BROOKHAVEN NATIONAL LABORATORY

BNL-75214-2005-JA-R1

ANALYTICAL EXPRESSION FOR THE RELATIVE DISPERSION OF THE CLOUD DROPLET SIZE DISTRIBUTION

Y. Liu, P. H. Daum, and S. S. Yum

Published in
Geophysical Research Letters
[vol. 33, L02810, doi:10.1029/2005GL024052 (2006)]

December 2005
Revised: February 2006

Environmental Sciences Department/Atmospheric Sciences Division

Brookhaven National Laboratory
P.O. Box 5000
Upton, NY 11973-5000
www.bnl.gov

Analytical expression for the relative dispersion of the cloud droplet size distribution

Yangang Liu,[1] Peter H. Daum,[1] and Seong Soo Yum[2]

Received 12 July 2005; revised 9 November 2005; accepted 14 December 2005; published 26 January 2006.

[1] An analytical expression that relates the relative dispersion (ratio of standard deviation to mean radius) of the cloud droplet size distribution to CCN spectra and updraft velocity is derived from adiabatic growth theory of cloud droplets. Coupled with the Twomey expression for droplet concentration, the analytical expression is used to examine the relationship of relative dispersion to droplet concentration under different combinations of CCN spectra and updraft velocities. These analytical results compare favorably with the corresponding simulations of an adiabatic parcel model. The analytical expression theoretically demonstrates that an increase in aerosol loading (CCN concentration) leads to concurrent increases in the droplet concentration and relative dispersion whereas a larger updraft velocity leads to a higher droplet concentration but a smaller relative dispersion. **Citation:** Liu, Y., P. H. Daum, and S. S. Yum (2006), Analytical expression for the relative dispersion of the cloud droplet size distribution, *Geophys. Res. Lett.*, *33*, L02810, doi:10.1029/2005GL024052.

1. Introduction

[2] The indirect effects of anthropogenic aerosols caused by alteration of microphysical properties of clouds are poorly understood. It is well known that an increase in aerosol loading results in an increase in cloud condensation nuclei (CCN), and hence an increase in the cloud droplet number concentration (N). The increased N decreases droplet sizes and enhances cloud albedo [*Twomey*, 1977]. The decreased droplet sizes suppress drizzle, which in turn increases the cloud liquid water content (L), cloud thickness, cloud lifetime, and cloud coverage, and further enhances cloud albedo [*Albrecht*, 1989]. Furthermore, it has been shown that an increase in aerosol loading also leads to an increase in the relative dispersion (ε = standard deviation/mean radius) of the cloud droplet size distribution [*Martin et al.*, 1994; *Hudson and Yum*, 1997; *Miles et al.*, 2000; *Liu and Daum*, 2002]. Contrary to the effects of increased N, the enhanced dispersion exerts a warming effect on climate (dispersion effect) [*Liu and Daum*, 2002]. The dispersion effect, which has been largely ignored in global climate models, is in part responsible for the large uncertainty in estimates of the indirect aerosol effects and the discrepancy between model estimates of the indirect aerosol effects and those constrained by observations [*Liu and Daum*, 2002; *Peng and Lohmann*, 2003; *Rotstayn and Liu*, 2003]. Microphysical modeling studies have also confirmed the increase of ε with aerosol loading [*Wood*, 2002; *Feingold and Chuang*, 2002; *Xue and Feingold*, 2004; *Yum and Hudson*, 2005]. Despite the progress, quantitative understanding of ε and its relationship to N and aerosol properties is in its infancy.

[3] To improve quantification of indirect aerosol effects in GCMs, it is desirable to derive analytical expressions from first principles that relate N and ε to pre-cloud aerosol properties. Over the last few decades, a number of analytical expressions have been developed for N [*Squires*, 1958; *Twomey*, 1959; *Ghan et al.*, 1993]. However, an analytical expression that clearly embodies the physics behind the increase of ε with aerosol loading is still lacking. To fill this gap, here we derive an analytical expression that relates ε to CCN spectra and updraft velocity (w). The relationship of ε to N calculated from the analytical expressions under different combinations of CCN spectra and w are then compared with simulations of an adiabatic parcel model [*Yum and Hudson*, 2005].

