Relating Aerosol Properties to Cloud Condensation Nuclei Spectra and Cloud Properties

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Introduction

The indirect aerosol effect is probably the most uncertain among known climate forcings, and much of the uncertainty arises from poor understanding and quantification of the relationship of relevant aerosol properties to cloud condensation nuclei (CCN) and further to cloud properties of climate importance. Many studies have been devoted to parameterizing the cloud droplet number concentration (N_c) as power-law functions of CCN concentrations (Twomey 1959), the aerosol number concentration (N_a) (Chuang et al. 2000) or aerosol mass loading (m_a) (Lowenthal et al. 2004). The various power-law relationships have been widely used in relevant studies because of their simplicity.

Some studies have demonstrated the need for parameterizing the CCN spectrum and hence N_c as functions of N_a, M_a and the spectral shape of the pre-cloud aerosol size distributions by assuming lognormal aerosol size distributions (Ghan et al. 1995; Cohard et al. 2000). The importance of accounting for the spectral shape of the aerosol size distribution in representing droplet activation in climate models is reinforced by a recent finding that anthropogenic aerosols also enhance the relative dispersion of the cloud droplet size distribution (ε_c), and that the enhanced ε_c leads to warming effect on climate (Liu and Daum 2002; Peng and Lohmann 2003; Rostatyan and Liu 2003). The value of ε_c has been shown to depend on N_a, M_a, as well as the relative dispersion of the aerosol size distribution (ε_α) (Liu et al. 2006). However, studies on the effects of the relationship between N_a, M_a, and ε_a on the parameterization of droplet activation are still in their infancy; especially, power-law formulation along this line is still lacking.

This work revisits the power-law formulation because of its simplicity and widespread use. Previous results are extended by expressing the CCN spectrum and N_c as power-law functions of N_a, m_a and ε_a. It is shown that the relationships between N_a, m_a, and ε_a are critically important for representing N_c and evaluating indirect aerosol effects.

Power-Law CCN Spectrum and Relationship to Aerosol Size Distribution Moments

The CCN spectrum can be derived from the Kohler theory, and a power-law aerosol size distribution leads to a power-law CCN spectrum (Twomey 1977; Jiusto and Lala 1981). Here we first recapitulate the major steps to derive the power-law CCN spectrum, and then relate the CCN spectrum to N_a, M_a, and ε_a and use the new expression to examine the relationship between the parameters of the power-law CCN spectrum.

Derivation of Power-Law CCN Spectrum

According to the classical Kohler theory, the equilibrium supersaturation (S) of a spherical aerosol particle is given by

$$S = ar_a^{-1} - br_a^{-3}, \tag{1}$$

where r_a is the radius of the particle, a the curvature coefficient, and b the solution coefficient b. Maximization of S with respect to r_a yields the critical radius (r_c) and critical supersaturation (S_c) given by

$$r_c = \sqrt{\frac{3b}{a}}, \tag{2a}$$

$$S_c = \frac{2a}{3r_c} = \frac{2a^{3/2}}{3\sqrt{3}} b^{-1/2}. \tag{2b}$$

The solute coefficient b is related to the dry radius, r_d, by a power-law relationship (Khvorostyanov and Curry 1999)

$$b = \alpha_b r_d^{\beta_b}, \tag{3}$$

where α_b and β_b depend on the physicochemical properties of the soluble part of the aerosol particle. The exponent β_b vary from 3 when the soluble fraction is proportional to the particle volume, to 2 when the soluble fraction is proportional to the particle surface area, to 0 when the soluble fraction is independent of the radius. Substitution of Eq. (3) into Eq. (2) then relates the dry radius to the supersaturation:

$$r_d = \left(\frac{2a^{3/2}}{\sqrt{27\alpha_b}} \right)^{2/\beta_b} S_c^{-2/\beta_b} \quad . \tag{4}$$

