Height Dependency of Aerosol-Cloud Interaction Regimes

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Key Points:

1. New features in the updraft-limited regime improve the understanding of the height dependency of aerosol-cloud interaction regimes.
2. Particle equilibrium supersaturation plays an important role in understanding the new features of the updraft-limited regime.
3. A new parameterization is proposed for the dependence of rain initiation height on aerosol number concentration and vertical velocity.
Abstract.

This study investigates the height dependency of aerosol-cloud interaction regimes in terms of the joint dependence of the key cloud microphysical properties (e.g. cloud droplet number concentration, cloud droplet relative dispersion, etc.) on aerosol number concentration ($N_a$) and vertical velocity ($w$). The three distinct regimes with different microphysical features are the aerosol-limited regime, the updraft-limited regime, and the transitional regime. The results reveal two new phenomena in updraft-limited regime: 1) The “condensational broadening” of cloud droplet size distribution in contrast to the well-known “condensational narrowing” in the aerosol-limited regime; 2) Above the level of maximum supersaturation, some cloud droplets are deactivated into interstitial aerosols in the updraft-limited regime whereas all droplets remain activated in the aerosol-limited regime. Further analysis shows that the particle equilibrium supersaturation plays important role in understanding these unique features. Also examined is the height of warm rain initiation and its dependence on $N_a$ and $w$. The rain initiation height is found to depend primarily on either $N_a$ or $w$ or both in different $N_a$-$w$ regimes, suggesting a strong regime dependence of the second aerosol indirect effect.
1. Introduction

Aerosols influence weather and climate through directly scattering and absorbing radiation (termed as the direct aerosol effect) and indirectly serving as cloud condensation nuclei (CCN) and ice nuclei (termed as the indirect aerosol effect) [Albrecht, 1989; Haywood and Boucher, 2000; Lohmann and Feichter, 2005; Twomey, 1974; 1977]. Twomey [1974; 1977] pointed out that an increase in aerosol number concentration ($N_a$) leads to increases in 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 $N_c$ increasing with increasing $N_a$ 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 report [IPCC, 2013]. 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 science community after decades of research.

Two microphysical factors have been proposed to be partially responsible for the tenacious problem. The first is the regime-dependence of $N_c$ on $N_a$. It is well known that for a given updraft velocity ($w$), the dependence of $N_c$ on $N_a$ is non-linear and regime-dependent: $N_c$ increases linearly with $N_a$ when $N_a$ is low, but the $N_c$ - $N_a$ relationship becomes sublinear when $N_a$ is high [Feingold et al., 2001]. Reutter et al. [2009] classified the non-linear $N_c$-$N_a$ relationship into three distinct regimes using a detailed cloud parcel model:
1) the aerosol-limited regime is characterized by high $w/N_a$ ($\geq 10^{-3}$ m s$^{-1}$ cm$^3$), high parcel supersaturation ($S_p$), and strong (linear) dependence of $N_c$ on $N_a$ but weak dependence of $N_c$ on $w$; 2) the updraft-limited regime is characterized by low $w/N_a$ ($\leq 10^{-4}$ m s$^{-1}$ cm$^3$), low $S_p$, and weak dependence of $N_c$ on $N_a$ but strong dependence of $N_c$ on $w$; and 3) the transitional regime falls between the aerosol-limited and updraft-limited regimes with sub-linear dependence of $N_c$ on both $N_a$ and $w$. They also suggested further studies in the less emphasized updraft-limited regime with more aerosols.

The second microphysical factor is the dispersion effect whereby changes in aerosol properties alter the spectral shape of the cloud droplet size distribution in addition to droplet number concentration. Liu and Daum [2002] showed, by analyzing data from marine clouds under a variety of aerosol conditions with $N_c$ ranging from tens to a few hundred, that increased $N_a$ leads to concurrent increases of $N_c$ and cloud droplet relative dispersion ($\varepsilon$) (i.e. the ratio of cloud droplet standard deviation to cloud droplet mean radius); and the enhanced $\varepsilon$ 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 [Kumar et al., 2016; Peng and Lohmann, 2003; Penner et al., 2006; Rotstyn and Liu, 2003; 2009]. This finding of $\varepsilon$ increasing with $N_a$ is consistent with some 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; 2016; Peng et al., 2007; Wood et al., 2002; Yum and Hudson, 2005], and theoretical analysis [Liu et al., 2006b]. On the other hand, several studies [Berg et al., 2011; Hudson et al., 2012; Lu et al., 2012; Ma et al., 2010; Martins and Dias, 2009] reported conflicting observations of decreasing $\varepsilon$ with increasing $N_a$. It is noteworthy that the studies reporting decrease of $\varepsilon$ with increasing $N_a$ are mainly in clouds affected by heavy pollution (e.g.,
several thousand aerosols per cc and several hundred of mean $N_c$ from Ma et al. [2010]), heavy biomass burning (e.g. up to $N_c=1284$ cm$^{-3}$ in Martins and Dias [2009]) or Amazonian clouds (e.g. $N_a$ up to 4093 cm$^{-3}$ in Cecchini et al. [2017]) , as opposed to the increase of $\varepsilon$ with increasing $N_a$ being found mostly in clean marine clouds.

Chen et al. [2016] systematically examine the co-dependence of $N_c$ and $\varepsilon$ on $N_a$ and $w$ using an adiabatic parcel model. It was shown that given $w$, $\varepsilon$ increases with increasing $N_a$ in the aerosol-limited regime, peaks in the transitional regime, and decreases with further increasing $N_a$ in the updraft-limited regime. This finding reconciles contrasting observations in literature and reinforces the compensating role of dispersion effect. The non-monotonic behavior of $\varepsilon$ further quantifies the relationship between the transitional $N_a$ and $w$ that separates the aerosol- and updraft-limited regimes.

