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An analytical expression for predicting the critical radius in the autoconversion parameterization

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[1] An analytical expression for the critical radius associated with Kessler-type parameterizations of the autoconversion process is derived. The expression can be used to predict the critical radius from the cloud liquid water content and the droplet number concentration, eliminating the need to prescribe the critical radius as an empirical constant in numerical models. Data from stratiform clouds are analyzed, indicating that on average continental clouds have larger critical radii than maritime clouds. This work further suggests that anthropogenic aerosols affect the autoconversion process by increasing the critical radius and decreasing the characteristic radius, which in turn inhibits the initiation of embryonic raindrops, and by decreasing the autoconversion rate after the initiation process. The potential impact of this work on the evaluation of the second indirect aerosol effect is discussed. *INDEX TERMS:* 1600 Global Change; 1640 Global Change: Remote sensing; 1655 Global Change: Water cycles (1836); 1704 History of Geophysics: Atmospheric sciences; 1854 Hydrology: Precipitation (3354). **Citation:** Liu, Y., P. H. Daum, and R. McGraw (2004), An analytical expression for predicting the critical radius in the autoconversion parameterization, *Geophys. Res. Lett.*, *31*, L06121, doi:10.1029/2003GL019117.

1. Introduction

[2] Accurate representation of cloud and precipitation processes in models of various scales such as global climate models (GCMs) is crucial for understanding the interactions between cloud microphysics and cloud dynamics [e.g., *Chen and Cotton*, 1987], for forecasting freezing drizzle and aircraft icing [*Rasmussen et al.*, 2002], and for improving GCMs [*Stokes and Schwartz*, 1994]. A key process that must be parameterized is the autoconversion process whereby large cloud droplets collect small ones and become embryonic raindrops [*Kessler*, 1969; *Manton and Cotton*, 1977; *Tripoli and Cotton*, 1980; *Liou and Ou*, 1989; *Baker*, 1993; *Boucher et al.*, 1995; *Liu and Daum*, 2004]. Accurate parameterization of this process is especially important for studies of the second indirect aerosol effect [*Boucher et al.*, 1995; *Lohmann and Fleichter*, 1997; *Rotstayn*, 2000].

[3] *Kessler* [1969] proposed a parameterization that linearly relates the autoconversion rate to the cloud liquid water content (L), and assumes a critical value for L below which no autoconversion occurs. Later Kessler-type parameterizations explicitly account for the droplet concentration (N) as well as L [*Manton and Cotton*, 1977; *Tripoli and Cotton*, 1980; *Liou and Ou*, 1989; *Baker*, 1993; *Liu and Daum*, 2004]. The inclusion of N allows for studying the second indirect aerosol effect. It has also been recognized that the threshold process should be determined by a critical radius (r_c) rather than by a critical L as conceived by Kessler. Considering autoconversion as a threshold process is a distinctive feature that sets Kessler-type parameterizations apart from other types of autoconversion parameterizations [e.g., *Berry*, 1968; *Beheng*, 1994; *Khairoutdinov and Kogan*, 2000].

[4] All the later Kessler-type parameterizations can be generically written as

$$P = fH(r_m - r_c), \tag{1}$$

where P is the autoconversion rate, f a generic function, and r_m the characteristic radius. The Heaviside function $H(r_m - r_c)$ is introduced to describe the threshold process such that there is no autoconversion when $r_m < r_c$. The characteristic radius is the volume-mean radius given by *Manton and Cotton* [1977], *Tripoli and Cotton* [1980], *Liou and Ou* [1989], and *Baker* [1993], the mean radius of the 4th moment given by *Boucher et al.* [1995], and the mean radius of the 6th moment in the parameterization that we have recently derived [*Liu and Daum*, 2004]. The function f is also different for different parameterizations.

[5] Although model results are sensitive to r_c [*Boucher et al.*, 1995; *Rotstayn*, 1999], the idea of threshold process embedded in Kessler-type parameterizations has been loosely used, and r_c has been largely prescribed as an empirical parameter that is arbitrarily tuned to match model simulations with observations. Here we derive an analytical expression for r_c by coupling the Liu-Daum parameterization with a new theory on rain formation that we have recently formulated [*McGraw and Liu*, 2003]. The expression can be used to predict r_c from L and N. Data from continental and marine stratiform clouds are examined, and implications for the evaluation of the autoconversion rate and the second indirect aerosol effect are discussed.

