

Analytical three-moment autoconversion parameterization based on generalized gamma distribution

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[1] Autoconversion of cloud droplets to embryonic raindrops is one of the most important microphysical processes in warm clouds. From first principles, a three-moment theoretical expression is analytically derived for the autoconversion rates of the number concentration, mass content, and radar reflectivity based on the generalized gamma distribution function for cloud droplet size distributions. Furthermore, the influence of the liquid water content L, droplet concentration N, shape parameter μ , and tail parameter q on the autoconversion rate are investigated, respectively. It is found that the autoconversion rate increases significantly with decreasing value μ , no matter how high or low the liquid water content is, but the parameter q only plays an important role at low liquid water content. These results may have many potential applications, especially to studies of the indirect aerosol effect and the influence of μ and q on cloud and precipitation.

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1. Introduction

- [2] With increasing computer power, accurate representation of microphysical processes becomes more and more important in various atmospheric models such as global climate models and cloud-resolving models. Autoconversion of cloud droplets to embryonic raindrops is a key microphysical process, which determines the onset of the precipitation associated with warm clouds. Over the last several decades, a series of parameterization schemes have been successively proposed and developed to describe the autoconversion process [Berry, 1968; Kessler, 1969; Berry and Reinhardt, 1974; Manton and Cotton, 1977; Sundqvist et al., 1989; Liou and Ou, 1989; Baker, 1993; Del Genio et al., 1996; Rotstayn, 1997], and most of them are successfully applied to atmospheric models of various scales.
- [3] As we know, all of the autoconversion parameterizations are one-moment schemes which describe the autoconversion rate of cloud liquid mass content. With increasing interest in the indirect aerosol effects, the multimoment microphysics parameterization for the autoconversion process has received extensive attention. A number of two-moment parameterization schemes have been addressed with great efforts which formulate predictive equations for both the mass content and the total number concentration of the hydrometeor categories [Zeigler, 1985; Ferrier, 1994;

Beheng, 1994; Meyers et al., 1997; Reisner et al., 1998; Cohard and Pinty, 2000; Khairoutdinov and Kogan, 2000; Seifert and Beheng, 2001, 2006; Chen and Liu, 2004; Milbrandt and Yau, 2005a; Morrison et al., 2005]. With predictive equations of mass content, total number concentration, and radar reflectivity, Milbrandt and Yau [2005b] recently proposed a three-moment parameterization scheme. Moreover, it was claimed that this three-moment version was successfully used to simulate a severe hailstorm with a mesoscale model, subsequently [Milbrandt and Yau, 2006a]. By comparing a series of sensitivity experiments using different one-moment, two-moment and three-moment versions, the three-moment scheme was found to be superior to other schemes in simulation of the maximum hail sizes [Milbrandt and Yau, 2006b].

[4] Recently, a useful analytical method for deriving autoconversion parameterization was proposed by applying the generalized mean value theorem for integrals to the general collection equation [Liu and Daum, 2004; Liu et al., 2005]. Moreover, the analytical formulation of two-moment scheme was derived for parameterizing the autoconversion process [Liu et al., 2007]. These autoconversion parameterizations have attracted increasing research interests and have been applied to global climate models and cloud resolving models [Guo et al., 2008; Li et al., 2008; Hsieh et al., 2009]. However, it should be noted that the cloud droplet size distribution in [Liu and Daum, 2004; Liu et al., 2005, 2007] is described in terms of the Weibull distribution function or the classical gamma distribution function. Actually, more and more researchers use the generalized gamma distribution function to describe the cloud droplet size distribution. The distribution function includes four parameters: the total droplet number concentration N, slope

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parameter λ , and two dispersion parameters μ and q. Cohard and Pinty [2000] has pointed out that this generalized gamma distribution function best describes the observed distribution of cloud droplets. In Seifert et al. [2006], the generalized gamma distribution has also been used to describe the various particle spectra in bulk mixed-phase cloud microphysics. In the present paper, using the generalized gamma distribution function to describe the cloud droplet size distribution, we analytically derive a three-moment theoretical expression for the autoconversion parameterization of the total number concentration, mass content and radar reflectivity. Starting from the obtained results, we further discuss the influence of the liquid water content L, droplet concentration N, shape parameter μ , and tail parameter q on the autoconversion rate.