According to the definition, the accumulative CCN concentration at supersaturation S (accumulative CCN spectrum) is all the particles with their critical supersaturations < S, i.e., all the particles larger than that activated at S:

$$N_{CCN} = \int_{r_c(S)}^{r_{d\max}} n(r_d) dr_d \quad , \tag{5a}$$

where $n(r_d)$ is the dry aerosol size distribution, r_{dmax} is the radius of the largest aerosol particle. According to Eq. (4),

$$r_c(S) = \left(\frac{2a^{3/2}}{\sqrt{27\alpha_b}} \right)^{2/\beta_b} S^{-2/\beta_b} \quad . \tag{5b}$$

Aerosol size distributions can often be described by a power-law function (Junge 1963; Twomey 1977)

$$n(r_d) = \alpha r_d^{-\beta} \quad , \tag{6}$$

where α and β are two empirical coefficients. Substitution of Eq. (6) into Eq. (5), and subsequent integration yields the power-law CCN spectrum given by

$$N_{CCN} = CS^k \quad , \tag{7a}$$

$$C = (1 - t^{1-\beta}) C_\infty \quad , \tag{7b}$$

$$C_\infty = \frac{\alpha}{\beta - 1} \left(\frac{2a^{3/2}}{\sqrt{27\alpha_b}} \right)^{2(1-\beta)/\beta_b} \quad , \tag{7c}$$

$$k = \frac{2(\beta - 1)}{\beta_b} \quad . \tag{7d}$$

where t is the ratio of r_{dmax} to the radius of the smallest dry particle activated at S.

Relationship of CCN Spectra to Aerosol Size Distribution Moments

The parameters of C and k in the CCN spectrum are intermediate quantities that are not predicted in climate models; it is practically desirable to further relate C and k (or α and β) to N_a, M_a and ε_a.

From Eq. (6), we can derive expressions for N_a and M_a, which leads to

$$\alpha = \frac{(\beta-1)}{\left(1-t^{1-\beta}\right)}\left[\frac{4\pi\rho_a(\beta-1)\left(t^{4-\beta}-1\right)}{3(4-\beta)\left(1-t^{1-\beta}\right)}\right]^{(1-\beta)/3} M_a^{(\beta-1)/3} N_a^{4-\beta} . \tag{8}$$

A combination of Eqs. (7) and (8) yields the expression that relates C to N_a and M_a:

$$C = \left(\frac{2a^{3/2}}{\sqrt{27\alpha_b}}\right)^{2(1-\beta)/\beta_b}\left[\frac{4\pi\rho_a(\beta-1)\left(t^{4-\beta}-1\right)}{3(4-\beta)\left(1-t^{1-\beta}\right)}\right]^{(1-\beta)/3} N_a^{(4-\beta)/3} M_a^{(\beta-1)/3} \tag{9}$$

Equation (9) indicates that a liner relationship between N_a and M_a means a linear relationship between C and N_a or M_a. This assumption has been implicitly applied in many studies []. However, the relationship between N_a and M_a are generally much more complex because they are often determined by different physical processes (Van Dingenen et al. 2000).

Similarly, for the power-law aerosol size distribution described by Eq. (6), the exponent β is related to ε_a by

$$\varepsilon_a = \left[\frac{(2-\beta)^2\left(1-t^{1-\beta}\right)\left(1-t^{3-\beta}\right)}{(1-\beta)(3-\beta)\left(1-t^{2-\beta}\right)^2}-1\right]^{1/2}, \tag{10a}$$

which has the following limits

$$\varepsilon_{a1} \equiv \varepsilon_a(\beta\to 1) = \left[\frac{(t+1)\ln t}{2(t-1)}-1\right]^{1/2}, \tag{10b}$$

$$\varepsilon_{a2} \equiv \varepsilon_a(\beta\to 2) = \left[\frac{(t-1)^2}{t\ln^2 t}-1\right]^{1/2}, \tag{10c}$$

$$\varepsilon_{a3} \equiv \varepsilon_a\left(\beta \to 3\right) = \left[\frac{(t+1)\ln t}{2(t-1)} - 1\right]^{1/2}. \tag{10d}$$

