BNL-72030-2004-JA

AN ANALYTICAL EXPRESSION FOR PREDICTING THE CRITICAL RADIUS IN THE AUTOCONVERSION PARAMETERIZATION

Yangang Liu, Peter H. Daum, and Robert McGraw
Atmospheric Sciences Division
Brookhaven National Laboratory
Upton, NY 11973-5000

January 2004

Submitted to
Geophysical Research Letters

By acceptance of this article, the publisher and/or recipient acknowledges the U.S. Government's right to retain a nonexclusive, royalty-free license in and to any copyright covering this paper.

Research by BNL investigators was performed under the auspices of the U.S. Department of Energy under Contract No. DE-AC02-98CH10886.

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

Yangang Liu, Peter H. Daum and Robert McGraw

Atmospheric Sciences Division, Brookhaven National Laboratory, Upton, New York

Abstract. An analytical expression for the critical radius associated with Kessler-type parameterizations of the autoconversion process is derived by coupling the kinetic potential theory on rain formation with a parameterization of the autoconversion rate. 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. Data from stratiform clouds are analyzed, indicating that on average continental clouds tend to have larger critical radii than their maritime counterparts. 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.

1. Introduction

Accurate representation of cloud and precipitation processes in atmospheric models of various scales [from large eddy simulations (LESs) to cloud resolving models (CRMs) to 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 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. The change from the critical L to r_c makes physical sense because a cloud with a small mean radius will not rain no matter how large L is. 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 later Kessler-type parameterizations can be generically written as

$$P = fH(r_m - r_c) \qquad [1]$$

where P is the autoconversion rate; f is a function of L and N; r_m is 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 the characteristic radius is less than r_c. The characteristic 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]. The function f is also different for different parameterizations.

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 has been used rather loosely, and r_c has been largely prescribed as an empirical parameter that is arbitrarily tuned to match model simulations with observations. The primary objective of this work is to derive 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. The derived 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

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

The key elements of the kinetic potential theory are recapitulated below. By analogy to the kinetic theory on nucleation, the kinetic potential $\Phi(j)$ for a droplet consisting of j water molecules is given by

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

$$\Lambda_{col}(g) = \frac{\Delta v}{\Delta_w^2} g^2 L, \qquad [2b]$$

$$\Lambda_{eva}(g) = \exp\left(\frac{vN}{L}\right) \Lambda_{con}(g), \qquad [2c]$$

where Ω_{con} (s^{-1}), Ω_{col} (s^{-1}), and Ω_{eva} (s^{-1}) denote the condensation, collection, effective evaporation rate constants, respectively, for a droplet consisting of g water molecules; μ = 3.0 x 10^{-23} (g) is the mass per water molecule; Γ = 1.1 x 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 then calculated by applying the relation $g = \frac{4\pi\rho_w}{3v} r^3$ to Eqs. (2a,b,c). Figure 1 shows a typical example of the change of the kinetic potential with the droplet radius. The kinetic potential first increases with increasing droplet radius because (Ω_{con} + Ω_{col}) < Ω_{eva}, and then decreases after reaching a peak because (Ω_{con} + Ω_{col}) > Ω_{eva} [Note that $\frac{d\Phi}{dg} = \ln\left(\frac{\Omega_{con} + \Omega_{col}}{\Omega_{eva}}\right)$].

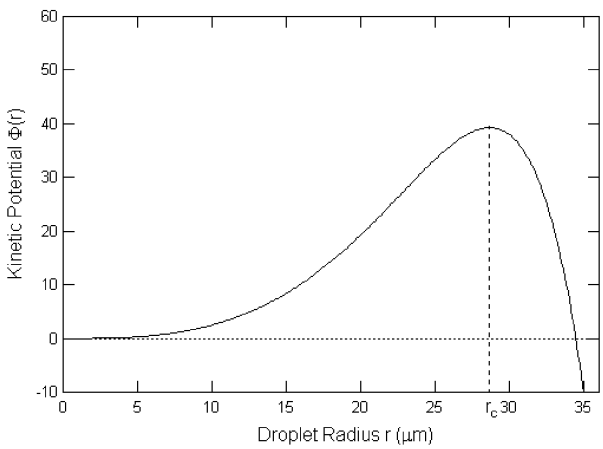

Figure 1. An example of the kinetic potential as a function of the droplet radius. The results are for a cloud liquid water content of 0.5 g m^{-3}, the droplet concentration of 300 cm^{-3}, and the condensation rate constant of 10^{24} s^{-1}.

