Expression for Critical Radius and Generalization to Consider Spectral Dispersion

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Introduction

Representation of cloud and precipitation processes is crucial for improving atmospheric models of various scales ranging from large eddy simulations (LES) to cloud resolving models (CRMs) to global climate models (GCMs) (Stokes and Schwartz 1994). A key process that must be parameterized is the so-called 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).

Kessler (1969) proposed a simple 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. One major improvement in later Kessler-type parameterizations is explicitly accounting 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 in the autoconversion parameterization allows for modeling studies of 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).

Without loss of generality, all the improved Kessler-type parameterizations can be generically written as

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

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

Furthermore, although model results are very sensitive to the value of r_c (Boucher et al. 1995, Rotstayn 1999), the idea of threshold process embedded in Kessler-type parameterizations have been used rather loosely, and r_c has been largely considered an empirical parameter that is arbitrarily tuned to match model simulations with observations. Liu et al. (2004) have derived an analytical expression for r_c by coupling a new theory on the rain formation that we have recently formulated (McGraw and Liu 2003) with the Liu-Daum parameterization. Here we first briefly discussed the Liu-Daum parameterization and the derivation of the expression for r_c, and then introduce a new Kessler-type parameterization by coupling Liu-Daum parameterization with the expression for r_c. Implications of the new parameterizations for the evaluation of the autoconversion rate and the second indirect aerosol effect are discussed.

Liu-Daum Parameterization

As detailed in Liu and Daum (2004), the Liu-Daum parameterization is given by

$$P = \eta N^{-1} L^3 H\left(r_6 - r_c\right) \tag{2a}$$

$$\eta = \left(\frac{3}{4\pi\rho_w}\right)^2 \kappa\beta_6^6 \tag{2b}$$

$$\beta_6 = \left[\frac{\left(1+3\varepsilon^2\right)\left(1+4\varepsilon^2\right)\left(1+5\varepsilon^2\right)}{\left(1+\varepsilon^2\right)\left(1+2\varepsilon^2\right)}\right]^{1/6} \tag{2c}$$

where $\kappa = 1.1 \times 10^{10}$ $cm^{-3}s^{-1}$ is an empirical coefficient in the Long collection kernel for $r < 50$ μm (Long 1974), ρ_w is the water density, ε is the relative dispersion of the cloud droplet size distribution, and r_6 is the mean radius of the 6th moment of the droplet size distribution. Heaviside function $H(r_6 - r_c)$ is introduced to consider the threshold process such that the autoconversion rate is negligibly small when $r_6 < r_c$.

Rewritten in the form of the commonly used previous Kesser-type parameterizations (e.g., Boucher et al. 1995), the Liu-Daum parameterization becomes

$$P = \alpha N^{-1/3} L^{7/3} H\left(r_6 - r_c\right) \tag{3a}$$

$$\alpha = \left(\frac{3}{4\pi\rho_w}\right)^2 \kappa\beta_6^6 \left(\frac{L}{N}\right)^{2/3} \tag{3b}$$

The following points should be emphasized. First, the Liu-Daum parameterization exhibits stronger dependence of the autoconversion rate on both the liquid water content (L^3), the droplet concentration (N^{-1}) and the relative dispersion; this change affects the evaluation of the precipitation and the indirect aerosol effect after the onset of the autoconversion Second, the control radius is the mean radius of the 6^{th} moment instead of 3^{rd} or 4^{th} moments; this change affects the evaluation of the onset of the autoconversion. These improvements comes from the elimination of the incorrect assumption of fixed collection efficiency inherent in the previous parameterizations (Liu and Daum 2004) Examination of the Liu-Daum parameterization provides an explanation for a number of long-standing issues associated with previous parameterizations. For example, such a wide range of values have been assigned to the coefficient a_k in studies using the original Kessler parameterization that, in practice, it has been often considered to be arbitrarily tunable (e.g., Kessler 1969, Liu and Orville 1969, Ghosh et al. 2000). It is evident from the Liu and Daum parameterization that the wide range of values assumed for a_k may stem from the variabilities in the liquid water content, droplet concentration and relative dispersion that are not properly accounted for in the original Kessler parameterization. Similar to the arbitrary tunability of the coefficient a_k in the original Kessler parameterization, a wide range of values have been also assigned to the α coefficient in modeling studies using the traditional R_4 parameterizations (Baker 1993, Boucher et al. 1995). The Liu-Daum parameterization again suggests that the wide range of the coefficient α in the early parameterizations may be largely due to the variability of droplet concentration, liquid water content and relative dispersion. This is demonstrated in Figure 1. The result also suggests that the substantial effects of variation of the liquid water content and droplet concentration are masked by the tunable coefficient α in the previous Kessler-type parameterization that has been commonly used.

