solution becomes increasingly difficult). The CPU time of the simplified parameterization is about the same as the adiabatic formulation of Fountoukis and Nenes [2005]; this is expected as the two parameterizations share the same expression for the condensation integral (section 2.3).

### 4. Conclusions and Summary

This study presents a physically based prognostic parameterization of droplet formation in which entrainment effects are explicitly considered. The parameterization is the first of its kind and it extents the works of Nenes and Seinfeld [2003] and Fountoukis and Nenes [2005]. The concept of “population splitting” (Nenes and Seinfeld, 2003) was used to obtain a closed solution for maximum supersaturation and cloud droplet number. Two formula-

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**Figure 7.** Cloud droplet number concentration as predicted by the parameterization and the parcel model. Simulations presented are for (top left) background, (top right) marine, (bottom left) continental, and (bottom right) urban aerosols. Nomenclature and conditions are the same as in Figure 5.
tions are presented, one for sectional and one for lognormal aerosol size distribution. Entrainment of ambient air is parameterized in terms of a per-length entrainment rate which implicitly assumes that all mixing processes occur homogeneously, at a timescale much faster than that of activation. Therefore the model presented here implies a lateral mixing mechanism, and instantaneous mixing of the entrained air. Since this work is an extension of the Nenes and Seinfeld [2003] and Fountoukis and Nenes [2005] parameterizations, by design it can treat the effects of externally mixed aerosol, CCN containing surfactants and slowly growing droplets (expressed by changes in the water vapor mass transfer coefficient). Even slightly soluble compounds can easily be accounted for during the computation of $F(s)$, and $n'(s)$.

[41] We also introduced the concept of critical entrainment rate, a value at which mixing completely prevents activation. The critical entrainment rate was shown to largely influence the characteristics of parcels experiencing intense mixing, and it is useful to determine whether or not entrainment effects are important for droplet number calculations. The critical entrainment rate is also useful for determining the characteristic $T$ and $p$ for cloud droplet formation.

**Figure 8.** Same as Figure 7 but classified according to the discriminant criterion and $e < 0.75 e_c$. Open symbols are for $\Delta < 0$ (kinetically limited regime), and solid symbols are for $\Delta \geq 0$ (lack of significant kinetic limitations).

**Figure 9.** Relative error in cloud droplet number concentration as function of $s_{\text{max}}$, for the data presented in Figures 7 and 5 ($e < 0.75 e_c$). Crosses indicate kinetically limited regimes ($\Delta < 0$), and dots denote lack of significant kinetic limitations ($\Delta \geq 0$).
The new parameterization was extensively tested against a detailed entraining numerical parcel model. The evaluation was considered for a wide range of relative humidity, entrainment rate, updraft velocity, water vapor uptake coefficient, ambient temperature, and aerosol characteristics. The parameterization closely followed the parcel model results with a mean relative error below 3% and mostly within 20%. The new parameterization requires approximately 30% more CPU time than the adiabatic formulation.

Finally, we also have proposed simplified version of the parameterization which for most conditions of cloud droplet formation, predict droplet with the same accuracy as the comprehensive formulation. This simplified formulation requires effectively the same CPU time as the Fountoukis and Nenes [2005] and Nenes and Seinfeld [2003] adiabatic droplet formation parameterizations.

In summary, the new parameterization framework offers for the first time a comprehensive and computationally efficient treatment of cloud droplet formation that can couple complex chemical effects on aerosol activation together with entrainment. The parameterization is ideally suited for large-scale regional and global models, with explicit aerosol-cloud interactions.

Acknowledgments. This study was supported by NASA MAP, NASA EOS-IDS-CACTUS, and a NASA New Investigator Award. The thoughtful comments of two anonymous reviewers are greatly appreciated.

References

Figure 10. Comparison between the simplified and comprehensive parameterizations. Simulations presented are for (top left) background, (top right) marine, (bottom left) continental, and (bottom right) urban aerosols. \( V = 0.1 \text{ m s}^{-1} \) (white), \( V = 1.0 \text{ m s}^{-1} \) (gray), and \( V = 5.0 \text{ m s}^{-1} \) (dark gray). All other conditions are presented in Tables 2 and 3.


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