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Effect of the droplet activation process on microphysical properties of warm clouds

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Abstract

This study investigates the effect of the droplet activation process on microphysical characteristics of warm clouds represented by correlation statistics between cloud droplet effective radius r_e and cloud optical thickness τ_c . The conceptual adiabatic model is first employed to interpret satellite-observed r_e - τ_c correlation statistics over two different regions and to reveal distinctively different increasing patterns of droplet number concentration between these regions. This difference is attributed to a different behavior of the droplet activation process induced by differing microphysical conditions of aerosols. Numerical experiments of changing aerosol size spectrum are then performed with a spectral bin microphysics cloud model. The results show that the slope of the size spectrum controls the r_e - τ_c relationship through its effect on increasing pattern of droplet number concentration due to the activation process. The simulated results are also found to reproduce the r_e - τ_c correlation statistics closely resembling those observed when the slope parameter and the aerosol amount are appropriately chosen. These results suggest that the r_e - τ_c correlation statistics observed by remote sensing studies contains a signature of how the droplet activation process takes place in the real clouds.

Keywords: cloud microphysics, aerosol indirect effect, satellite remote sensing, numerical modeling

1. Introduction

Warm (liquid) clouds exert significant effects on the radiation budget and hydrological cycle in the Earth's climate. These effects of the clouds are characterized by their microphysical structures, which are controlled by particle formation and growth processes. The liquid cloud droplets are formed through activation process from aerosols serving as cloud condensation nuclei (CCNs) and then grow through condensation and coalescence processes that are dominant for non-drizzling and drizzling stages of cloud development, respectively (e.g., Rogers and Yau 1989). These microphysical processes determine the physical properties of the clouds represented by parameters such as cloud droplet effective radius r_e and cloud optical thickness τ_c , which have

been observed by air-borne and space-borne remote sensing techniques (Nakajima *et al* 1991, Han *et al* 1994, Nakajima and Nakajima 1995, Kawamoto *et al* 2001).

Characteristic relationships have been found between these two quantities by previous studies. Nakajima *et al* (1991) found two distinct relationships between them, i.e. positive and negative correlation patterns. The former and the latter were found for non-drizzling and drizzling clouds, respectively, suggesting distinctively different microphysical characteristics between these two stages of cloud development. These correlation statistics also have an important implication for distinct climatic effects of the clouds with different correlation patterns, which should have very different radiative properties (Slingo 1989).

These analyses were extended to wider regions of the First International Satellite Cloud Climatology Project (ISCCP) Regional Experiment (FIRE) and the Atlantic Stratocumulus Transition Experiment (ASTEX) by Nakajima and Nakajima (1995), who also reported these positive and negative correlation patterns that are found in a more complicated manner. Shown in figure 1 are typical examples of their results illustrating the correlation statistics between r_e and τ_c obtained from the FIRE (figure 1(a)) and ASTEX (figure 1(b)) regions. Figure 1 illustrates that the joint histogram is dominated by positive and negative correlation patterns for the ASTEX and FIRE regions, respectively. Fundamental mechanisms responsible for these correlation statistics were explored by our recent studies (Suzuki *et al* 2006, 2010) using a detailed microphysics cloud model. These studies showed that the positive and negative correlation patterns are formed by condensation and coalescence processes, respectively, and also showed how the correlation statistics systematically change with differing conditions of aerosol amount and static stability through their effects on these microphysical processes.

Although these results well explain basic characters of the correlation statistics obtained by previous remote sensing studies, observed joint histograms shown in figure 1 contain more complicated relationships between r_e and τ_c . The example for the FIRE statistics (figure 1(a)) includes a tiny fragment of positive correlation below $r_e \sim 12 \mu\text{m}$ underneath the large negative correlation branch, illustrating a ‘high-heeled’ shape as a whole. The ASTEX plot (figure 1(b)), in contrast, appears to include only the positive correlation branch without the negative correlation part. This contrast between these two cases is associated with different behaviors of the droplet activation process as demonstrated in this study, where we investigate how the aerosol microphysical conditions influence the r_e - τ_c correlation statistics through the droplet activation process and thereby complement our previous studies that examined condensation and coalescence processes.