However, previous Aerosol-Cloud Interaction (ACI) studies, including ours in Chen et al. [2016], have been primarily focused on the height of maximum supersaturation (See Appendix B for details). Although $N_c$ being a constant above the height of maximum supersaturation seems to be a reasonable assumption, it may be problematic under certain conditions, for example, higher $N_a$, low $w$, larger mode radius, and/or anthropogenically-influenced aerosol type [Nenes et al., 2001].

Furthermore, it is well known that $\varepsilon$ depends strongly on the height above cloud base [Çelik and Marwitz, 1999; Peng et al., 2007]. Traditional condensational theory predicts that condensational growth leads to narrowing of cloud droplet size distribution with height [Lamb and Verlinde, 2011]. Observational studies show the variation of $\varepsilon$ with height depends on the pollution level of the clouds [Pawlowska et al., 2006]. Also, the cloud radiative effects are determined by the whole cloud layer above cloud base. Thus, the
complete ACI regime is likely height-dependent. However, systematic study of the ACI regime dependence on height is still lacking, and thus the first objective of this study is to extend Chen et al. [2016] to investigate the height dependence of the ACI regime.

The second objective of this study is to examine the dependence of rain initiation height on aerosol properties and vertical velocity. Many studies have shown that aerosol-induced precipitation suppression not only horizontally increases the cloud fraction as conceived by [Albrecht, 1989] and [Liou and Ou, 1989], but also may vertically increase the cloud top height depending on thermodynamic conditions of atmosphere [Pincus and Baker, 1994; Wood, 2007]. Satellite studies have found higher cloud tops are in polluted cumulus clouds [Yuan et al., 2011] and open cell stratocumulus clouds [Christensen and Stephens, 2011] than in clean clouds. But conflicting results have been reported. For example, Segrin et al. [2007] and Christensen and Stephens [2011] found that pollution only led to insignificant cloud-top changes in close cell stratocumulus clouds. Li et al. [2011] found little changes of warm cloud tops over the U. S. Southern Great Plain with aerosols. Our systematic examination of the regime dependence of rain initiation height on $N_a$ and $w$ can provide physical insight into this important issue, because rain initiation and the subsequent drizzling process is one of the primary factors that determine the cloud top height and cloud fraction.

The remainder of this paper is organized as follows. Section 2 describes numerical experimental settings and calculations. Section 3 presents the results and reveals the unique features of cloud microphysical properties in aerosol- and updraft-limited regimes. Key findings are summarized and discussed in Section 4.

2. Model and Numerical Experiments
The cloud parcel model used in this study [Chen et al., 2016] contains the full treatment of droplet nucleation and condensation following the widely used method of “Lagrangian bins” [Heymsfield and Sabin, 1989; Howell, 1949; Leitch et al., 1986]. When the parcel rises and cools, water vapor is condensed on the particles. To simplify the problem, the cloud parcel is treated as a closed system without exchange of air, particles, and energy. Entrainment-mixing, sedimentation, and collision-coalescence processes are not considered in this study (See Appendix A for detailed descriptions of the equations used in the model).

The initial aerosol size distribution is assumed to be lognormal with geometric mean radius of 0.06μm and geometric standard deviation of 1.5 [Reutter et al., 2009]. The hygroscopicity is 0.61 assuming ammonium sulfate [Petters and Kreidenweis, 2007]. The effect of different initial aerosol size distributions on the results is also examined and is discussed in Supporting Information. Simulations are performed with 1000 Lagrangian particle size bins to represent the cloud droplet size distribution. Time step is $\frac{1.0 m}{w}$, except between cloud base and maximum supersaturation where a shorter step ($\frac{0.1 m}{w}$) is used to identify the level of maximum supersaturation more accurately. The vertical resolution of model output is 10 m from cloud base to 600 m, and 100 m above 600 m; the higher resolution in the lower part of clouds facilitates detailed examination of physical processes in the early stage of droplet formation. Parallel computing (OpenMP) is used to facilitate a large number of simulations.

3. Results

3.1 Regime Dependency of Vertical Profiles

Based on Reutter et al. [2009] and Chen et al. [2016], the dependency of key cloud properties on $N_a$ and $w$ are classified into three different regimes at maximum
supersaturation height. Figure 1 is a schematic figure summarizing the key microphysical features of each regime. The information superscripted by (1) and (2) is summarized in this paragraph based on previous studies, and the information superscripted by (3) will be discussed in detail in this paper. First, the aerosol-limited regime is characterized by low N_a/w, high S_p, high activation fraction, a strong dependence of N_c on N_a, and a positive relation between ε and N_a. Second, the updraft-limited regime is characterized by high N_a/w, low S_p, low activation fraction, a strong dependence of N_c on w, and a negative relation between ε and N_a. Third, the transitional regime is located between the aerosol-limited and updraft-limited regimes, exhibits a non-linear dependence of N_c on N_a and w, and the largest values of ε are also in transitional regime. In general, the ratio of N_a to w is a good metric gauging the aerosol-cloud interaction regimes; the aerosol-limited regime, transitional regime, and updraft-limited regime correspond to N_a/w < x, N_a/w = x and N_a/w > x, respectively, where x = 5.7 × 10^{-4} m s^{-1} cm^{3} is estimated by curve-fitting the relationship between the transitional w and the N_a corresponding to the maximum ε (e.g., Figure 3 of Chen et al. [2016]). Note the slight difference of this fitting coefficient from that (5.3 × 10^{-4} m s^{-1} cm^{3}) in Chen et al. [2016], which is due to the differences in the model settings (e.g. number of particle bins, time step) and air heat capacity used (dry air in Chen et al. [2016] and moist air in this study).

