

A New Parameterization of Cloud Droplet Activation Applicable to General Circulation Models

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ABSTRACT

A new parameterization is proposed to link the droplet number concentration to the size distribution and chemical composition of aerosol and updraft velocity. Except for an empirical assumption of droplet growth, the parameterization is formulated almost entirely on first principles to allow for satisfactory performance under a variety of conditions. For a series of updraft velocity ranging from 0.03 to 10.0 m s⁻¹, the droplet number concentrations predicted with the parameterization are in good agreement with the detailed parcel model simulations with an average error of $-4 \pm 26\%$ (one standard deviation). The accuracy is comparable to or better than some existing parameterizations. The parameterization is able to account for the effects of droplet surface tension and mass accommodation coefficient on activation without adjusting the empirical parameter. These desirable attributes make the parameterization suitable for being used in the prognostic determination of the cloud droplet number concentration in general circulation models (GCMs).

1. Introduction

Serving as nucleating sites for water vapor, aerosol particles may be activated into droplets when a rising air parcel reaches supersaturation, giving rise to various types of clouds in the atmosphere. On the global scale, anthropogenic aerosols indirectly affect the earth's radiation balance through modifying cloud properties. One of the key linkages in aerosol–cloud interaction is the cloud droplet number concentration, as it plays an important role in determining the effective radius (albedo) and precipitation rate (lifetime) of clouds. Recent global-scale simulations showed that both albedo and lifetime effects of aerosols (also termed as the first and second indirect effects, respectively) exert negative radiative forcing (cooling), though considerable divergences in its magnitude still persist among studies due to the generally poor characterization of aerosol–cloud interaction in general circulation models (GCMs) (Houghton et al. 2001).

Cloud droplet activation is a highly nonlinear process in nature. Only a fraction of particles can grow beyond

their critical sizes to form droplets depending on size distribution, chemical composition, and updraft velocity, while the unactivated ones exist as interstitial particles. An example is that the presence of surface-active organic components enhances activation through decreasing droplet surface tension (Facchini et al. 1999). In addition, the growth of a particle may lag behind the equilibrium path due to inadequate water condensation, thus imposing kinetic limitation on, and further complicating, the activation process (Chuang et al. 1997). As a standard tool, numerical integration of the analytical equations governing droplet activation (usually in the form of Lagrangian parcel simulation) has been widely utilized to study various aspects of droplet activation (e.g., Ming and Russell 2004). However, from the perspective of a GCM, such a detailed simulation incurs expensive computational cost and is infeasible. Therefore, computationally efficient, yet accurate, approaches must be developed to represent the activation process in GCMs.

In the last few years, some GCMs (e.g., Ghan et al. 1997; Lohmann et al. 1999) migrated from diagnosing the cloud droplet number concentration solely from aerosol mass using empirical relationships (e.g., Boucher and Lohmann 1995) to prognostic treatment by explicitly accounting for the sources and sinks of

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droplets. As a major source, droplet activation occurs both in stratiform and in convective clouds, and its rate was calculated with the parameterizations of Chuang and Penner (1995) and Abdul-Razzak and Ghan (2000). These formulations rely heavily on correlating empirical parameters from parcel model results. Though both parameterizations can be satisfactorily applied within the tested ranges, their generality is seriously impaired as a result of empirical fitting. For example, because all the published parameters were based specifically on data for ammonium sulfate aerosol, the parameterizations are not applicable to organic aerosol, whose activation efficiency is significantly different.

As one of the early attempts to develop parameterizations based on first principles, Twomey (1959) applied a lower bound of particle growth to power-law cloud condensation nuclei (CCN) spectra and derived an analytical expression for the cloud droplet number concentration. Though this approach completely eliminates the need for empirical parameters, its applicability to kinetically limited activation is unclear. Built on the novel concept of population splitting (differentiating droplets close to the critical diameters from those much larger), the parameterization of Nenes and Seinfeld (2003) substantially reduced the reliance on correlation by achieving a reliable prediction of the droplet number concentration in some conditions without resorting to empirical parameters and was able to successfully handle the surface tension effect of organic aerosol (Facchini et al. 2000). Efforts have been underway to apply it to deal with the variations in microphysical parameters, such as mass accommodation coefficients, and are described in a recently published paper (Fountoukis and Nenes 2005).

The study proposes a new parameterization, which is formulated almost entirely on first principles except for an empirical assumption of droplet growth. Its accuracy is assessed by comparison with parcel model results and other physically based parameterizations. Its capability in handling the kinetics of the activation process is also discussed.

