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Key Points:
- Considering spectral shape offers new understanding of aerosol-cloud interaction regimes
- A new expression is proposed to distinguish between aerosol- and updraft-limited regimes
- The results help to reconcile discrepancy between previous studies

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New understanding and quantification of the regime dependence of aerosol-cloud interaction for studying aerosol indirect effects

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Abstract Aerosol indirect effects suffer from large uncertainty in climate models and among observations. This study focuses on two plausible factors: regime dependence of aerosol-cloud interactions and the effect of cloud droplet spectral shape. We show, using a new parcel model, that combined consideration of droplet number concentration (N_c) and relative dispersion (ε, ratio of standard deviation to mean radius of the cloud droplet size distribution) better characterizes the regime dependence of aerosol-cloud interactions than considering N_c alone. Given updraft velocity (w), ε increases with increasing aerosol number concentration (N_a) in the aerosol-limited regime, peaks in the transitional regime, and decreases with further increasing N_a in the updraft-limited regime. This new finding further reconciles contrasting observations in literature and reinforces the compensating role of dispersion effect. The nonmonotonic behavior of ε further quantifies the relationship between the transitional N_a and w that separates the aerosol- and updraft-limited regimes.

1. Introduction

Twomey [1974, 1977] pointed out that an increase in aerosol number concentration (N_a) leads to increases in cloud condensation nuclei (CCN) and cloud droplet number concentration (N_c), which in turn reduces droplet sizes and enhances cloud albedo when liquid water remains unchanged. Although the notion of droplet concentration increasing with increasing aerosol concentration (N_a) is well understood qualitatively and several parameterizations have been developed (hereafter number effect) (see *Ghan et al.* [2011] for a recent review), the so-called aerosol indirect effects remain among the most uncertain climate forcings according to the latest *Intergovernmental Panel on Climate Change* [2013] report. Furthermore, climate models tend to overestimate the cooling of aerosol indirect effects and are more susceptible to aerosols compared to observations [*Lohmann and Lesins*, 2002; *Ruckstuhl et al.*, 2010]. Reducing model uncertainty and reconciling models with observations continue to be a major challenge facing the climate community after decades of research.

Two microphysical factors have been proposed to be partially responsible for the tenacious problem. First, it is well known that for a given updraft velocity (w), the dependence of N_c on N_a is nonlinear and regime dependent: N_c increases linearly with N_a when N_a is low, but the N_c-N_a relationship becomes sublinear and levels off when N_a is high. Using an ensemble of detailed parcel model simulations over wide ranges of N_a and w, *Reutter et al.* [2009] further classified the nonlinear N_c-N_a relationship into three distinct regimes according to the ratio of w to N_a: aerosol-limited regime, transitional regime, and updraft-limited regime. Briefly, the aerosol-limited regime is characterized by high w/N_a ($\geq 10^{-3}$ m s^{-1} cm^3), high supersaturation, and strong (linear) dependence of N_c on N_a but weak dependence of N_c on w; the updraft-limited regime is characterized by low w/N_a ($\leq 10^{-4}$ m s^{-1} cm^3), low supersaturation, and weak dependence of N_c on N_a but strong dependence of N_c on w; the transitional regime falls between the aerosol-limited and updraft-limited regimes with sublinear dependence of N_c on both N_a and w. Evidently, the magnitude and importance of the aerosol indirect effect depend highly on the aerosol-cloud interaction regime [*Stevens*, 2013].

