



Towards an enhanced droplet activation scheme for multi-moment bulk microphysics schemes

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ABSTRACT

Initial droplet spectra produced upon activation impact the ensuing chain of microphysical processes and therefore play a crucial role in cloud evolution. This work re-examines dependencies of newly formed cloud droplet size distribution (CSD) characteristics on environmental and aerosol properties via parcel model simulations that serve as the basis for a multi-moment bulk microphysics droplet activation scheme suitable for a cloud-resolving model (CRM). It is found that applying a fixed size threshold to define activated droplets versus employing physical considerations can lead to erroneous activation and overly broad CSDs for high aerosol concentration and weak updraft conditions. Aerosol distributions characterized by larger median sizes and/or increased solubility can result in greater activated droplet numbers, whereas impacts of these parameters on CSD spectral width depend on both aerosol number concentration and updraft velocity. An expansion of the activation scheme to include CSD spectral width is proposed to aid efforts to extend high-order moment prediction to cloud droplet categories in CRMs as well as better represent variability in the activation process on the cloud scale.

1. Introduction

Advancements in understanding aerosol impacts on clouds and precipitation are aided by employing numerical models to simulate the underlying physical processes given the difficulty in observing such processes. Many cloud-resolving models (CRMs) continue to parameterize sub-grid microphysical processes using bulk approaches as bin (spectral) schemes remain computationally expensive. One such process is the activation of aerosol particles to cloud droplets. Because this process directly links cloud microphysics and aerosols, it is arguably the root of aerosol-cloud microphysical interactions. Liquid droplet formation in the atmosphere is primarily controlled by the numbers and sizes of aerosol particles that serve as potential cloud condensation nuclei (CCN), and the environmental supersaturation (*SS*) (Pruppacher and Klett, 1997). Aerosol chemical composition and hygroscopicity (or solubility) are generally less important (Dusek et al., 2006; Ward et al., 2010, hereafter W10), although they can be relevant early in the activation process as well as in highly polluted and/or low *SS* environments (Feingold, 2003; Reutter et al., 2009, hereafter R09).

Numerous studies have investigated sensitivities of cloud droplet formation to various aspects of aerosol properties (e.g., Chen et al.,

2016; Ghan et al., 2011; Peng et al., 2007; R09; W10; Yum and Hudson, 2005) and generally support the notion that increased aerosol number concentrations (N_a) result in increased cloud droplet number concentrations (N_c) and reduced droplet sizes, all else being equal (Twomey, 1977). R09 used parcel model simulations to conceptualize three droplet activation regimes based on the ratio of upward vertical velocity (w) to N_a . In the aerosol-limited (AL) regime, characterized by ratios of $w/N_a \geq 10^{-3} \text{ m s}^{-1} \text{ cm}^3$ and relatively large *SS* values, large fractions of aerosol particles serve as CCN and activate to cloud droplets such that N_c is proportional to N_a . The updraft-limited (UL) regime, defined by ratios of $w/N_a \leq 10^{-4} \text{ m s}^{-1} \text{ cm}^3$, features relatively low *SS* values, lower fractions of aerosols serving as CCN, and a nearly linear dependence of N_c on w . In between these two regimes lies the transition (TR) regime, in which N_c exhibits a non-linear dependence on both N_a and w .

Aerosol impacts on the spectral width of cloud drop size distributions (CSDs) have recently received greater attention given the dependence of cloud optical properties and microphysical processes on this parameter (e.g., Hudson and Yum, 1997; Hudson et al., 2012; Igel and van den Heever, 2017; Liu and Daum, 2002; Lu and Seinfeld, 2006; Peng et al., 2007). Chen et al. (2016, hereafter C16) used parcel model

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simulations to investigate the regime dependence of the relative dispersion (d)¹ of newly activated CDSs, where d is the ratio of droplet radius standard deviation (σ) to the mean radius (\bar{r}). C16 demonstrated that increasing N_a resulted in increasing (decreasing) d values via reduced condensational narrowing (spectral broadening) rates within the AL (UL) regime, with d values peaking in the TR regime. Their findings suggest a similar regime dependence for d as R09 noted for N_c and help explain reportedly conflicting relationships between N_a and CDS spectral characteristics (cf. Hudson and Noble, 2014; Liu et al., 2014), although the applicability of these results within bulk microphysical schemes was not addressed.

Simulating aerosol-cloud interactions with CRMs employing bulk microphysics requires that the scheme minimally predict two CDS parameters, namely mass and number concentrations, and represent the droplet activation process. Various activation schemes aim to determine N_c from aerosol and environmental properties and include analytical expressions (e.g., Abdul-Razzak et al., 1998; Morrison et al., 2005) as well as lookup tables (LUTs) based on detailed parcel model calculations (e.g., Saleeby and Cotton, 2004, hereafter SC04; Segal and Khain, 2006; Thompson and Eidhammer, 2014). Expressions to diagnose CDS spectral width from N_c (Grabowski, 1998; Liu et al., 2006; Morrison and Grabowski, 2007) or cloud water content (Geoffroy et al., 2010) have also been developed, although more robust methods to obtain CDS spectral width upon activation are presently lacking. This latter point is relevant for triple-moment (3M) bulk microphysics that aim to predict distribution spectral width alongside number and mass concentrations (e.g., Loftus et al., 2014; Milbrandt and Yau, 2005).

