



Theoretical expression for the autoconversion rate of the cloud droplet number concentration

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Received 16 April 2007; revised 21 June 2007; accepted 3 August 2007; published 29 August 2007.

[1] Accurate parameterization of the autoconversion rate of the cloud droplet concentration (number autoconversion rate in $\text{cm}^{-3} \text{s}^{-1}$) is critical for evaluating aerosol indirect effects using climate models; however, existing parameterizations are empirical at best. A theoretical expression is presented in this contribution that analytically relates the number autoconversion rate to the liquid water content, droplet concentration and relative dispersion of the cloud droplet size distribution. The analytical expression is theoretically derived by generalizing the analytical formulation previously developed for the autoconversion rate of the cloud liquid water content (mass autoconversion rate in $\text{g cm}^{-3} \text{s}^{-1}$). Further examination of the theoretical number and mass autoconversion rates reveals that the former is not linearly proportional to the latter as commonly assumed in existing parameterizations. The formulation forms a solid theoretical basis for developing multi-moment representation of the autoconversion process in atmospheric models in general. **Citation:** Liu, Y., P. H. Daum, R. L. McGraw, M. A. Miller, and S. Niu (2007), Theoretical expression for the autoconversion rate of the cloud droplet number concentration, *Geophys. Res. Lett.*, 34, L16821, doi:10.1029/2007GL030389.

1. Introduction

[2] Microphysical processes of clouds and precipitation occur on scales smaller than grid sizes of most atmospheric models such as climate models, and need to be accurately parameterized. One such process is autoconversion whereby cloud droplets grow into embryonic raindrops. Since the late 1960s, great effort has been devoted to developing and improving parameterization of the autoconversion rate of the liquid water content (mass autoconversion rate hereafter) [Berry, 1968; Kessler, 1969; Manton and Cotton, 1977; Liou and Ou, 1989; Baker, 1993; Liu and Daum, 2004; Liu et al., 2004, 2005, 2006a, 2006b].

[3] However, the autoconversion rate for the cloud droplet concentration (number autoconversion rate, hereafter) has received little attention. With growing recognition of the importance of droplet concentration and relative dispersion in cloud-related phenomena, along with advances in computer power, two-moment schemes for microphysical parameterizations that considers the mass and number autoconversion rates have found increasing applications [Beheng, 1994; Khairoutdinov and Kogan, 2000; Cohard and Pinty, 2000; Seifert and Beheng, 2001; Chen and Liu,

2004; Morrison et al., 2005; Zhang et al., 2007]. The pressing need for accurate parameterization of the number autoconversion rate has been reinforced by the increasing interest in cloud-climate interactions, and aerosol indirect effects [Boucher et al., 1995; Lohmann and Feichter, 2005; Rotstayn, 2000; Rotstayn and Liu, 2005].

[4] Virtually all existing parameterizations for the number autoconversion rate have essentially followed an earlier study by Berry and Reinhardt [1974], assuming that the number autoconversion rate is linearly proportional to the corresponding mass autoconversion rate, which itself is empirically obtained by curve-fitting numerical simulations from detailed microphysical models [Ziegler, 1985; Beheng, 1994; Khairoutdinov and Kogan, 2000; Seifert and Beheng, 2001]. Therefore, existing parameterizations for the number autoconversion rate suffer from all the deficiencies of simulation-based expressions for the mass autoconversion rate (see Liu and Daum [2004] and Liu et al. [2004, 2005, 2006a, 2006b] for details about the deficiencies), for example, lacking clear physics. It is desirable to have a theoretical expression for the number autoconversion rate derived from first principles. Furthermore, the linear proportionality between the number and mass autoconversion rates commonly assumed in existing parameterizations is questionable as well and warrants rigorous examination.

[5] In a series of publications [Liu and Daum, 2004; Liu et al., 2004, 2005, 2006a, 2006b], we have presented a theoretical formulation for the mass autoconversion rate. The primary objective of this contribution is to generalize the formulation for the mass autoconversion rate to derive an analytical expression for the number autoconversion rate. The secondary objective is to combine the theoretical number and mass autoconversion rates to examine the validity of the common assumption of the linear proportionality between these two rates.