2. Description of parameterization

For a closed adiabatic air parcel at an updraft velocity V , the supersaturation, s , varies with time, t , following (Leitch et al. 1986)

$$\frac{ds}{dt} = \alpha V - \gamma \frac{dW}{dt}, \quad (1)$$

where W is the liquid water content (LWC) and dW/dt is the condensation rate of water. The coefficients α and γ are

$$\alpha = \frac{gM_w\Delta H_v}{C_{pa}RT^2} - \frac{gM_a}{RT} \quad (2)$$

$$\gamma = \frac{RT}{p_s M_w} + \frac{M_w\Delta H_v}{C_{pa}pM_a T}, \quad (3)$$

where the physical parameters include the parcel temperature T , the universal gas constant R , the gravitational constant g , the molecular weight of water M_w , the latent heat of water ΔH_v , the heat capacity of air C_{pa} , the molecular weight of air M_a , and the saturated vapor pressure of water p_s . It is clear from Eq. (1) that, under the regulation of mass and energy balance, the supersaturation is driven by the adiabatic cooling of the parcel (the first term on the rhs), while being suppressed by the depletion of water vapor due to condensation (the second term). At maximum parcel supersaturation, s_{\max} , $ds/dt = 0$, leading to the following expression for the condensation rate:

$$\frac{dW}{dt} = \frac{\alpha V}{\gamma}. \quad (4)$$

For a droplet with a diameter of D_p , the difference between the parcel supersaturation, s , and the equilibrium supersaturation of the droplet, s_{eq} , determines the direction and scale of water mass transport between the vapor and droplet (liquid) phases:

$$\frac{dD_p}{dt} = \frac{G(D_p)}{D_p} (s - s_{\text{eq}}). \quad (5)$$

As a function of D_p , the growth coefficient, G , is related to the mass and heat accommodation coefficients (α_C and α_T , respectively) and the vapor and thermal jump coefficients (Δ_v and Δ_T , respectively) [see Eqs. (13.14), (13.20), and (13.28) of Pruppacher and Klett (1997)].

The integral form of Eq. (5) is formulated as

$$D_p^2 = D_p^2(\tau) + 2 \int_{\tau}^t G(D_p')(s - s_{\text{eq}}) dt', \quad (6)$$

which depicts the continuous growth of a droplet starting at a diameter of $D_p(\tau)$ at time τ and subsequently growing to D_p at time t .

Analogous to the lower bound for droplet growth proposed by Twomey (1959), the integral term in Eq. (6) is hypothetically expressed as

$$\int_{\tau}^t G(D'_p)(s - s_{\text{eq}}) dt' = \frac{G[D_p(\tau)]}{2\alpha V} (s_t^k - s_{\tau}^k), \quad (7)$$

where s_{τ} and s_t are the parcel supersaturation at time τ and t , respectively, and k is an empirical coefficient that controls the kinetic droplet growth. Equation (7) evolves into Twomey's lower bound when $k = 2$.

If the critical droplet diameter D_{pc} based on the Köhler equation is chosen to be the initial droplet diameter $D_p(\tau)$, D_p at the maximum supersaturation of s_{max} , $D_{p\text{max}}$, is derived from Eqs. (6) and (7):

$$D_{p\text{max}}^2 = D_{pc}^2 + \frac{G(D_{pc})}{\alpha V} (s_{\text{max}}^{2.4} - s_c^{2.4}), \quad (8)$$

where s_c is the critical supersaturation; k is the only empirical parameter of the parameterization. The value (2.4) is chosen based on fitting parcel model simulations (see the next section) and reflects the fact that the actual droplet diameter at s_c may be less than D_{pc} owing to kinetic limitation. This necessitates a growth term even smaller than Twomey's lower bound to compensate for the overestimation of the initial diameter. A precise treatment of the size dependence of G requires averaging over droplet growth from D_{pc} to $D_{p\text{max}}$. Because the growth path cannot be determined, Eq. (8) approximately takes into account size dependence by evaluating G at D_{pc} .

For a population of aerosol, whose size distribution is characterized by the number concentration, $n(D_{pd})$, of particles with a dry diameter between D_{pd} and $D_{pd} + dD_{pd}$, the collective condensation rate can be expressed as

$$\frac{dW}{dt} = \frac{\pi}{2} \rho_w \int_{D_{\text{min}}}^{D_{\text{max}}} D_p^2 \frac{dD_p}{dt} n(D_{pd}) dD_{pd}, \quad (9)$$

where D_{min} and D_{max} are the lower and upper bounds of the dry aerosol size distribution, respectively.

