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Droplet activation parameterization: the population splitting concept revisited

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Abstract

In this work we postulate, implement and evaluate modifications to the “population splitting” concept introduced by Nenes and Seinfeld (2003) for calculation of water condensation rates in droplet activation parameterizations. The modifications introduced here lead to an improved accuracy and precision of the parameterization-derived maximum supersaturation, s_{\max} , and droplet number concentration, N_d , as determined by comparing against those of detailed numerical simulations of the activation process. A numerical computation of the first-order derivatives $\partial N_d / \partial \chi_j$ of the parameterized N_d to input variables χ_j was performed, and compared against the corresponding parcel model derived sensitivities, providing a thorough evaluation of the impacts of the introduced modifications in the parameterization ability to respond to aerosol characteristics. The proposed modifications require only minor changes for their numerical implementation in existing codes based on the population splitting concept.

1 Introduction

During the process of cloud formation, preexisting aerosol particles act as cloud condensation nuclei (CCN) upon which cloud droplets first form and subsequently grow. Changes in either the amount or composition of atmospheric aerosol can alter cloud microphysical and optical properties, indirectly impacting the planetary radiation balance and hydrological cycle. Aerosol-cloud interactions constitute some of the most uncertain aspects of anthropogenic climate change estimates (Intergovernmental Panel on Climate Change, 2007).

Calculation of droplet number in atmospheric models requires the computation of new droplet formation (i.e., droplet activation), which occurs at subgrid scales and its representation is computationally expensive if done explicitly using numerical parcel models. For this reason, parameterizations of the activation process have been developed. In these formulations, the fraction of atmospheric aerosol that activates into

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cloud droplets is determined for an air parcel that ascends with an updraft velocity, w . These activation parameterizations use a Lagrangian parcel model approach to study the detailed process of water vapor condensation on the population of growing droplets. A thorough review of activation parameterizations can be found in Ghan et al. (2011).

5 Most of these activation schemes follow the framework proposed by the seminal work of Twomey (1959) which involves two conceptual steps. First, the availability of CCN is determined as function of supersaturation (e.g., using Köhler theory or adsorption activation theory, together with aerosol size distribution and chemical composition), and second, by approximately solving the water vapor balance in the ascending cloud parcel to determine the maximum supersaturation, s_{\max} , attained in it. After this is done, the number of activated cloud droplets, N_d , is equal to the concentration of CCN with a critical supersaturation, s_c , lower than s_{\max} . A number of activation parameterizations have been developed using this approach (e.g., Feingold and Heymsfield, 1992; Ghan et al., 1993; Nenes and Seinfeld, 2003; Pinsky et al., 2012), and many have
10 been incorporated into GCM and regional models to compute aerosol indirect effects (e.g., Abdul-Razzak and Ghan, 2000; Fountoukis and Nenes, 2005; Ming et al., 2006; Shipway and Abel, 2010).

The central problem these schemes need to address is the correct estimation of the size of the growing droplets at the time of peak supersaturation. The condensation rate of water vapor onto activated droplets in the parcel is proportional to the integral diameter of the growing droplet population, and therefore it plays an important role in defining s_{\max} . This task is particularly problematic for the largest particles in the CCN population. As noted by Chuang et al. (1997), a portion of the CCN population, those with relatively low s_c are “inertially-limited” (Nenes et al., 2001) and their size does not equilibrate instantaneously with the ambient supersaturation. Therefore, the equilibrium assumption is not adequate for computing the sizes for these particles.
20 This limitation would likely affect particles larger than approximately $0.2 \mu\text{m}$ in diameter, therefore impacting the coarse mode as well as a sizable fraction of accumulation mode particles.

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Even though coarse mode particles typically contribute a small number concentration to the CCN population, they represent an important sink for water vapor, effectively modulating the parcel s_{\max} (e.g., Ghan et al., 1998; Barahona et al., 2010; Morales Betancourt and Nenes, 2013). This means that even modest increases in either the number or the hygroscopicity of these large particles can cause a significant decrease in s_{\max} , often leading to lower droplet concentrations (Morales Betancourt and Nenes, 2013). Furthermore, because of the large contribution of accumulation mode particles to the total CCN active population, accurately accounting for the water uptake of the inertially limited portion of accumulation mode CCN, is of great importance in determining s_{\max} and N_d .

Within the parameterization framework first proposed by Nenes and Seinfeld (2003), different approaches have been incrementally adopted to improve their ability to capture the supersaturation across a large set of conditions. Fountoukis and Nenes (2005) extended this framework to include the effect of mass transfer limitations in the non-continuum regime through an effective water vapor accommodation coefficient. Kumar et al. (2009) introduced changes in the CCN spectra to allow for adsorption activation. Barahona and Nenes (2007) introduced a framework to account for the impact of entrainment and mixing in decreasing the condensation rate on the droplets to sub-adiabatic levels. The prediction of N_d with Fountoukis and Nenes (2005) parameterization is typically within $\pm 20\%$ when compared to parcel model simulations for a wide range of aerosol conditions and vertical velocity, and is capable of reproducing observed cloud droplet data (Fountoukis et al., 2007; Meskhidze et al., 2005). However, when the population of “inertially limited” CCN is large, it tends to slightly overestimate N_d and s_{\max} . Barahona et al. (2010) noted this and introduced a novel way of approximating the condensation rate on the large particles, to better account for their contribution to depleting the available water vapor. This new approach corrected the overprediction issue of Fountoukis and Nenes (2005) in conditions where there is a significant presence of large CCN. As we show in the present work, the modifications