2. Generalized Expression for Autoconversion Rate

[6] According to Liu et al. [2004, 2005, 2006b], the autoconversion rate for any bulk quantity Y can be generically written as

$$P_Y = P_{Y0} T_Y, \quad (1)$$

where P_Y is the autoconversion rate; P_{Y0} is the rate function describing the conversion rate after the onset of the autoconversion process, and $0 \leq T_Y \leq 1$ is the threshold function describing the transition behavior of the autoconversion process. The analytical expressions for P_{Y0} and T_Y will be derived below.

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2.1. General Expression for Rate Function

[7] Without loss of generality, we consider the quantity Y that is related to the δ -th power moment of the droplet size distribution such that

$$y = \alpha r^\delta, \quad (2a)$$

$$Y = \alpha \int r^\delta n(r) dr = \alpha N r_\delta^\delta, \quad (2b)$$

where r is the droplet radius, $n(r)$ is the droplet size distribution, N is the droplet concentration, α and δ are parameters indicative of the characteristics of Y and the order of the power moment, and r_δ is the δ -th mean radius of the droplet population. For example, the pair of $\alpha = (4/3\pi\rho_w)$ and $\delta = 3$ indicates that Y is the cloud liquid water content; the pair of $\alpha = 1$ and $\delta = 0$ indicates that Y represents the cloud droplet concentration N . Similar to the derivation of the mass rate function presented by *Liu and Daum* [2004], the rate function for Y is readily expressed as

$$P_{Y0} = \alpha \int n(r_1) dr_1 \int K(r_1, r_2) r_2^\delta n(r_2) dr_2, \quad (3)$$

where $r_{1,2}$ represent the radii of the collector and collected droplets, respectively, K is the collection kernel, and the integration is over all the droplets. Application to equation (3) of the Long collection kernel for $r_1 < 50 \mu\text{m}$, $K(r_1, r_2) = \kappa_2 r_1^6$, and subsequent integration yields

$$P_{Y0} = \alpha \kappa_2 N^2 r_6^6 r_\delta^\delta, \quad (4)$$

where the coefficient $\kappa_2 \approx 1.9 \times 10^{11}$ in $\text{cm}^{-3}\text{s}^{-1}$, r_1 is in cm, and the collection kernel K is in $\text{cm}^3 \text{s}^{-1}$ [Long, 1974]. Further application to equation (4) of the linear relationship between the mean radius of any order (r_p) and the mean volume radius (r_3), $r_p = \beta_p r_3$, leads to

$$P_{Y0} = \alpha \left(\frac{3}{4\pi\rho_w} \right)^{(6+\delta)/3} \kappa_2 \beta_6^6 \beta_\delta^\delta N^{-\delta/3} L^{(6+\delta)/3}, \quad (5)$$

where ρ_w is the water density, L is the liquid water content, and β_6 and β_δ are dimensionless parameters depending on the relative dispersion of the cloud droplet size distribution.

2.2. General Expression for Threshold Function

[8] As treated for the mass autoconversion rate [Liu et al., 2005, 2006b], the threshold function for Y is given by

$$T_Y = \frac{P_Y}{P_{Y0}} = \frac{\int_{r_c}^{\infty} r^6 n(r) dr}{\int_0^{\infty} r^6 n(r) dr} \frac{\int_{r_c}^{\infty} r^\delta n(r) dr}{\int_0^{\infty} r^\delta n(r) dr}, \quad (6)$$

where r_c is the critical radius beyond which the collection process starts to dominate the growth of cloud drops [Liu et al., 2004]. Further evaluation of equation (6) requires specifying the mathematical form of the cloud droplet size distribution. It has been shown that cloud droplet size distributions are well described by the general Weibull

droplet size distribution given by [Liu and Hallett, 1997; Liu and Daum, 2000]

$$n(r) = \frac{qN}{r_0^q} r^{q-1} \exp\left[-\left(\frac{r}{r_0}\right)^q\right], \quad (7a)$$

where the parameter q is related to the relative dispersion (ε) of the cloud droplet size distribution through

$$\varepsilon = \left[\frac{2q\Gamma(2/q)}{\Gamma^2(1/q)} - 1 \right]^{1/2} \approx q^{-1}. \quad (7b)$$