2. Kinetic Potential Theory, Threshold Process and Critical Radius

[6] Although it has been well established that three physical processes (condensation, evaporation and collection) are involved in the formation of warm rain, many issues regarding the initiation of warm rain remain unsolved [*Beard and Ochs*, 1993; *Telford*, 1996]. Recently, we developed a new theory on rain formation by extending the theory of statistical crossing of a kinetic potential barrier in nucleation to the processes of condensation, evaporation

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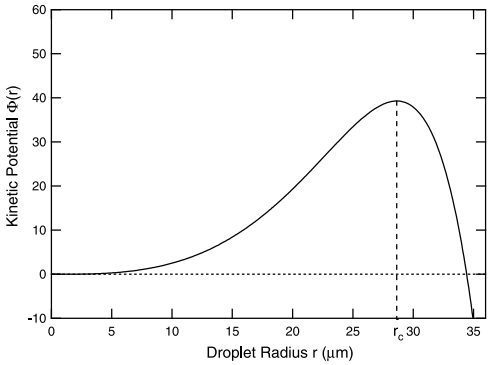

Figure 1. An example of the kinetic potential as a function of the droplet radius. The results are for L = 0.5 g m^{-3}, N = 300 cm^{-3}, and $\beta_{con} = 10^{24}$ s^{-1}.

and collection occurring in warm clouds [*McGraw and Liu*, 2003].

[7] Briefly, by analogy to the kinetic theory on nucleation, the kinetic potential Φ(j) for a droplet consisting of j water molecules is given by

$$\Phi(j) \equiv -\ln\left[\prod_{g=1}^{j-1} \frac{\beta_{con}(g) + \beta_{col}(g)}{\gamma_{eva}(g+1)}\right], \tag{2a}$$

$$\beta_{col}(g) = \frac{\kappa v}{\rho_w^2} g^2 L, \tag{2b}$$

$$\gamma_{eva}(g) = \exp\left(\frac{vN}{L}\right)\beta_{con}(g), \tag{2c}$$

where β_{con} (s^{-1}), β_{col} (s^{-1}), and γ_{eva} (s^{-1}) denote the condensation, collection, effective evaporation rate constants, respectively, for a droplet consisting of g water molecules; $\nu = 3.0 \times 10^{-23}$ (g) is the mass per water molecule; $\kappa = 1.1 \times 10^{10}$ cm^{-3} s^{-1} is a constant in the Long collection kernel [*Long*, 1974]; ρ_w is the water density (g cm^{-3}). The kinetic potential as a function of droplet radius (r) can be calculated by applying the relation $g = \frac{4\pi\rho_w}{3v}r^3$ to equations (2a)–(2c). Figure 1 shows a typical example. The kinetic potential first increases with increasing droplet radius because ($\beta_{con} + \beta_{col}$) < γ_{eva}, and then decreases after reaching a peak because ($\beta_{con} + \beta_{col}$) > γ_{eva} [Note that $\frac{d\Phi}{dg} = -\ln\left(\frac{\beta_{con}+\beta_{col}}{\gamma_{eva}}\right)$].

[8] The point where the kinetic potential reaches its maximum is worth emphasizing because it physically defines a critical point. As in nucleation theory, the maximum kinetic potential is referred to as the "barrier"; the corresponding droplet radius defines r_c. Before reaching the critical point, the droplet system is in a stable state because more potential is needed to climb the "hill". Once the barrier is passed, the system becomes unstable down the "hill", and embryonic raindrops spontaneously form. Therefore, the idea of a threshold process and r_c inherent in Kessler-type parameterizations emerges naturally from the kinetic potential theory.

3. Analytical Expression for Critical Radius

[9] It is desirable to relate r_c to L and N because these two variables are predicted/diagnosed in state-of-the art GCMs [*Ghan et al.*, 1997a, 1997b; *Rotstayn*, 1997]. At the critical point, the forward and reverse rate constants are equal [*McGraw and Liu*, 2003], i.e.,

$$\beta_{con} + \beta_{col} = \gamma_{eva} \tag{3}$$

Substituting equations (2b) and (2c) into equation (3) and using the relation $g = \frac{4\pi\rho_w}{3v}r^3$, we obtain

$$r_c = \left\{\left(\frac{3}{4\pi}\right)^2 \frac{v}{\kappa}\beta_{con}\left[\exp\left(\frac{vN}{L}\right) - 1\right]\frac{1}{L}\right\}^{1/6}. \tag{4a}$$

Because (νN/L) $\ll$ 0, and $\exp\left(\frac{vN}{L}\right) \approx 1 + \frac{vN}{L}$, equation (4a) can be simplified to

$$r_c = \left\{\left(\frac{3}{4\pi}\right)^2 \frac{v^2}{\kappa}\beta_{con}\frac{N}{L^2}\right\}^{1/6} \approx 4.09 \times 10^{-4}\beta_{con}^{1/6}\frac{N^{1/6}}{L^{1/3}}, \tag{4b}$$

where r_c is in μm, L in g m^{-3}, N in cm^{-3}, and β_{con} in s^{-1}.