[5] The outline of the paper is as follows. In section 2 we present the general theoretical expression for the autoconversion rate including the rate function and threshold function, followed in section 3 by giving a three-moment parameterization of the autoconversion rate. Section 4 discusses the influence of the liquid water content L, droplet concentration N, shape parameter μ , and tail parameter q on the autoconversion rate. Finally, a summary and conclusion are given in section 5.

2. Theoretical Expression for Autoconversion Parameterization

[6] One of the most general analytical distribution functions used to describe size distributions of cloud droplets is the so-called four-parameter generalized gamma distribution [Cohard and Pinty, 2000; Seifert et al., 2006]:

$$n(r) = \frac{Nq\lambda^{\frac{\mu+1}{q}}}{\Gamma\left(\frac{\mu+1}{q}\right)} r^{\mu} \exp(-\lambda r^{q}), \tag{1}$$

where r is the radius of a cloud droplet. n(r) is the total number concentration per unit volume of droplet radius r, N is the total droplet number concentration, λ is the slope parameter, μ and q are dispersion parameters. It is noted that μ is the shape parameter which represents the shape of the whole distribution while q is the tail parameter mainly determining the distribution of larger droplets. Γ is the classical gamma function. The relative dispersion of the size distribution ε (ratio of the standard derivation to the mean radius) is the function of μ and q, and decreases with an increase in μ or q (Appendix A). By setting q = 1, the distribution function reduces to the classical gamma distribution function. Satisfying the relation $\mu = q - 1$, it becomes the Weibull distribution function. These two special distribution functions are widely used to describe the cloud droplet size distributions. Without loss of generality, we will use the generalized gamma distribution function to derive the theoretical expression for autoconversion parameterization.

[7] All the autoconversion parameterizations that have been developed so far can be generically written as [*Liu and Daum*, 2004; *Liu et al.*, 2005, 2007]

$$P_Y = P_{Y0}T_Y, \tag{2}$$

where P_Y is the autoconversion rate for any bulk quantity Y, P_{Y0} is the rate function describing the conversion rate after the onset of the autoconversion process, and T_Y is the threshold function describing the transition behavior of the autoconversion process.

[8] The generalized gamma distribution function in equation (1) can be integrated analytically over the entire cloud droplet size domain. This property is especially useful in obtaining any moment of the size distribution. Applying the generalized mean value theorem for integrals, the quantity *Y* that is related to the *p*th power moment of the droplet size distribution is given by

$$Y = \alpha \int r^p n(r) dr = \alpha N r_p^p, \tag{3}$$

where α is the parameter indicative of the characteristics of Y, p is the order of the power moment, and r_p is the pth mean radius of the cloud droplet population (Appendix B). For example, the pair of $\alpha=1$ and p=0 indicates that Y represents the cloud droplet concentration; the pair of $\alpha=(\frac{4}{3}\pi\rho_w)$ and p=3 indicates that Y is the cloud liquid water content; the pair of $\alpha=64$ and p=6 indicates that Y is the radar reflectivity.

[9] The rate function for the quantity *Y* is expressed by *Liu et al.* [2007]

$$P_{Y0} = \alpha \int n(R)dR \int K(R,r)r^p n(r)dr, \qquad (4)$$

where R is the radius of the collector droplet, while r is the radius of the collected droplet. K(R, r) is the collection kernel, and the integration is over all the droplets. Long (1974) showed that for $R < 50~\mu\mathrm{m}$, the collection kernel can be well approximated that by

$$K(R,r) = k_2 R^6, (5)$$

where the coefficient $k_2 \approx 1.9 \times 10^{17}$ is in m⁻³s⁻¹. Substitution of equation (5) into the rate function (4), we have the following expression

$$P_{Y0} = \alpha k_2 N^2 r_6^6 r_p^p, (6)$$

and we assume a general linear relation between the mean volume radius r_3 and the mean radius of any order r_p , $r_p = \beta_p r_3$ (Appendix B). It is readily shown that