Expression for the Critical Radius

Kinetic Potential Theory

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. McGraw and Liu (2003) have recently 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 and collection occurring in warm clouds. Briefly, by analogy to the kinetic theory on nucleation, the kinetic potential $\Phi(j)$ for a droplet consisting of j water molecules is given by

$$\Phi(j) \equiv -\ln\left[\prod_{g=1}^{j-1}\frac{\beta_{con}(g)+\beta_{col}(g)}{\gamma_{eva}(g+1)}\right] \tag{4a}$$

$$\beta_{col}(g) = \frac{\kappa\nu}{\rho_w^2}g^2L \tag{4b}$$

$$\gamma_{eva}(g) = \exp(\frac{\nu N}{L})\beta_{con}(g) \tag{4c}$$

$$g = \frac{4\pi\rho_w}{3\nu}r^3 \tag{4d}$$

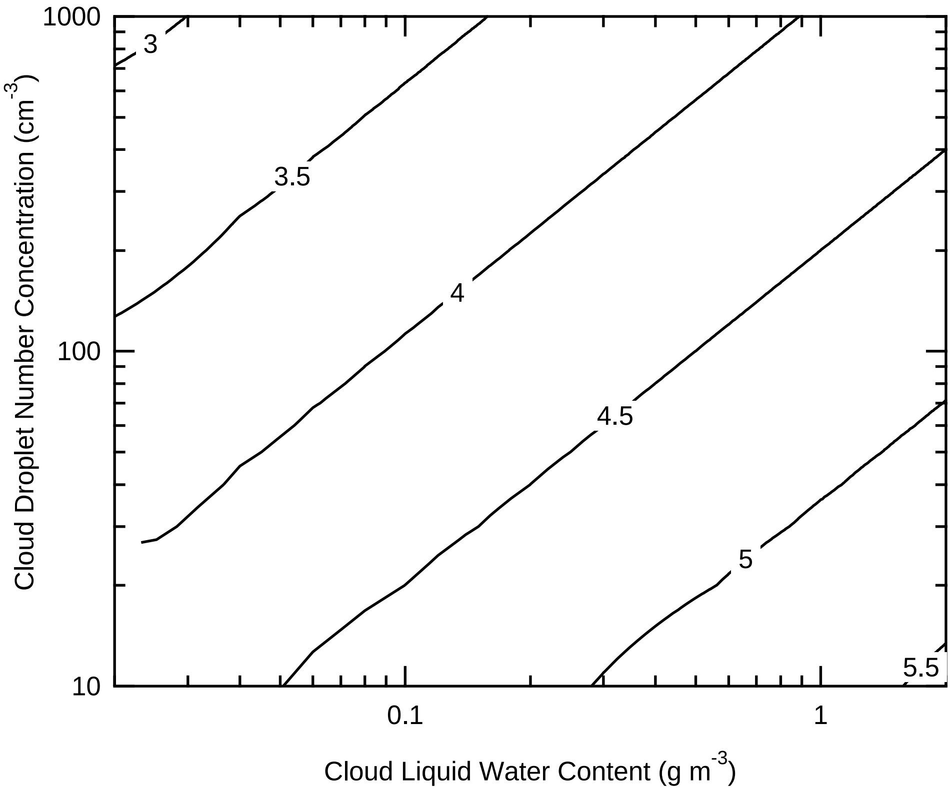

Figure 1. The empirical coefficient α as a function of the liquid water content and droplet concentration as calculated from Equation (3) for a monodisperse cloud droplet size distribution. The contour value denotes the log(α). The α coefficient varies by three orders of magnitude depending on the combination of the droplet concentration and the liquid water content. The effect of the liquid water content and droplet concentration on the conversion coefficient in the original Kessler scheme is even larger, depending the choice of the threshold liquid water content Lc.

where β_{con} (s^{-1}), β_{col} (s^{-1}), and γ_{eva} (s^{-1}) denote the condensation, collection, effective evaporation rate constants, respectively; $\nu = 3.0 \times 10^{-23}$ (g) is the mass per water molecule; s^{-1} is a constant in the Long collection kernel; ρ_w is the water density (g cm^{-3}). The kinetic potential as a function of droplet radius (r) can be then calculated using Equation (4). Figure 2 shows a typical example of the change of the kinetic potential with the droplet radius. The kinetic potential first increases with increasing droplet radius and then decreases after reaching a peak.

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 threshold process and r_c inherent in Kessler-type parameterizations of the autoconversion process emerges naturally from the kinetic potential theory.