Less understood is the second factor—dispersion effect whereby changes in aerosol properties alter the spectral shape of the cloud droplet size distribution in addition to droplet number concentration (dispersion effect hereafter). *Liu and Daum* [2002] showed, by analyzing data from marine clouds under clean and polluted conditions, that increased N_a leads to concurrent increases of N_c and relative dispersion (ε) of the cloud droplet size distribution defined as the ratio of standard deviation to mean radius of the droplet size distribution, and the enhanced ε negates the number effect and may be partly responsible for the overestimated indirect aerosol effect and the discrepancy between model estimates of the indirect aerosol effect and those constrained by observations [*Peng and Lohmann*, 2003; *Penner et al.*, 2006; *Rotstayn and Liu*, 2003, 2009]. This finding of ε increasing with N_a has been confirmed by subsequent observational studies [*Chen et al.*, 2012; *Lu et al.*, 2007; *Pandithurai et al.*, 2012; *Peng and Lohmann*, 2003], parcel model simulations [*Ching et al.*, 2012; *Peng et al.*, 2007; *Wood et al.*, 2002; *Yum and Hudson*, 2005], and theoretical analysis [*Liu et al.*, 2006]. The theoretical expression by *Liu et al.* [2006] extends the Twomey formulation [*Twomey*, 1959] by adding an analytical expression that relates ε to the power law CCN spectrum and vertical velocity (w), clearly revealing that increasing N_a leads to concurrent increases of ε and N_c whereas increasing w increases N_c but decreases ε. *Lu et al.* [2012] reported observational evidence for the increase of N_c and decrease of ε with increasing w. On the other hand, several studies [*Berg et al.*, 2011; *Hudson et al.*, 2012; *Ma et al.*, 2010; *Martins and Dias*, 2009] reported conflicting observations of decreasing ε with increasing aerosols. The seemingly conflicting observations are still awaiting reconciliation [*Hudson and Noble*, 2014; *Liu et al.*, 2014].

It is noteworthy that the studies reporting decrease of ε with increasing aerosols are mainly on clouds affected by heavy pollution [e.g., *Ma et al.*, 2010] or heavy biomass burning [*Martins and Dias*, 2009], as opposed to the increase of ε with increasing N_a being found mostly in clean or marine clouds. Thus, the contrasting observational dispersion effect seems to support the suggestion that the response of ε to aerosol changes may be like that of N_c, exhibiting different behaviors of distinct regimes [*Liu et al.*, 2014]. However, systematic consideration of ε in classification of aerosol-cloud interaction regime is lacking, and virtually all the theoretical and modeling studies on dispersion effect have been limited to aerosol-limited and transitional regimes. Filling this important gap is the primary objective of this paper. We systematically examine the codependence of ε and N_c on N_a and w using an adiabatic parcel model. Our work extends *Reutter et al.* [2009] in two aspects. First, *Reutter et al.* [2009] only examined the regime dependence of N_c; we add the dispersion effect and consider N_c and ε together. Second, *Reutter et al.* [2009] focused on pyroconvective clouds with N_a ranging from 200 to 10^5 cm^{-3} and w from 0.25 to 20 m s^{-1}. We extend the ranges of both N_a (10 to 10^5 cm^{-3}) and w (0.05 to 20 m s^{-1}) to cover the clouds under pristine conditions and with lower w as observed in stratus clouds as well. As will become evident, these extensions permit a more complete understanding and characterization of aerosol-cloud interaction regimes and reconcile the conflicting observations on dispersion effect; the results have vital implications for the roles of regime dependence and dispersion effect in resolving the conundrum of aerosol indirect effects.

2. Description of Model and Simulation Setup

The new parcel model follows the widely used concept of "Lagrangian bin" [*Howell*, 1949] and contains full treatment of droplet nucleation and condensation processes with the flexibility of user-specified aerosol size distribution and detailed aerosol chemistry composition [*Heymsfield and Sabin*, 1989; *Leaitch et al.*, 1986]. The key physics of this model using in this study is same to other cloud parcel models: adiabatic updraft cooling leads to supersaturation in the parcel, which drives water vapor to condense on existing wet particles. Vapor diffusion process is described by condensational growth equation [*Lamb and Verlinde*, 2011]. The κ-Kohler model is used to treat aerosol growth and droplet nucleation [*Petters and Kreidenweis*, 2007; *Pöschl et al.*, 2010]. The processes of droplet collision and coalescence, sedimentation and entrainment-mixing are not considered in this study. The numerical scheme follows the ordinary differential equation solver released in 2013 in Fortran 90 (VODE-F90), which is an extension of the well-known VODE [*Brown et al.*, 1989], and improves the performance of this model.