$$P_{Y0} = \alpha \left(\frac{3}{4\pi\rho_w}\right)^{(6+p)/3} k_2 \beta_6^6 \beta_p^p N^{-p/3} L^{(6+p)/3},\tag{7}$$

where

$$\beta_{6}^{6} = \frac{\Gamma\left(\frac{\mu+1}{q}\right)\Gamma\left(\frac{\mu+7}{q}\right)}{\Gamma^{2}\left(\frac{\mu+4}{q}\right)},$$

$$\beta_{p}^{p} = \left[\frac{\Gamma\left(\frac{u+p+1}{q}\right)}{\Gamma\left(\frac{u+1}{q}\right)}\right]\left[\frac{\Gamma\left(\frac{u+4}{q}\right)}{\Gamma\left(\frac{u+1}{q}\right)}\right]^{-p/3}.$$
(8)

According to the above expression, then we have the following expression of the rate function,

$$P_{Y0} = \alpha \left(\frac{3}{4\pi\rho_{w}}\right)^{(6+p)/3} \cdot k_{2} \frac{\Gamma^{p/3}\left(\frac{\mu+1}{q}\right)\Gamma\left(\frac{\mu+7}{q}\right)\Gamma\left(\frac{\mu+p+1}{q}\right)}{\Gamma^{2+p/3}\left(\frac{\mu+4}{q}\right)} N^{-p/3}L^{(6+p)/3},$$
(9)

where ρ_w is the water density, and L is the cloud liquid water content.

[10] Consider the effect of truncating the cloud droplet size distribution on the autoconversion rate [$Liu\ et\ al.$, 2005], the threshold function for the bulk quantity Y is given by

$$T_{Y} = \frac{P_{Y}}{P_{Y0}} = \left[\frac{\int_{r_{c}}^{\infty} r^{6} n(r) dr}{\int_{0}^{\infty} r^{6} n(r) dr} \right] \left[\frac{\int_{r_{c}}^{\infty} r^{p} n(r) dr}{\int_{0}^{\infty} r^{p} n(r) dr} \right], \tag{10}$$

where r_c is the critical radius beyond which the autoconversion process become dominant, corresponds to the kinetic potential barrier of the droplet population and is a function of the number concentration and the mass content [Liu et al., 2004].

[11] Application of the generalized gamma distribution function to equation (10) leads to the following expression describing the threshold function of Y

$$T_Y = \gamma \left(\frac{\mu + 7}{q}, x_{cq}\right) \gamma \left(\frac{\mu + p + 1}{q}, x_{cq}\right),\tag{11}$$

$$x_{cq} = \lambda r_c^q = \left[\frac{\Gamma\left(\frac{\mu+4}{q}\right)}{\Gamma\left(\frac{\mu+1}{q}\right)} \right]^{q/3} x_c^{q/3}, \tag{12}$$

$$x_c = 9.7 \times 10^{-20} N^{3/2} L^{-2},$$
 (13)

where $\Gamma(n, a) = \int_a^\infty x^{n-1} e^{-x} dx$ is the incomplete gamma function; $\gamma(n, a) = \int_a^\infty x^{n-1} e^{-x} dx / \int_a^\infty x^{n-1} e^{-x} dx$ is the normalized incomplete gamma function; x_c is the ratio of the critical to mean masses and the expression of x_{cq} is given in Appendix C. From equations (9) and (11), the theoretical expression for autoconversion rate of the bulk quantity Y is given by

$$P_{Y} = \alpha \left(\frac{3}{4\pi\rho_{w}}\right)^{(6+p)/3} \\ \cdot k_{2} \frac{\Gamma^{p/3}\left(\frac{\mu+1}{q}\right)\Gamma\left(\frac{\mu+7}{q}, x_{cq}\right)\Gamma\left(\frac{\mu+p+1}{q}, x_{cq}\right)}{\Gamma^{2+p/3}\left(\frac{\mu+4}{q}\right)} \\ \cdot N^{-p/3}L^{(6+p)/3}. \tag{14}$$