In this study, the conceptual adiabatic model is at first employed to interpret the satellite-observed correlation statistics and to underscore the significance of the droplet activation process in understanding the distinct difference between the FIRE and ASTEX statistics. We then perform numerical experiments of changing the aerosol size spectrum with a spectral bin microphysics cloud model and demonstrate how the different behaviors of activation process induced by differing aerosol microphysical assumptions lead to the difference in shape of the correlation statistics.

2. Implication from adiabatic model

Figure 1 includes the contour curves associated with specified values of the number concentration N_c and the liquid water path W given by the adiabatic model. This conceptual model introduced by previous studies (e.g., Brenguier *et al* 2000) conveniently describes how the cloud droplets tend to grow vertically through adiabatic condensational growth processes.

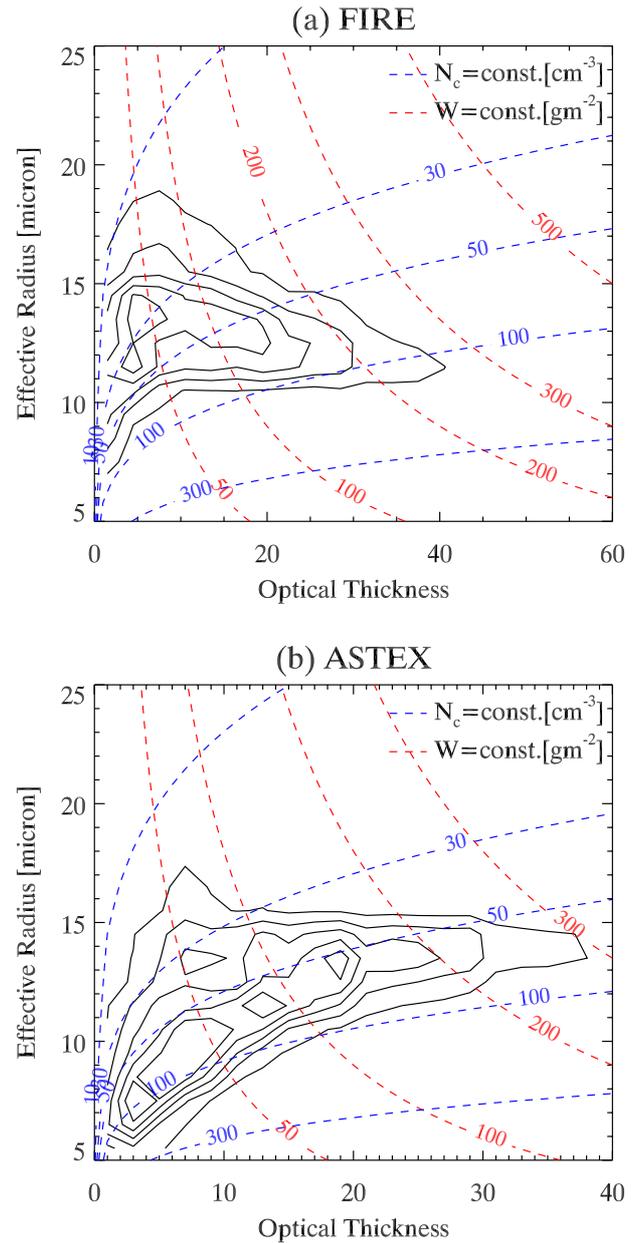


Figure 1. Joint probability distribution functions between cloud droplet effective radius r_e and cloud optical thickness τ_c over the (a) FIRE and (b) ASTEX regions obtained by Nakajima and Nakajima (1995) with theoretical relationships given by the adiabatic model superimposed. The contours are for 10, 30, 50, 70 and $90\% \mu\text{m}^{-1}$. The theoretical relationships are shown for specified values of number concentration $N_c = 10, 30, 50, 100$ and 300 cm^{-3} as given by (1) (blue) and liquid water path $W = 50, 100, 200$ and 300 g m^{-2} as given by (2) (red).