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 of the autoconversion process emerges naturally from the kinetic potential theory.

3. Analytical Expression for Critical Radius

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

$$\Omega_{con} + \Omega_{col} = \Omega_{eva} \qquad [3]$$

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

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

Because (ΓN/L) << 0, and $\exp\left(\frac{vN}{L}\right) \approx 1 + \frac{vN}{L}$, Eq. (4a) can be simplified to

$$r_c = \left(\frac{3}{4\pi}\right)^2 \frac{v^2}{\Gamma} \Omega_{con} \frac{N}{L^2}\Bigg)^{1/6} \approx 4.09 \times 10^{-4} \Omega_{con}^{1/6} \frac{N^{1/6}}{L^{1/3}} , \qquad [4b]$$

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

In general, Ω_{con} is a function of cloud 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], the autoconversion process starts when the mean radius of the 6th moment reaches r_c. The mean radius of the 6th moment (r_6) is given by

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

where the dimensionless parameter κ = 1.12. Equating r_6 with r_c, we obtain an expression for Ω_{con}

$$\Omega_{con} = \frac{k\kappa^6 L_*^4}{(v\rho_w)^2 N_*^3} = 2.37 \times 10^{31} \frac{L_*^4}{N_*^3} , \qquad [6]$$

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 Ω_{con} in s^{-1}.

Equation (6) is used to estimate Ω_{con} from concurrent measurements of L and N in drizzling clouds reported in *Yum and Hudson* [2001, 2002]. As shown in Figure 2, the majority of Ω_{con} values fall between 10^{22} and 10^{23} (s^{-1}); the mean, minimum and maximum of Ω_{con} are 1.15 x 10^{23}, 1.02 x 10^{23} and 1.67 x 10^{24} (s^{-1}), respectively. These estimates seem consistent with our modeling study [*McGraw and Liu*, 2003].

4. Critical Radius of Ambient Clouds and Important Implications

Equation (4b) indicates that r_c is a function of L and N given Ω_{con}, 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 Eq. (4b) using the mean Ω_{con} = 1.15 x 10^{23} s^{-1}, and the summary of data on L and N from stratiform clouds given in *Miles et al.* [2000]. It is clear from Figure 3 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 LESs and CRMs than in GCMs.

It is interesting to compare r_6 with r_c because the former, or its "equivalent" such as the volume-mean radius, is often used in studies of both the first and the second indirect aerosol effects [*Twomey*, 1991; *Albrechet*, 1989]. The equation relating r_c to r_6 can be derived by using Eq. (5) to eliminate N in Eq. (4b),

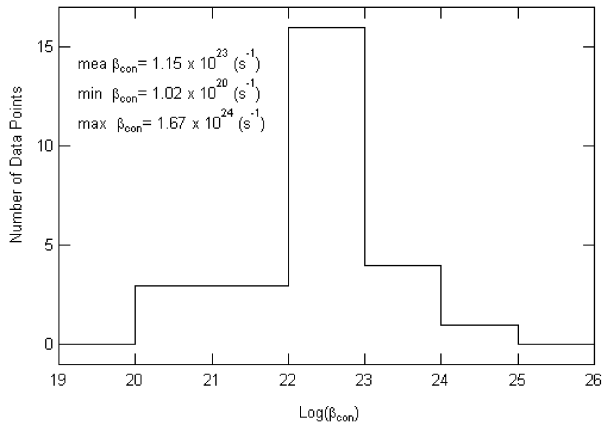

Figure 2. Histogram of the condensate rate constant estimated by applying Eq. (6) to concurrent measurements of liquid water content and droplet concentration in drizzling clouds. The data are from *Yum and Hudson* [2001, 2002].