To investigate the height dependency, we partition the 2500 simulations (50 values of N_a logarithmically distributed between 10 and 10^5 cm^{-3} and 50 values of w logarithmically distributed between 0.05 and 20 m s^{-1}) into six groups according to the ratio N_a/w ranging from aerosol-limited regime to updraft-limited regime. Figure 2 shows the vertical variations of mean values of S_p, N_c, and ε in different N_a/w groups. The dashed warm color represents aerosol-limited regime; the solid cold color represents the updraft-limited regime; the thick
The black line denotes the transitional regime which is defined by the maximum $\varepsilon$ for each $w$ at the height maximum supersaturation as shown in Chen et al. [2016]. The height relative to the cloud base is hereafter referred to as height for convenience. Note that the simulation is stopped when air temperature reaches -8°C to strictly concentrate on warm cloud processes [Crawford et al., 2012; Li et al., 2013].

The profiles in Figure 2a show that $S_p$ remains larger in the aerosol-limited regime than in the updraft-limited regime during parcel rising. It is also evident that $N_c$ almost holds constant above the level of maximum supersaturation in the aerosol-limited regime as commonly assumed. However, in the updraft-limited regime, $N_c$ first increases with height, peaks at a certain height above the level of maximum supersaturation, and then decreases with further increasing heights. The highest $N_c$ occurs near the transitional regime. As will be detailed in Section 3.2, such “unconventional” change of $N_c$ with height is due to the so-called “kinetic effect” [Chuang et al., 1997; Nenes et al., 2001].

The height dependency of $\varepsilon$ (Figure 2c) is more interesting: $\varepsilon$ decreases rapidly with height above 10m in the aerosol-limited regime, but decreases much slower or even increases in the updraft-limited regime. Above 110 m, $\varepsilon$ decreases with height in both regimes. To understand the behavior of Figure 2c, Figures 3a and 3b further show the cloud droplet size standard deviation ($\sigma$) and mean radius ($R_{\text{mean}}$) as a function of height, respectively. Figure 3a shows decreasing $\sigma$ in the aerosol-limited regime but increasing $\sigma$ in the updraft-limited regime. In the aerosol-limited regime, larger particles grow slower than smaller particles and the droplet size distribution narrows with height, because particle equilibrium supersaturation ($S_k$) is much smaller than $S_p$ and can be ignored. In contrast, in the updraft-limited regime, larger particles grow faster than smaller particles and the size
distribution broadens due to the dependence of $S_k$ on droplet size (See Appendix in Chen et al. [2016]). Grabowski et al. [2011] also showed that $\sigma$ increases with time at lower $w$ in their Lagrangian simulations. The condensational narrowing in the aerosol-limited regime have been abundantly studied in the published literature and textbooks (e.g. [Pruppacher and Klett, 1997]); however the condensational broadening in the updraft-limited regime has not received much attention because both $S_k$ and the variations of $S_k$ with droplet size are usually assumed to be small and ignored (See appendix C for more quantitative analysis).

The increasing $\sigma$ and slower growing $R_{\text{mean}}$ (Figure 3b) lead to the slower decrease of $\epsilon$ with height in the updraft-limited regime. Based on the analysis of the data at 1000m height, $\epsilon$, $\sigma$ and $\frac{1}{R_{\text{mean}}}$ in the extreme updraft-limited regime ($1.9 \times 10^5 \, cm^{-3} \, m^{-1} \, s < N_a/w \leq 2.0 \times 10^6 \, cm^{-3} \, m^{-1} \, s$) are larger than the corresponding values in the extreme aerosol-limited regime ($0.5 \, cm^{-3} \, m^{-1} \, s < N_a/w \leq 7.6 \, cm^{-3} \, m^{-1} \, s$) by the factor of 448.70, 187.23 and 2.51, respectively.

To further investigate how the dependence of $S_k$ on droplet size influences cloud properties, Figure 4 show the results from a suite of simulations which are like those shown in Figure 2 and Figure 3, except that the $S_k$ of the median cloud droplet bin (e.g. the $S_k$ of the 300th activated bin if the number of activated particle bins are 600) has applied to all the cloud droplets when solving the condensational growth equation (Equation A1) in the code. The purpose of this experiment is to dissect the effect from the dependence of $S_k$ on droplet size. Comparison of Figure 4a and Figure 2a shows that larger values of $N_a/w$ correspond to larger values of the differences of $S_p$ between Figure 4a and Figure 2a in the updraft-limited regime, which suggests that ignoring the dependence of $S_k$ on droplet size overestimates the $S_p$, especially when $N_a/w$ is large. Comparison between Figure 4b and Figure 2b shows that
droplets do not evaporate and deactivate in the higher part of clouds when the dependence of $S_k$ on droplet radius is ignored in the updraft-limited regime. Compared to Figure 2c, Figure 4c shows a smaller $\varepsilon$ for all cases and faster decreasing at high levels in the updraft-limited regime. Figure 4d and 4e show that ignoring dependence of $S_k$ on droplet size leads to a smaller $\sigma$ for all the cases. The decreasing $\sigma$ with height in the updraft-limited regime when change of $S_k$ with radius is ignored confirms that the "condensational broadening" in the updraft-limited regime is the result of variations of $S_k$ with particle sizes. Smaller values of $R_{\text{mean}}$ in the updraft-limited regime in Figure 4e than those in Figure 3b are consistent with the $N_c$ profiles (Figure 4b and 2b): more activated cloud droplets lead to smaller cloud droplet radius. Based on the analysis of the data of Figure 4 at 1000m, $\varepsilon$, $\sigma$, and $\frac{1}{R_{\text{mean}}}$ in the extreme updraft-limited regime are larger than the corresponding values in the extreme aerosol-limited regime by the factor of 137.47, 11.58 and 12.31, respectively. A comparison of Figure 2 and Figure 4 indicates that the vertical profiles of $\varepsilon$ are due to more $\sigma$ than $R_{\text{mean}}$ when considering the dependence of $S_k$ on droplet size because the regime variation of $\sigma$ is stronger than $R_{\text{mean}}$. These features suggest that dependence of $S_k$ on droplet size causes evaporation/deactivation of small particles, and thus spectral broadening in the updraft-limited regime. Also, $\varepsilon$ becomes 1-2 order smaller if the dependence of $S_k$ on droplet size is neglected, which implies that $S_k$ is critical for all cases even in the aerosol-limited regime.