According to this model, the cloud water content q tends to grow linearly with height h from the cloud base as

$$q = \lambda h,$$

where λ represents the adiabaticity of the atmosphere and is assumed to be $5.2 \times 10^{-3} \text{ g m}^{-4}$ in this study. Under the assumption of constant number concentration N_c for the condensation process, the effective particle radius r_e can be

represented as

$$r_e(h) = k^{-1/3} \left(\frac{3}{4\pi\rho_w} \frac{q(h)}{N_c} \right)^{1/3},$$

where ρ_w denotes the liquid water density and k represents the factor relating the effective radius to volume-mean radius and is assumed as $k^{-1/3} = 1.1$. These profiles of q and r_e determine the optical thickness τ_c , the liquid water path W and the effective radius $r_e(H)$ at the cloud top when the geometrical thickness H of the cloud is given. The relationships between τ_c and $r_e(H)$ for given values of N_c and W are obtained as (see Suzuki *et al* (2010) for derivations)

$$\tau_c = \frac{8}{5} (\pi k)^2 \frac{\rho_w}{\lambda} N_c^2 r_e(H)^5 \quad (1)$$

and

$$\tau_c = \frac{9}{5} \frac{1}{\rho_w} \frac{W}{r_e(H)}, \quad (2)$$

respectively.

Equation (1) represents the condensational growth curves for specified values of N_c and is shown by blue contours in figure 1. The liquid water path W , representing the vertical extent of the cloud as (2), is shown by red contours in figure 1. These theoretical curves offer a hint at how N_c and W tend to change associated with particle growth. Figure 1(a) shows that the lowest edge of the joint histogram crosses the contour curves for specified N_c values, illustrating a rapid increase in N_c with increasing r_e for the incipient stage of cloud formation over the FIRE region, whereas the joint histogram in figure 1(b) shows a more gradual change in N_c as r_e increases over the ASTEX region. This difference in changing pattern of N_c characterizes the contrast in shape of the correlation statistics between these two regions.

The values of N_c implied here represent equivalent values of the droplet number concentration determined from given effective radius and optical thickness when the adiabatic condensational growth process is assumed. The change in N_c therefore means the change in this equivalent quantity due to the processes other than the adiabatic condensational growth. A plausible mechanism responsible for the increase in number concentration is provided by the droplet activation process that forms liquid cloud particles from aerosols serving as CCNs. The activation process operates to add the cloud droplets to an existing cloud as determined by the shape of the aerosol size spectrum and a typical increase in supersaturation just above the cloud base. Different increasing patterns of N_c found between figures 1(a) and (b) suggest that the activation process takes place in a different manner so the liquid particles increase in number at different rates between these two cases. The droplet activation process occurs in a manner that depends on aerosol size spectrum and updraft velocity, the former and the latter being associated with aerosol microphysics and static stability, respectively. Although the updraft velocity is of importance in determining the supersaturation value that affects how many aerosol particles are activated, the aerosol size spectrum is examined here as a factor controlling the way the activation process occurs. Since the droplet activation

starts to occur from the largest-sized aerosol particles and proceeds toward smaller sizes, the shape of the aerosol size spectrum characterizes how the number concentration N_c increases through the activation process. We then investigate this effect on r_e and τ_c with a bin-type detailed microphysics cloud model. Numerical experiments of changing the aerosol size distribution assumptions are performed as described in the next section.

3. Bin model simulation

The model used for the experiments is a non-hydrostatic spectral bin microphysics cloud model developed by Suzuki *et al* (2006, 2010) based on the original model of Khain and Sednev (1996) and Khain *et al* (1999). This model predicts an explicit change in size distribution function of cloud particles taking into account the microphysical processes of warm clouds, i.e. activation from aerosols, condensational growth and coalescence processes as described in Suzuki *et al* (2010).