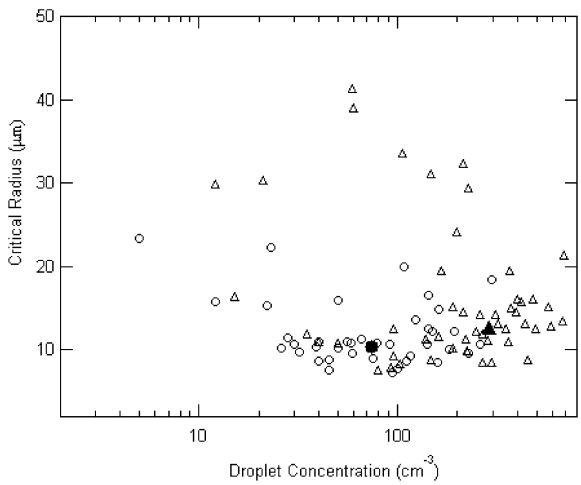

Figure 3. Relationship of the critical radius to the droplet concentration. Data are from *Miles et al.* [2000]. The open triangles and dots represent continental and marine stratiform clouds, respectively. The larger solid triangle and dot represent the average of continental and marine clouds, respectively. The dot and dash lines represent two calculations from Eq.(7) at L=0.001 and 2 gm^{-3}, respectively.

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

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 as assumed in studies of the first indirect aerosol effect. This is illustrated by the dotted and dashed 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 seem to support this notion. Because of higher droplet concentrations, the continental clouds have an average r_c (12.71 μm) larger than that of the marine clouds (10.32 μm) (Note that there is little difference in the averaged L because the average points fall along virtually the same line of a constant L). These values are close to those prescribed in CRMs [Tripoli and Cotton, 1980], but larger than those normally assigned in GCMs [*Boucher and Lohmann*, 1995; *Rasch and Kristjansson*, 1998; *Rotstayn*, 1999]. A smaller r_c for GCMs has often been 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 Eq. (4b) supports this argument.

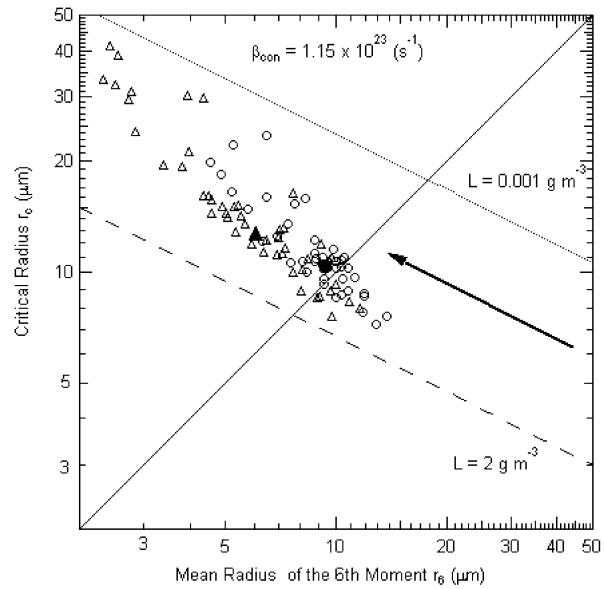

Figure 4. Dependence of the critical radius on the mean radius of the 6th moment. The meanings of symbols are the same as in Figure 3. The arrow denotes the direction of increasing aerosol loading.

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 [*Albrechet*, 1989]. However, there is rain ($r_6 > r_c$) in some continental clouds whereas there is no rain ($r_6 < r_c$) in some marine clouds, 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 further suggests that anthropogenic aerosols inhibit the initiation of embryonic raindrops by increasing r_c as well. Comparative quantification of the three effects has not been adequately investigated, and our work provides a novel way to address this issue.

5. Concluding Remarks

An analytical expression for r_c is derived for the first time by coupling the kinetic potential theory on the formation of warm rain with the parameterization of the autoconversion rate. The new expression gives the threshold process assumed in Kessler-type parameterizations and r_c a solid physical basis. The expression can be used to predict r_c from L and N, eliminating the need to prescribe r_c in numerical models. Examination of data from strati-

form clouds indicates that r_c varies from cloud to cloud, and that on average, continental clouds tend to have larger r_c than their maritime counterparts. It is further suggested that anthropogenic aerosols have the effect of increasing r_c and decreasing the mean radius concurrently, inhibiting the onset of embryonic raindrops. We believe that this new approach of specifying r_c will significantly improve the representation of precipitation in numerical models and our ability to model the second indirect aerosol effect.