[10] In general, β_{con} is a function of turbulence that is unknown at present [*McGraw and Liu*, 2003]. Nevertheless, it can be estimated from microphysical measurements in drizzling clouds as follows. According to *Liu and Daum* [2004], autoconversion starts when the mean radius of the 6th moment reaches r_c. The mean radius of the 6th moment (r_6) is given by

$$r_6 = \left(\frac{3}{4\pi\rho_w}\right)^{1/3}\alpha\left(\frac{L}{N}\right)^{1/3}, \tag{5}$$

where α = 1.12. Equating r_6 with r_c, we obtain

$$\beta_{con} = \frac{k\alpha^6 L_*^4}{(v\rho_w)^2 N_*^3} \approx 2.37 \times 10^{31}\frac{L_*^4}{N_*^3}, \tag{6}$$

where the subscript "*" denotes the corresponding variables at threshold where the two radii are equal; L* is in g m^{-3}, N in cm^{-3}, and β_{con} in s^{-1}.

[11] Equation (6) is used to estimate β_{con} from concurrent measurements of L and N in drizzling clouds reported by *Yum and Hudson* [2001, 2002]. As shown in Figure 2, the majority of β_{con} values fall between 10^{22} and 10^{23} (s^{-1}); the

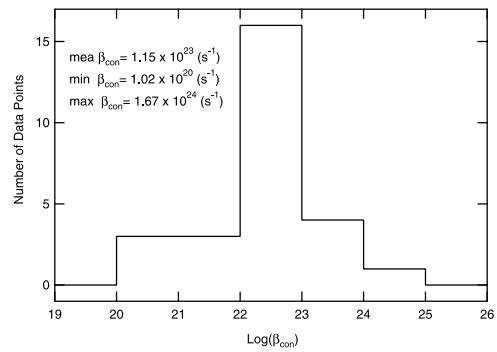

Figure 2. Histogram of the condensation rate constants estimated from microphysical measurements in drizzling clouds.

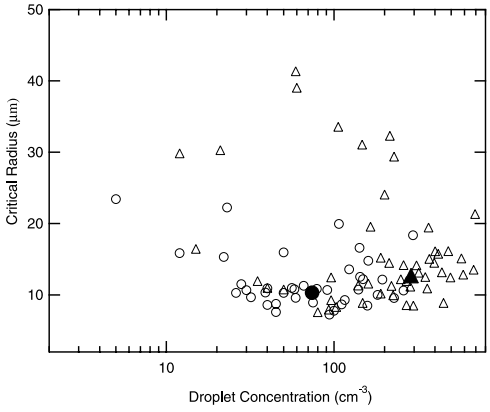

Figure 3. Relationship of the critical radius to the droplet concentration. The open triangles and dots denote continental and marine clouds, respectively. The solid triangle and dot denote the average of continental and marine clouds, respectively. The dot and dash lines represent two calculations from equation (7) at L = 0.001 and 2 gm^{-3}, respectively.

mean, minimum and maximum of β_{con} are 1.15×10^{23}, 1.02×10^{23} and 1.67×10^{24} (s^{-1}), respectively.

4. Critical Radius of Ambient Clouds and Important Implications

[12] Equation (4b) indicates that r_c is a function of L and N, varying from cloud to cloud, even from place/time to place/time in the same cloud. To demonstrate this, Figure 3 shows r_c calculated from equation (4b) using the mean β_{con} and the data on L and N from stratiform clouds given by *Miles et al.* [2000]. It is clear that r_c varies significantly, from *circa* 6 μm to 40 μm. Note that since each point in Figure 3 actually represents an average of many samples, variation in r_c is expected to be even larger for individual clouds. This suggests that prescribing r_c as a constant is more troublesome in small-scale models than in GCMs.