The droplet activation process, a main focus of this study, is computed from the size distribution function of hygroscopic aerosols $f_{CCN}(r)$, which is also predicted in the model. The critical size of aerosol particle is determined from the supersaturation S according to the Köhler theory and the part of the aerosol spectra greater than the critical size is activated to grow into cloud particles. The cloud particles thus formed are subject to further growth due to condensation and coalescence processes (Suzuki *et al* 2010).

The droplet activation process therefore takes place in a manner that depends on the aerosol size spectrum, which is initially assumed to have a power law form as

$$f_{CCN}(z, r) = f_0(z) \left(\frac{r}{r_0} \right)^{-p}, \quad (3)$$

where $r_0 = 0.1 \mu\text{m}$ and $f_0(z)$ is given by an exponential decay profile as

$$f_0(z) = f_{\text{sfc}} \exp\left(-\frac{z}{H}\right),$$

where f_{sfc} and H denote the surface value of f_0 and the scale height set as 1 km here, respectively. The parameter p represents the slope of the size spectrum and determines how the number concentration of cloud particles potentially increases in the course of cloud formation: the larger the value of p is, the more rapidly the number concentration tends to increase. Note that the case of $p = 4$ is referred to as the Junge distribution. To examine this effect on r_e - τ_c correlation statistics, we perform sensitivity experiments of changing this parameter p in the simulation of warm cloud formation. We conduct idealized two-dimensional experiments to form a single low-level warm cloud by the experimental design described in Suzuki *et al* (2010). This simulation generates low-level clouds with updraft velocities of around $1\text{--}2 \text{ m s}^{-1}$. The computational domain is 30 km in the horizontal and 5 km in the vertical direction and the resolutions are 250 m in the horizontal and 50 m in the vertical direction. The unstable and stable layers are assumed for