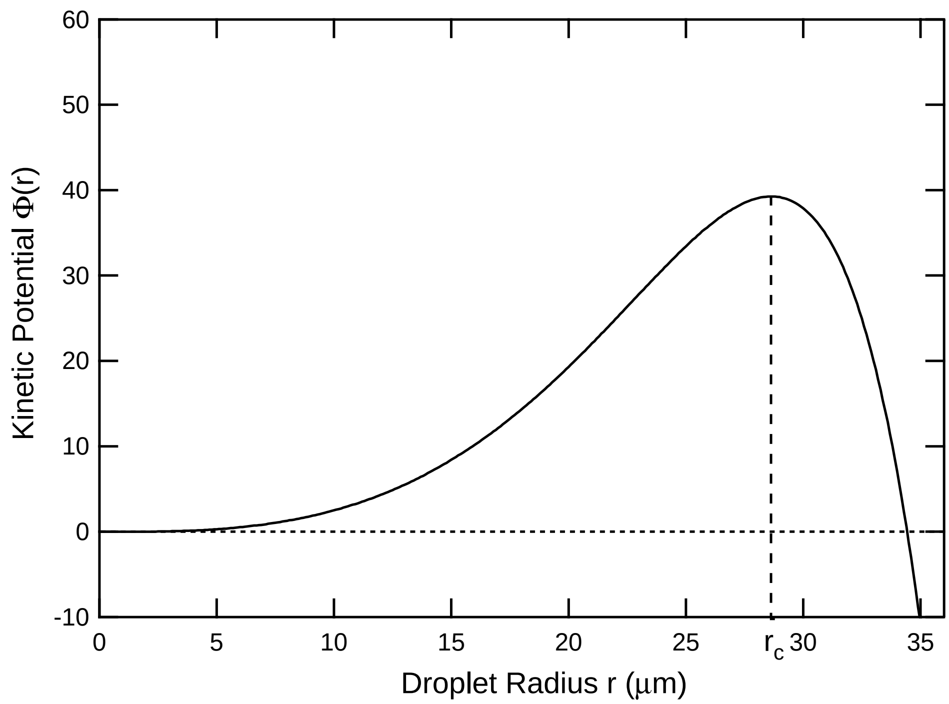

Figure 2. An example of the kinetic potential as a function of the droplet radius. The results are for a cloud liquid water content L = 0.5 g m^{-3}, the cloud droplet number concentration N = 300 cm^{-3}, and the condensation rate constant $\beta_{con} = 9 \times 10^{24}$ s^{-1}.

Analytical Expression for Critical Radius

In state-of-the art GCMs, L and N are predicted/diagnosed. It is therefore desirable to have an analytical expression that relates r_c to these two variables. It is known from McGraw and Liu (2003) that at the critical point, the forward and reverse rate constants are equal, i.e.,

$$\beta_{con} + \beta_{col} = \gamma_{eva} \tag{5}$$

Therefore, the critical radius is given by

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

Because (νN/L) << 1, Equation (6a) can be simplified to

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

In general, β_{con} is a complex function of cloud turbulence that is unknown at present (McGraw and Liu 2003). The mean condensation rate constant of $\beta_{con} = 1.15 \times 10^{23}$ s^{-1} estimated in Liu et al. (2004) from microphysical measurements is used in this work.

Critical Radius of Ambient Clouds

Equation (6b) 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 (6b) using the mean β_{con} and the data on L and N from stratiform clouds given in Miles et al. (2000). It is clear that r_c varies significantly, from ~ 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.