[13] It is interesting to compare r_6 with r_c because r_6, or its "equivalent", is often used in studies of both the indirect aerosol effects [*Twomey*, 1991; *Albrecht*, 1989]. The equation relating r_c to r_6 can be derived by using equation (5) to eliminate N in equation (4b),

$$r_c = \left[\left(\frac{3}{4\pi}\right)^3 \frac{\nu^2 \beta_{con}}{\kappa \rho_w L}\right]^{1/6} \frac{\alpha^{1/2}}{r_6^{1/2}} \approx \frac{23.72}{L^{1/6} r_6^{1/2}}, \qquad (7)$$

where L and the radii are in g m^{-3} and μm, respectively. Equation (7) indicates that r_c decreases with increasing r_6 when L remains constant. This is illustrated by the dot and dash lines in Figure 4, which correspond to the lower and upper limits of L = 0.001 g m^{-3} and L = 2 g m^{-3} in the Miles et al. data, respectively. It is well known that an increase in aerosol loading leads to an increase in N and a decrease in the mean radius. Accordingly, anthropogenic aerosols tend to move clouds up the line, diminishing r_6 but increasing r_c. The data presented in Figure 4 support this notion. Because of higher droplet concentrations, continental clouds have an average r_c (12.71 μm) larger than that of marine clouds (10.32 μm). These values are close to those prescribed in cloud-scale models [*Tripoli and Cotton*, 1980], but larger than those assigned in GCMs [*Boucher and Lohmann*, 1995; *Rasch and Kristjansson*, 1998; *Rotstayn*, 1999]. A smaller r_c for GCMs is often attributed to a coarser model resolution and subgrid variabilities of cloud properties [*Rotstayn*, 2000; *Pincus and Klein*, 2000; *Zhang et al.*, 2002]. The nonlinear dependence of r_c on L and N revealed by equation (4b) supports this argument.

[14] Figure 4 also shows that on average, continental clouds are more likely to have $r_6 < r_c$ than marine clouds, suggesting that rain onset is inhibited in continental clouds by increased anthropogenic aerosols. This result is consistent with the assumption of the second indirect aerosol effect [*Albrecht*, 1989]. However, some continental clouds have $r_6 > r_c$ whereas some marine clouds have $r_6 < r_c$, suggesting the complexity of the second indirect aerosol effect. It is known that anthropogenic aerosols affect precipitation by decreasing the characteristic radius (e.g., r_6) and by decreasing the conversion rate from cloud water to rain water after the onset of the autoconversion process. This study suggests that anthropogenic aerosols also inhibit the initiation of embryonic raindrops by increasing r_c.

5. Concluding Remarks

[15] An analytical expression for r_c is derived by coupling the kinetic potential theory on rain formation with the Liu-Daum autoconversion parameterization. The expression can be used to predict r_c from L and N, eliminating the need to prescribe r_c in models. Examination of data from stratiform clouds indicates that r_c varies from cloud to cloud, and that on average, continental clouds have larger r_c than maritime clouds. It is shown that anthropogenic aerosols have the effect of increasing r_c and decreasing the mean radius concurrently, inhibiting the onset of embryonic raindrops. Use of this new expression will improve the representation of precipitation in numerical models and our ability to model the second indirect aerosol effect.

[16] Three points are worth noting. First, there is uncertainty in our estimate for β_{con} (hence r_c) due to limited

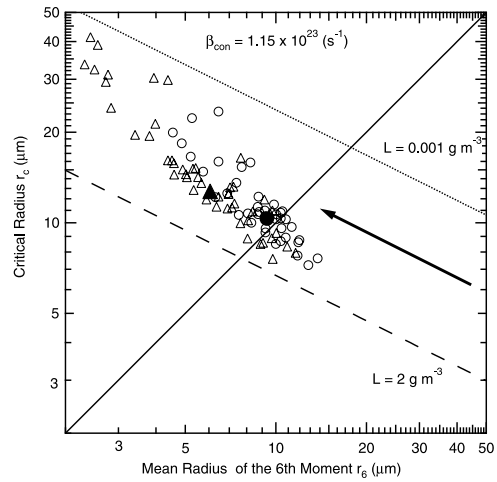

Figure 4. Dependence of the critical radius on the mean radius of the 6th moment. The meanings of symbols are the same as in Figure 3. The arrow denotes the direction of increasing aerosol loading.

measurements of L and N used in the calculation, and to the uncertainty as to if the measurements were taken at threshold points as required by equation (6). Furthermore, β_{con} is expected to depend on cloud turbulence and droplet radius. It would be desirable to derive it from the first principles. Second, relative dispersion of the cloud droplet size distribution is not considered an independent variable in the current formulation. However, this quantity is also crucial for evaluating cloud radiative properties and indirect aerosol effects [*Liu and Daum*, 2000, 2002; *Peng and Lohmann*, 2003; *Rotstayn and Liu*, 2003]. Further progress requires extending the formulation to explicitly account for the relative dispersion. Finally, the expression for r_c is essentially local. The effect of subgrid variabilities of cloud properties on r_c needs to be addressed in GCMs. As a first order approximation, this effect could be simply accounted for by adjusting β_{con} because r_c is proportional to $\beta_{con}^{1/6}$ (equation (4b)).

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