growth of a droplet with diameter D_p is given by (Nenes and Seinfeld, 2003),

$$D_p \frac{dD_p}{dt} = Gs \quad (2)$$

where G is the mass transfer coefficient of water to the droplets (Appendix A). Since q_l is proportional to the total volume concentration of the droplet population, the condensation rate in Eq. (1) can be expressed in terms of D_p by using the growth rate Eq. (2),

$$\frac{dq_l}{dt} = \frac{\pi \rho_w}{2 \rho_a} Gs \int n(d_p) D_p(d_p, t) dd_p \quad (3)$$

where $D_p(d_p, t)$ is the wet diameter at a time t after in-cloud ascent, of a droplet growing on an aerosol particle of dry size d_p . Equation (3) indicates that the condensation rate is proportional to the integral diameter of the droplet size distribution. Using Köhler theory (e.g., Nenes and Seinfeld, 2003) or adsorption activation theory (Kumar et al., 2009) to relate the dry size of the aerosol, d_p , to s_c , the integral in Eq. (3) can be expressed in terms of the critical supersaturation s_c . Following Nenes and Seinfeld (2003), the integral diameter (also termed condensation integral) in s_c space is defined here as,

$$I(0, s) \equiv \int_0^s n(s_c) D_p(s_c, t) ds_c \quad (4)$$

where the first and second arguments in $I(a, b)$, represent the lower and upper integration limits respectively. The maximum supersaturation can be found by setting $ds/dt = 0$ in Eq (1). Using Eq. (4) and after some manipulation, the supersaturation equation at the moment of maximum supersaturation can be written as

$$s_{\max} I(0, s_{\max}) = \beta \quad (5)$$

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with $\beta = 2\rho_a\alpha w/(\pi\rho_w\gamma G)$. Equation (5) cannot, in general, be solved analytically. The diameter of the growing droplets at peak supersaturation is necessary to calculate the condensation integral, $I(0, s_{\max})$, and still requires a formulation in terms of the dry aerosol size distribution. The “population splitting” approach (Nenes and Seinfeld, 2003) provides such framework to approximate D_p and compute $I(0, s_{\max})$ by identifying different asymptotic regimes, and is briefly explained below.

2.1 The “population splitting” concept

A solution to the supersaturation balance Eq. (5) requires to express the condensation rate, proportional to $I(0, s_{\max})$, in terms of the dry aerosol size distribution and the size of droplets at the time of maximum supersaturation, t_m . To estimate the size $D_p(s_c, t_m)$, Eq. (2) is often integrated from the activation time, τ_{s_c} , defined as $s(\tau_{s_c}) = s_c$, to the time when s reaches a maximum, i.e.,

$$D_p^2 = D_p(\tau_{s_c})^2 + 2G \int_{\tau_{s_c}}^{t_m} s dt \quad (6)$$

Two assumptions, each representing asymptotic growth limits, have been often adopted to obtain an approximate expression for D_p in Eq. (6). One such approximation, denoted here $D_p^{(1)}$, consists of neglecting droplet growth after activation, and that the droplet diameter at s_{\max} is given by the critical wet diameter D_{pc} , i.e., $D_p^{(1)} = D_p(\tau_{s_c}) = D_{pc}$ (e.g., Ghan et al., 1993). Using Köhler theory, D_{pc} (hence $D_p^{(1)}$) can be written as a function of s_c (see Appendix A),

$$D_p^{(1)} = \frac{2A}{3s_c} \quad (7)$$

Although adequate for the smallest CCN, Eq. (7) overestimates the wet diameter when applied to the largest particles in the CCN population. Due to their size, droplets

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growing on aerosol particles with a dry diameter larger than $\sim 0.2\mu\text{m}$ cannot grow in equilibrium with the ambient supersaturation (Chuang et al., 1997). As a consequence of this “inertial limitation” (Nenes et al., 2001), these droplets fall far behind their equilibrium diameter as the parcel supersaturation increases, and therefore application of Eq. (7) leads to a large overestimation of their size. This in turn leads to overestimating the condensation rate, biasing s_{max} and N_{d} low (Ghan et al., 1993).

Another approximation for D_{p} in Eq. (6), which we will denote here as $D_{\text{p}}^{(2)}$, first introduced by Twomey (1959), considers that particle growth after exposure to their critical supersaturation is the main contributor to particle size. This approach, effectively neglects the initial size of the particles when exposed to s_{c} , $D_{\text{p}}(\tau_{s_{\text{c}}})$, and considers only the contribution of the growth term in Eq. (6). Twomey (1959) further proposed a lower bound for the supersaturation integral relating it to s_{c} , namely

$$\int_{\tau_{s_{\text{c}}}}^{t_{\text{m}}} s dt = \frac{s_{\text{max}}^2 - s_{\text{c}}^2}{2\alpha w} \quad (8)$$

However, neglecting $D_{\text{p}}(\tau_{s_{\text{c}}})$ can cause a large underestimation of D_{p} , and therefore, of the surface area for water vapor condensation, particularly for large CCN. When this approximation is adopted, the droplet size $D_{\text{p}}(s_{\text{c}}, t_{\text{m}})$ can be found by replacing Eq. (8) into Eq. (6), i.e.,

$$D_{\text{p}}^{(2)} = \left(\frac{G}{\alpha w} \right)^{1/2} \left(s_{\text{max}}^2 - s_{\text{c}}^2 \right)^{1/2} \quad (9)$$

Subsequent approaches to the problem have acknowledged that in actuality both regimes occur within the same CCN population. Abdul-Razzak et al. (1998) identified these regimes based on the proximity of s_{c} to s_{max} , proposing that for particles with $s_{\text{c}} \ll s_{\text{max}}$ the growth term was dominant, while for those with $s_{\text{c}} \sim s_{\text{max}}$ the effect of growth was negligible, and their size was close to their activation size.

reflecting the fact that in this region the critical diameter $D_p^{(1)}$ is always larger than the growth term, $D_p^{(2)}$.