2. Derivation of the Analytical Expression for Relative Dispersion

[4] Droplet condensational growth is described by the following equations [*Pruppacher and Klett*, 1997],

$$\frac{dr}{dt} = \frac{S}{Gr}, \tag{1a}$$

$$G = \left(\frac{L_v}{R_v T} - 1\right)\frac{L_v \rho_w}{k_d T} + \frac{\rho_w R_v T}{D_v e_s} \tag{1b}$$

$$D_v = 2.11 \times 10^{-5}\left(\frac{T}{273.15}\right)^{1.94}\left(\frac{101325}{P}\right), \tag{1c}$$

$$k_d = 4.18 \times 10^{-3}[5.69 + 0.017(T - 273.15)], \tag{1d}$$

$$L_v = 2.5 \times 10^6\left(\frac{273.15}{T}\right)^{\left(0.167+3.26\times10^{-4}T\right)}, \tag{1e}$$

$$e_s(T) = 6.112 \times 10^2 \exp\left[\frac{17.67(T - 273.15)}{T - 29.65}\right], \tag{1f}$$

where r is the droplet radius, S the water vapor supersaturation, ρ_w the water density, T the air temperature in

[1]Brookhaven National Laboratory, Upton, New York, USA.
[2]Department of Atmospheric Sciences, Yonsei University, Seoul, South Korea.

0094-8276/06/2005GL024052$05.00

Kelvin degree, D_v (m^2s^{-1}) the vapor diffusivity, k_d the thermal conductivity, L_v (J kg^{-1}) the latent heat of vapor condensation, R_v = 461 J $kg^{-1}K^{-1}$ the specific gas constant of water vapor, and P and e_s are the air and saturation vapor pressures, respectively, in Pascal.

[5] To derive the equation that describes the relative dispersion ε, we introduce a new variable

$$x = \left(\frac{r - r_1}{r_1}\right), \tag{2a}$$

which is a fractional measure of the radius deviation of each droplet from the mean radius, r_1, of the droplet population. In terms of x, r can be expressed as

$$r = r_1(1 + x). \tag{2b}$$

Application of equation (2b) to equation (1a) results in

$$(1 + x)\frac{dr_1}{dt} + r_1\frac{dx}{dt} = \frac{S}{G}\frac{1}{r_1(1 + x)} \approx \frac{S}{Gr_1}(1 - x). \tag{3}$$

(The Taylor expansion $\frac{1}{1+x} \approx 1 - x$ is used to simplify the equation). Multiplication of both sides of equation (3) by r_1 yields

$$\frac{(1 + x)}{2}\frac{dr_1^2}{dt} + r_1^2\frac{dx}{dt} = \frac{S}{G}(1 - x). \tag{4}$$

Averaging all the terms of equation (4) over the droplet population leads to an equation for the mean radius

$$\frac{1}{2}\frac{dr_1^2}{dt} = \frac{S}{G}. \tag{5}$$

Substitution of equation (5) back to equation (4) yields

$$r_1^2\frac{dx}{dt} = -\frac{2S}{G}x. \tag{6}$$

Multiplication of equation (6) by x and averaging over the droplet population leads to

$$\frac{r_1^2}{2}\frac{d\overline{x^2}}{dt} = -\frac{2S}{G}\overline{x^2}. \tag{7}$$

Division of equation (7) by equation (5) gives

$$\frac{d\overline{x^2}}{\overline{x^2}} = -\frac{2dr_1^2}{r_1^2}. \tag{8}$$

Noting that $\varepsilon^2 = \overline{x^2}$, we have

$$\varepsilon = \varepsilon_0 r_{10}^2 r_1^{-2}, \tag{9}$$

where the subscript "0" denotes the initial time. Equation (9) indicates that ε decreases when droplets grow, which is consistent with the adiabatic condensation theory.