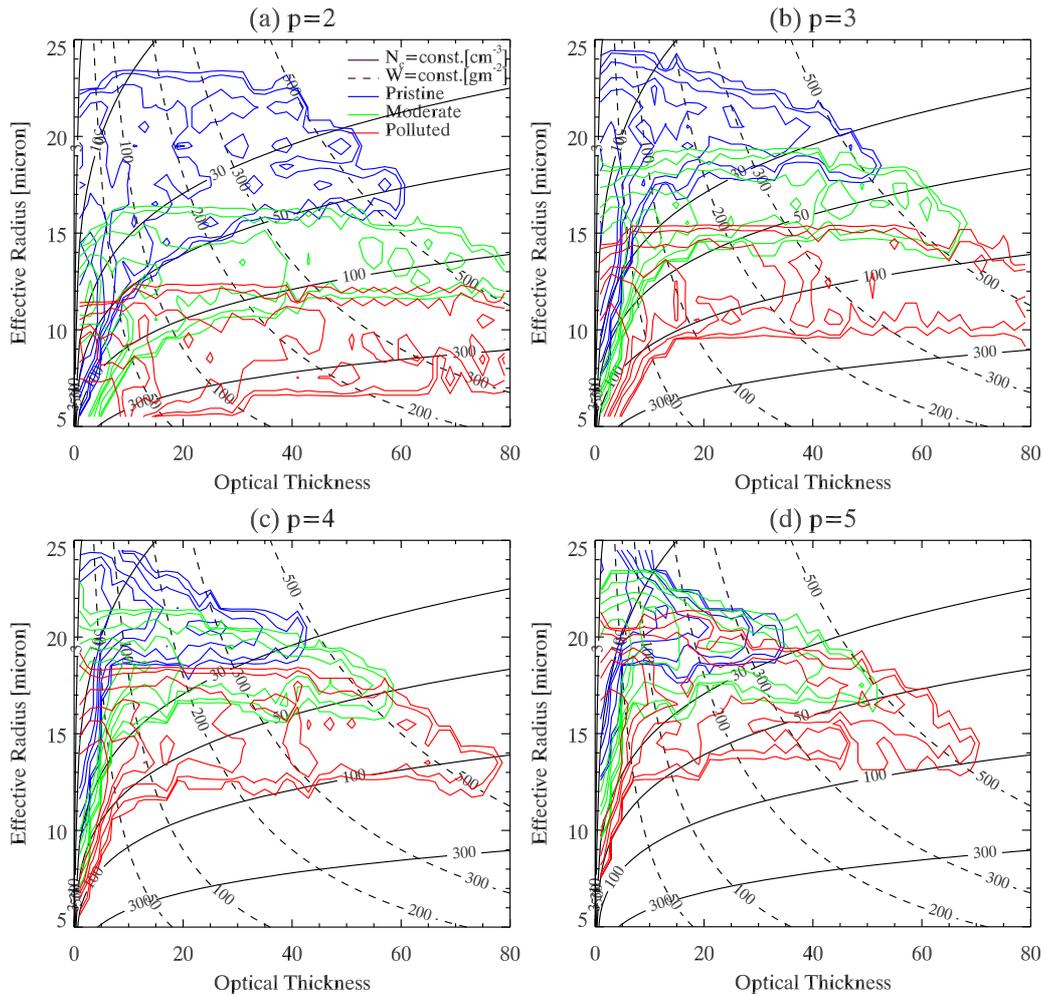


Figure 2. Joint probability distribution functions between r_e and τ_c simulated by the model for pristine (blue), moderate (green) and polluted (red) conditions under the assumptions of (a) $p = 2$, (b) $p = 3$, (c) $p = 4$ and (d) $p = 5$. The contours are for 1, 3, 10, 30, 50 and 90% μm^{-1} . Theoretical relationships given by the adiabatic model are also shown.

the lowest 1 km and the upper 4 km layers, respectively, in a manner similar to Suzuki *et al* (2010). The aerosol amount represented by f_{sfc} , as well as p , is also changed to examine the combined effect of amplitude and slope of size spectrum on cloud microphysics. The parameter f_{sfc} is changed so that the surface aerosol number concentration N_a ranges approximately from 10^7 to 10^{10} m^{-3} , and the results are classified according to N_a values as ‘pristine’ for $N_a = 10^7$ – 10^8 m^{-3} , ‘moderate’ for $N_a = 10^8$ – 10^9 m^{-3} and ‘polluted’ for $N_a = 10^9$ – 10^{10} m^{-3} .

Figure 2 shows the simulated results in the form of r_e – τ_c joint histograms for pristine (blue), moderate (green) and polluted (red) aerosol amount conditions under the assumption of $p = 2$ (figure 2(a)), $p = 3$ (figure 2(b)), $p = 4$ (figure 2(c)) and $p = 5$ (figure 2(d)). These figures also include theoretical relationships given by the adiabatic model in a manner similar to figure 1. Figure 2 indicates that r_e and τ_c tend to become smaller and larger, respectively, as the aerosol amount increases from pristine to polluted conditions as found by Suzuki *et al* (2010) for all cases of p values. These tendencies are found to occur in a similar way to changing

number concentration N_c : the plot for the more polluted case coincides with the contours for larger N_c values.

Besides this common tendency with changing aerosol amount, the correlation statistics are also found to vary in shape systematically with changing p values. The result for $p = 2$ (figure 2(a)) indicates that the correlation plot for each aerosol amount tends to follow the contour curve for specified N_c values more closely than do those for larger values of p . This difference is clearly found when the lowest edges of the plots are compared: the shape of the lowest edge for smaller p values is found to be closer to N_c contours. This means that the number concentration tends to stay more constant or increase more gradually when p values are smaller whereas the number concentration tends to increase more rapidly when p values are larger, as expected from the activation mechanisms represented in the model described above.