[9] Application of the general Weibull droplet size distribution to equation (6) leads to the following expressions describing the general threshold function:

$$T_Y = \gamma\left(\frac{6+q}{q}, x_{cq}\right) \gamma\left(\frac{\delta+q}{q}, x_{cq}\right), \quad (8a)$$

$$x_{cq} = \left(\frac{r_c}{r_0}\right)^q = \Gamma^{q/3} \left(\frac{3+q}{q}\right) x_c^{q/3}, \quad (8b)$$

$$x_c = 9.7 \times 10^{-17} N^{3/2} L^{-2}, \quad (8c)$$

where x_c is the ratio of the critical to mean masses, Γ and γ are the complete and incomplete gamma function, respectively (see Liu et al. [2004, 2005, 2006b] for more discussions about x_c). Combination of equations (5) and (8) yields the general expression for the autoconversion rate of Y :

$$P_Y = \alpha \left(\frac{3}{4\pi\rho_w} \right)^{(6+\delta)/3} \kappa_2 \gamma\left(\frac{6+q}{q}, x_{cq}\right) \gamma\left(\frac{\delta+q}{q}, x_{cq}\right) \beta_6^6 \beta_\delta^\delta N^{-\delta/3} L^{(6+\delta)/3} \quad (9)$$

3. Number Autoconversion Rate

3.1. Theoretical Expression

[10] Equations (5), (8) and (9) suggests that the rate function, threshold function, and the autoconversion rate of any moment Y can be expressed as functions of liquid water content, droplet concentration and relative dispersion. And the general expressions are reduced to those previously derived for the mass autoconversion rate when $\alpha = (4/3\pi\rho_w)$ and $\delta = 3$. The number autoconversion rate is readily obtained by applying of $\alpha = 1$ and $\delta = 0$ to the general expressions, i.e.,

$$P_{N0} = \left(\frac{3}{4\pi\rho_w} \right)^2 \kappa_2 \frac{\Gamma\left(\frac{6+q}{q}\right)}{\Gamma^2\left(\frac{3+q}{q}\right)} L^2, \quad (10a)$$

$$T_N = \gamma\left(\frac{6+q}{q}, x_{cq}\right) \gamma(1, x_{cq}), \quad (10b)$$

$$P_N = \left(\frac{3}{4\pi\rho_w} \right)^2 \kappa_2 \frac{\Gamma\left(\frac{6+q}{q}, x_{cq}\right) \Gamma(1, x_{cq})}{\Gamma^2\left(\frac{3+q}{q}\right)} L^2. \quad (10c)$$

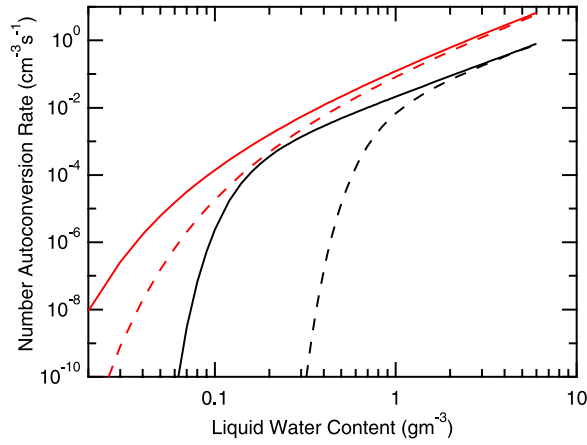


Figure 1. Dependence of the number autoconversion rate on liquid water content. The solid and dashed lines represent those for droplet concentration $N = 50$ and 500 cm^{-3} , respectively. The black and red colors represent those for relative dispersion $\varepsilon = 0.33$ ($q = 3$) and 1 ($q = 1$), respectively. Note that the solid and dashed lines overlap with each other when the liquid water content is sufficient high.

The derivation of the above equations uses the expression for β_p ,

$$\beta_p^p = \Gamma\left(\frac{p+q}{q}\right) \Gamma^{-2}\left(\frac{3+q}{q}\right)$$