Three points are worth noting. First, there is uncertainty in our estimate for Φ_{con} (hence r_c) due to limited measurements of L and N used in the calculation, and to the uncertainty to ascertain if these measurements were taken exactly at threshold points as required by Eq. (6). More research is needed to reduce the uncertainty involved. Furthermore, Φ_{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 kinetic potential theory and the expression for r_c. 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 Φ_{con} because r_c is proportional to $\Phi^{1/6}_{con}$ [Eq. (4b)].

Acknowledgments. This research was supported by the Atmospheric Radiation Measurements Program of the US Department of Energy under contract DE-AC03-98CH10886, a LDRD grant from the Brookhaven National Laboratory under contract LDRD 03-026, and the Atmospheric Chemistry Program under contract DE-AC02-98CH10886. The authors thank Dr. S. E. Schwartz at BNL for stimulating discussions, and Dr. S. J. Ghan at PNL and an anonymous reviewer for their insightful comments.

References

Albrecht, B., Aerosols, cloud microphysics, and fractional cloudiness, *Science, 245*, 1227-1230, 1989.

Baker, M. B., Variability in concentrations of CCN in the marine cloud-top boundary layer, *Tellus*, *45B*, 458-472, 1993.

Beard, K. V., and H. T. Ochs, Warm-rain initiation: An overview of microphysical mechanisms, *J. Appl. Meteor.*, *32*, 608-625, 1993.

Beheng, K. D., A parameterization of warm cloud microphysical conversion processes, *Atmos. Res.*, *33*, 193-206, 1994.

Berry, E. X., Modification of the warm rain process. Preprints 1st Nat. Conf. Weather Modification, Albany, Amer. Meteor. Soc., 81-88, 1968:

Boucher, O., and U. Lohmann, The sulfate-CCN-cloud albedo effect. A sensitivity study with two general circulation models, *Tellus*, *47B*, 281-300, 1995.

Boucher, O., H. L. Treut, and M. B. Baker, Precipitation and radiation modeling in a general circulation model: Introduction of cloud microphysical process, *J. Geophys. Res.*, *D100*, 16395-16414, 1995.

Chen, C., and W. R. Cotton, The physics of the marine stratocumulus-capped mixed layer, *J. Atmos. Sci.*, *44*, 2951-2977, 1987.

Ghan, S. J., L. R. Leung, and Q. Hu, Application of cloud microphysics to NCAR community climate model, *J. Geophys. Res.*, *D102*, 16507-16527, 1997a.

Ghan, S. J., L. R. Leung, and R. C. Easter, Prediction of cloud droplet number in a general circulation model, *J. Geophys. Res.*, *D102*, 21777-21794, 1997b.

Kessler, E., On the distribution and continuity of water substance in atmospheric circulation, *Meteoro. Monogr., 10*, 84, 1969.

Khairoutdinov, M, and Y. Kogan, A new cloud physics parameterization in a large-eddy simulation model of marine stratocumulus, *Mon. Wea. Rev.*, *128*, 229-243, 2000.

Liou, K. N., and S. C. Ou, The role of cloud microphysical processes in climate: An assessment from a one-dimensional perspective, *J. Geophys. Res.*, *D94*, 8599-8607, 1989.

Liu, Y., and P. H. Daum, Spectral dispersion of cloud droplet size distributions and the parameterization of cloud droplet effective radius, *Geophys. Res. Lett.*, *27*, 1903-1906, 2000.

Liu, Y., and P. H. Daum, Indirect warming effect from dispersion forcing, *Nature*, *419*, 580-581, 2002.

Liu, Y., and P. H. Daum, On the parameterization of the autoconversion process. Part I: Analytical formulation of the Kessler-type parameterizations, *J. Atmos. Sci.*, in press.