Nenes and Seinfeld (2003) used the clues provided by this classification to define rules for the estimation of D_p . For those CCN with $s_{\max} > s_c > s_p^+$ (termed here population I), D_p was approximated by D_{pc} . This is a reasonable assumption since these small particles are the most likely to equilibrate instantaneously with the ambient supersaturation, and as discussed before, they have little time to grow. For those CCN with $s_p^+ > s_c > s_p^-$ (termed here population II), approximation $D_p^{(2)}$ was used. This still leaves a third population out, the large CCN with $s_c < s_p^-$. Despite the rules of Eq. (10) dictate that for this population $D_p^{(1)} > D_p^{(2)}$, it is well known that in actuality they are generally not capable of growing at equilibrium, so their size at s_{\max} is much smaller than their D_{pc} . Using these arguments, the large CCN particles were merged together into population II by using approximation $D_p^{(2)}$ for all particles with $s_c < s_p^+$, i.e., discarding s_p^- (Fig. 1a).

The approach was completed by defining an empirically derived s_p for the regime where $s_{\max} < \xi_c$ (and Eq. (10) admits only imaginary solutions), this is:

$$\frac{s_p^+}{s_{\max}} = \frac{2 \times 10^7}{3} A s_{\max}^{-0.3824} \quad (11)$$

The population splitting formulation has been shown to have great skill in capturing the behavior of s_{\max} under a large set of aerosol and updraft inputs. The Fountoukis and Nenes (2005) parameterization (FN hereafter) which is based on the framework described above has also been capable of reproducing observed cloud droplet concentrations (e.g., Meskhidze et al., 2005; Fountoukis et al., 2007).

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2.1.1 Correction for inertially-limited CCN

Based on detailed numerical simulations of the activation process, Barahona et al. (2010) noted that when the activation process occurs in situations of weak updrafts, and the aerosol contains a significant number of large CCN, the FN parameterization exhibited a tendency to overestimate s_{\max} and N_d . It was shown that this behavior originated in the assumptions made regarding the size of the inertially limited CCN. By analyzing the first-order derivatives of the FN parameterization with respect to input parameters, Morales Betancourt and Nenes (2013) further confirmed a lack of sensitivity of N_d computed with FN to perturbations in the properties of coarse mode particles i.e., to number concentration, mode diameter, and hygroscopicity parameter. This indicated that although the total droplet number was not drastically affected by the coarse mode aerosol properties, the slight overestimation of N_d and s_{\max} above mentioned was due to the underestimation of the water vapor depletion by the large CCN population.

A simple correction for these “inertially limited” droplets was introduced by Barahona et al. (2010). As the timescale for large soluble particles (whose equilibrium supersaturation follows the Köhler Eq. A4) to grow to D_{pc} is many times larger than the timescale of cloud formation, it was proposed that the condensation rate on this population could be estimated by approximating their size at s_{\max} with their equilibrium diameter at $s = 0$, D_{p0} . Using Köhler theory, the equilibrium diameter has been shown to be equal to $D_{pc}/\sqrt{3}$ (Barahona et al., 2010), and is denoted here by $D_p^{(3)}$. This correction term, which improves the predictions of the parameterization when a significant number of large CCN are present, was applied to all the population II particles. In this work we show that the approach of Barahona et al. (2010) inadvertently overestimates the size for the population II particles by adding $D_p^{(2)}$ and $D_p^{(3)}$ concurrently to estimate their size at s_{\max} . Equation (6) for D_p involves the square root of the sum of the growth term and the initial size, therefore directly adding both terms results in an overestimation bias for D_p , and a corresponding overestimation of the contribution of this population to the

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condensation rate. Therefore, it is necessary to revise the population splitting concept to consistently combine the contributions from all CCN to the condensation rate.

Up until now, we have assumed particles activate in accordance with Köhler theory. However, insoluble particles, such as uncoated mineral dust and volcanic ash, for which activation follows the adsorption activation theory (Sorjamaa and Laaksonen, 2007; Kumar et al., 2009; Latham et al., 2011), tend to uptake considerably less water before activation than Köhler particles. As shown by Kumar et al. (2009), the ratio between the critical wet diameter D_{pc} and the dry aerosol diameter d_p for insoluble particles is less than two for most conditions, and this ratio is only weakly dependent on the size of the dry particle (see Appendix A). For this reason, insoluble particles that activate via adsorption activation are typically capable of growing at equilibrium with the ambient supersaturation, reaching their D_{pc} , and the mechanisms of kinetic limitations are different than those outlined in Barahona et al. (2010). Furthermore, the behavior of insoluble particles as explored by Kumar et al. (2009) considers that independently of their size, all insoluble particles are capable of reaching D_{pc} , and the contribution from growth dominates at all particles sizes, which implies that the population splitting concept is not necessary for these particles. For these reasons, the revision of the population splitting concept is limited to particles activating in accordance with Köhler theory.

2.2 The “population splitting” concept revisited

We aim to improve two main aspects of the parameterization framework of Nenes and Seinfeld (2003) and Barahona et al. (2010). First, to better account for the size of inertially limited CCN, so their contribution to supersaturation depletion can be quantified correctly. The second goal is to avoid the discontinuity in s_p^\pm introduced in Eq. (11).