Figure 1 shows ε_a as a function of β for different values of t. It is clear that ε_a reaches its maximum at $\beta = 2$ and symmetric around this peak. The symmetry around $\beta = 2$ is also evident from the three limit equations. As shown in Figure 1, Eq. (10a) is well approximated by

$$\varepsilon_a = \varepsilon_{a2} \exp\left[-\ln\left(\frac{\varepsilon_{a2}}{\varepsilon_{a1}}\right)(\beta - 2)^2\right] \tag{10e}$$

Or

$$\beta = 2 \pm \ln\left(\frac{\varepsilon_a}{\varepsilon_{a2}}\right) \ln^{-1}\left(\frac{\varepsilon_{a1}}{\varepsilon_{a2}}\right) \tag{10f}$$

According to Eqs. (7, 9, and 10), given aerosol chemical properties (known a and b) and t, C depends on N_a, m_a and ε_a whereas k is determined only by ε_a (k is an increasing [decreasing] function of ε_a: (k is an increasing (decreasing) function of ε_a when $\beta<2$ [$\beta \geq 2$]).

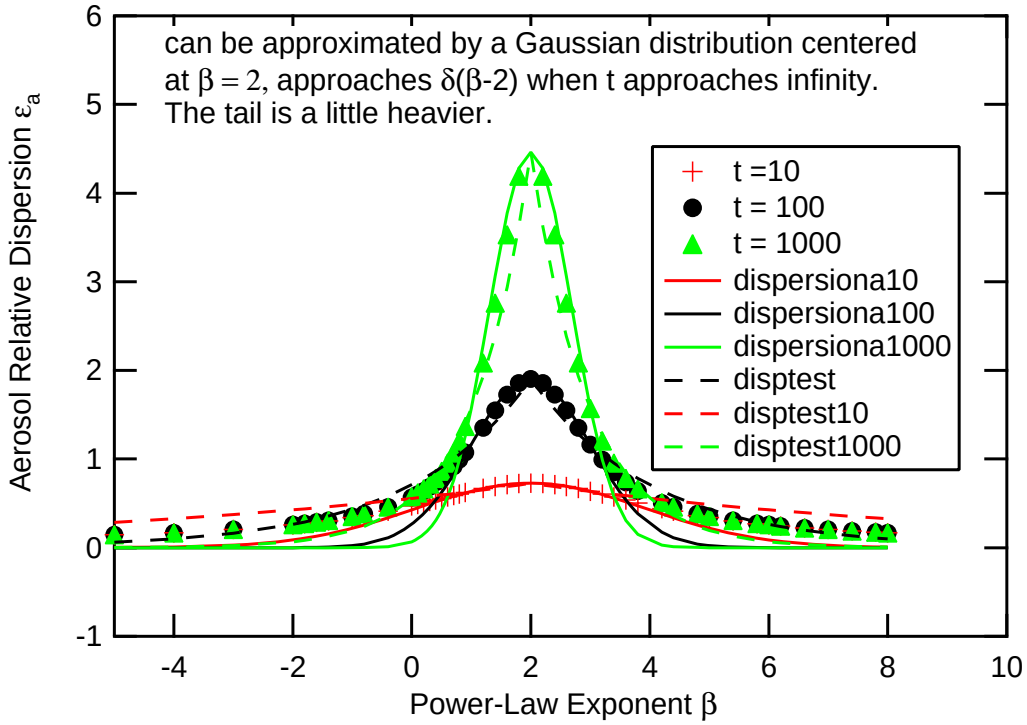

Figure 1. Relationship between the aerosol relative dispersion and the power-law exponent β. The different symbols denote results calculated from Eq. (10a) for different values of t. The dashed lines are the corresponding approximations.