Lohmann, U., and J. Fleichter, Impact of sulfate aerosols on albedo and lifetime of clouds: A sensitivity study with the ECHAM 4 GCM, *J. Geophys. Res.*, *D102*, 13685-13700, 1997.

Long, A. B., Solutions to the droplet collection equation for polynomial kernels, *J. Atmos. Sci.*, *31*, 1040-1052, 1974.

Manton, M. J., and W. R. Cotton, Formulation of approximate equations for modeling moist deep convection on the mesoscale, Atmos. Sci. Paper No. 266, Dept. of Atmos. Sci., Colorado State University, 1977.

McGraw, R., and Y. Liu, Kinetic potential and barrier crossing: A model for warm cloud drizzle formation, *Phys. Rev. Lett., 90*, 018501-1-4, 2003.

Miles, L. N., J. Verlinde, and E. E. Clothiaux, Cloud droplet size distributions in low-level stratiform clouds, *J. Atmos. Sci.*, *57*, 295-311, 2000.

Peng, Y., and U. Lohmann, Sensitivity study of the spectral dispersion of the cloud droplet size distribution on the indirect aerosol effect, *Geophys. Res. Lett.*, *30*, 14-1-14-4, doi: 10.1029/2003GL017192, 2003.

Pincus, R., and S. A. Klein, Unresolved spatial variability and microphysical process rates in large-scale models, *J. Geophys. Res.*, *D105*, 27059-27065, 2000.

Rasch, P. J., and J. E. Kristjansson, A comparison of the CCM3 model climate using diagnosed and predicted condensate parameterization, *J. Climate*, *11*, 1587-1614, 1998.

Rasmussen, R. M., I. Geresdi, G. Thompson, K. Manning, and E. Karplus, Freezing drizzle formation in stratified layer clouds: The role of radiative cooling of cloud droplets, cloud condensation nuclei, and ice initiation, *J. Atmos. Sci., 59*, 837-859, 2002.

Rotstayn, L. D., A physically based scheme for the treatment of stratiform clouds and precipitation in large-scale models. I: Description and evaluation of the microphysical processes, *Q. J. R. Meteorol. Soc.*, *123*, 1227-1282, 1997.

Rotstayn, L. D., Indirect forcing by anthropogenic aerosols: A global climate model calculation of the effective radius and cloud-lifetime effects, *J. Geophys. Res.*, *D104*, 9369-9380,1999.

Rotstayn, L. D., On the "tuning" of the autoconversion parameterizations in climate models, *J. Geophys. Res.*, *D105*, 15495-15507, 2000.

Rotstayn, L. D., and Y. Liu, Sensitivity of the indirect aerosol effect to the parameterization of cloud droplet spectral dispersion, *J. Climate*, *16*, 3476-3481, 2003.

Stokes, G. M., and S. E. Schwartz, The Atmospheric Radiation Measurement (ARM) Program: Programmatic background and design of the cloud and radiation test bed, *Bull. Amer. Meteoro. Soc.*, *75*, 1201-1221, 1994.

Telford, J. W., Clouds with turbulence; the role of entrainment, *Atmos. Res.*, *40*, 261-282, 1996.

Tripoli, G. J., and W. R. Cotton, A numerical investigation of several factors contributing to the observed variable intensity of deep convection over South Florida, *J. Atmos. Sci.*, *19*, 1037-1063, 1980.

Twomey, S., Aerosols, clouds, and radiation, *Atmos. Environ.*, *25*, 2435-2442, 1991.

Yum, S. and J. G. Hudson, Microphysical relationships in warm clouds, *Atmos. Res.*, *57*, 81-104 2001.

Yum, S. and J. G. Hudson, Maritime/continental microphysical contrasts in stratus, *Tellus*, *54B*, 61-73, 2002.

Zhang, J., U. Lohmann, and B. Lin, A new statistically based autoconversion rate parameterization for use in large-scale models, *J. Geophys. Res.*, *D107*, AAC3-1-16, doi: 10.1029/2001JD001484, 2002.

Y. Liu, P. H. Daum, and R. McGraw, Atmospheric Sciences Division, Brookhaven National Laboratory, Upton, NY 11973-5000. (e-mail: lyg@bnl.gov; phdaum@bnl.gov; rlm@bnl.gov.)