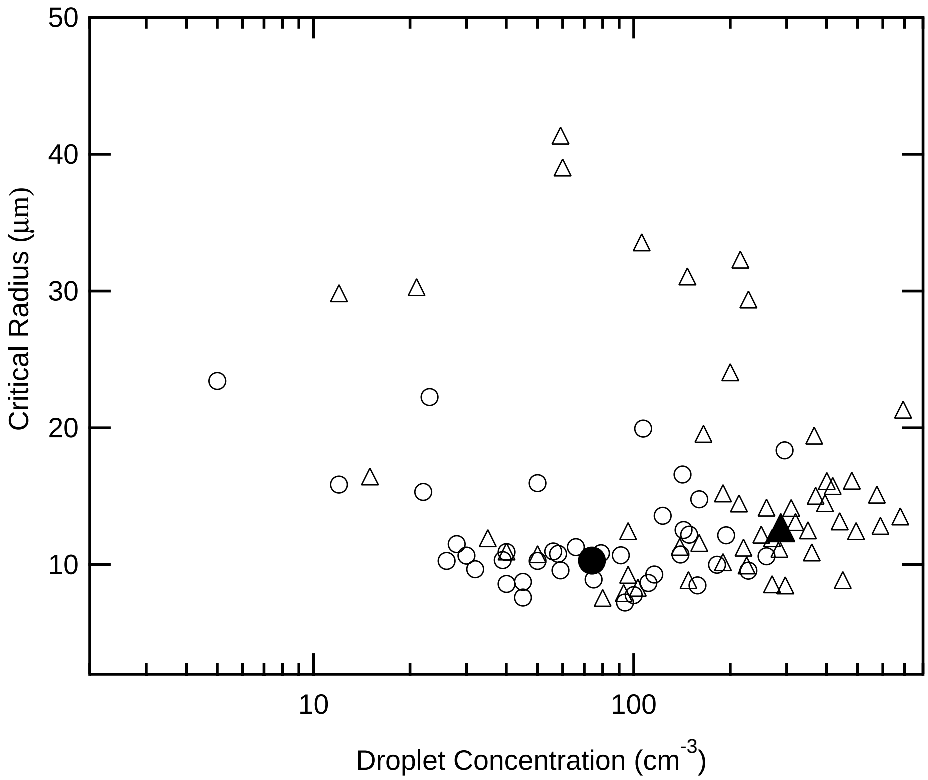

Figure 3. Relationship of the critical radius to the droplet concentration. The open triangles and dots denote continental and marine clouds, respectively. The solid triangle and dot denote the average of continental and marine clouds, respectively.

New Parameterization and Dispersion Effect

A new parameterization for the autoconversion rate is readily obtained by substituting the expression for r_c into the Liu-Daum parameterization. In the form of Equation (1), the new parameterization is

$$f = \left(\frac{3}{4\pi\rho_w}\right)^2 \kappa \left[\frac{\left(1+3\varepsilon^2\right)\left(1+4\varepsilon^2\right)\left(1+5\varepsilon^2\right)}{\left(1+\varepsilon^2\right)\left(1+2\varepsilon^2\right)}\right] N^{-1} L^3 H\left(r_6 - r_c\right) \tag{7a}$$

$$r_6 = \left(\frac{3}{4\pi\rho_w}\right)^{1/3} \left[\frac{\left(1+3\varepsilon^2\right)\left(1+4\varepsilon^2\right)\left(1+5\varepsilon^2\right)}{\left(1+\varepsilon^2\right)\left(1+2\varepsilon^2\right)}\right]^{1/6} \left(\frac{L}{N}\right)^{1/3} \tag{7b}$$

$$r_c = 4.09\times10^{-4}\beta_{con}^{1/6}\frac{N^{1/6}}{L^{1/3}} \tag{7c}$$

The new parameterization has the following important implications for the evaluation of the second indirect aerosol effect. First, anthropogenic aerosols inhibit the onset of embryonic raindrops by decreasing the control radius r_6 and by increasing the critical radius. This phenomenon is self-evident from the relation between r_6 and r_c, (Liu et al. 2004a)

$$r_c = \left[\left(\frac{3}{4\pi}\right)^3 \frac{\nu^2\beta_{con}}{\kappa\rho_w L}\right]^{1/6} \frac{\alpha^{1/2}}{r_6^{1/2}} \approx \frac{23.72}{L^{1/6} r_6^{1/2}} \tag{8a}$$

or from the equation

$$\Delta = r_6 - r_c = r_6 - \frac{23.72}{L^{1/6} r_6^{1/2}} \tag{8b}$$

Second, an increase in aerosol loading also decreases the conversion rate from cloud water to rain water after the onset of the autoconversion process. This phenomenon is evident from Equation (7a). Note that the new parameterization accounts for the effects of relative dispersion on the control radius and the part after the autoconversion starts; the effect of the dispersion on the critical radius can be addressed by generalizing the kinetic potential theory for the drizzle formation (Liu et al. 2004b).

Conclusions

The Liu-Daum parameterization for the autoconversion rate is briefly introduced, and is used to explain the wide range of the empirical coefficient α that has been commonly used in previous Kessler-type parameterizations. An analytical expression for r_c is introduced and discussed by coupling the kinetic potential theory on the formation of warm rain with the Liu-Daum parameterization. A new parameterization for the autoconversion rate is presented by substituting the expression for r_c into the Liu-Daum parameterization It is shown that anthropogenic aerosols have the effect of increasing r_c but decreasing the control radius concurrently, inhibiting the onset of embryonic raindrops. An increase in aerosol loading also decreases the conversion rate from cloud water to rain water after the onset of the autoconversion.

The effect of the relative dispersion on the autoconversion rate after the onset of this process manifest itself in Equation (7a). Part of the effect of the relative dispersion on the onset of the autoconversion process is embodied in the equation for the control radius (Equation [7b]). However, a complete solution is awaiting the inclusion of the relative dispersion in the formulation of the expression for r_c.

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