Because the focus of this study is on the effects of N_a and w on cloud properties, for simplicity the input aerosol size distribution is assumed to be a lognormal distribution with geometric mean radius of 0.06 μm and geometric standard deviation of 1.5 [*Reutter et al.*, 2009], and the aerosol chemical composition is assumed to be sulfate ammonium with hygroscopicity parameter (κ) of 0.61 [*Petters and Kreidenweis*, 2007]. The parcel starts at the altitude with air temperature of 10°C, air pressure of 919 hPa, and air relative humidity of 95% according to similar studies [*Ghan et al.*, 2011; *Reutter et al.*, 2009; *Xue and Feingold*, 2004]. The number of size bin is 200, and the time step is determined by the relationship of 1.0 m/w [*Saleeby and Cotton*, 2004]. The size bins are distributed logarithmically between 0.01 μm and 1 μm. As *Reutter et al.* [2009], this study focuses primarily on the results at the level of maximum supersaturation.

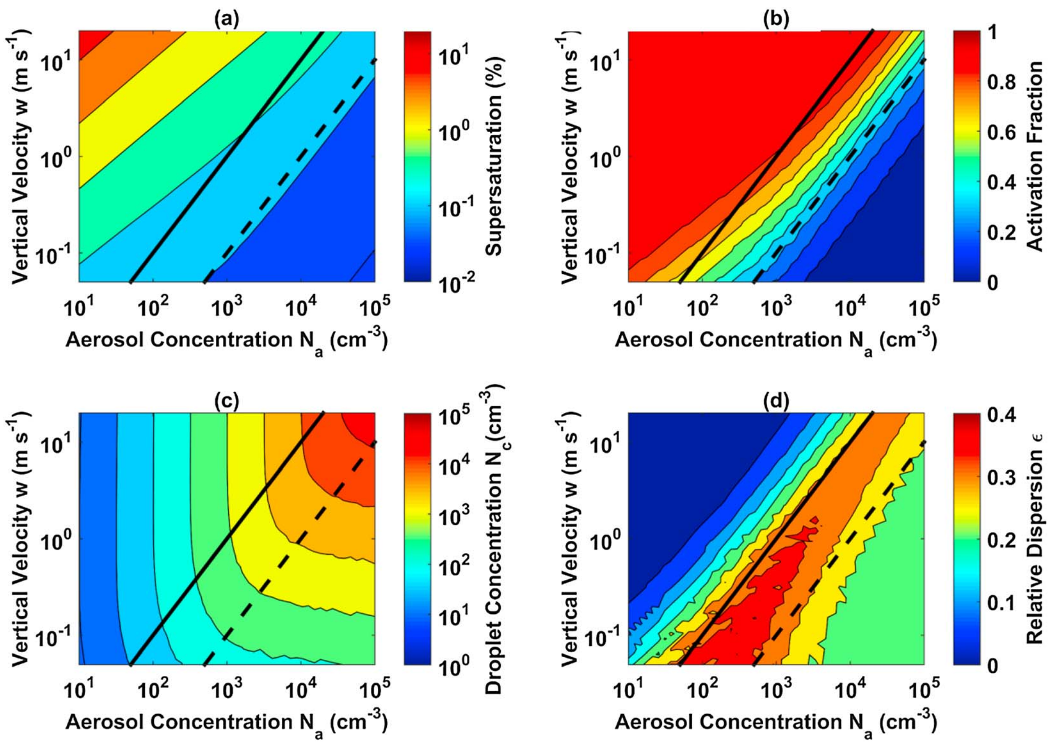

Figure 1. Joint dependence on aerosol number concentration (N_a) and vertical velocity (w) of (a) maximum supersaturation, (b) activation fraction, (c) cloud droplet number concentration (N_c), and (d) cloud droplet relative dispersion ε. The solid and dashed black lines denote the expressions obtained by *Reutter et al.* [2009] to distinguish between the different regimes: solid black line: $w = 10^{-3}\,N_a$; dashed black line: $w = 10^{-4}\,N_a$.