As s_{max} approaches ξ_c from the right, s_p^\pm from Eq. (10) approaches $1/\sqrt{2}$. However, the value of s_p^+ for $s_{max} = \xi_c$ in Eq. (11) is in general, not equal to $1/\sqrt{2}$. This implies a discontinuity in the calculation of the surface area of droplets, which in turn, creates

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a discontinuity in the parameterization response in scenarios where s_{\max} shifts from the $s_{\max} < \xi_c$ regime, to the $s_{\max} > \xi_c$.

The first goal is attained by recognizing, as Barahona et al. (2010), that neither $D_p^{(1)}$ or $D_p^{(2)}$ are appropriate approximations for the size of the largest CCN particles. However, instead of merging all CCN with $s_c < s_p^+$ in the same population (Population I in Fig. 1a) we consider that only the largest particles, those with $s_c < s_p^-$, should be approximated as in Barahona et al. (2010), i.e., $D_p^{(3)} \approx D_{pc}/\sqrt{3}$ (Fig. 1b). Similarly, and to maintain consistency and avoid overestimation of the water uptake, D_p for CCN with $s_p^+ > s_c > s_p^-$ are approximated with Eq. (9)

$$D_p^{(1)} \approx D_{pc}(s_c) = \frac{2A}{3s_c} \quad (12a)$$

$$D_p^{(2)} \approx \left(\frac{G}{\alpha W}\right)^{1/2} (s_{\max}^2 - s_c^2)^{1/2} \quad (12b)$$

$$D_p^{(3)} \approx D_{p0}(s_c) = \frac{2A}{3\sqrt{3}s_c} \quad (12c)$$

and the integral $I(0, s_{\max})$ is naturally split in the different components:

$$s_{\max} \left[I(0, s_p^-) + I(s_p^-, s_p^+) + I(s_p^+, s_{\max}) \right] = \beta \quad (13)$$

The second goal is achieved by defining the partition supersaturation for $s_{\max} < \xi_c$ such that it transitions smoothly to the regime where CCN is completely dominated by inertially limited particles. Noting that as $s_{\max} \rightarrow \xi_c$, both roots become identical $s_p^+ = s_p^-$, and both approach the value $1/\sqrt{2}$, we define s_p^\pm as:

$$\frac{s_p^\pm}{s_{\max}} = \frac{2A \times 10^7}{3} (s_{\max}^{-0.3824} - \xi_c^{-0.3824}) + \frac{1}{\sqrt{2}} \quad (14)$$

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which maintains the same empirically-derived dependence on s_{\max} , but solves the discontinuity issue in the original framework of Nenes and Seinfeld (2003). From this expression, the vanishing of the term $I(s_p^-, s_p^+)$ emerges naturally for $s_{\max} < \xi_c$, since both roots collapse to the same value. The regions where each approximation should be used are depicted in Fig. 1b.

2.3 Numerical implementation

The modifications proposed here can be implemented in the existing Barahona et al. (2010) framework without the need of any major changes. Using the functions $I_1(0, s_p)$ and $I_2(s_p, s_{\max})$ whose formulas are given in Nenes and Seinfeld (2003) for sectional, and in Fountoukis and Nenes (2005) for lognormal aerosol size distribution (see Appendix B), $I(0, s_{\max})$ is simply given by the following expression,

$$I(0, s_{\max}) = \frac{1}{\sqrt{3}} I_2(0, s_p^-) + \left[I_1(0, s_p^+) - I_1(0, s_p^-) \right] + I_2(s_p^+, s_{\max}) \quad (15)$$

which can be implemented with minimal adjustments to codes that use the original population splitting concept. This expression can be extended to the formulation of Barahona and Nenes (2007) that includes the effects of entrainment and mixing in the supersaturation development. If subsaturated air entrains the air parcel at a fractional entrainment rate μ , the condensation rate onto the droplets is reduced, and Eq. (5) transforms to

$$s_{\max} I(0, s_{\max}) = \beta(1 - \mu/\mu_c) \quad (16)$$

where μ_c is the “critical entrainment rate” defined in Barahona and Nenes (2007) as the entrainment rate that prevents the cloud parcel to generate water vapor supersaturation, and is given by

$$\mu_c = \frac{\alpha}{1 - RH} \left(1 - \frac{L_v M_w \Delta T}{RT^2} \right)^{-1} \quad (17)$$

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where RH is the relative humidity of the entrained air, and $\Delta T = T - T'$ is the difference between the parcel and entrained air temperatures.

3 Results

3.1 Parameterization evaluation

5 The augmented parameterization was tested against computations of N_d and s_{\max} from a detailed numerical parcel model of the condensational growth of droplets (Nenes et al., 2001). In order to explore the parameterization in the conditions typically encountered in a GCM simulation, we employed off-line annual average aerosol fields from a climatological simulation performed by Morales Betancourt and Nenes (2013) with the Community Atmospheric Model 5.1 (CAM5.1). The simulations correspond to present-day aerosol emissions from the Lamarque et al. (2010) emission inventory. CAM5.1 includes a 3-mode lognormal aerosol model (MAM3). The aerosol fields correspond to the 930 hPa pressure level, and only the grid cells between 75° N and 75° S were used, totaling 9504 instances of aerosol size distributions and chemical composition corresponding to each grid cell considered. The ranges over which the characteristics of the test aerosol fields are reported in Table 1.