Power-Law Expression For N_c

Twomey (1959) derived a power-law expression for N_c

$$N_c = 10^{4k/(k+2)} \left[\frac{2\left(c_1 G\right)^{3/2}}{4\pi\rho_w k c_2 B\left(\frac{k}{2}, \frac{3}{2}\right)} \right]^{k/(k+2)} w^{3k/2(k+2)} c^{2/(k+2)}, \tag{11}$$

where B() is the complete Beta function, w the cloud updraft velocity, and c_1, c_2 and G are functions of the cloud base temperature and pressure. Although the Twomey expression has been widely used in studies of aerosol indirect effects, a drawback of the Twomey expression is that both C and k are intermediate quantities, not aerosol properties available in climate models. This deficiency can be eliminated by substituting Eqs. (7d), (9), and (10) into Eq. (11), which leads to

$$N_c = f\left(\beta\right) w^{3(\beta-1)/2(\beta+\beta_b-1)} N_a^{(4-\beta)\beta_b/3(\beta+\beta_b-1)} M_a^{(\beta-1)\beta_b/3(\beta+\beta_b-1)} \tag{12a}$$

$$f\left(\beta\right) = 10^{4(\beta-1)/(\beta+\beta_b-1)} \left[\frac{(c_1 G)^{3/2}\beta_b}{4\pi\rho_w c_2(\beta-1)B\left(\frac{\beta-1}{\beta_b}, \frac{3}{2}\right)} \right]^{(\beta-1)/(\beta+\beta_b-1)} \left[\left(\frac{2a^{3/2}}{\sqrt{27\alpha_b}} \right)^{-2(\beta-1)/\beta_b} \left[\frac{4\pi\rho_a(\beta-1)\left(t^{4-\beta}-1\right)}{3(4-\beta)\left(1-t^{1-\beta}\right)} \right]^{(1-\beta)/3} \right]^{\beta_b/(\beta+\beta_b-1)} \tag{12b}$$

Equation (12), together with Eq. (10c), suggests that N_c is a combined function of N_a, M_a, and ε_a, not N_a, or M_a alone. Similarly, we can relate the relative dispersion of the cloud droplet size distribution to N_a, M_a and ε_a (Liu et al. 2006).

Implications for Indirect Aerosol Effects

Generally, N_a and M_a are neither independent of each other, nor exhibits a linear relationship as commonly assumed (van Dingenen et al. 1995, 1999, 2000; Hegg and Haufman 1998). To examine the effect of a nonlinear N_a-M_a relationship on evaluation of indirect aerosol effects, we assume a power-law relationship

$$N_a \propto M_a^{\delta}, \tag{13}$$

where δ depends on the physical processes such as condensational growth, coagulation and entrainment (van Dingenen et al. 2000). Equation (13) includes no ($\delta = 0$) and linear ($\delta = 1$) dependence of N_a and M_a as its special cases. Substitution of Eq. (13) into Eq. (12) yields

$$N_c \propto N_a^{\left[(4-\beta)\beta_b/3(\beta+\beta_b-1)+(\beta-1)\beta_b/3\delta(\beta+\beta_b-1)\right]} \tag{14a}$$

When $\beta_b = 3$, Eq. (14a) becomes

$$N_c \propto N_a^{\left[(4-\beta)/(\beta+2)+(\beta-1)/\delta(\beta+2)\right]} \tag{14b}$$

According to Eq. (14a), there is no dependence of N_c on N_a, if δ and β satisfies

$$\delta_0 = \frac{1-\beta}{4-\beta} \tag{15}$$

Under this condition, N_c does not depend on M_a neither, and is only a function of ε_a. The same conclusion applies to the CCN parameter C. It is noteworthy that β_b does not have any effect on the "no-dependence" curve. Furthermore, N_c depends linearly on N_a, if δ and β satisfies

$$\delta_1 = \frac{\beta_b}{3+\beta_b} \tag{16a}$$

Unlike the "no-dependence" curve, the "linear dependence" depends on β_b, but independent of β. When $\beta_b = 3$, Eq. (16a) becomes

$$\delta_1 = \frac{1}{2} \tag{16b}$$

Equations (15) and (16b) can be used as references in analysis of data from ambient aerosols. As shown in Figure 2, the "no-dependence" curve consists of two branches around $\beta = 4$: when $\beta < 4$, δ_0 decreases to minus infinity β whereas δ_0 decreases from infinity to 1 when β increases from 4 to infinity. When $\delta_1 = 0.5$, N_c depends linearly on N_a regardless of β values. Unfortunately, there are no simultaneous measurements of β and δ to compare against these theoretical results.

Figure 2. Need a Figure 2.