3. Regime Dependence of Aerosol-Cloud Interaction

3.1. Regime Classification

Current understanding and classification of the aerosol-cloud interaction regime is based predominantly on the response of N_c to N_a and w [*Reutter et al.*, 2009]. Thus, to lend confidence in our model, we first examine the dependence of N_c on N_a and w and compare the results with *Reutter et al.* [2009]; we then examine the dependence of ε on N_a and w and analyze the results to improve regime classification. A total of 2500 cases with different combinations of N_a (50 values between 10 and $10^5\,\mathrm{cm}^{-3}$) and w (50 values between 0.05 and $20\,\mathrm{m\,s}^{-1}$) are simulated. The results are summarized in Figure 1, which shows (a) maximum supersaturation, (b) activation fraction, (c) N_c, and (d) ε as a joint function of N_a and w. Also shown for comparison are the two equations provided by *Reutter et al.* [2009] to distinguish between three different regimes of N_c dependence according to their numerical simulations: solid black line for $w/N_a = 10^{-3}\,\mathrm{m\,s^{-1}\,cm^3}$ and dashed black line for $w/N_a = 10^{-4}\,\mathrm{m\,s^{-1}\,cm^3}$. According to *Reutter et al.* [2009], the aerosol-limited regime and updraft-limited regime correspond to $w/N_a > 10^{-3}\,\mathrm{m\,s^{-1}\,cm^3}$ and $w/N_a < 10^{-4}\,\mathrm{m\,s^{-1}\,cm^3}$, respectively; between the two lines lies the transitional regime. Evidently, the aerosol-limited regime is characterized by high w/N_a ($>10^{-3}\,\mathrm{m\,s^{-1}\,cm^3}$), high supersaturation, and high activation fraction. In this regime, N_c is linearly proportional to N_a and largely independent of w. The updraft-limited regime is characterized by low w/N_a ($<10^{-4}\,\mathrm{m\,s^{-1}\,cm^3}$), low supersaturation, and low activation fraction. In this regime, N_c depends mainly on w, and largely independent of N_a. In the transitional regime, the variables fall intermediary between the aerosol- and updraft-limited regimes, and N_c depends nonlinearly on both N_a and w. These results are largely consistent with *Reutter et al.* [2009].

Figure 1d shows that not only the ε-N_a relationship exhibits distinct regimes but also the regime dependence is nonmonotonic: for a given value of w: ε first increases with increasing N_a in the aerosol-limited regime but decreases with increasing N_a in the updraft-limited regime, with a peak occurring in the transitional regime. The nonmonotonic regime dependence of ε on N_a is worth emphasizing, as opposed to the nonlinear yet still