As shown by Morales Betancourt and Nenes (2013), the first-order derivatives of the parameterized N_d with respect to input parameters are useful in understanding the parameterization ability to respond to perturbations to the input variables. Therefore in addition to evaluating the parameterized N_d and s_{\max} against those of parcel model simulations, we also performed calculations of the first-order derivatives of the parameterized N_d with respect to any input variable χ_j , $\partial N_d / \partial \chi_j$. These sensitivity calculations were then compared against finite difference approximation to the derivatives with the numerical cloud parcel model. We report the mean of the relative error ϵ and the standard deviation of the errors σ_ϵ between both quantities for each of the 9504 cases considered. Three different versions of the parameterization framework of

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Nenes and Seinfeld (2003) are evaluated here: the Fountoukis and Nenes (2005), FN, the Barahona et al. (2010), BN, and the modification proposed in this paper.

The results show a significant improvement in the accuracy and precision of the parameterized N_d and s_{\max} values, without any appreciable increase in the computational cost. Figure 2a and Table 2 summarize the results of the parameterization evaluation. When FN was used, the relative error in s_{\max} was on average $+31\% \pm 25\%$ while in N_d was of $+7.8\% \pm 9.7\%$. Both numbers indicate a moderate overestimation in both fields for the conditions explored. Figure 2b shows a similar analysis for the Barahona et al. (2010) parameterization, with a relative error for s_{\max} of $-24\% \pm 7\%$, and N_d of $-10\% \pm 7.8\%$, showing a small underestimation of both fields under these conditions. For both fields there is a marked decrease in both the average error (a measure of parameterization bias) and in the dispersion of the errors (a measure of the parameterization accuracy). Figure 3 shows the results of the comparison against parcel model for the case of N_d . The relative error when applying the modifications proposed in this work were considerably lower, being $-6.0\% \pm 6.2\%$ for s_{\max} , and $-2.7\% \pm 4.8\%$ for N_d . The sensitivity to total aerosol perturbation, dN_d/dn_a , exhibited a similar improvement.

A summary of the mean relative errors of the sensitivities $\partial N_d / \partial \chi_j$ for the Barahona et al. (2010) and for the parameterization presented in this work are shown in Fig. 4. The modifications introduced here result in a higher sensitivity to aerosol number concentration when compared to BN for the 3 modes considered. Figure 4a suggests that most of the improvement in the ability to predict s_{\max} , N_d , and dN_d/dn_a , is due to a better representation of the response to accumulation mode particles. As such the mean error for $\partial N_d / \partial n_a$ of the accumulation mode went from an average of -9.4% for BN, to only -0.6% . Since this mode represents the bulk of the CCN population any changes to the representation of its water uptake have great impact on s_{\max} and N_d . Figure 4 also shows that the magnitude of the mean errors for the Aitken and the accumulation mode sensitivity to n_a , κ_a , and d_g are smaller for the parameterization presented here. Nevertheless, it can also be seen that the modifications introduced here result in an overestimation of the sensitivities of these variables for the coarse mode particles.

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4 Summary and conclusions

The “population splitting” concept of Nenes and Seinfeld (2003) and Barahona et al. (2010) was further developed to consistently account for the condensation rate of inertially-limited CCN. The modifications to this parameterization framework were shown to improve the accuracy and precision for predictions of maximum supersaturation s_{\max} , and cloud droplet number concentration N_d . Similarly, the sensitivity of the parameterized N_d to aerosol number concentration, dN_d/dn_a , was found to be in better agreement when compared to detailed numerical simulations of the activation process. The first-order derivatives $\partial N_d/\partial \chi_j$ of the parameterized N_d were also compared against numerical parcel model estimates. This analysis showed that the modifications presented here result in a more consistent response to perturbations to the characteristics of Aitken and Accumulation mode particles, while revealing a slight overrepresentation of the response to coarse mode aerosol properties. Implementation of these modifications to the population splitting framework is straightforward and does not require any major modifications to the previous formulations. This minor code change comes at no additional computational expense, and produces virtually identical results to a numerical parcel model, both in terms of N_d and sensitivities $\partial N_d/\partial \chi_j$. The impact of these changes is expected to be larger in environments dominated by highly hygroscopic coarse mode aerosol, such as marine environments far from pollution sources, as well as regions with a large number of accumulation mode particles.

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Appendix A

Notation

The functions α and γ from Eq. (1) are given by,

$$\alpha = \frac{gL_v M_w}{c_p RT^2} - \frac{gM_a}{RT} \quad (\text{A1})$$

and,

$$\gamma = \frac{L_v^2 M_w}{c_p RT^2} + \frac{M_a p}{M_w e_s}, \quad (\text{A2})$$

where T is the temperature of the air parcel, e_s is the saturation vapor pressure, g is the gravitational constant, L_v is the latent heat of vaporization of water, c_p is the heat capacity of air, R , the universal gas constant, and M_a and M_w are the molecular weights of air and air respectively.