3. Analytical Three-Moment Expression for Autoconversion Rate

3.1. Autoconversion Rate of Number Concentration

[12] It is shown that the rate function P_{Y0} , the threshold function T_Y and the autoconversion rate P_Y of any moment Y

can be expressed as functions of liquid water content L, number concentration N and dispersion parameters μ , q of cloud droplets from equations (9), (11), and (14). To derive the autoconversion rate of number concentration, we apply $\alpha=1$ and p=0 to the general expressions of the rate function, threshold function, and autoconversion rate, and then they can be simplified as

$$P_{N0} = \left(\frac{3}{4\pi\rho_w}\right)^2 k_2 \frac{\Gamma\left(\frac{\mu+1}{q}\right)\Gamma\left(\frac{\mu+7}{q}\right)}{\Gamma^2\left(\frac{\mu+4}{q}\right)} L^2, \tag{15}$$

$$T_N = \gamma \left(\frac{\mu+1}{q}, x_{cq}\right) \gamma \left(\frac{\mu+7}{q}, x_{cq}\right), \tag{16}$$

$$P_{N} = \left(\frac{3}{4\pi\rho_{w}}\right)^{2} k_{2} \frac{\Gamma\left(\frac{\mu+1}{q}, x_{cq}\right) \Gamma\left(\frac{\mu+7}{q}, x_{cq}\right)}{\Gamma^{2}\left(\frac{\mu+4}{q}\right)} L^{2}, \quad (17)$$

with the expression $x_{cq} = \left[\frac{\Gamma\left(\frac{\mu+4}{q}\right)}{\Gamma\left(\frac{\mu+1}{q}\right)}\right]^{q/3} x_c^{q/3}$.

3.2. Autoconversion Rate of Mass Content

[13] The general expressions are reduced to those previously derived for the mass autoconversion rate when $\alpha = (\frac{4}{3}\pi\rho_w)$ and p=3. The theoretical expression for mass content is readily obtained

$$P_{L0} = \left(\frac{3}{4\pi\rho_w}\right)^2 k_2 \frac{\Gamma\left(\frac{\mu+1}{q}\right)\Gamma\left(\frac{\mu+7}{q}\right)}{\Gamma^2\left(\frac{\mu+4}{q}\right)} N^{-1} L^3, \tag{18}$$

$$T_L = \gamma \left(\frac{\mu + 4}{a}, x_{cq}\right) \gamma \left(\frac{\mu + 7}{a}, x_{cq}\right),\tag{19}$$

$$P_{L} = \left(\frac{3}{4\pi\rho_{w}}\right)^{2} k_{2} \frac{\Gamma\left(\frac{\mu+1}{q}\right) \Gamma\left(\frac{\mu+4}{q}, x_{cq}\right) \Gamma\left(\frac{\mu+7}{q}, x_{cq}\right)}{\Gamma^{3}\left(\frac{\mu+4}{q}\right)} N^{-1} L^{3}.$$
(20)

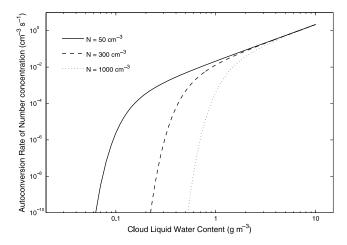
3.3. Autoconversion Rate of Radar Reflectivity

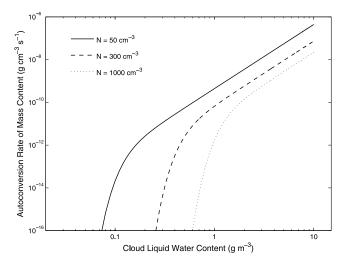
[14] For p = 6, and $\alpha = 64$, the expressions of the rate function, the threshold function, and the autoconversion rate for radar reflectivity are given by

$$P_{Z0} = 64 \left(\frac{3}{4\pi\rho_w}\right)^4 k_2 \frac{\Gamma^2 \left(\frac{\mu+1}{q}\right) \Gamma^2 \left(\frac{\mu+7}{q}\right)}{\Gamma^4 \left(\frac{\mu+4}{q}\right)} N^{-2} L^4, \quad (21)$$

$$T_Z = \left[\gamma\left(\frac{\mu+7}{q}, x_{cq}\right)\right]^2,\tag{22}$$

$$P_{Z} = 4 \left(\frac{3}{2\pi\rho_{w}}\right)^{4} k_{2} \frac{\Gamma^{2}\left(\frac{\mu+1}{q}\right) \Gamma^{2}\left(\frac{\mu+7}{q}, x_{cq}\right)}{\Gamma^{4}\left(\frac{\mu+4}{q}\right)} N^{-2} L^{4}. \quad (23)$$