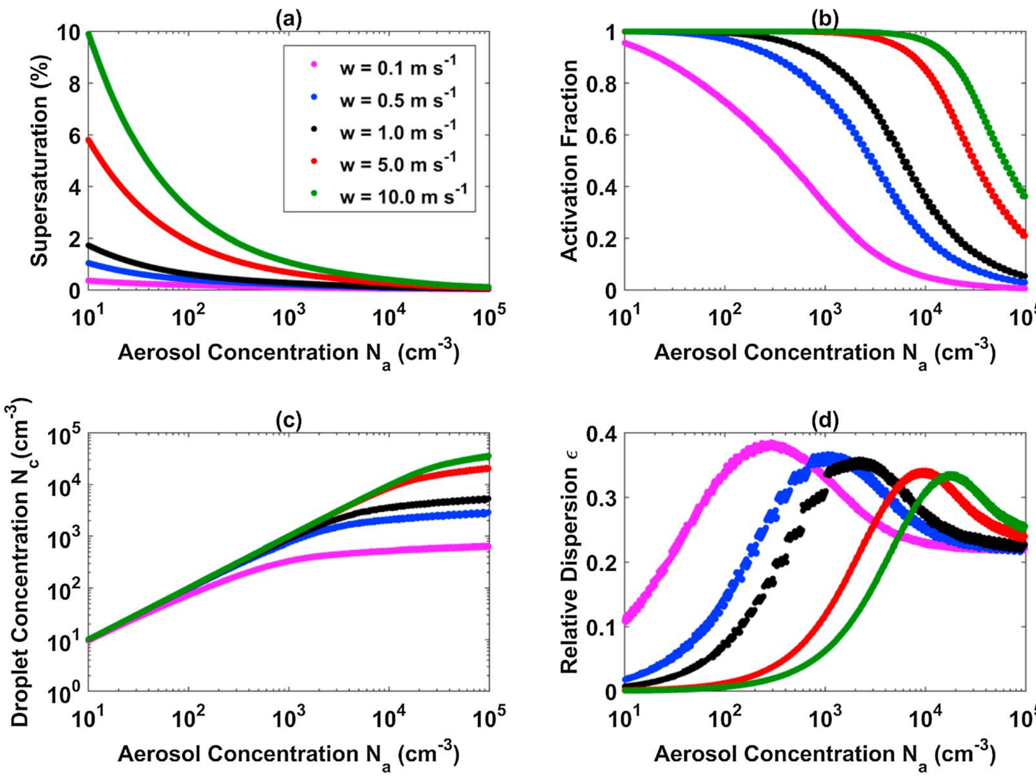

Figure 2. Dependence on aerosol number concentration (N_a) of (a) maximum supersaturation, (b) activation fraction, (c) cloud droplet number concentration (N_c), and (d) cloud droplet relative dispersion (ε) at selected values of updraft velocity (w).

monotonic dependence of N_c on N_a shown in Figure 1c. Equally worth emphasizing is that ε always peaks in the transitional regime confined by the two expressions obtained by *Reutter et al.* [2009] based on the response of N_c. The dependence of ε on w for a given N_a exhibits stark difference with that of N_c as well; ε decreases with increasing w when N_a is low in the aerosol-limited regime but increases with increasing w when N_a is high in the updraft-limited regime. These unique features of ε-N_a relationship as compared to N_c-N_a relationship can be better seen in Figure 2, which shows the dependence on N_a of (a) maximum supersaturation, (b) activation fraction, (c) N_c, and (d) ε at several selected values of w representative of the wide range examined. Note that to improve the accuracy of simulated ε shown in this figure, time steps are 0.01 s for w of 5 m s^{-1} and 10 m s^{-1} and 1 s for other values of w. Five hundred size bins are used when w is 0.1 m s^{-1}.

3.2. New Regime Separation Equation

The distinct dependence of aerosol-cloud interaction regimes calls for simple expressions that can be used to identify which regime the cloud in question lies in. The two expressions proposed by *Reutter et al.* [2009] based on the N_c-N_a relationship can be used for this purpose. Here we propose to simplify the problem by taking advantage of the unique feature of ε peaking at a certain value of N_a for a given w, which indicates that the regime transition of the ε-N_a relationship is much sharper than the N_c-N_a relationship. The point of peak ε can be defined as the transitional point, and the relationship between the pair of N_a and w at the transitional point can be used to simply separate aerosol-limited from updraft-limited regimes.

Figure 3 shows the relationship between the transitional aerosol concentration (N_a*) and updraft velocity (w*) obtained from all the simulations, along with a line representing the linear regression equation that divides the aerosol-limited and updraft-limited regimes:

$$w^* = 5.298\times10^{-4} N_a^* \tag{1}$$

where w* is in unit m s^{-1} and N_a* is in unit cm^{-3}. The aerosol-limited and updraft-limited regimes lie above and below the line described by equation (1), respectively. Also shown for comparison are the two bordering