To illustrate the detailed regime dependence on $N_a$ and $w$ at different heights, Figure 5 shows regime dependence of $N_c$ and $\varepsilon$ at three typical heights of 20m (a, b, upper panel), 560m (c, d, middle panel), and 2110m (e, f, lower panel) above cloud base. At 20m, only some cases (1412 out of 2500) have reached their maximum supersaturation. At 560m, all the cases just have reached their levels of maximum supersaturation. At 2110m, the
variation of $\varepsilon$ with height become stable (relative changes of the mean values in each $N_a/w$ group is less than 3% per 100m). Besides the similar characteristics to those at the maximum supersaturation, it is interesting to note that $N_c$ starts to decrease with increasing $N_a$ at the lower part of the cloud when $N_a$ is larger than about $10^4$ cm$^{-3}$ (Figure 5a). This is because $S_p$ decreases with increasing $N_a$ faster just above cloud base than in higher levels. The patterns at the two other altitudes are similar to each other as shown in Figure 5c and 5e, except that $N_c$ increases more rapidly with increasing $N_a$ in updraft-limited regime at 560m than at 2110m, which implies decreasing of $N_c$ with height above 560m. Figure 5b, 5d, and 5f show the dependence of $\varepsilon$ on $N_a$ and $w$ at 20m, 560m and 2110m, respectively. The similar pattern of regime dependence remains at different altitudes, with the peak $\varepsilon$ shifting to higher $N_a$ as the cloud parcel rises higher. After a few hundred meters above the maximum supersaturation, the branch of decreasing $\varepsilon$ with increasing $N_a$ disappears within the ranges of $N_a$ and $w$ examined.

Figure 6 examines $\sigma$ and $R_{\text{mean}}$ at the same levels as Figure 5. The plots of $\sigma$ substantiate the spectral narrowing in the aerosol-limited regime and spectral broadening in the updraft-limited regime from 20m to 2110m. For the same $N_a$, a larger (smaller) $w$ corresponds to a narrower (broader) size distribution. These phenomena are consistent with Figure 3a and can be explained by the different magnitudes of $S_p$ minus $S_k$ in the different regimes. Plots of $R_{\text{mean}}$ show that particle growth from 20m to 2110m in the aerosol-limited regime is more prominent than those in the updraft-limited regime, which can be explained by the theory that the enhanced competition among particles for available water vapor in the updraft-limited regime.

3.2 “Kinetic effects” and their impacts on cloud droplet number concentration
The decrease of \( N_c \) with increasing \( N_a \) above the level of maximum supersaturation in the updraft-limited regime as shown in Figure 2b is more evident in Figure 7, which shows \( N_c \) normalized by their maximum values in each regime group. The three profiles from the aerosol-limited regime converge and decrease with height due to the parcel volume expansion. The three profiles from the updraft-limited regime show that the strengths of evaporation and deactivation at higher levels depend on regimes. This subsection examines the physical mechanism behind this phenomenon.

*Nenes et al.* [2001] demonstrated three types of kinetic effects termed as "inertial mechanism," "evaporation mechanism," and "deactivation mechanism". Briefly, "inertial mechanism" mostly influences those particles growing from large dry aerosols. These particles always have a large critical radius, which needs sufficient time to reach and thus mostly they are unable to reach their critical radius in the lower part of clouds, but their size is large enough to play important role in radiation and precipitation. Evaporation mechanism applies to those particles that could be potentially activated but evaporate to form interstitial aerosols because the time they are exposed to high supersaturation is not long enough. Deactivation mechanism applies to those activated particles that evaporate into interstitial aerosols because \( S_p \) drops below \( S_k \). The kinetic effects can be further illustrated by examining the differences between \( S_p \) and \( S_k \) that drives condensation/evaporation of particles

Figure 8a and 8b show two examples of vertical variations of \( S_k \) and \( S_p \) at different size bins in the aerosol-limited regime and updraft-limited regime, respectively. The black lines are the particle bins with the smallest activated particles at maximum supersaturation; the green circles represent the particle bins with smallest activated particles at about 1000m.
Initially, dry aerosols are assigned to each particle bins in the order of increasing sizes, and the same order remains when particles grow by condensation. In the typical aerosol-limited regime, $S_p$ is much larger than $S_k$ even when the parcel rises high; thus, the smallest activated particle at maximum supersaturation always remains activated. However, in the typical updraft-limited regime, $S_p$ is only slightly larger than $S_k$ near the level of maximum supersaturation. Small differences between $S_p$ and $S_k$ cause small “driving force” for growth.