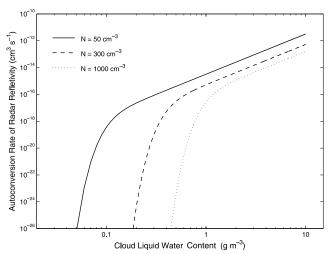


Figure 1. Dependence of the autoconversion rate of (top) number concentration, (middle) mass content, and (bottom) radar reflectivity on liquid water content. The dispersion parameters are set to $\mu = 2$ and q = 3.

These three-moment theoretical expressions of the rate functions, threshold functions and autoconversion rates have been given. Then we can derive the autoconversion rate of three-moment for cloud droplets (small drops) and raindrops (large drops),

$$2\frac{dN_r}{dt} = -\frac{dN_c}{dt} = P_N,$$

$$\frac{dL_r}{dt} = -\frac{dL_c}{dt} = P_L,$$

$$a\frac{dZ_r}{dt} = -\frac{dZ_c}{dt} = P_Z,$$
(24)

the indices c and r stand for cloud droplets and raindrops, the coefficient a has the range $0.5 \le a < 1$ (when the two cloud droplets through collision/coalescence have the same radius, the coefficient becomes the minimum value). It should be emphasized that all the quantities are in SI units including P_N (m⁻³s⁻¹), P_L (kg m⁻³ s⁻¹), and P_Z (m³ s⁻¹). To have further applications in a bulk scheme, it is noteworthy that the dispersion parameters μ and q have the range $\mu \ge 0$ and q > 0, respectively; only one of them is a free parameter in three-moment parameterization because of the constraint of the added moment-radar reflectivity. These theoretical expressions should be readily applicable to multimoment bulk microphysical schemes.

[15] Noteworthily, a typical cloud droplet size distribution of the Weibull distribution function with q=3 is considered. Under this condition, the theoretical expressions of autoconversion rates for the number concentration, mass content, and radar reflectivity are simplified as

$$P_N = \left(\frac{3}{4\pi\rho_w}\right)^2 k_2 (x_c^2 + 2x_c + 2)e^{-2x_c}L^2, \tag{25}$$

$$P_L = \left(\frac{3}{4\pi\rho_w}\right)^2 k_2 \left(x_c^2 + 2x_c + 2\right) (x_c + 1)e^{-2x_c} N^{-1} L^3, \qquad (26)$$

$$P_Z = 4\left(\frac{3}{2\pi\rho_w}\right)^4 k_2 \left(x_c^2 + 2x_c + 2\right)^2 e^{-2x_c} N^{-2} L^4.$$
 (27)

It should be pointed out that the formulas (25) and (26) are just the two-moment theoretical expression by $Liu\ et\ al.$ [2007]. In the next section, we will discuss the influence of the liquid water content L, droplet concentration N, shape parameter μ , and tail parameter q on the autoconversion rate.

4. Discussions

[16] According to equations (17), (20), and (23), these three-moment autoconversion rates are all dependent on the liquid water content L, droplet concentration N, shape parameter μ , and tail parameter q. It is necessary to discuss the influence of L, N, μ , and q on the autoconversion rate, respectively. Figure 1 shows the autoconversion rates of the number concentration, mass content, and radar reflectivity are functions of L for different values of N ($N = 50 \text{ cm}^{-3}$, $N = 300 \text{ cm}^{-3}$, and $N = 1000 \text{ cm}^{-3}$) at $\mu = 2$ and q = 3. It is evident