3.2. Further Examination

[11] Equation (10c) coupled with equations (7b), (8b), and (8c) suggests that the number autoconversion rate depends on liquid water content, droplet concentration, and relative dispersion. Figure 1 illustrates the dependence of the number autoconversion rate on liquid water content calculated from equation (10c) at different values of droplet concentration and relative dispersion (solid and dashed curves for $N = 50 \text{ cm}^{-3}$ and $N = 500 \text{ cm}^{-3}$; black and red curves for $\varepsilon = 0.33$ ($q = 3$) and $\varepsilon = 1$ ($q = 1$)). Evidently, the number autoconversion rate generally increases with increasing liquid water content. The dependence can be characterized in two distinct regimes, which are dominated by the threshold function and rate function, respectively (threshold-dominated and rate-dominated hereafter). The number autoconversion rate increases faster in the threshold-dominated regime than that in the rate-dominated regime. A smaller relative dispersion (black curves) leads to a smaller number autoconversion rate in both regimes, but the threshold-dominated regime exhibits a steeper transition. The dependence of the number autoconversion rate on droplet concentration is more interesting. A smaller droplet concentration (dashed curves) gives rise to a larger number autoconversion rate in the threshold-dominated regime; but the dependence on droplet concentration diminishes in the rate-dominated regime where the curves for different droplet concentrations converge into a single curve. In short, except for its independence of droplet concentration in the rate-dominated regime, all the features of the number autoconversion rate are similar to those for the mass autoconversion

rate reported previously [Liu and Daum, 2004; Liu et al., 2004, 2005, 2006a, 2006b]. The feature that the number autoconversion rate should be described by two different functions is worth emphasizing, suggesting that existing parameterizations that have been often obtained by using a single function such as a power-law to fit detailed model results may distort the number autoconversion rate.

[12] Furthermore, existing parameterizations for the number autoconversion rate assume that the number autoconversion rate is linearly proportional to the mass autoconversion rate. This assumption of linear proportionality is equivalent to assuming that all new “drizzle” drops have the same radius r_* (typical drop radius hereafter [Beheng, 1994; Khairoutdinov and Kogan, 2000; Seifert and Beheng, 2001], i.e.,

$$P_N = \frac{3}{4\pi\rho_w r_*^3} P_L. \quad (11)$$

[13] Differences between different parameterizations lie in the differences in their parameterizations for mass autoconversion rate, and especially in their choices of different values assigned to the typical drop radius. For example, $r_* = 32, 25,$ and $40 \mu\text{m}$ were chosen by Beheng [1994], Khairoutdinov and Kogan [2000], and Seifert and Beheng [2001], respectively. Despite its widespread use, this linear proportionality assumption and the wide range of r_* values used by different authors remain unexamined.

[14] The new theoretical expression for number autoconversion rate, coupled to that for the mass autoconversion rate previously presented by Liu and Daum [2004] and Liu et al. [2004, 2005, 2006a, 2006b], allows a rigorous examination of this assumption of linear proportionality, or if r_* is a constant.

[15] By relating the theoretical number autoconversion rate (equation (10c)) to the mass autoconversion rate presented previously [Liu and Daum, 2004; Liu et al., 2006a], we obtain a theoretical expression for r_* ,

$$r_* = \left[\frac{\gamma\left(\frac{3+q}{q}, x_{cq}\right)}{\gamma(1, x_{cq})} \right]^{1/3} \tilde{r}_3 \quad (12)$$

[16] Figure 2 shows some results calculated from equation (12). It is clear from Figure 2 that instead of being a constant as commonly assumed in existing parameterizations, r_* varies substantially with droplet concentration, liquid water content, and relative dispersion. Furthermore, the dependency also features two distinct regimes: r_* first decreases with increasing mean volume radius, and then linearly increases with increasing mean volume radius beyond some point. Careful inspection of equations (10) and (12) indicates that the first and second regimes are dominated by the threshold function and rate function, respectively. The dependence of r_* on liquid water content, droplet concentration and relative dispersion may be the reason for the various values of r_* used in existing parameterizations.

[17] Two points are noteworthy. First, in the foreseeable future, the two-moment parameterization scheme that predicts the liquid water content and droplet concentration but fixes relative dispersion will occupy the center stage during

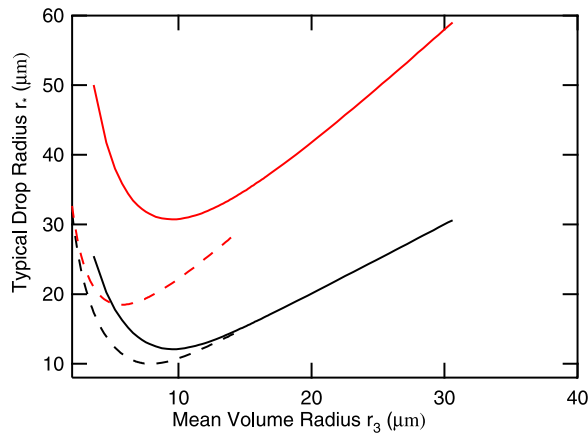


Figure 2. Same as Figure 1, except for the dependence of the typical drop radius r^* on the mean-volume radius r_3 . Note that the solid and dashed lines overlap with each other when the mean-volume radius is sufficiently large.