Bringing Eq. (5) into Eq. (9) results in the following expression for the condensation rate at s_{max} :

$$\frac{dW}{dt} = \frac{\pi}{2} \rho_w \int_{D_{\text{min}}}^{D_{\text{max}}} G[D_{p\text{max}}(D_{pd})](s_{\text{max}} - s_{\text{eq}}) D_{p\text{max}}(D_{pd}) \times H(D_{p\text{max}} - D_{pc}) n(D_{pd}) dD_{pd}. \quad (10)$$

The Heaviside function $H(D_{p\text{max}} - D_{pc})$ implies that only the droplets that exceed D_{pc} and undergo free growth contribute to condensation at s_{max} , an assumption that can be justified on the ground that since the

particles whose $D_{p\text{max}}$ is smaller than D_{pc} are unactivated, their sizes are determined only by the ambient supersaturation, and thus must be stationary at s_{max} ($ds/dt = 0$). So, the contribution to the condensation rate is zero. Also note that Eq. (10) evaluates G exactly at $D_{p\text{max}}$ and s_{eq} is calculated from $D_{p\text{max}}$ using the Köhler equation. This counteracts possible overestimation of the size dependence of G due to evaluating G at D_{pc} in Eq. (8).

For a sectional representation of size distribution, Eq. (10) can be rewritten as

$$\frac{dW}{dt} = \frac{\pi}{2} \rho_w \sum_{j=1}^J G(D_{p\text{max}j})(s_{\text{max}} - s_{\text{eq}j}) D_{p\text{max}j} \times H(D_{p\text{max}j} - D_{pcj}) N_j, \quad (11)$$

where J is the total number of size sections; $D_{p\text{max}j}$, D_{pcj} and N_j are the droplet diameter at s_{max} , critical droplet diameter and total number concentration for section j , respectively.

Based on the bisection method, the implementation of the parameterization begins with initial lower (s_L) and upper (s_U) limits of s_{max} , which are set to 0 and 50%, respectively, in this study. Then at a guess of $s_{\text{max}} = (s_L + s_U)/2$, the droplet diameter $D_{p\text{max}}$ can be calculated using Eq. (8). The resulting $D_{p\text{max}}$ is brought into Eq. (10) to evaluate the condensation rate dW/dt , which is then compared to the target rate determined by Eq. (4). Either s_L or s_U is reset to s_{max} depending on whether dW/dt is lower or higher than the target value. The above steps are iterated until the calculated dW/dt is sufficiently close to the target value. The particles, whose critical supersaturation is lower than the calculated maximum supersaturation, are counted as activated droplets (Nenes et al. 2001). Our calculations show that s_{max} usually converges within six iterations. This represents a substantial saving in computer time, as detailed parcel model simulations often need hundreds of iterations of comparable complexity.

3. Results

a. Comparison with parcel model

With the purpose to assess the performance of the new parameterization, the predicted cloud droplet number concentrations are compared with the detailed simulations with an adiabatic parcel model, which implements complete aerosol and cloud microphysics (Russell and Seinfeld 1998) and was utilized to study the roles of organic aerosol in the formation of marine clouds (Russell et al. 2000) and polluted fogs (Ming and

TABLE 1. Size distribution parameters (N : number concentration in cm^{-3} ; D_g : mean diameter in μm ; σ : standard deviation) of single-modal (SM) and trimodal (TM) test cases. All TM distributions are from Whitby (1978); D_{\min} and D_{\max} are the lower and upper bounds of dry diameter in μm .

Test case	Mode 1			Mode 2			Mode 3			D_{\min}	D_{\max}
	N_1	$D_{g,1}$	σ_1	N_2	$D_{g,2}$	σ_2	N_3	$D_{g,3}$	σ_3		
SM1	200	0.02	2.5							0.001	5
SM2	1000	0.02	2.5							0.001	5
SM3	1000	0.02	1.5							0.005	0.1
SM4	200	0.2	2.5							0.03	15
SM5	10 000	0.02	2.5							0.001	5
TM1-M	340	0.010	1.6	60	0.070	2.0	3.1	0.62	2.7	0.003	100
TM2-M	680	0.010	1.6	120	0.070	2.0	6.2	0.62	2.7	0.003	100
TM1-C	1000	0.016	1.6	800	0.068	2.1	0.72	0.92	2.2	0.003	40
TM2-C	2000	0.016	1.6	1600	0.068	2.1	1.44	0.92	2.2	0.003	40
TM1-B	6400	0.016	1.7	2300	0.076	2.0	3.2	1.02	2.16	0.003	40
TM2-B	12 800	0.016	1.7	4600	0.076	2.0	6.4	1.02	2.16	0.003	40
TM1-U	106 000	0.014	1.8	32 000	0.054	2.16	5.4	0.86	2.21	0.003	40
TM2-U	212 000	0.014	1.8	64 000	0.054	2.16	10.8	0.86	2.21	0.003	40

Russell 2004). A set of test cases featuring combinations of various size distributions of aerosol and a wide range of updraft velocity are used to examine the effectiveness of the parameterization in handling these two key variables in the activation process. To facilitate the intercomparison among existing parameterizations, the size distributions of the test cases employed by Nenes and Seinfeld (2003) are largely retained and are summarized in Table 1 for the completeness of this work.