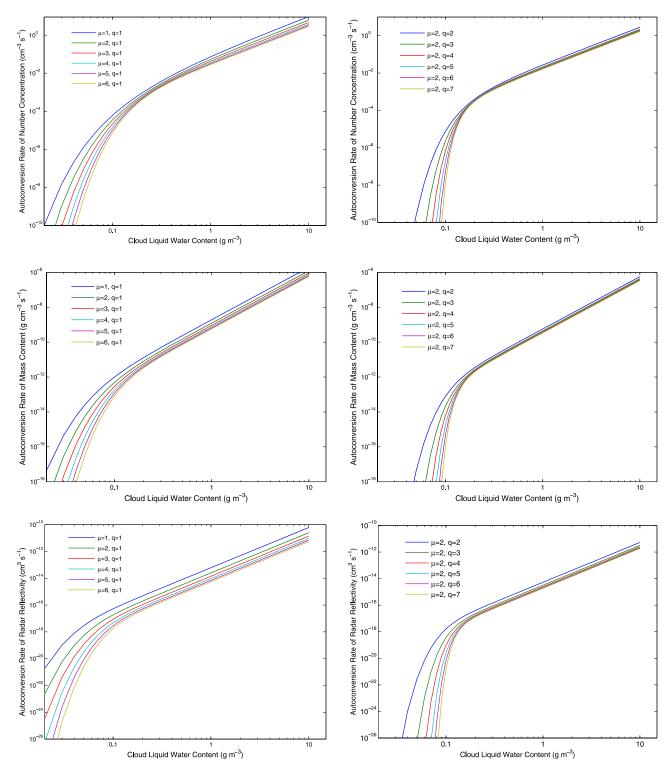


Figure 2. Dependence of the autoconversion rate of (top) number concentration, (middle) mass content, and (bottom) radar reflectivity on liquid water content (left) at different μ values and (right) at different q values. Note that the droplet concentration N is for 50 cm⁻³ in all the six panels.

that the three-moment autoconversion rates generally increase with increasing liquid water content L. The dependence of the autoconversion rates on N illustrates that the higher droplet concentration results in a lower autoconversion rate at given liquid water content. In fact, it can be comprehended that the smaller droplets resulting from higher

number concentration (given liquid water content) reduce the collision and coalescence efficiency by increasing colloidal stability. It is known that the droplet concentration is changed by anthropogenic aerosol emissions, hence these autoconversion expressions related to droplet concentration can be used to study the indirect aerosol effect. Additionally, for the

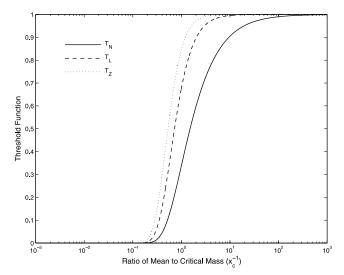


Figure 3. Comparison with the threshold functions of number concentration, mass content, and radar reflectivity. The dispersion parameters are at $\mu = 2$ and q = 3.

autoconversion rate of number concentration, we can see that the three curves from different droplet concentration converge together at a higher liquid water content in the first panel. This phenomena has also been reported previously by *Liu et al.* [2007].

[17] Figure 2 shows the three-moment autoconversion rates as a function of the liquid water content L for different values μ and q. The droplet concentration N is set to 50 cm^{-3} in all the panels. We can see that the three-moment autoconversion rates become lower with the increased parameters μ or q (implying the decrease in the relative dispersion). It can be understood that the smaller relative dispersion makes collision and coalescence processes less efficient. The left three panels show that the autoconversion rate significantly increases with decreasing value μ , no matter how high or low the liquid water content is. As for the influence of tail parameter q in the right three panels, the autoconversion rate only changes obviously at low liquid water content. These results indicate that the higher concentration of larger droplets resulting from a smaller q enhance the efficiency of collision and coalescence processes substantially at low liquid water content, which may be helpful to understand the effect of giant aerosol. From the preceding discussion, we can draw that the parameters μ and q have different influence on the autoconversion rate. It is noteworthy that the influence of μ and q cannot be distinguished for the autoconversion rate of the classical gamma distribution and the Weibull distribution, owing to exiting constraint q = 1 or $\mu = q - 1$.