The function G in the droplet growth equation is given by,

$$G = 4 \left[\frac{\rho_w RT}{e_s D_v M_w} + \frac{L_v \rho_w}{k_a T} \left(\frac{L_v M_w}{RT} - 1 \right) \right]^{-1} \quad (\text{A3})$$

where ρ_a and ρ_w are the density of air and water respectively, D_v is the water vapor diffusivity, and k_a is the thermal conductivity of air. The equation describing the equilibrium supersaturation over the surface of a water droplet containing a solute is given by the Köhler equation,

$$s_{\text{eq}} = \frac{A}{D_p} - \kappa \frac{d_p^3}{D_p^3} \quad (\text{A4})$$

where κ is the hygroscopicity parameter (Petters and Kreidenweis, 2007), and the coefficient A is related to the droplet surface tension σ as $A = 4M_w\sigma/RT$. The critical supersaturation s_c , i.e., the maximum of Eq. (A4), also defines the critical diameter, D_{pc} , $s_{eq}(D_{pc}) = s_c$. By setting $ds_{eq}/dD_p = 0$ and solving for D_p , it can be seen that the critical diameter is related to s_c as $D_{pc} = 2A/3s_c$. Similarly, the dry diameter d_p can be related to its corresponding critical supersaturation s_c , (e.g., Seinfeld and Pandis, 2006),

$$s_c = \left(\frac{4A^3}{27\kappa} \right)^{1/2} d_p^{-3/2} \quad (\text{A5})$$

The power law relationship between s_c and d_p of Eq. (A5) implies that D_{pc} grows as $d_p^{3/2}$ for soluble particles (following Köhler theory), and the ratio D_{pc}/d_p increases with aerosol size as $\sim d_p^{1/2}$. For insoluble particles such as dust, a few layers of water molecules are adsorbed onto the aerosol surface at subsaturated conditions, resulting in equilibrium wet diameters that are similar to the dry aerosol diameter. Kumar et al. (2009) derived a relation equivalent to Eq. (A5) for insoluble particles, and expressed it as $s_c \approx cd_p^{-x}$, with c and x being empirically derived quantities, the exponent x ranging between 0.8 and 1. This lower value for the exponent x compared to Köhler particles implies that the ratio D_{pc}/d_p decreases slightly with increasing d_p .

Appendix B

Summary of changes in existing codes

The conceptual approach for all the parameterization discussed here involve the same steps and require an iterative solution of Eq. (5). Fundamental to the computation of N_d is to determine the number of particles that would activate as a function of

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supersaturation, N_{CCN} , and is represented by a cumulative CCN spectrum $F(s)$. In the case where the aerosol size distribution is described by n_m lognormal modes $F(s)$ is given by,

$$F(s) = \sum_i^{n_m} \frac{n_{ai}}{2} \text{erfc}(u_i(s)) \quad (\text{B1})$$

where $\text{erfc}(x) = 1 - \text{erf}(x)$ is the complement error function, n_m is the number of modes in the aerosol size distribution, and n_{ai} is the number concentration corresponding to mode i . The function u_i is given by,

$$u_i(s) = \frac{2 \ln(s_{g_i}/s)}{3\sqrt{2} \ln \sigma_{g_i}} \quad (\text{B2})$$

where s_{g_i} is the critical supersaturation corresponding to the geometric mean diameter d_{g_i} of the mode.

The conceptual steps in the solution are as follows:

1. Guess an initial value for s_{max} .
2. Compute $\xi_c = (16A^2 \alpha w / 9G)^{1/4}$ (or equivalently $\Delta = 1 - \xi_c^4 / s_{\text{max}}^4$).
3. Evaluate ξ_c (or Δ) to determine the corresponding partitioning supersaturations s_p^\pm .
4. Compute the integral $I(0, s_{\text{max}})$.
5. Evaluate the expression: $s_{\text{max}} I(0, s_{\text{max}}) \stackrel{?}{=} \beta$ (Eq. 5).
6. If convergence is met in step 5, $N_d = F(s_{\text{max}})$. If convergence is not met, repeat steps 1 to 5.

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Existing parameterization codes involve minimal changes in steps 3 and 4 alone. We specifically address the necessary changes to codes that follow the Fountoukis and Nenes (2005) and the Barahona et al. (2010). Alternatively, codes are available upon request to the authors.

5 This formulation of Fountoukis and Nenes (2005) involves the computation of only one partitioning supersaturation, s_p^+ , corresponding to the larger of the two roots in Eq. (10). If $\xi_c > s_{\max}$ then s_p^+ is computed from the positive root in Eq. (10). If $\xi_c < s_{\max}$ then s_p^+ computed using Eq. (11). The integral $I(0, s_{\max})$ takes the form:

$$10 \quad I(0, s_{\max}) = \left[I_1(0, s_p^+) + I_2(s_p^+, s_{\max}) \right] \quad (\text{B3})$$

where for lognormal aerosol,

$$I_1(0, s_p) = \frac{n_{a_i}}{2} \left(\frac{G}{\alpha w} \right)^{1/2} s_{\max} \left[\operatorname{erfc}(u_i(s_p)) - \frac{g_i}{2} \left(\frac{s_{g_i}}{s_{\max}} \right)^2 \operatorname{erfc} \left(u_i(s_p) + \frac{3 \ln \sigma_i}{\sqrt{2}} \right) \right] \quad (\text{B4})$$

$$I_2(s_{p_1}, s_{p_2}) = \frac{n_{a_i}}{2} D_{g_i} k_i \left[\operatorname{erf} \left(u_i(s_{p_1}) - \frac{3 \ln \sigma_i}{\sqrt{2}} \right) - \operatorname{erf} \left(u_i(s_{p_2}) - \frac{3 \ln \sigma_i}{\sqrt{2}} \right) \right] \quad (\text{B5})$$

15 with $g_i = \exp(\frac{9}{2} \ln^2(\sigma_i))$, $k_i = \exp(\frac{9}{8} \ln^2(\sigma_i))$, and $D_{g_i} = 2A/3s_{g_i}$ is defined as the critical wet diameter corresponding to the geometric mean diameter d_{g_i} for mode i .