Single-modal cases SM1, SM2, and SM5 share the same mean dry diameter and standard deviation, while the total number concentration increases by as much as 50 times. The standard deviation of SM3 (1.5) is smaller than that of SM2 (2.5), representing a much narrower size distribution. The mean diameter shifts from 0.02 μm of SM1 to 0.2 μm of SM4, representing a 1000 time increase in dry aerosol mass, as the number concentration remains unchanged.

Trimodal cases TM1-M, TM1-C, TM1-B, and TM1-U are representative of marine, clean continental, average background, and urban aerosols, respectively (Whitby 1978). Each distribution consists of a nucleation, an accumulation, and a coarse mode. In the corresponding TM2 cases, the number concentration of each mode is arbitrarily doubled to strengthen kinetic limitations.

Each test case is run at six updraft velocities (0.03, 0.1, 0.5, 1.0, 5.0, and 10.0 m s^{-1}), which cover the whole range encountered in stratiform (a few cm s^{-1}) and convective (a few m s^{-1}) clouds. All particles are assumed to be composed of pure ammonium sulfate $[(\text{NH}_4)_2\text{SO}_4]$. The same physical parameters are used both for the parameterization and for the parcel model. Some key ones are the parcel temperature T (283 K),

the parcel pressure p (800 mbar), the mass accommodation coefficient α_C (1), and the heat accommodation coefficient α_T (0.96) (Abdul-Razzak and Ghan 2000). The lower and upper bounds of dry aerosol distribution (D_{\min} and D_{\max}) in Eq. (10) are carefully chosen so that the 200 logarithmically spaced equal size bins between them cover more than 99% of aerosol number and mass (Table 1).

The droplet number concentrations predicted with the parameterization are compared with the parcel model simulations in Fig. 1. Over a wide range of the concentrations that spans three orders of magnitude (10–20 000 cm^{-3}), the parameterization closely tracks the detailed simulations. The data points lie evenly above and below the 1:1 line. The deviations from the 1:1 line are minimal when the concentrations are approximately higher than 100 cm^{-3} . By contrast, at velocities less than 0.1 m s^{-1} , both predicted and simulated concentrations are lower than 100 cm^{-3} . In these conditions, while underestimating the concentrations for TM1-M and TM2-M, the parameterization slightly overestimates for cases SM1, SM2, and SM5, showing no sign of systematic errors. Despite the increase in the total particle number concentration, the parameterization handles cases SM1, SM2, and SM5 equally well, overpredicting the droplet number concentrations by 14%. It shows no difficulty in dealing with the narrow size distribution in case SM3 and the high aerosol mass in case SM5 with an average error of 6%. The statistics of all cases yield an average error of -4% with a standard deviation of 26%.

The activation ratio is defined as the fraction of particles that are activated. Interestingly, the activation ratios from the parameterization and parcel model simu-