[6] Under the steady-state assumption, r_1 is given by [*Cooper*, 1989]

$$r_1 = \frac{aGw}{4\pi\rho_w bS_m N}, \tag{10a}$$

$$a = \left(\frac{gL_v}{c_pR_vT^2} - \frac{g}{R_dT}\right), \tag{10b}$$

$$b = \frac{R_vT}{e_s} + \frac{R_dL_v^2}{c_pR_vPT}, \tag{10c}$$

where c_p = 1005 J kg^{-1} K^{-1} is the specific heat of dry air at constant pressure, R_d = 287 J $kg^{-1}K^{-1}$ the specific gas constant of dry air, and S_m is the maximum supersaturation. Subsequent substitution of equation (10a) into equation (9) yields

$$\varepsilon = \varepsilon_0 r_{10}^2\left(\frac{4\pi\rho_w bS_m}{aGw}\right)^2 N^2. \tag{11}$$

[7] To further relate S_m and N to CCN properties and updraft velocities, we use the Twomey formulation for droplet activation [*Twomey*, 1959]. Briefly, the CCN spectrum is given by

$$N_{CCN} = c(100S)^k. \tag{12}$$

where N_{CCN} is the cumulative CCN concentration, c and k are two empirical coefficients that depend on aerosol properties, and "100" is introduced because CCN spectra are expressed as a function of percent supersaturation instead of fractional supersaturation. The Twomey expressions for S_m and N are

$$S_m = 10^{-2k/(k+2)}\left[\frac{2(aG)^{3/2}}{4\pi\rho_w kbB\left(\frac{k}{2},\frac{3}{2}\right)}\right]^{1/(k+2)} w^{3/2(k+2)} c^{-1/(k+2)}, \tag{13}$$

$$N = 10^{4k/(k+2)}\left[\frac{2(aG)^{3/2}}{4\pi\rho_w kbB\left(\frac{k}{2},\frac{3}{2}\right)}\right]^{k/(k+2)} w^{3k/2(k+2)} c^{2/(k+2)}, \tag{14}$$

where B() is the complete Beta function. A combination of equations (11), (13) and (14) yields the expression for ε

$$\varepsilon = \varepsilon_0 r_{10}^2 10^{4k/(k+2)}\left(\frac{4\pi\rho_w b}{G}\right)^2 \cdot \left[\frac{2(aG)^{3/2}}{4\pi\rho_w kbB\left(\frac{k}{2},\frac{3}{2}\right)}\right]^{2(k+1)/(k+2)} w^{(k-1)/(k+2)} c^{2/(k+2)}. \tag{15}$$

Equations (14) and (15) together demonstrate that others being the same, an increase in aerosol loading (hence c) leads to concurrent increases in N and ε. Equation (15) also indicates that a smaller k yields larger values N and ε,

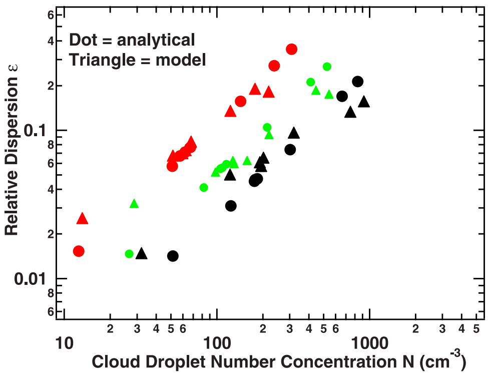

Figure 1. Relationship of relative dispersion (ε) and the cloud droplet number concentration (N) for droplets growing by condensation under adiabatic and uniform conditions. The circles and triangles represent the results from the analytical expression derived in this paper and the adiabatic parcel model simulations, respectively. The red, green, and black colors represent results for updraft velocity w = 0.1 ms^{-1}, 0.5 ms^{-1}, and 2 ms^{-1}, respectively.

which is consistent with parcel model results [*Hudson and Yum*, 1997].