the transition from one-moment to multi-moment parameterization schemes, as reflected in the current modeling activities [e.g., Seifert *et al.*, 2006; Zhang *et al.*, 2007]. According to both theoretical [Liu *et al.*, 1995] and observational [Costa *et al.*, 2000] studies, we consider a typical droplet size distribution with $q = 3$ ($\varepsilon = 0.33$) for the purpose of the two-moment schemes. Under this condition, the theoretical expressions for number and mass autoconversion rates is simplified as

$$P_N = \left(\frac{3}{4\pi\rho_w}\right)^2 \kappa_2 (x_c^2 + 2x_c + 2) e^{-2x_c} L^2 \quad (13a)$$

$$P_L = \left(\frac{3}{4\pi\rho_w}\right)^2 \kappa_2 (x_c^2 + 2x_c + 2) (1 + x_c) e^{-2x_c} N^{-1} L^3. \quad (13b)$$

These two theoretical expressions should be readily applicable to two-moment schemes for parameterizing the autoconversion process in atmospheric models.

[18] Second, the number autoconversion rate discussed in this paper is referred to the loss rate of cloud droplets; a relevant quantity is the formation rate of embryonic raindrops caused by the autoconversion process. Under the approximation of binary collisions, it takes two cloud droplets to form one embryonic raindrop, and therefore the autoconversion-induced increasing rate of embryonic raindrops is half of the number autoconversion rate given here.

4. Concluding Remarks

[19] The analytical formulation previously derived for the mass autoconversion rate is first generalized to consider the rate of change of any moment of the cloud droplet size distribution caused by the autoconversion process. The general formulation is then applied to theoretically derive an analytical expression for the number autoconversion rate. It is shown that like the mass autoconversion rate, the number autoconversion rate depends on the liquid water content, droplet concentration and relative dispersion. The

dependency is characterized by two distinct regimes: one is dominated by the threshold function and the other by the rate function. A single function such as a power-law as often used in existing parameterizations cannot fully describe such two-function behaviors. It is also shown that the number autoconversion rate is not linearly proportional to the mass autoconversion rate as commonly assumed in existing parameterizations.

[20] It should be emphasized that although only the number autoconversion rate is examined in detail in this work, the extension to autoconversion rates for other quantities such as radar reflectivity is straightforward using the general formulation. It is interesting to examine the impact of replacing existing parameterizations with the theoretical one on model results. The result is useful for differentiating precipitating from non-precipitating clouds using remote sensing techniques as well, which will be addressed in another paper.

[21] **Acknowledgments.** Liu, Daum, McGraw, and Miller are supported by the Atmospheric Radiation Measurements Program and Atmospheric Sciences Program of the U.S. Department of Energy. Niu is supported by the Natural Science Foundation of China and Natural Science Key Projects of Jiangsu Universities. Discussions with Steve Ghan, Wei-Kuo Tao, Leon Rotstajn, and Yali Luo stimulated this work.