[18] Threshold functions are of importance to autoconversion process, which considerably affect climatically important properties: cloud fraction, liquid water path, and aerosol indirect effect [*Guo et al.*, 2008]. Based on our three-moment parameterization, the threshold functions of the number concentration, mass content, and radar reflectivity have been given according to equations (16), (19), and (22), respectively. We present the three threshold functions at $\mu = 2$ and q = 3 in

Figure 3. It shows that these threshold functions are distinctly different. However, in existing multimoment parameterization schemes, the threshold functions of any moment are identical; such as, the Heaviside function in Kessler-type parameterizations and the exponential function in Sundqvist-type parameterizations. Since the number concentration, mass content, and radar reflectivity don't always have the linear relation, it is more physically reasonable to prescribe the threshold functions as different forms.

5. Conclusions

[19] Based on the generalized gamma distribution function of cloud droplet size distributions, we analytically derived a three-moment parameterization for autoconversion rates of the number concentration, mass content, and radar reflectivity from first principles. Furthermore, we discuss the influence of the liquid water content L, droplet concentration N, shape parameter μ , and tail parameter q on the autoconversion rate, respectively. We find that the dispersion parameters μ and q affect autoconversion rate differently: the autoconversion rate significantly increases with decreasing value μ , no matter how high or low the liquid water content is; but the parameter q only plays an important role at low liquid water content.

[20] Since the multimoment autoconversion parameterization in this work is analytically derived from theoretical studies, it would be interesting to examine the impact of replacing existing empirical parameterizations with these theoretical expressions on modeling studies of aerosol-cloud-precipitation interactions.

Appendix A: Relative Dispersion of Generalized Gamma Distribution

[21] The four-parameter generalized gamma distribution function is given by:

$$n(r) = \frac{Nq\lambda^{\frac{\mu+1}{q}}}{\Gamma(\frac{\mu+1}{q})} r^{\mu} \exp(-\lambda r^{q}). \tag{A1}$$

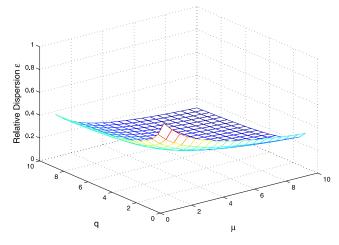


Figure 4. The relative dispersion ε is connected with the parameters μ and q.

[22] The relative dispersion of the size distribution ε is defined as the ratio of the standard deviation and the mean radius of the droplet size distribution,

$$\varepsilon = \frac{\sigma}{\overline{r}} = \frac{\sqrt{\int (r - \overline{r})^2 n(r) dr}}{N\overline{r}}.$$
 (A2)

[23] From the generalized gamma distribution (A1), the corresponding relative dispersion can be expressed as

$$\varepsilon = \frac{\left[\Gamma\left(\frac{1+\mu}{q}\right)\Gamma\left(\frac{3+\mu}{q}\right) - \Gamma\left(\frac{2+\mu}{q}\right)^2\right]^{1/2}}{\Gamma\left(\frac{2+\mu}{q}\right)}.$$
 (A3)

Hence one knows the relative dispersion is the function of the parameters μ and q, and it decreases with an increase in μ or q from Figure 4.

Appendix B: p-th Mean Radius r_p and Expressions for β_p and β_p^p

[24] The pth moment of the cloud droplet size distribution M_p is defined as

$$M_{p} = \frac{\int r^{p} n(r) dr}{N} = \left(\frac{1}{\lambda}\right)^{p/q} \frac{\Gamma\left(\frac{\mu + p + 1}{q}\right)}{\Gamma\left(\frac{\mu + 1}{q}\right)}, \quad (B1)$$

where N is the total droplet number concentration, $p = 0, 1, 2, 3, \dots$, and n(r) is the generalized gamma distribution function (A1). The pth mean radius of the cloud droplet population is defined as

$$r_p = M_p^{1/p} = \left(\frac{1}{\lambda}\right)^{1/q} \left[\frac{\Gamma\left(\frac{\mu+p+1}{q}\right)}{\Gamma\left(\frac{\mu+1}{q}\right)} \right]^{1/p}.$$
 (B2)