Since $S_p$ decreases with height faster than $S_k$ does, at 1000m (Figure 8b), the particles in the 597th bin evaporates and deactivated into interstitial particles, and the 617th bin become the smallest activated particle bin. Similarly, the 617th size bin is no longer the smallest activated particle bins when the parcel further rises higher than 1000m, because particles of that size bin have $S_k$ larger than $S_p$, evaporate and eventually deactivate. This is the same as “evaporation mechanism” or “deactivation mechanism” identified by Nenes et al. [2001]:

Both “evaporation mechanism” and “deactivation mechanism” applies to the particles with their critical supersaturation lower than $S_{max}$ but higher than local $S_p$. Thus, they evaporate from cloud droplets to interstitial aerosols ultimately. When local $S_p$ becomes smaller than $S_k$, “evaporation mechanism” applies to the particles of radius smaller than the Köhler critical radius while “deactivation mechanism” applies to the particles of radius larger than their Köhler critical radius. Thus “deactivation mechanism” is the primary reason for decreasing $N_c$ above maximum supersaturation. Note that in this study, the definition of cloud droplets follows that given by Reutter et al. [2009]: particles are larger than the critical radius $r_{c,\text{cld}}$ as Equation 1 shown.

$$r_{c,\text{cld}} = \frac{2a}{3 \ln(S_p)}$$  \hspace{1cm} (1a)
where $\sigma_s$ is surface tension, $\rho_w$ is the water density, $R_w$ is gas constant for water, and $T$ is air temperature. Likewise, droplets will deactivate into particles when their radii are smaller than $r_{c,cld}$. Although $r_{c,cld}$ is not exactly the Köhler critical radius, “deactivation mechanism” is still the most plausible mechanism to cause the decrease of $N_c$. Because decreasing $S_p$ leads to increasing $r_{c,cld}$ (Equation 1), the evaporated particle has their radius larger than $r_{c,cld}$ but smaller than the $r_{c,cld}$ at the next time step. This requires the evaporated particle has larger $S_k$ and hence smaller radius growth rate. Smaller particle size contributes to larger $S_k$ (Equation A9). Thus the evaporated particle are mostly the ones with their radii slightly larger than $r_{c,cld}$ and with their Köhler critical radius smaller than the particle size, which corresponds to the “deactivation mechanism”.

Figure 8c and 8d show the radius growth rate of the same cases as figure 8a and 8b. In the typical aerosol-limited regime, larger particles have smaller growth rate than smaller particles. While in the typical updraft-limited regime, larger particles have larger growth rate than smaller particles. Note that, the differences of radius growth rate of the 700th bin and 900th bin are not significant below 200m because the supersaturation differences “$S_p-S_k$” are large.

To summarize, the smaller differences between $S_k$ and $S_p$ in the updraft-limited regime causes loss of some small cloud droplets and thus decrease of $N_c$ through the “deactivation mechanism”.

3.3 Regime dependence of rain initiation height
Rain initiation is critical for understanding warm rain processes [Beard and Ochs, 1993; Magaritz-Ronen et al., 2016; Wang and Grabowski, 2009]. Recent studies investigated the rain initiation height by assuming a certain effective radius \( r_e \) (or volume mean radius) as an empirical threshold of rain initiation [Freud and Rosenfeld, 2012]. However, systematic examination of the dependence of the rain initiation height on the \( N_a \) and \( w \) (or regime dependence of rain initiation height) is still lacking. Liu et al. [2005] derived a physically based threshold function as a function of droplet concentration, liquid water content, and relative dispersion. Liu et al. [2008] further related this threshold function to threshold radar reflectivity and reported observational evidence for the dependence of the threshold radar reflectivity on \( N_c \). Here we take advantage of this theoretical autoconversion threshold function to examine the regime dependence of rain initiation height.

Briefly, the autoconversion threshold function \( T_{\text{auto}} \) [Liu et al., 2006a; Liu et al., 2005] is given as

\[
T_{\text{auto}} = \frac{\int_{r_{c,\text{rain}}}^{\infty} (\int_{r_{c,\text{rain}}}^{\infty} K(r_1, r_2) r_2^3 n(r_2) dr_2) n(r_1) dr_1}{\int (\int K(r_1, r_2) r_2^3 n(r_2) dr_2) n(r_1) dr_1} \quad \text{or} \quad (2a)
\]

\[
T_{\text{auto}} = \left[ \frac{\int_{r_{c,\text{rain}}}^{\infty} r^6 n(r) dr}{\int_{0}^{\infty} r^6 n(r) dr} \right] \left[ \frac{\int_{r_{c,\text{rain}}}^{\infty} r^3 n(r) dr}{\int_{0}^{\infty} r^3 n(r) dr} \right] \quad \text{or} \quad (2b)
\]

\[
r_{c,\text{rain}} = 2.8522 \frac{N_c^{\frac{1}{6}}}{LWC^{\frac{1}{2}}} \quad \text{or} \quad (2c)
\]

where \( K(r_1, r_2) \) is the collection kernel between droplets with radius as \( r_1 \) and \( r_2 \), \( r \) is droplet radius, \( n(r) \) is the droplet size distribution, \( r_{c,\text{rain}} \) is the critical radius of rain initiation given by Liu et al. [2004], and LWC is the liquid water content. The threshold function \( T_{\text{auto}} \) ranges between 0 and 1, and measures the strength of collection process relative to the diffusional growth; a larger \( T_{\text{auto}} \) indicates a relatively stronger occurrence of collection process.
Equation 2 reveals that LWC and thus $T_{auto}$ increases with height as the parcel rises, eventually leading to drizzle formation or the start of collection process. In this study, $1 \geq T_{auto} \geq 0.9$ is used to define the rain initiation height. Note that our sensitivity study using different values of $T_{auto}$ (i.e. 0.1, 0.3, 0.5, 0.7, 0.9) shows that the results reported in this study are not sensitive to the choice of $T_{auto}$ value (not shown).