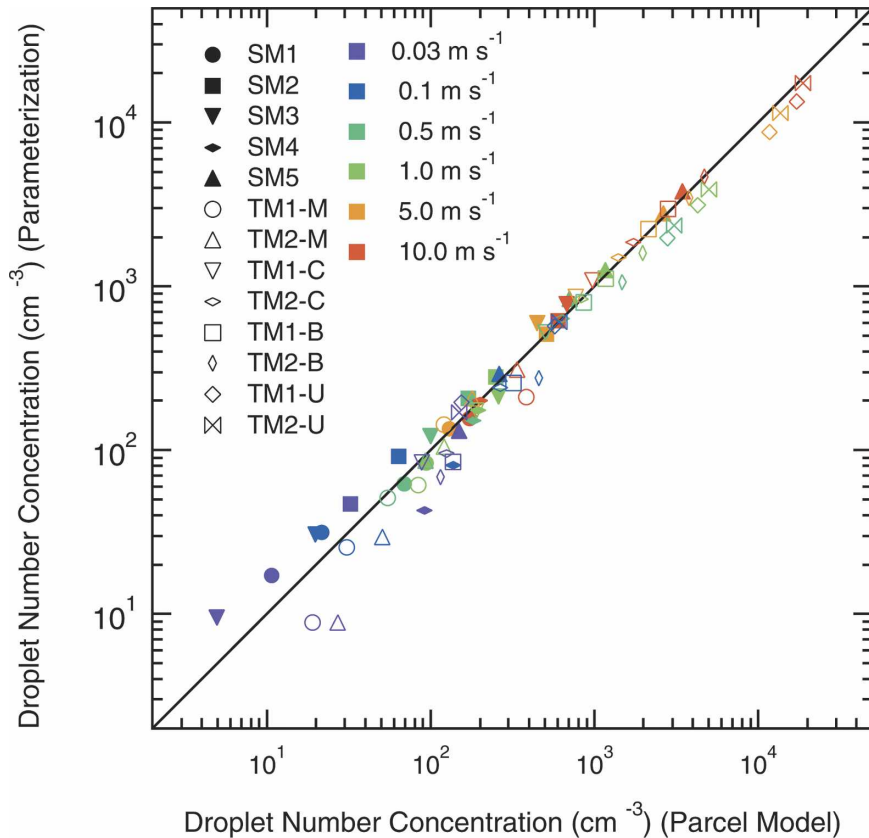


FIG. 1. Comparison of the droplet number concentrations predicted with the parameterization and calculated with the parcel model.

lations are in excellent agreement when either relatively strong (ratio >0.1) or very weak activation (ratio <0.004) occurs (Fig. 2). The errors of the parameterization at low droplet number concentrations ($<100 \text{ cm}^{-3}$) all correspond to intermediate activation ratios.

b. Sensitivity of parameterization

A desirable quality of a parameterization is insensitivity to empirical parameters to a certain extent, thus allowing it to be reliably applied to untested conditions. As the only empirical parameter of the new parameterization, k in Eq. (7) controls droplet growth, and subsequently affects the calculated condensation rate and maximum supersaturation. Higher values of k impede droplet growth and lower condensation rates, thus resulting in higher maximum supersaturation and droplet number concentrations.

A sensitivity analysis is made through running the parameterization at a series of k values between 2.0 and 3.0. The percentage errors are shown in Fig. 3. The predicted droplet number concentrations at $k = 2.0$ and 3.0 are compared with the parcel model simulations in

Fig. 4. The parameterization at $k = 2.0$ consistently underestimates the droplet number concentrations as a result of overestimation of droplet growth, representing an average decrease of 11% compared to the base case at $k = 2.4$. At $k = 3.0$, the droplet growth is less significant than in the base case and enhances the droplet number concentrations by 22% over the base case. The analysis also shows that varying k from 2.3 to 2.5 causes only a difference of less than 3% relative to the base case. Therefore, it does not require fitting the parcel model results extensively for the parameterization to achieve satisfactory performance.

4. Discussion

a. Comparison with other parameterizations

The analytical formula for the droplet number concentration derived by Twomey (1959) is based on the assumption that, once a particle is activated [i.e., the parcel supersaturation exceeding the supersaturation required for activation (s_a), as prescribed by power-law CCN spectra], the droplet size is much greater than that

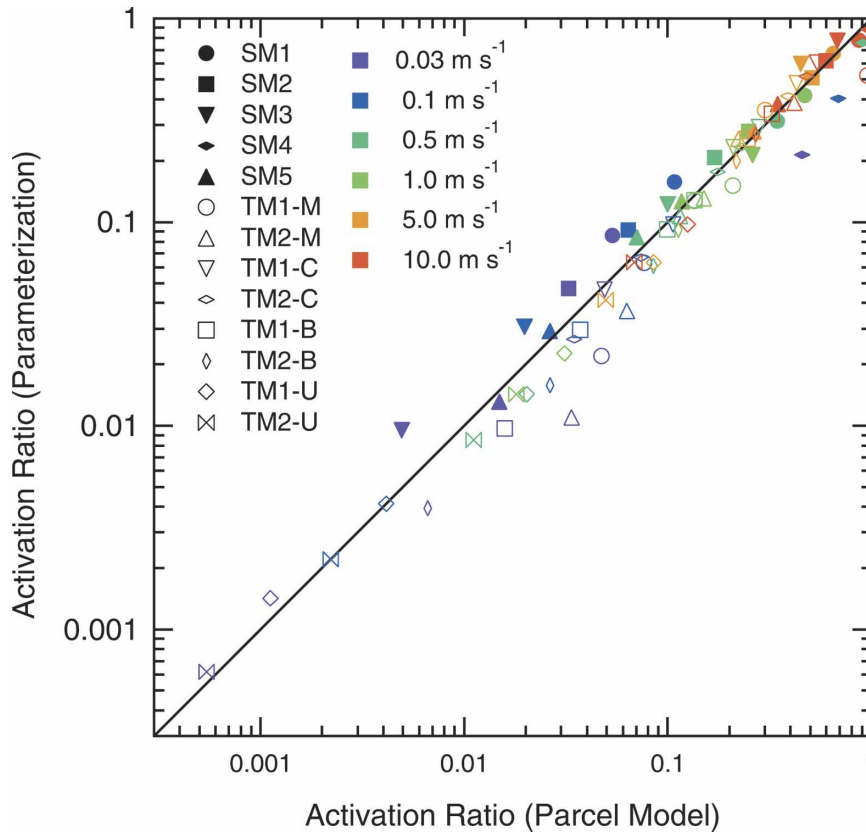


FIG. 2. Comparison of the activation ratios predicted with the parameterization and calculated with the parcel model.