The current work extends the findings of C16 to the current LUT-based aerosol activation scheme used in the Regional Atmospheric Modeling System (RAMS) (Cotton et al., 2003; SC04; Saleeby and van den Heever, 2013, hereafter SvdH13) and additionally examines aerosol size and solubility impacts on newly activated CDS properties. Because early cloud development processes such as condensational growth, evaporation, and droplet self-collection depend on and impact CDS spectral width (Hudson and Yum, 1997; Seifert and Beheng 2001; Lu and Seinfeld, 2006; Igel and van den Heever, 2017), an expansion of the activation LUTs to include CDS spectral width is proposed as a preliminary step for extending 3M prediction to CDSs in CRMs for improved simulations of aerosol-cloud interactions.

2. Methodology

The current RAMS two-moment microphysics module determines the fractional number of aerosol particles that activate to cloud droplets from five-dimensional LUTs based on model predicted air temperature (T), w , N_a , and the geometric median radius (r_g) and soluble fraction (ϵ) of the aerosol size distribution (SvdH13). These LUTs are created offline using a one-dimensional Lagrangian adiabatic parcel model (Feingold and Heymsfield, 1992; Heymsfield and Sabin, 1989; SC04) to simulate explicit droplet activation and initial CDS growth for a range of ambient atmospheric conditions [T , w] and binned lognormal aerosol size distributions given by

$$N(r) = \frac{N_a}{r\sqrt{2\pi} \ln \sigma_g} \exp \left[-\frac{[\ln(r/r_g)]^2}{2(\ln \sigma_g)^2} \right] \quad (1)$$

where r is the dry aerosol particle bin radius and σ_g is the geometric standard deviation of the distribution. As the parcel model simulations focus on the activation process, other processes such as coalescence, sedimentation, and mixing are not considered. Details of the parcel model can be found in SC04 and SvdH13, and only a brief description is provided here. At the onset of parcel model calculations, the initially dry aerosol particles in all bins first deliquesce and reach their

¹ Chen et al. (2016) use the symbol ϵ to denote relative dispersion in their study.

equilibrium diameters in a sub-saturated environment based on the Köhler equation for solution droplets. The parcel is then lifted at a fixed upward velocity w , and particle growth by vapor diffusion, along with concurrent changes in the ambient environment, are iteratively computed using the Variable-coefficient Ordinary Differential Equation (VODE) solver (Brown et al., 1989). The time resolution of these calculations is determined within the VODE solver, and the frequency at which the solver is called is controlled by a longer model time step based on fixed upward parcel displacement increments (Δz) at the specified w ($\Delta t = \Delta z/w$). Model calculations proceed until the parcel reaches a height 50 m beyond the level of maximum saturation ratio (S_{max}) or total parcel displacement exceeds 2 km. Upon model termination, S_{max} and the fractional number of aerosols (f_{actv}) resulting in newly formed cloud droplets, defined as particles having diameters of at least 2 μm , are cataloged in the LUTs according to the specified T , w , N_a , r_g , and ϵ parameter values.

A critical point regarding the creation of these LUTs is the use of a fixed minimum diameter (D_{min}) to define cloud droplets in the parcel model, which can produce erroneous CDS characteristics, particularly within the UL regime. For aerosol distributions with large r_g values under low SS conditions, for example, deliquesced aerosols within the large tail of the distribution can exceed 2 μm in diameter yet remain unactivated as ‘haze’ particles (Levin and Cotton, 2009; McFiggans et al., 2006). For this study, aerosol particles activate to cloud droplets based on the critical diameter D_{crit} as a function of parcel supersaturation ratio (S_r) as in R09:

$$D_{crit} = \frac{8\sigma_{sol}M_w}{3 \ln(S_r)RT\rho_w} \quad (2)$$

where σ_{sol} is the surface tension of a solution droplet, M_w and ρ_w are the molar mass and density of liquid water, respectively, and R is the universal gas constant. Additionally, at relatively large w values within the AL regime, N_c stabilizes shortly after reaching supersaturation. However, parcel ascent and condensational growth continue beyond the level of S_{max} , potentially causing additional narrowing of the CDS. In the current work, model calculations terminate upon reaching S_{max} as changes in N_c are negligible with continued ascent (Peng et al., 2007; R09).

Parcel model simulations are performed to examine the sensitivities of CDS characteristics to w , N_a , r_g , and ϵ , with the ranges for these parameters listed in Table 1. Aerosols are assumed to be a mix of soluble and insoluble material of equal density, specified by ϵ , where fully soluble aerosols correspond to ammonium sulfate with hygroscopicity parameter $\kappa = 0.61$ (Petters and Kreidenweis, 2007). Following SvdH13, aerosol geometric standard deviation is fixed at $\sigma_g = 1.8$, and aerosol distributions (Eq. 1) are partitioned into 100 logarithmically-spaced bins spanning a size range specific to each r_g value. For all simulations, $\Delta z = 1$ m, and initial values of relative humidity, air temperature and pressure are set to RH = 0.99, $T = 10^\circ\text{C}$ and $p = 900$ hPa, respectively.

3. Results

3.1. Cloud droplet definition impacts

Comparisons of the resulting S_{max} and CDS properties using $D_{min} = 2 \mu\text{m}$ [Method- D_{min}] versus D_{crit} [Method- D_{crit}] to define cloud droplets in the parcel model are displayed in Fig. 1 as functions of w and N_a for aerosol distribution parameters $r_g = 0.06 \mu\text{m}$ and $\epsilon = 1.0$. Note that panels a, b, e, and f graphically depict subsets of the S_{max} and f_{actv} LUTs. For both methods, the AL (UL) regime exhibits high (low) S_{max} and f_{actv} values, consistent with results of R09. However, at low w values within the UL regime, N_c becomes increasingly dependent on N_a at larger N_a values with Method- D_{min} (Fig. 1c) as opposed to being largely independent of N_a with Method- D_{crit} (Fig. 1g) as found by R09 and C16. With Method- D_{min} , d values generally increase monotonically from the

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