Therefore we derive

$$\beta_{p} = \frac{r_{p}}{r_{3}} = \left[\frac{\Gamma\left(\frac{\mu+p+1}{q}\right)}{\Gamma\left(\frac{\mu+1}{q}\right)} \right]^{1/p} \left[\frac{\Gamma\left(\frac{\mu+4}{q}\right)}{\Gamma\left(\frac{\mu+1}{q}\right)} \right]^{-1/3}, \quad (B3)$$

and for β_p^p

$$\beta_p^p = \left[\frac{\Gamma\left(\frac{\mu+p+1}{q}\right)}{\Gamma\left(\frac{\mu+1}{q}\right)} \right] \left[\frac{\Gamma\left(\frac{\mu+4}{q}\right)}{\Gamma\left(\frac{\mu+1}{q}\right)} \right]^{-p/3}, \tag{B4}$$

$$\beta_0^0 = \beta_3^3 = 1, \tag{B5}$$

$$\beta_6^6 = \frac{\Gamma\left(\frac{\mu+7}{q}\right)\Gamma\left(\frac{\mu+1}{q}\right)}{\Gamma^2\left(\frac{\mu+4}{q}\right)}.$$
 (B6)

It is noteworthy that, for the classical gamma distribution function and the Weibull distribution function, β_p and β_p^p are directly derived from the above general expressions.

Appendix C: Expression for x_{cq} Related With x_c

[25] For the generalized gamma distribution function of cloud droplet size distributions, the mean mass of cloud droplets is given by

$$\overline{m} = \frac{m_{total}}{N} = \frac{\frac{4}{3}\pi\rho_w \int_0^\infty r^3 n(r)dr}{\int_0^\infty n(r)dr} = \frac{4}{3}\pi\rho_w \left(\frac{1}{\lambda}\right)^{3/q} \frac{\Gamma\left(\frac{\mu+4}{q}\right)}{\Gamma\left(\frac{\mu+1}{q}\right)},\tag{C1}$$

and the critical mass is

$$m_c = \frac{4}{3}\pi \rho_w r_c^3,\tag{C2}$$

where r_c is the critical radius beyond which the autoconversion process become dominant. So the ratio of the critical to mean masses x_c is derived as

$$x_c = \frac{m_c}{\overline{m}} = \lambda^{3/q} \frac{\Gamma\left(\frac{\mu+1}{q}\right)}{\Gamma\left(\frac{\mu+4}{q}\right)} r_c^3.$$
 (C3)

From the equation (C3), we have

$$r_c = \left(\frac{1}{\lambda}\right)^{1/q} \left[\frac{\Gamma\left(\frac{\mu+4}{q}\right)}{\Gamma\left(\frac{\mu+1}{q}\right)}\right]^{1/3} x_c^{1/3}.$$
 (C4)

Hence the expression for x_{cq} is given by

$$x_{cq} = \lambda r_c^q = \left[\frac{\Gamma\left(\frac{\mu+4}{q}\right)}{\Gamma\left(\frac{\mu+1}{q}\right)} \right]^{q/3} x_c^{q/3}.$$
 (C5)

For the classical gamma distribution function q = 1, the expression of x_{cq} becomes

$$x_{ca} = \lambda r_c = [(\mu + 1)(\mu + 2)(\mu + 3)]^{1/3} x_c^{1/3}.$$
 (C6)

For the Weibull distribution function $\mu = q - 1$, it can be written as

$$x_{cq} = \lambda r_c^q = \Gamma^{q/3} \left(\frac{q+3}{q} \right) x_c^{q/3}. \tag{C7}$$

Note that when we choose the Weibull distribution function with q = 3, it is shown that

$$x_{cq} = \lambda r_c^3 = x_c, \tag{C8}$$

which is necessary to derive the theoretical expressions for the three-moment parameterization (25), (26), and (27). [26] Acknowledgments. We thank the three anonymous reviewers for their constructive comments that help greatly improve the earlier manuscript. The first author is also thankful to Yangang Liu for many helpful discussions. This work was supported by the Special Foundation for China Nonprofit Industry (GYHY200706036) and the NSFC National Excellent Young Scientists Fund (40825008) and State Key Laboratory of Loess and Quaternary Geology (LQ0701).

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