at s_a and thus is determined solely by water condensation:

$$D_{pmax}^2 = \frac{G}{\alpha V} (s_{max}^{2.0} - s_a^{2.0}). \tag{12}$$

If the same assumption is applied to Eq. (8) and the critical supersaturation of a particle (s_c) is known from chemical composition, then

$$D_{pmax}^2 = \frac{G(D_{pc})}{\alpha V} (s_{max}^{2.4} - s_c^{2.4}). \tag{13}$$

To examine the validity of Twomey’s assumption, Eq. (13) is used to substitute for Eq. (8) of our parameterization. By keeping the other equations the same, we are able to formulate a new parameterization, which is similar to Twomey’s formula in the case of power-law CCN spectra. It is clear from Fig. 5 that the positive biases of the new parameterization propagate with increasing aerosol loading and decreasing updraft velocity, both factors contributing to kinetic limitation on the activation process. The average error is more than a factor of 2 for all TM cases. A comparison of Eqs. (13)

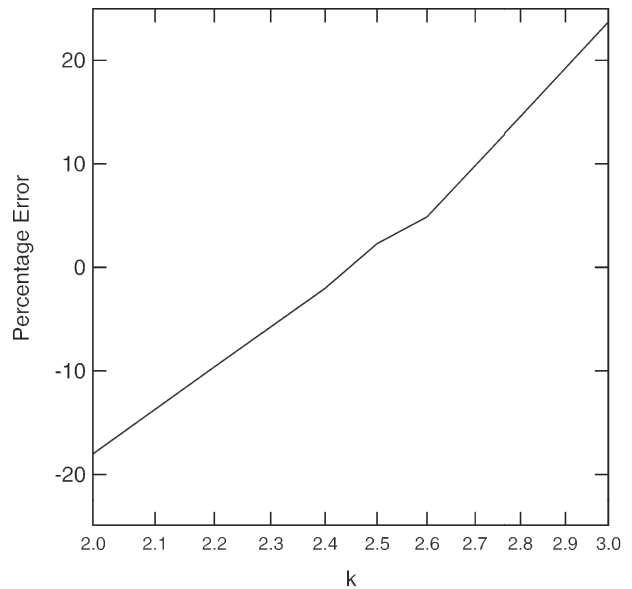


FIG. 3. Percentage errors of the parameterization at different k values.

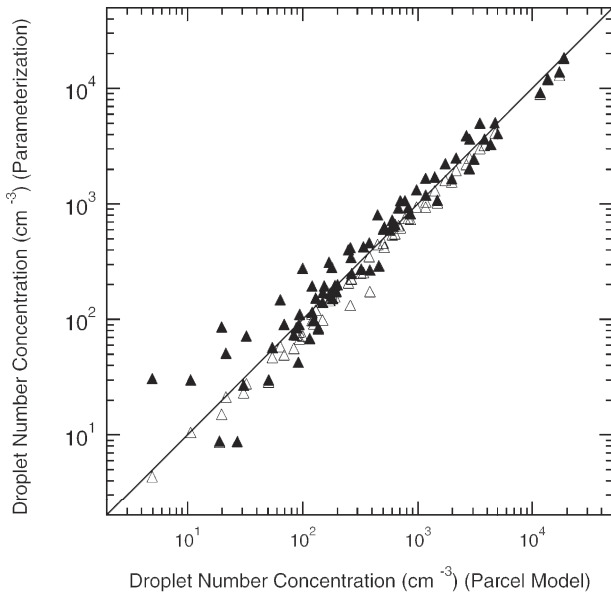


FIG. 4. Comparison of the droplet number concentrations predicted with the parameterization at $k = 2.0$ (open triangles) and 3.0 (solid triangles) and calculated with the parcel model.

and (8) reveals that, in the kinetically limited cases, droplet growth after being activated is not significant enough to justify the omission of the initial size (i.e., the critical size), as assumed by Twomey (1959).