3. Relationship Between Relative Dispersion and Droplet Concentration

[8] Equations. (14) and (15) can be used to study the effects of CCN property and w on N, ε, and the relationship of ε to N. Figure 1 shows some results calculated under different combinations of CCN spectra and w derived from the same measurements that were described by *Yum and Hudson* [2005]. Also shown in Figure 1 are the corresponding simulations using an adiabatic parcel model [*Yum and Hudson*, 2005]. It is clear from Figure 1 that for a given w, N and ε increase simultaneously when aerosol loading (CCN) increases, providing theoretical support for the previously reported observations [*Hudson and Yum*, 1997; *Liu and Daum*, 2002]. On the other hand, for a given CCN spectrum, an increase in w increases N but decreases ε. These analytical results agree reasonably well with parcel model simulations. It is interesting to note that the opposite changes of the relative dispersion with increasing CCN and w arise from the same physical reason: the relative change in water vapor available for condensation per droplet. The dependence of N and ε on w may be one of the reasons for the large scatter in observed ε-N relationships [e.g., *Liu and Daum*, 2002, Figure 1]. In fact, an analytical relationship between ε and N can be derived from equations (14) and (15):

$$\varepsilon = \frac{8\pi\rho_w\varepsilon_0 r_{10}^2 ba^{3/2}}{G^{1/2}kB\left(\frac{k}{2},\frac{3}{2}\right)}w^{-1/2}N. \tag{16}$$

4. Concluding Remarks

[9] An analytical expression that relates the relative dispersion of the cloud droplet size distribution to CCN spectra and updraft velocities is derived from the adiabatic growth theory of cloud droplets. Coupled with the Twomey expression for droplet concentration, the analytical expression is applied to examine the relationship of relative dispersion to droplet concentration under different combinations of CCN spectra and updraft velocities. The analytical results compare favorably with the corresponding simulations of an adiabatic parcel model, and theoretically demonstrates that an increase in aerosol loading (CCN concentration) leads to concurrent increases in the droplet concentration and the relative dispersion whereas a larger updraft velocity leads to a higher droplet concentration but a smaller relative dispersion. Note that equation (16) also indicates that in addition to updraft velocity, aerosol properties (e.g., k) and thermodynamic properties (temperature and pressure) also affect the adiabatic slope of the relationship between droplet concentration and relative dispersion.

[10] The following points are noteworthy. First, several approximations (e.g., neglected curvature and solute effects and the Taylor expansion) are made in the derivation of the analytical expression. The slight discrepancy between the analytical expression and model simulations may be due to these approximations. Second, this paper only addresses the effect of competition for available water vapor on the droplet concentration, relative dispersion, and their relationship under adiabatic and uniform conditions. Many other factors/processes are expected to affect the competition process and hence modify the relationship of droplet concentration to relative dispersion, including accommodation coefficient, surface tension, solubility, ripening, gaseous adsorption, turbulent entrainment/mixing, and droplet collection. Nevertheless, the basic features of this work should hold, and the analytical expression should be a useful reference for more detailed investigation, especially for non-drizzling clouds where water vapor condensation is the dominant growth mechanism. Third, besides the Twomey power-law expressions, several other parameterizations for CCN spectra, droplet concentration and maximum supersaturation have been developed over the last few years, with different levels of complexity [*Ghan et al.*, 1993; *Abdul-Razzak et al.*, 1998; *Nenes and Seinfeld*, 2003]. The corresponding expressions for the relative dispersion can be similarly obtained by applying these more advanced expressions to equation (11). Such extensions seem necessary considering that the exponent of k in CCN spectra is not a constant, but usually decreases with increasing supersaturation [*Hudson*, 1984; *Ji and Shaw*, 1998; *Yum and Hudson*, 2001]. Because of their close relationship, it is desirable to address these issues together.

[11] It is also noteworthy that the larger relative dispersion for polluted clouds seems to run counter to the conventional wisdom of the cloud physics community that polluted clouds have narrower spectra. However, the conventional wisdom has been often based on some measure of absolute spectral dispersion (e.g., standard deviation) of the cloud droplet size distribution, instead of the relative dispersion. A larger relative dispersion for polluted clouds is due in part to the corresponding smaller mean radius.