References

- Baker, M. B. (1993), Variability in concentrations of CCN in the marine cloud-top boundary layer, *Tellus, Ser. B.*, *45*, 458–472.
- Beheng, K. D. (1994), A parameterization of warm cloud microphysical conversion processes, *Atmos. Res.*, *33*, 193–206.
- Berry, E. X. (1968), Modification of the warm rain process, paper presented at 1st National Conference on Weather Modification, Am. Meteorol. Soc., Albany, N. Y.
- Berry, E. X., and R. L. Reinhardt (1974), An analysis of cloud drop growth by collection: part II. Single initial distributions, *J. Atmos. Sci.*, *31*, 1825–1831.
- Boucher, O., H. L. Treut, and M. B. Baker (1995), Precipitation and radiation modeling in a general circulation model: Introduction of cloud microphysical process, *J. Geophys. Res.*, *100*, 16,395–16,414.
- Chen, J., and S. Liu (2004), Physically based two-moment bulkwater parameterization for warm-cloud microphysics, *Q. J. R. Meteorol. Soc.*, *130*, 51–78.
- Cohard, J., and J. Pinty (2000), A comprehensive two-moment warm microphysical bulk scheme. I: Description and tests, *Q. J. R. Meteorol. Soc.*, *126*, 1815–1842.
- Costa, A., C. Oliveira, J. Oliveira, and A. Sampaio (2000), Microphysical observations of warm cumulus clouds in Ceara, Brazil, *Atmos. Res.*, *54*, 167–199.
- Kessler, E. (1969), On the distribution and continuity of water substance in atmospheric circulation, *Meteorol. Monogr.* *10*, 84 pp. Am. Meteorol. Soc., Boston, Mass.
- Khairoutdinov, M., and Y. Kogan (2000), A new cloud physics parameterization in a large-eddy simulation model of marine stratocumulus, *Mon. Weather Rev.*, *128*, 229–243.
- Liou, K. N., and S. C. Ou (1989), The role of cloud microphysical processes in climate: An assessment from a one-dimensional perspective, *J. Geophys. Res.*, *94*, 8599–8607.
- Liu, Y., and P. H. Daum (2000), Spectral dispersion of cloud droplet size distributions and the parameterization of cloud droplet effective radius, *Geophys. Res. Lett.*, *27*, 1903–1906.
- Liu, Y., and P. H. Daum (2004), Parameterization of the autoconversion process. part I: Analytical formulation of the Kessler-type parameterizations, *J. Atmos. Sci.*, *61*, 1539–1548.
- Liu, Y., and J. Hallett (1997), The “1/3” power-law between effective radius and liquid water content, *Q. J. R. Meteorol. Soc.*, *123*, 1789–1795.
- Liu, Y., L. You, W. Yang, and F. Liu (1995), On the size distribution of cloud droplets, *Atmos. Res.*, *35*, 201–216.
- 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.
- Liu, Y., P. H. Daum, and R. McGraw (2005), Size truncation effect, threshold behavior, and a new type of autoconversion parameterization, *Geophys. Res. Lett.*, *32*, L11811, doi:10.1029/2005GL022636.

- Liu, Y., P. H. Daum, and R. McGraw (2006a), Parameterization of the autoconversion process. part II: Generalization of Sundqvist-type parameterizations, *J. Atmos. Sci.*, *63*, 1103–1109.
- Liu, Y., P. H. Daum, R. McGraw, and M. Miller (2006b), Generalized threshold function accounting for effect of relative dispersion on threshold behavior of autoconversion process, *Geophys. Res. Lett.*, *33*, L11804, doi:10.1029/2005GL025500.
- Lohmann, U., and J. Feichter (2005), Global indirect aerosol effects: A review, *Atmos. Chem. Phys.*, *5*, 715–737.
- Long, A. B. (1974), Solutions to the droplet collection equation for polynomial kernels, *J. Atmos. Sci.*, *31*, 1040–1052.
- Manton, M. J., and W. R. Cotton (1977), Formulation of approximate equations for modeling moist deep convection on the mesoscale, *Atmos. Sci. Pap.* 266, Dep. of Atmos. Sci., Colo. State Univ., Fort Collins.
- Morrison, H., J. A. Curry, and V. I. Khvorostyanov (2005), A new double-moment microphysics parameterization for application in cloud and climate models. part I: Description, *J. Atmos. Sci.*, *62*, 1665–1677.
- Rotstajn, L. D. (2000), On the “tuning” of the autoconversion parameterizations in climate models, *J. Geophys. Res.*, *105*, 15,495–15,507.
- Rotstajn, L. D., and Y. Liu (2005), A smaller global estimate of the second indirect aerosol effect, *Geophys. Res. Lett.*, *32*, L05708, doi:10.1029/2004GL021922.
- Seifert, A., and K. D. Beheng (2001), A double-moment parameterization for simulating autoconversion, accretion and self-collection, *Atmos. Res.*, *59–60*, 265–281.
- Seifert, A., A. Khain, A. Pokrovsky, and K. D. Beheng (2006), A comparison of spectral bin and two-moment bulk mixed-phase cloud microphysics, *Atmos. Res.*, *80*, 46–66.
- Zhang, R., G. Li, J. Fan, D. L. Wu, and M. J. Molina (2007), Intensification of Pacific storm track linked to Asian pollution, *Proc. Natl. Acad. Sci. U.S.A.*, *104*, 5295–5299.
- Ziegler, C. L. (1985), Retrieval of thermal and microphysical variables in observed convective storms. part I: Model development and preliminary testing, *J. Atmos. Sci.*, *42*, 1487–1509.

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