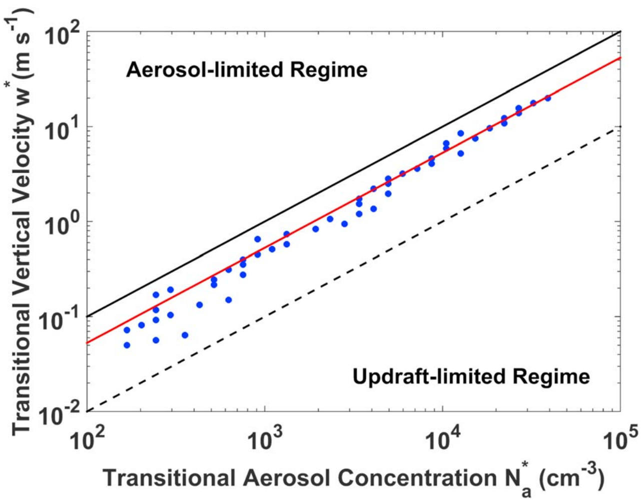

Figure 3. Relationship between the transitional aerosol number concentration (N_a*) and the transitional vertical velocity (w*). The blue dots denote the model simulations from Figure 1, and the red solid line represents the linear fit $w^* = 5.298 \times 10^{-4} N_a^*$. The solid and dashed black lines are the same as Figure 1.

expressions (black solid and dashed lines) given by *Reutter et al.* [2009] based on the dependence of N_c on N_a. It is obvious that the new ε-based regime equation falls intermediately between the two N_c-based regime expressions bordering the aerosol-limited and updraft-limited regimes, supporting the use of equation (1) to separate aerosol-limited regimes from updraft-limited regimes.

3.3. New Physical Understanding

The nonlinear dependence of N_c on N_a and w is well studied and understood physically. Less well studied and understood is the dependence of ε on N_a and w. *Liu and Daum* [2002] relates the behavior of increasing ε with increasing N_a but decreasing w to enhance competition for water vapor and slowdown of condensational narrowing in the presence of high aerosol loading or weak updraft. A number of subsequent studies with adiabatic parcel models [*Peng et al*., 2007; *Yum and Hudson*, 2005] have confirmed this mechanism by showing that as N_a increases, the increase of ε with increasing N_a arises from simultaneous increase of standard deviation and decrease of mean radius of the droplet population. As an extension, *Liu et al.* [2006] further put this mechanism on a theoretical footing by presenting an analytical formulation that extends the Twomey analytical expression for N_c to include ε as well.

However, this mechanism only works for the aerosol-limited regimes; the decrease of ε with further increasing N_a in the updraft-limited regime seems conflicting with the established explanation and somewhat counterintuitive, calling for deeper exploration. It is known that droplet nucleation and subsequent condensational growth depends on the balance between the parcel supersaturation and the particle equilibrium supersaturation, and the droplet size distribution is highly related to the size distribution of radius growth rate [*Srivastava*, 1991]. To understand the regime behaviors, Figure 4a shows particle radius growth rate as a function of radius at three typical value of N_a: $N_a = 50\,\text{cm}^{-3}$ in aerosol-limited regime, $N_a = 2.2 \times 10^3\,\text{cm}^{-3}$