Figure 9 presents the joint dependency of rain initiation height ($H_{rain}$) on $N_a$ and $w$ (a); also shown are the regime dependence of the corresponding LWC (b), $N_c$ (c), and $\epsilon$ (d). The blank area indicates that the air temperature is lower than -8°C and the simulation stops after $T_{auto}$ reaches 0.9. Evidently, the basic pattern of $H_{rain}$ is like that of $N_c$: $H_{rain}$ depends primarily on $N_a$ when $N_a/w$ is low in the aerosol-limited regime and on $w$ when $N_a/w$ is high in the updraft-limited regime. The close resemblance of the corresponding LWC and $H_{rain}$ reflects the linear relationship between LWC and height [Albrecht et al., 1990]. The general pattern of $N_c$ is same as those at other levels. Figure 9d shows that $\epsilon$ has low values in the aerosol-limited regime and high values in the updraft-limited regime at $H_{rain}$, consistent with the condensational narrowing in the aerosol-limited regime and condensational broadening in the updraft-limited regime.

Further, it is desirable to have an analytical expression for quantifying the regime dependence of $H_{rain}$. We find that the relationship between $H_{rain}$ and $N_a$ normalized by the corresponding transitional regime values (superscripted by *) is virtually collapsed onto a single curve (Figure 10). The transitional regime values correspond to maximum relative dispersion at each $w$ at the height of maximum supersaturation (Equation 3b and 3c). It can be seen that $H_{rain}/H_{rain}^*$ first increases rapidly with increasing $N_a/N_a^*$ in the aerosol-limited regime ($N_a/N_a^* < 1$), and gradually levels off in the updraft-limited regime ($N_a/N_a^* > 1$). The
“universal” curve can be described by

\[
\frac{H_{\text{rain}^*}}{H_{\text{rain}}} = \begin{cases} 
-3.926 & 1 + \exp\left(0.6339 \times \ln\left(\frac{N_a}{N_a^*}\right) - 1.195\right) + 3.934 \\
0.3644 \times \left(\ln\left(\frac{N_a}{N_a^*}\right)\right)^{0.7711} + 0.9398 & \frac{N_a}{N_a^*} \leq 1 \\
\frac{N_a}{N_a^*} & \frac{N_a}{N_a^*} > 1
\end{cases}
\]

\(H_{\text{rain}^*} = 1.570 \times 10^3 \, w^{0.781}\) \hspace{1cm} (3b)

\(N_a^* = 1.752 \times 10^3 \, w\) \hspace{1cm} (3c)

where the expressions for \(H_{\text{rain}^*}\) and \(N_a^*\) are obtained by fitting the numerical simulations. The units of \(H_{\text{rain}}, w\) and \(N_a\) are \(\text{m}, \text{m s}^{-1}\) and \(\text{cm}^3\), respectively. To the best of our knowledge, Equation 3 is the first expression that quantifies the co-dependence of rain initiation height on \(N_a\) and \(w\).

It is worth noting that the collision coalescence process is not considered in the parcel model, some changes of the results are expected if collision coalescence process is considered in view of its role in determining the cloud droplet size distribution [Feingold et al., 1996; Wood, 2006]. The effect of collision-coalescence process is expected to be larger in the aerosol-limited regime more than in updraft-limited regime because of larger \(R_{\text{mean}}\) (Figure 3b). In view of the limitations of this study (e.g., adiabatic parcel model without considering collision-coalescence process), it needs to be evaluated against more sophisticated numerical simulations such as large-eddy simulations and measurements.

4. Conclusions and discussions

This work extends our previous study to examine the height dependence of aerosol-cloud interaction regime, esp., height dependency of \(N_c\) and \(\varepsilon\) as a joint function of \(N_a\) and \(w\). Also investigated, for the first time, is the regime dependence of \(H_{\text{rain}}\) on \(N_a\) and \(w\).
Analysis shows that cloud droplet size distributions experience spectral narrowing with height in the aerosol-limited regime, but spectral broadening with height in the updraft-limited regime. The unique behavior of spectral broadening in the updraft-limited regime arises from the $S_k$ and its dependence on droplet radius (See Appendix C). In the updraft-limited regime, $S_p$ and $S_k$ are comparable; together with the size dependence of the $S_k$, larger particles “win” the competition for water vapor between particles with different radius. This new feature does not exist if the dependence of $S_k$ on size is ignored. It is also found that although $N_c$ is constant and determined by that at the level of maximum supersaturation in the aerosol-limited regime as conventionally assumed, it varies nonlinearly with height in the updraft-limited regime (first increases with increasing height, peaks at certain height and then decreases with further increasing height). Also, the reason of $N_c$ behavior is related to the critical role of $S_k$, which causes some small particles to evaporate and eventually deactivated into interstitial particles. However, droplet deactivation likely has a minor role in the spectral broadening in the updraft-limited regime, since only up to 0.08% cloud droplets have negative growth rate.