Computation of the partitioning supersaturation following the correction for inertially limited CCN by Barahona et al. (2010) is identical as in the Fountoukis and Nenes (2005). The integral $I(0, s_{\max})$ however involves an extra term, and takes the form,

$$20 \quad I(0, s_{\max}) = I_1(0, s_p^+) + I_2(s_p^+, s_{\max}) + \frac{1}{\sqrt{3}} I_2(0, s_p^+) \quad (\text{B6})$$

where the extra term $I_2(0, s_p^+)/\sqrt{3}$ can be derived from Eq. (B5),

$$I_2(0, s_p) = \frac{n_{a_i}}{2} D_{g_i} k_i \left[\operatorname{erfc} \left(u_i(s_p) - \frac{3 \ln \sigma_i}{\sqrt{2}} \right) \right] \quad (\text{B7})$$

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The modifications introduced in this manuscript involve the computation of the partitioning supersaturations s_p^\pm . This computation is done in the following way,

$$s_p^\pm = \begin{cases} \xi_c > s_{\max} \longrightarrow s_p^\pm \text{ from Eq. (10)} \\ \xi_c < s_{\max} \longrightarrow s_p^\pm \text{ from Eq. (14)} \end{cases} \quad (\text{B8})$$

5 Computation of the integral $I(0, s_{\max})$ can be achieved by applying Eq. (15) using the expressions provided in Eqs. (B4), (B5), and (B7).

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Table 1. Size distribution parameters for the 3-modal aerosol size distribution used for evaluation of the parameterization. Each log-normal mode is characterized by the number concentration n_{a_i} , geometric standard deviation σ_{g_i} , geometric mean diameter d_{g_i} , and the hygroscopicity of the mode κ_{a_i} .

Aerosol mode	σ_{g_i}	n_{a_i} (cm^{-3})	d_{g_i} (μm)	κ_{a_i}
(1) – Aitken	1.6	40–200	0.004–0.055	0.37–0.72
(2) – Accumulation	1.8	30–510	0.13–0.35	0.18–1.05
(3) – Coarse	1.8	0.1–5.0	1.0–4.0	0.11–1.16

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Table 2. Summary of comparisons against parcel model simulations expressed as $\epsilon \pm \sigma_\epsilon$.

Activation Parameterization		s_{\max}	N_d	dN_d/dn_a
Fountoukis and Nenes (2005)	FN	+31% ± 25%	+7.8% ± 9.7%	+8.7% ± 30.2%
Barahona et al. (2010)	BN	−24% ± 7%	−10% ± 7.8%	−19.5% ± 17.6%
This work		−6.0% ± 6.2%	−2.7% ± 4.8%	−9.3% ± 12.1%

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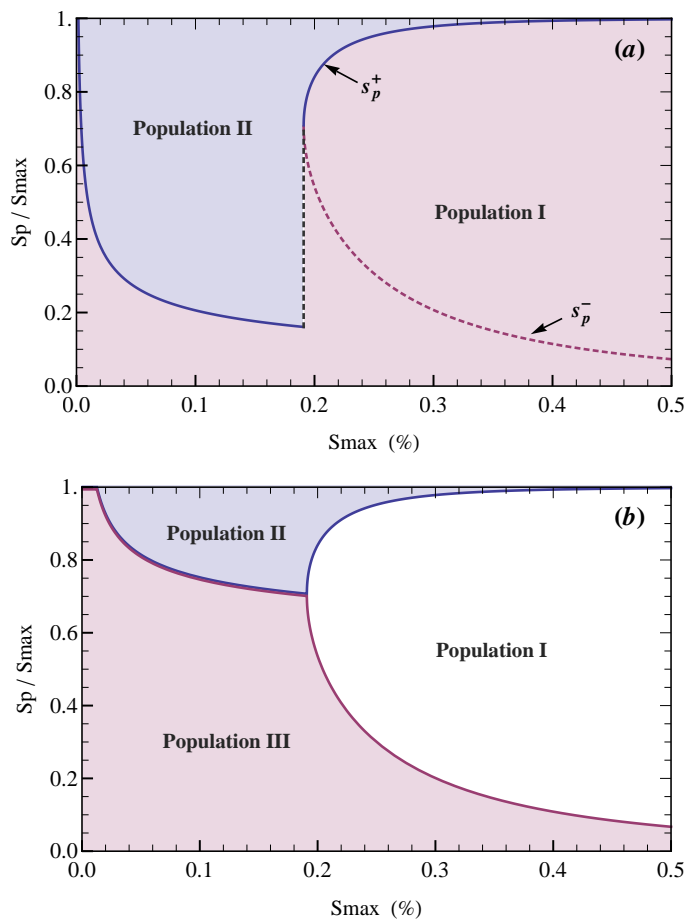
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Fig. 1. The “partitioning supersaturations” s_p^\pm illustrated in the $s_c - s_{\max}$ space as used in Nenes and Seinfeld (2003) and Barahona et al. (2010); and (b) as used in this study. The example here is for a vertical velocity $w = 0.1 \text{ ms}^{-1}$.