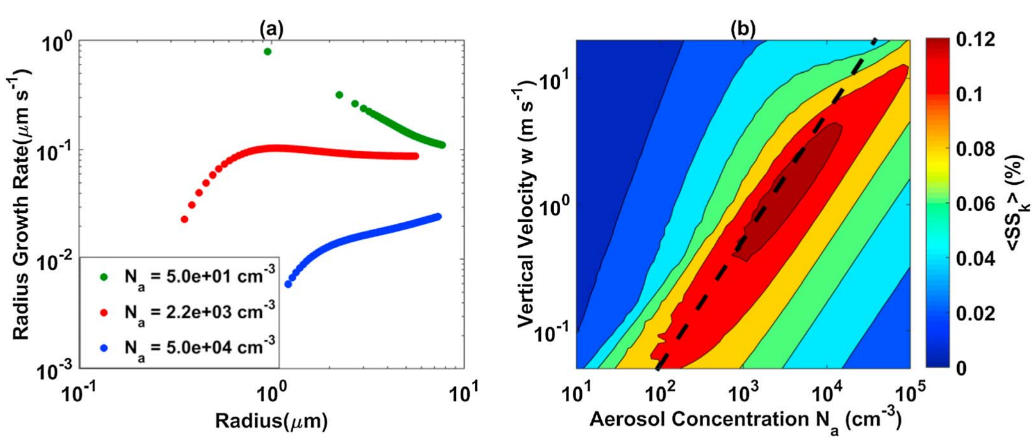

Figure 4. (a) Radius growth rate as a function of radius at three typical values of aerosol concentration (N_a) representing aerosol-limited regime (green), updraft-limited regime (blue), and at the transitional point (red), respectively. Vertical velocity (w) is $1.0\,\text{m s}^{-1}$. (b) Joint dependence on N_a and w of mean droplet equilibrium supersaturation. The dashed line separates the regimes based on equation (1).

in transitional regime and $N_a = 5.0 \times 10^4\ \mathrm{cm}^{-3}$ in updraft-limited regime. Figure 4a shows that when N_a is low in aerosol-limited regime, the droplet radius growth rate decreases with increasing radius and when N_a is high in the updraft-limited regime, the droplet radius growth rate increases with increasing radius. Based on condensational growth theory (see detailed derivation in Appendix A), the first derivative of radius growth rate to radius for each particle relies on the "driving force"—the differences between parcel supersaturation S and particle equilibrium supersaturation S_k. Comparison between Figures 1a and 4b shows that in aerosol-limited regime S is much larger than S_k, which leads to a large driving force, negative dependence of growth rate on radius (the dominance of the first term in equation (A3) in Appendix A) and condensational narrowing. As N_a increases, condensational narrowing in aerosol-limited regime slows down, leading to increasing ε with increasing N_a [*Liu et al.*, 2006; *Peng et al.*, 2007; *Yum and Hudson*, 2005]. However, in updraft-limited regime, S and S_k are comparable, which leads to a small driving force, positive dependence of growth rate on radius (the dominance of the second term in equation (A3) in Appendix A), and spectral broadening. The spectral broadening in updraft-limited regime is suppressed with increasing N_a, which causes decrease of ε with increasing N_a. The radius growth rate as a function of particle radius at the transitional point exhibits an intermediate behavior. These results reinforce the importance to consider the curvature and solute effects and resemble somewhat the so-called ripening process [*Çelik and Marwitz*, 1999; *Wood et al.*, 2002].

4. Important Implications for Aerosol Indirect Effects

Stevens and Feingold [2009] pointed out that changes in the system in isolation may be canceled, or compensated for, by an opposing change that becomes evident when the system is looked at as a whole. *Liu and Daum* [2002] showed that a larger ε leads to a larger droplet effective radius, a smaller albedo, and thus warming effect on climate that negates part of the cooling effect from the increased droplet concentration. *Liu et al.* [2008] further showed that the magnitude of ε is proportional to that of the number effect, and thus accounting for dispersion effect likely reduces the intermodel discrepancy of aerosol indirect effect as well. Climate model simulations that consider the dispersion effect largely confirmed these results as well [*Peng and Lohmann*, 2003; *Penner et al.*, 2006; *Rotstayn and Liu*, 2003, 2009].

The new finding of unique regime dependence of N_c and ε in this study further reinforces and extends the compensating role of dispersion effect to the updraft-limited regime: dispersion effect is warming and offsets the cooling of the number effect when the number effect is strong in the aerosol-limited regime whereas it is cooling and enhances the cooling of the number effect when the number effect is weak in the updraft-limited regime.