Relative dispersion exhibits a stronger height dependence. At the level of maximum supersaturation, $\epsilon$ increases in the aerosol-limited regime but decreases in the updraft-limited regime as $N_a$ increases. However, this contrasting behavior gradually disappears at high altitudes within the examined range of $N_a$ and $w$, with the specific height of disappearance relying on the regime. At higher level, increasing $N_a/w$ retards the particle growth in the aerosol-limited regime, but enhances the particle growth in the updraft-limited regime (Figure 3b). Thus, condensational broadening in updraft-limited regime is enhanced with larger $N_a/w$ and the contrasting behavior disappears. The enhanced particle growth in updraft-limited regime can be explained below: larger $N_a/w$ causes more evident
“kinetic mechanism”, smaller $N_c$, and more evaporation of smaller particles; Thus, larger fraction of large droplets, which are better at water vapor consuming and grow faster in updraft-limited regime, leads to the enhancement of particle growth in the updraft-limited regime.

The regime dependence of rain initiation height is examined by coupling the autoconversion threshold function derived in Liu et al. [2006a] with the parcel model simulations. Distinct regime patterns illustrate that the height of rain initiation largely depends on $N_a$ when $N_a$ is small, and on $w$ when $N_a$ is large. Cloud microphysical parameters show significant variations and clear regime patterns at the onset of precipitation when the different combinations of $N_a$ and $w$ are considered. This feature suggests that simple parameterizations based on one cloud microphysical properties (e.g., LWC or $n^{th}$ moment mean radius) may cause large errors when there are large variations of $N_a$ and $w$ in the atmosphere. A new expression is proposed to quantify the dependence of $H_{\text{rain}}$ on $N_a$ and $w$.

Two points are noteworthy. First, the results are based on the simulations with fixed typical initial aerosol size distribution parameters, temperature, and relative humidity. The sensitivities to the input aerosol size distributions have been examined, and the main conclusions remain valid, with some subtle differences (see Supporting Information for details). The impacts of the initial relative humidity and air temperature are found to be even weaker (see Supporting Information for details). Effect of particle hygroscopicity or chemical composition should be examined as well to explore the complete variable space in the ACI regime studies. Second, this study is based on an adiabatic parcel that ignores turbulent entrainment-mixing and collision coalescence processes. An unresolved question
is how the regime patterns (especially for $\varepsilon$) will change when these processes are considered. These will be the subject of future study.

**Acknowledgement.** This study is supported by the US Department of Energy’s Atmospheric System Research (ASR) program. The simulation data has been uploaded as Supporting Information.
Appendix A: Key Model Equations and Further Analysis

Briefly, the cloud parcel model consists of the following key equations. The condensational growth is described by:

$$\frac{dm_i}{dt} = 4\pi r_i G \cdot (S_p - S_{K,i})$$

(A1a)

$$G = \left[ \frac{RT}{M_w D'_v e_s} + \frac{l_v}{M_w k'_T T} \left( \frac{l_v}{RT} - 1 \right)(1 + S_{K,i}) \right]^{-1}$$

(A1b)

where \(m\) is the particle mass, \(R\) is the gas constant, \(D'_v\) is the diffusion coefficient corrected with molecular kinetics, \(k'_T\) is the thermal coefficient corrected with molecular kinetics, \(e_s\) is the saturation water vapor pressure, \(l_v\) is the latent heat, and subscript “i” represents the \(i^{th}\) Lagrangian particle size bin. More details about this equation are in Lamb and Verlinde [2011]. The particle radius in each bin is calculated from the particle mass given by:

$$r_i = \left( \frac{3}{4 \pi \rho_{s,i}} \right)^{\frac{1}{3}}$$

(A2)

where \(\rho_s\) is the density of the solution. Liquid water mixing ratio \(q_l\), water vapor mixing ratio \(q_v\), air temperature and air pressure are described by the following equations:

$$\frac{dq_l}{dt} = \frac{1}{\rho_d} \sum_i n_i \frac{dm_i}{dt}$$

(A3)

$$\frac{dq_v}{dt} = -\frac{dq_l}{dt}$$

(A4)

$$\frac{dT}{dt} = -\frac{g_w}{c_p} + \frac{l_v}{c_p} \frac{dq_l}{dt}$$

(A5)

$$\frac{dP}{dt} = -\frac{P g_w}{R_a T}$$

(A6)

where \(\rho_d\) is dry air density, \(n_i\) is number concentration per volume air in the \(i^{th}\) particle bin, \(g\)
is gravity acceleration, \( c_p \) is air heat capacity and \( R_a \) is gas constant for air. Calculation of \( S_p \) is based on \( T \) and \( q_v \).

In the simulations presented in the main paper, the initial \( T \), \( P \) and \( RH \) are set to be 283.15K, 919hPa and 0.95, respectively. The initial aerosol size distribution is assumed to follow lognormal distribution given by

\[
\frac{dn_i}{dr} = \frac{N_a}{\sqrt{2\pi r_i \ln \sigma_g}} \exp \left( \frac{(\ln r_i - \ln r_g)^2}{2 \ln^2 \sigma_g} \right)
\]

where the geometric mean radius \( r_g \) is set to be 0.06\( \mu \)m; the geometric standard deviation \( \sigma_g \) is set to be 1.5.