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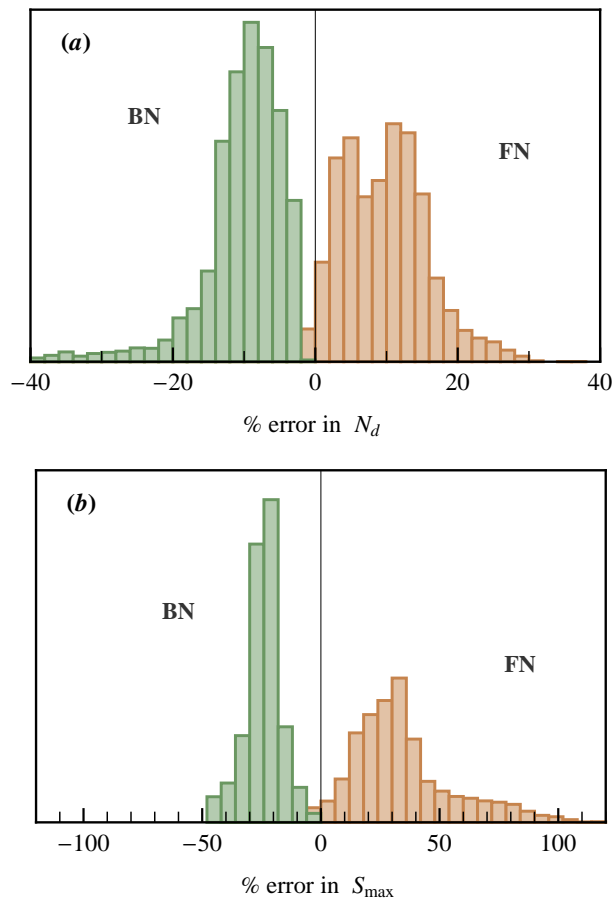
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Fig. 2. Histogram of the frequency of occurrence for the relative error $\epsilon = 1 - \chi_{\text{param}}/\chi_{\text{PM}}$, where χ_{param} is the parameterized value, and χ_{PM} is the value from parcel model simulations. **(a)** for the droplet number N_d , **(b)** for the maximum supersaturation s_{max} .

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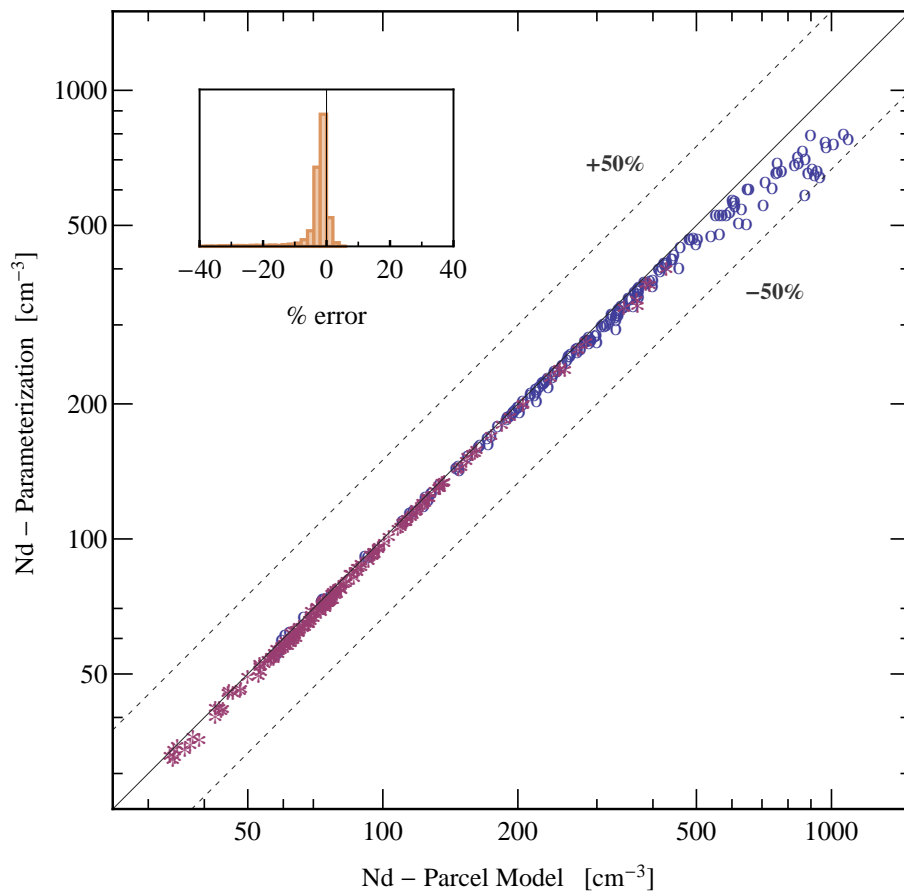


Fig. 3. Comparison between parcel model simulations and parameterization results. Blue circles correspond to continental aerosol while red stars are for marine aerosol. The inset is an histogram of the relative error between parcel model and parameterization derived N_d .

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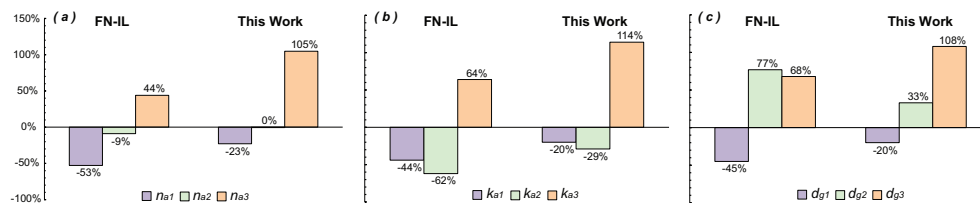


Fig. 4. Mean relative percent error, ϵ , between sensitivities computed with the detailed parcel model simulations and the parameterization results. Comparisons are shown here for (a) sensitivity to aerosol number $\partial N_d / \partial n_{a_i}$, (b) sensitivity to aerosol number $\partial N_d / \partial \kappa_{a_i}$, (c) sensitivity to aerosol number $\partial N_d / \partial d_{g_i}$. Comparisons are shown for the BN parameterization and the results of this work. Subindices follow the notation of Table 1.

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