Because the parameterizations of Abdul-Razzak and Ghan (2000) and Nenes and Seinfeld (2003) do not take into account the size dependence of G , this study makes no attempt to compare them directly with our parameterization. However, it is worth noting that Nenes and Seinfeld (2003) showed that the parameterization of Abdul-Razzak and Ghan (2000) does not perform well for some cases like marine aerosol owing to biases introduced by empirical correlation.

b. Applicability to surface-active organic aerosol

Aerosol particles in the atmosphere often consist of an array of inorganic and organic compounds that alter the thermodynamic and surface properties of cloud droplets when activated. Facchini et al. (2000) measured the surface tension of fog water collected in the Po Valley and at several other locations. The results showed that surface-active organic compounds can con-

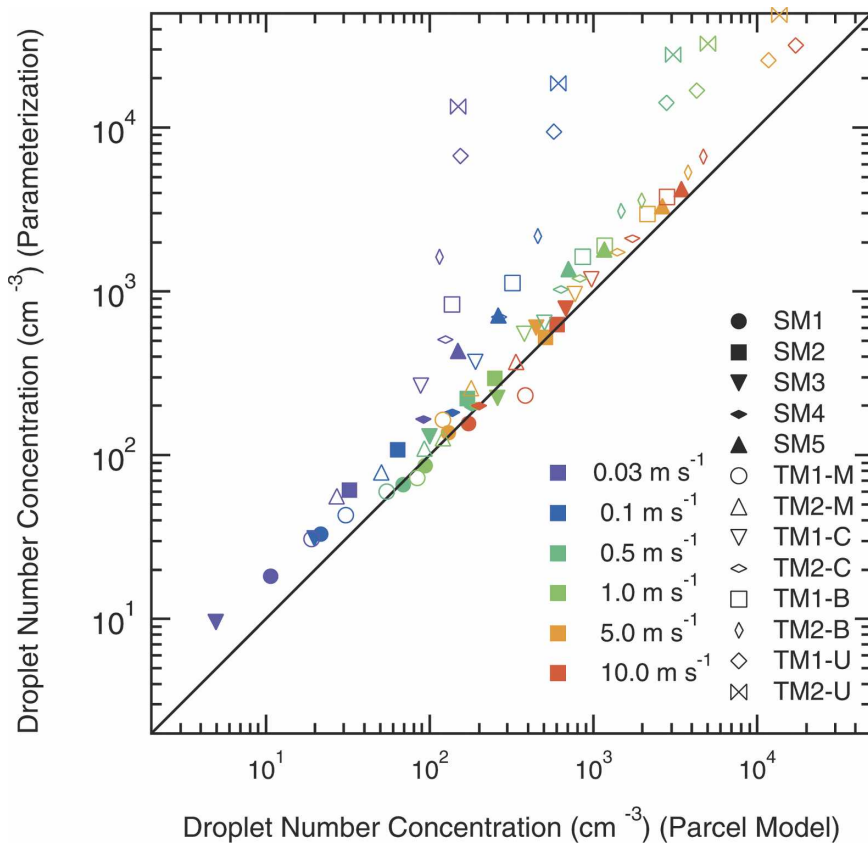


FIG. 5. Comparison of the droplet number concentrations predicted with Twomey's formulation and calculated with the parcel model.

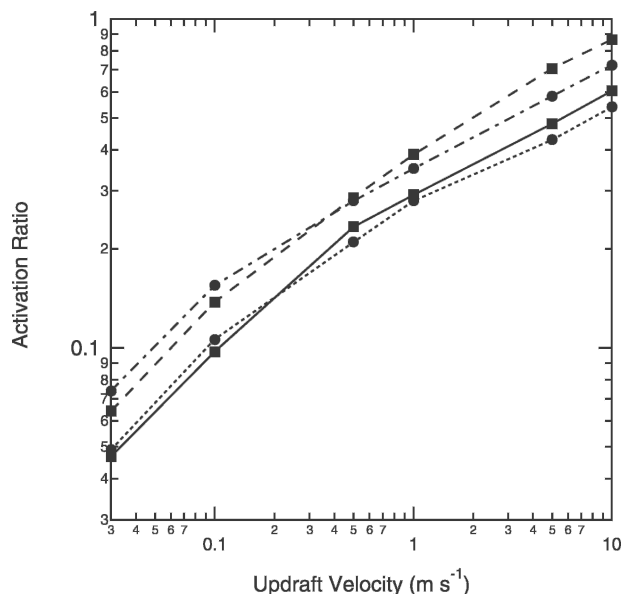


FIG. 6. The relationship between activation ratio and updraft velocity at reduced droplet surface tension using the parameterization (dashed line with squares) and parcel model (dotted-dashed line with circles). The base case uses the surface tension of water (solid line with squares for the parameterization; dotted line with circles for the parcel model).

siderably reduce droplet surface tension. Lower surface tension decreases the critical supersaturation of droplets, which is used in the parameterization as the threshold for activation. The chemical composition of the organic fraction of sea salt aerosol was studied by Ming and Russell (2001). Following that study, a test case uses four model compounds (*viz.*, malic acid, citric acid, glucose, and fructose) to represent the soluble organic mass, which is assumed to account for 20% of the total aerosol mass [the other 80% is $(\text{NH}_4)_2\text{SO}_4$] while the size distribution is the same as case TM1-C. The dependence of droplet surface tension on organic concentration is based on the measurements by Facchini *et al.* (2000).