5. Conclusions

The responses of cloud droplet number concentration and relative dispersion to changes in aerosol number concentration and vertical velocity are investigated together by performing parcel model simulations with wide ranges of aerosol concentration and updraft velocity that cover virtually all likely cases of ambient clouds, improving our understanding of regime dependence of aerosol-cloud interactions, reconciling conflicting observations on dispersion effect, and reducing intermodel uncertainties in aerosol indirect effects. It is shown that combined consideration of droplet number concentration and relative dispersion (i.e., ratio of standard deviation to the mean radius of the cloud droplet size distribution) provides a more complete description of regime dependence of aerosol-cloud interactions than considering droplet number concentration alone: relative dispersion increases with increasing aerosol concentration in the aerosol-limited regime, peaks at a certain aerosol concentration in the transitional regime, and decreases with further increasing aerosol concentration in the updraft-limited regime. This new finding further reconciles contrasting observations in literature as a manifestation of regime dependence of relative dispersion. The contrasting behaviors of dispersion effect between the aerosol-limited and updraft-limited regimes reinforce the compensating role of dispersion effect, which negates the cooling effect when the number effect is strong in the aerosol-limited regime but enhances the cooling when the number effect is weak in the updraft-limited regime, thus helping reduce the uncertainty in aerosol indirect effects in climate models. The conspicuous peak behavior of relative dispersion further defines a new expression that quantifies the relationship between the transitional aerosol number concentration and vertical velocity and separates the aerosol- and updraft-limited regimes.

The following points are noteworthy for the future study. First, in addition to the primary impacts from aerosol number concentration and vertical velocity, "aerosol secondary parameters (i.e., aerosol chemical composition, mean radius, and spectral shape)" are also expected to affect the regime classification [*Feingold and Chuang*, 2002; *Shantz et al*., 2003, 2008, 2010; *Xue and Feingold*, 2004]. Second, this study is mainly concerned with the results at the level of maximum supersaturation like most previous studies. It is worthwhile to examine the height dependence of the aerosol-cloud interaction regimes. Finally, this study focuses on adiabatic clouds. The effect of entrainment-mixing processes will be investigated with an entraining cloud parcel.

Appendix A: Theoretical Analysis of Dependence of Growth Rate on Droplet Radius

The diffusive growth rate of a cloud droplet is described by equation (A1):

$$\frac{\mathrm{d}r}{\mathrm{d}t} = \frac{1}{r}\frac{S - S_k}{G}, \tag{A1}$$

$$G = \left[\frac{RT\rho_w}{M_w D_v^{'} e_s(T)} + \frac{l_v \rho_w}{M_w k_T^{'} T}\left(\frac{l_v}{RT} - 1\right)(1 + S_k)\right]^{-1}, \tag{A2}$$

where S is the parcel supersaturation, S_k is the particle equilibrium supersaturation, r is the particle radius, t is time, R is the gas constant, T is the air temperature, M_w is the mole mass of water, ρ_w is the water density, e_s is the saturation vapor pressure, and l_v is the latent heat. D'_v and k'_T are the modified diffusion coefficient and thermal conductivity including near droplet surface modification [*Lamb and Verlinde*, 2011]. Note that S_k is often ignored in the calculation of G.

Taking the first derivative of equation (A1) with respect to droplet radius leads to equation (A3).

$$\frac{\mathrm{d}\left(\frac{\mathrm{d}r}{\mathrm{d}t}\right)}{\mathrm{d}r} = -\frac{1}{rG}\left[\frac{S - S_k}{r} + \frac{\mathrm{d}S_k}{\mathrm{d}r}\right] \tag{A3}$$

A negative value of the left-hand side (LHS) of equation (A3) indicates that the cloud droplet distribution narrows because larger droplet grows slower than smaller droplets whereas a positive LHS value indicates that the cloud droplet distribution broadens because larger droplets grow faster than smaller droplets. Neglecting the curvature and solute effects ($S_k \equiv 0$), cloud droplet distribution would narrow during condensational growth (termed as condensational narrowing). This assumption holds when S is much larger than S_k, and the first term dominates on the right-hand side (RHS) of equation (A3) as in aerosol-limited regime (see main body text). However, when S and S_k are comparable, the first term in the bracket on the right-hand side of equation (A3) is negligibly small and the second term dominates, which leads to droplet growth rate increasing with droplet radius, because S_k decreases with particle sizes when particles are larger than their critical radii based on the Köhler theory [*Köhler*, 1936; *Pruppacher and Klett*, 1997].

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