**Appendix B: Maximum Parcel Supersaturation**

The parcel supersaturation can be described by

\[
\frac{dS_p}{dt} = a w - b \frac{d\rho_l}{dt}
\]

\[
a = \left( \frac{g l_v}{c_p R_w T^2} - \frac{g}{R_d T} \right)
\]

\[
b = \frac{R_w T}{e_s} + \frac{R_d l_v^2}{c_p R_w P T}
\]

where \( \rho_l \) is the liquid water density, and \( R_d \) is the gas constant for dry air. The first term on the right-hand-side describes the updraft cooling is the source term of supersaturation whereas the second term describes condensation-induced depletion of water vapor and is the sink term of supersaturation. As the parcel rises, the source term dominates first and thus \( S_p \) increases with height. At the height of maximum supersaturation, the source and sink terms of the \( S_p \) are balanced with each other. Beyond the maximum supersaturation, \( S_p \) starts to decrease with further increasing height.
Appendix C. Role of Particle Equilibrium Supersaturation in Condensational Broadening

The condensational narrowing in the aerosol-limited regime have been abundantly studied in the published literature and textbooks (e.g. [Pruppacher and Klett, 1997]); however the condensational broadening in the updraft-limited regime has not received much attention because both $S_k$ and its size dependence are usually assumed to be small and ignored. This assumption is incorrect for the updraft-limited regime. We can further understand this by analyzing the expressions for $S_k$ and its variation with radius:

$$S_k = \frac{a_s}{r} - \frac{\kappa r_d^3}{r^3 - r_d^3}$$  \hspace{1cm} (A9a)$$

$$\frac{dS_k}{dr} = -a_s \frac{1}{r^2} + 3\kappa r_d^3 \frac{1}{r^4}$$  \hspace{1cm} (A9b)$$

$$a_s = \frac{2\sigma_s}{R_v\rho_w T}$$  \hspace{1cm} (A9c)$$

where $r_d$ is the dry particle size and $\kappa$ is the hygroscopicity of aerosols. In the derivation of Equation A9b from Equation A9a, $r \gg r_d$ is assumed. Equation A9b reveals that $dS_k/dr$ is negative since the second term in Equation A9b is very small due to $r \gg r_d$. The dependence of radius growth rate on radius is given below [Chen et al., 2016; Korolev, 1995].

$$\frac{d}{dr} \left( \frac{dr}{dt} \right) = -\frac{G}{\rho_s \tau} \left( \frac{S_p - S_k}{r} + \frac{dS_k}{dr} \right)$$  \hspace{1cm} (A10)$$

In the updraft-limited regime, $S_p$ is small and comparable to $S_k$. The negative $dS_k/dr$ leads to a positive contribution to $(dr/dt)/dr$, which leads to broadening of particle size distribution.

Reference:


Feingold, G., S. M. Kreidenweis, B. Stevens, and W. R. Cotton (1996), Numerical simulations of stratocumulus processing of cloud condensation nuclei through collision-


Figure 1. Schematic illustration of the main characteristics of aerosol-cloud interaction regimes. Details about the item with superscript (1), (2) and (3) are discussed in Reutter et al. [2009], Chen et al. [2016] and this paper, respectively. Note that the transitional regime defined by the $\epsilon$ behavior at the level of maximum supersaturation is a single line, i.e. for a fixed set of aerosol properties other than $N_a$. The region of transitional regime in this figure is used to schematically illustrate the range of aerosol properties.
Figure 2. Vertical profiles of (a) parcel supersaturation ($S_p$), (b) cloud droplet number concentration ($N_c$), and (c) cloud droplet relative dispersion ($\epsilon$) at different ratios of aerosol number concentration ($N_a$) to vertical velocity ($w$). The thick black line denotes the transitional regime, that divides the aerosol-limited (dashed) and updraft-limited regime (solid) (See text for details). The numbers of cases in each $N_a/w$ group are 179, 500, 722, 588, 376, 136, respectively.
Figure 3. Same as Figure 2, except for (a) cloud droplet standard deviation ($\sigma$) and (b) cloud droplet mean radius ($R_{\text{mean}}$).
Figure 4. Vertical profiles of (a) $S_p$, (b) $N_c$, (c) $\epsilon$, (d) $\sigma$ and (e) $R_{\text{mean}}$ when $S_k$ are assumed to be the same for all the cloud droplets.
Figure 5. Joint dependence on $N_a$ and $w$ of (a, c and e) cloud droplet number concentration, and (b, d, and f) cloud droplet relative dispersion at (a-b) 20 m, (c-d) 560m and (e-f) 2110m, respectively.
Figure 6. Regime dependency of (a, c and e) cloud droplet standard deviation and (b, d, and f) cloud droplet mean radius on aerosol number concentration and vertical velocity at (a-b) 20 m, (c-d) 560m and (e-f) 2110m.
Figure 7. Vertical profiles of normalized $N_c$ by the maximum $N_c$ for each profile. The meanings of each color are the same as Figure 2.
Figure 8. Typical examples of the vertical profile of particle equilibrium supersaturation (a-b) and radius growth rate (c-d) in aerosol-limited regime (a and c) and updraft-limited regime (b and d). The black line denotes the smallest bin activated at the maximum supersaturation. The green circles denote the smallest bin activated at 1000m. The red dash line denotes the parcel supersaturation. A higher bin number corresponds to a larger dry aerosol radius. $w$ is 0.1 m s$^{-1}$ for both examples.
Figure 9. Joint dependency on $N_a$ and $w$ of (a) rain initiation height, (b) liquid water content (LWC), (c) cloud droplet number concentration, and (d) relative dispersion at the height of rain initiation.
Figure 10. Relationship between the normalized height of rain initiation and the normalized aerosol based on the 2500 simulations. The corresponding values at peak relative dispersion at maximum supersaturation are used for the normalization. The black line is the piecewise fitting given by Equation 3.