As shown by the parcel simulations plotted in Fig. 6, as compared to the base case that uses the surface tension of water (76 mN m^{-2}), the lowering of droplet surface tension due to the presence of organic compounds enhances activation. At an intermediate updraft velocity of 1.0 m s^{-1} , the activation ratio increases from 0.28 in the base case to 0.35 in the reduced surface tension case. The parameterization only slightly overestimates activation for both cases. For this particular size distribution, the parameterization overestimates the absolute activation ratios when velocity exceeds 1.0 m s^{-1} , but correctly accounts for the relative increases due to lower surface tension.

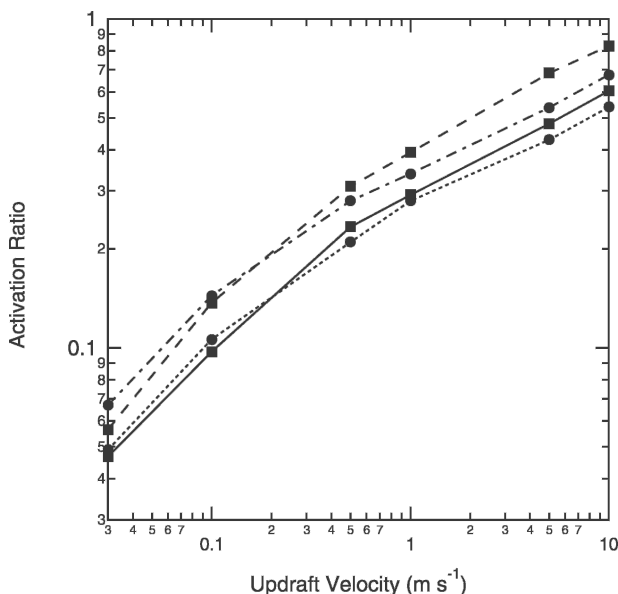


FIG. 7. The relationship between activation ratio and updraft velocity at a lower mass accommodation coefficient of 0.043 using the parameterization (dashed line with squares) and parcel model (dotted-dashed line with circles). The base case uses 1 (solid line with squares for the parameterization; dotted line with circles for the parcel model).

c. Applicability to low mass accommodation coefficient

The mass accommodation coefficient α_C is a microphysical parameter critical for determining the size dependence of the growth coefficient G in Eq. (5). A lower value of α_C slows down droplet growth and results in a higher maximum supersaturation at the same updraft velocity; α_C also affects the distribution of liquid water among different sizes. Despite its importance, significant divergences of several orders of magnitude exist among the measured α_C of pure water droplets (Seinfeld and Pandis 1998). Adding an extra layer of complexity to the problem, some studies reported that the film forming compounds (FFC) are able to coat droplets and decrease α_C to 1×10^{-5} (Feingold and Chuang 2002).

As shown in Fig. 7, for Case TM1-C, a change of α_C from 1 to 0.043 (Feingold and Chuang 2002) increases the activation ratio from 0.28 to 0.34 at 1.0 m s^{-1} . Explicitly considering the size dependence of G , the parameterization represents the trend well; such a change increases the predicted activation ratio from 0.29 to 0.39 at 1.0 m s^{-1} . Despite consistent overestimation at high velocities ($>1.0 \text{ m s}^{-1}$), the parameterization succeeds in capturing the relative effect of low α_C over the whole span of updraft velocity.

5. Conclusions

This study lays out the theoretical framework of a new parameterization, which does not attempt to treat the equilibrium-controlled small and kinetic-controlled large droplets in different ways. Instead, an empirical relationship that relates the droplet diameter at a particular supersaturation to the critical droplet diameter and supersaturation is developed so that both categories of droplets can be accommodated within one consistent framework. Its satisfactory performance is shown by comparing the predicted droplet number concentrations and activation ratios with the detailed parcel model simulations.

The lowering of droplet surface tension due to surface-active organic components of aerosol enhances activation. The parameterization utilizes the critical droplet diameter, which is affected by droplet surface tension, as the starting point of droplet growth. This approach satisfactorily incorporates the surface tension effect into the parameterization. Fully considering the kinetic nature of water condensation, the parameterization calculates the condensation rate using first principles, and is well equipped to handle kinetically limited cloud activation. Thus, effect of chemical composition on cloud droplet formation can be accounted for explicitly.

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