These results suggest that the change in aerosol size spectrum modifies the r_e – τ_c correlation statistics in a systematic way. The change in amplitude of the spectrum (or the aerosol amount), represented by f_{sfc} in the model, causes overall changes of r_e and τ_c in a manner corresponding to change in cloud droplet number concentration. The change in

slope of the spectrum represented by p in the present model, on the contrary, has an effect of modifying the pattern of increase in number concentration, resulting in the change in shape of the r_e - τ_c correlation plot.

Correlation statistics similar to those observed by Nakajima and Nakajima (1995) can be reproduced from the simulation results by choosing appropriate values of the aerosol amount and the slope parameter. Figure 3 shows the correlation plots obtained from the results in figure 2 when composited in a different way: figure 3(a) shows the results for moderate to polluted aerosol amount under the assumption of $p = 4$ and figure 3(b) shows those for moderate conditions with $p = 2$. Figure 3(a) illustrates that the lowest edge of the joint histogram tends to cross the contour curves for N_c values and the correlation plot depicts a ‘high-heeled’ shape similar to the feature in figure 1(a) observed over the FIRE region. The joint histogram in figure 3(b), in contrast, indicates a lowest edge that follows the contour curve for specific N_c values (100 cm^{-3} in this case) and closely resembles the ASTEX plots shown in figure 1(b). The similarity of figures 1 and 3 suggests that the typical difference in satellite-observed correlation statistics between the FIRE and ASTEX regions can be explained by difference in slope of the aerosol size spectrum as well as in aerosol amount.

4. Conclusion and discussion

This study investigates the effect of the droplet activation process on correlation statistics between r_e and τ_c . The adiabatic model applied to satellite-observed correlation plots shows that the number concentration tends to increase more rapidly with increasing r_e over the FIRE region whereas the increase is more gradual over the ASTEX region. This difference suggests that the activation process takes place in a different manner between these two cases. Numerical simulations of changing aerosol size spectrum with a spectral bin microphysics model are then performed to show that the shape of the correlation plot is significantly modified by changing the slope parameter of aerosol size spectrum. The amplitude of the size spectrum, representing the aerosol amount, is found to change the overall location of the correlation plot in a manner corresponding to the change in cloud droplet number concentration as found by our previous study. These results reveal how the r_e - τ_c statistics tend to vary in value and shape with differing conditions of aerosol size spectrum, and provide an understanding of the effect of aerosol microphysical conditions on cloud microphysical properties through the activation process. The correlation statistics is indeed shown to become closely similar to those observed over the FIRE and ASTEX regions when the slope parameter and the aerosol amount are appropriately chosen. These results suggest that the signature of the activation process is contained in the r_e - τ_c correlation statistics observed by remote sensing studies.

The effect of the aerosol size spectrum on the droplet activation process has also been represented by the CCN activity spectrum that describes the number concentration N_{CCN} of CCNs activated at supersaturation S as compiled from