[12] **Acknowledgments.** Y. Liu and P. H. Daum were supported by the DOE Atmospheric Radiation Measurements Program and Atmospheric Sciences Program. S. S. Yum was supported by KOSEF grant R14-2002-031-01000-0. The authors thank Y. Peng at Max Planck Institute of

Meteorology and the anonymous reviewers for their insightful comments. This manuscript has been authored by Brookhaven Science Associates, LLC under contract DE-AC02-98CH1-886 with the U. S. Department of Energy. The U. S. Government retains, and the publisher, by accepting the article for publications, acknowledges, a world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for the U. S. Government purposes.

References

Abdul-Razzak, H., S. J. Ghan, and C. Rivera-Carpio (1998), A parameterization of cloud droplet nucleation, I, Single aerosol type, *J. Geophys. Res.*, *103*, 6123–6131.

Albrecht, B. A. (1989), Aerosols, cloud microphysics, and fractional cloudiness, *Science*, *245*, 1227–1230.

Cooper, W. A. (1989), Effects of variable droplet growth histories on droplet size distributions. Part I: Theory, *J. Atmos. Sci.*, *46*, 1301–1311.

Feingold, G., and P. Y. Chuang (2002), Analysis of the influence of film-forming compounds on droplet growth: Implications for cloud microphysical processes and climate, *J. Atmos. Sci.*, *59*, 2006–2018.

Ghan, S. J., C. C. Chaung, and J. E. Penner (1993), A parameterization of cloud droplet nucleation, I, Single aerosol type, *Atmos. Res.*, *30*, 197–221.

Hudson, J. G. (1984), Cloud condensation nuclei measurements within clouds, *J. Clim. Appl. Meteorol.*, *23*, 42–51.

Hudson, J. G., and S. S. Yum (1997), Droplet spectral broadening in marine stratus, *J. Atmos. Sci.*, *54*, 2642–2654.

Ji, Q., and G. E. Shaw (1998), On the supersaturation spectrum and size distribution of cloud condensation nuclei, *Geophys. Res. Lett.*, *25*, 1903–1906.

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

Martin, G. M., D. W. Johnson, and A. Spice (1994), The measurement and parameterization of effective radius of droplets in warm stratocumulus clouds, *J. Atmos. Sci.*, *51*, 1823–1842.

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

Nenes, A., and J. H. Seinfeld (2003), Parameterization of cloud droplet formation in global climate models, *J. Geophys. Res.*, *108*(D14), 4415, doi:10.1029/2002JD002911.

Peng, Y., and U. Lohmann (2003), Sensitivity study of the spectral dispersion of the cloud droplet size distribution on the indirect aerosol effect, *Geophys. Res. Lett.*, *30*(10), 1507, doi:10.1029/2003GL017192.

Pruppacher, H. R., and J. D. Klett (1997), *Microphysics of Clouds and Precipitation*, 954 pp., Springer, New York.

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

Squires, P. (1958), The microstructure and colloidal stability of warm clouds, *Tellus*, *10*, 256–271.

Twomey, S. (1959), The nuclei of natural cloud formation. II: The supersaturation in natural clouds and the variation of cloud droplet concentration, *Geofis. Pura Appl.*, *43*, 243–249.

Twomey, S. (1977), The influence of pollution on the shortwave albedo of clouds, *J. Atmos. Sci*, *34*, 1149–1152.

Wood, R. (2002), How important is the spectral ripening effect in stratiform boundary layer clouds? Studies using simple trajectory analysis, *J. Atmos. Sci.*, *59*, 2681–2693.

Xue, H., and G. Feingold (2004), A modeling study of the effect of nitric acid on cloud properties, *J. Geophys. Res.*, *109*, D18204, doi:10.1029/2004JD004750.

Yum, S. S., and J. G. Hudson (2001), Vertical distributions of cloud condensation nuclei spectra over the springtime Arctic Ocean, *J. Geophys. Res.*, *106*, 15,045–15,052.

Yum, S. S., and J. G. Hudson (2005), Adiabatic predictions and observations of cloud droplet spectral broadness, *Atmos. Res.*, *73*, 203–223.

P. H. Daum and Y. Liu, Brookhaven National Laboratory, Building 815E, Upton, NY 11973-5000, USA. (lyg@bnl.gov)

S. S. Yum, Department of Atmospheric Sciences, Yonsei University, Seoul 120-749, South Korea.