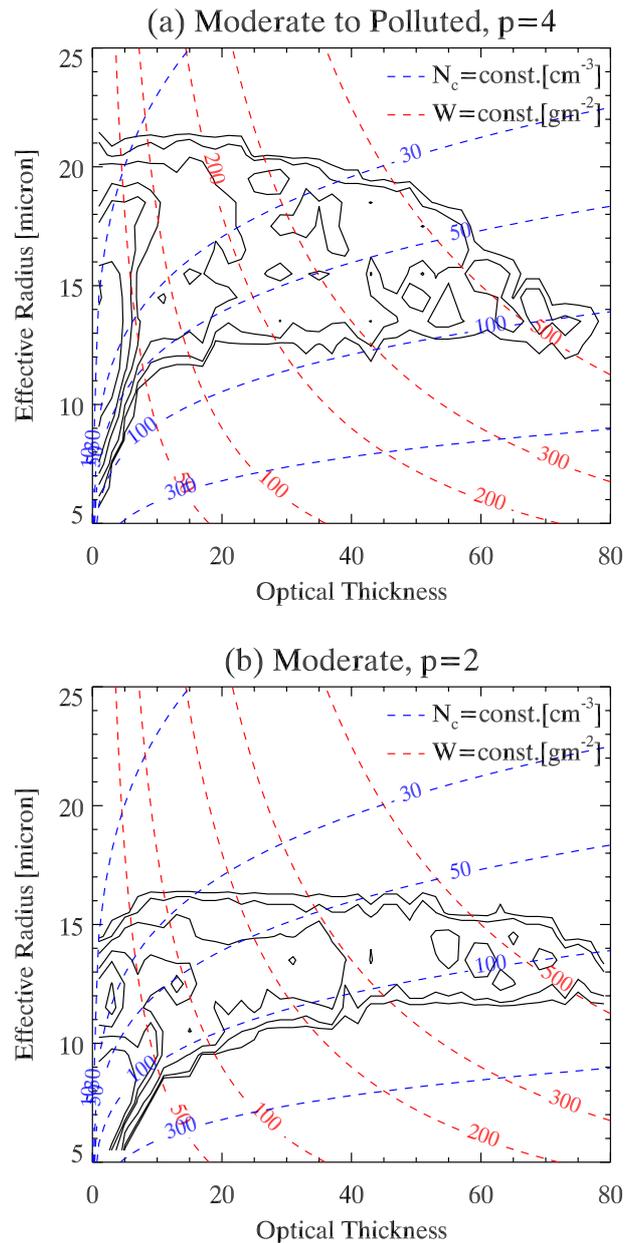


Figure 3. Joint probability distribution functions between r_e and τ_c simulated by the model for (a) moderate to polluted conditions with $p = 4$ and (b) moderate conditions with $p = 2$. The contours are for 1, 3, 10, 30, 50 and 90% μm^{-1} . Theoretical relationships given by the adiabatic model are also shown.

measurements of a variety of maritime and continental sites (see, e.g., Andreae and Rosenfeld 2008). This relationship is typically characterized by the form of $N_{\text{CCN}} = cS^k$, where values of parameters c and k have been reported by many previous studies since Twomey (1959) to vary with air-mass conditions reflecting the size distribution and the chemical composition of aerosols. For given chemical species, these parameters are determined by the size distribution function, where the parameter k directly relates to the size exponent p as $k = 2(p - 1)/3$. The range of p from 2 to 5 assumed in this study corresponds approximately to that of k from 0.7 to 2.7. This suggests that the clouds formed in air-mass characterized

by different CCN activity spectra may have different r_e - τ_c relationships.

The findings of this study, when combined with our previous study also demonstrating the stability effect on the correlation statistics, provide a theoretical basis for classifying the r_e - τ_c statistics in terms of aerosol microphysics and atmospheric stability conditions. As shown by Suzuki *et al* (2010), the condensation and the coalescence processes are responsible for the positive and negative correlation patterns between r_e and τ_c , respectively, in a manner that depends on environmental factors such as aerosol amount and static stability conditions. These basic tendencies of r_e and τ_c now turn out to be modified by the shape of the aerosol size spectrum through the droplet activation process: differing conditions of the slope parameter p in the present model cause the difference in increasing pattern of droplet number concentration, changing the r_e - τ_c correlation statistics in shape. The correlation statistics can therefore be understood as being formed from the aerosol size spectrum and static stability conditions through the combined effect of activation, condensation and coalescence processes.

Satellite remote sensing products of aerosols and clouds may offer an opportunity to investigate this hypothesis on the global scale since the satellite observables for aerosols, i.e. the optical thickness and the Ångström exponent, characterize the aerosol microphysical state. The optical thickness represents the aerosol amount and the Ångström exponent uniquely relates to the slope parameter p of the aerosol size spectrum (see, e.g., Nakajima and Higurashi 1997). These aerosol observables characterize the size distribution of aerosol particles acting as CCNs, which may also be inferred from the r_e - τ_c correlation statistics according to the findings of this study. These satellite-based analyses could be performed in future studies in an attempt to classify cloud properties on the global scale and help improve our understandings of climatic aerosol effect on clouds and its representations in climate models.

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