Equation (14b) can be also applied to examining remote sensing measurements of indirect aerosol effect. In remote sensing, the indirect aerosol effect is often quantified by (Kim et al. 2003, Feingold et al. 2003).

$$I_N = -\frac{d \ln r_e}{d \ln N_a}, \tag{17}$$

where r_e is the effective radius of the cloud droplet size distribution. This measure is less sensitive to the uncertainties involved with remote sensing techniques. Application of Eq. (14b) to Eq. (17) yields

$$I_N = \frac{1}{3}\left(\frac{4-\beta}{\beta+2} + \frac{\beta-1}{\delta(\beta+2)}\right), \tag{18}$$

which indicates that the first indirect aerosol effect as defined by Eq. (17) actually measures the combined effects of β and δ. For the linear N_a-M_a relationship (δ = 1), the indirect aerosol effect is given by

$$I_{N0} = \frac{1}{\beta+2} \tag{19}$$

The ratio of I_N to I_{N0} is given by

$$\xi_N = \frac{I}{I_0} = \frac{4-\beta}{3} + \frac{\beta-1}{3\delta} \tag{20}$$

Figure 3 shows the dependence of ξ_N on δ for different values of β. It is evident that, except for β = 1, δ has no effect on I, the assumption of a linear N_a-M_a relationship underestimates the indirect aerosol effect in the sublinear regime (δ<1), but overestimates the indirect aerosol effect in the superlinear regime (δ> 1).

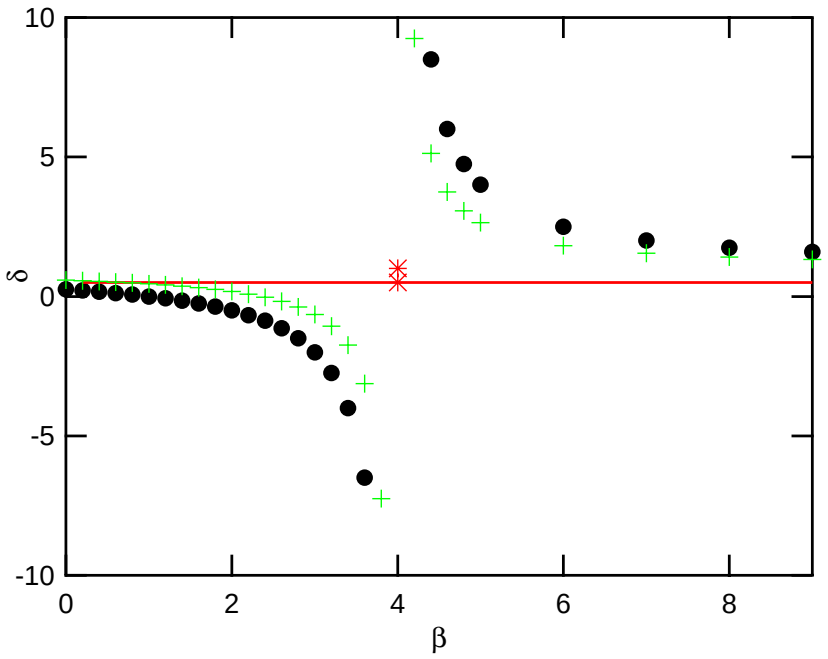

Figure 3. Characteristic δ-β curves. The black dots denote the no-dependence curve; the red * denotes the case where the cloud droplet number concentration is linearly related to the aerosol number concentration. The crosses are an example.

Note that if the indirect aerosol effect (I_M) is defined as the relative change of the effective radius with respect to the relative change of M_a instead of N_a, then we have

$$I_M = \delta I_N = \frac{\delta}{3}\left(\frac{4-\beta}{\beta+2} + \frac{\beta-1}{\delta(\beta+2)}\right),$$
$$\xi_M = \delta\xi_N = \frac{\delta(4-\beta)+\beta-1}{3}, \qquad (22)$$

where ξ_M is the ratio of I_M to I_{M0} (note $I_{M0} = I_{N0}$).

The results calculated from Eq. (22) are also shown in Figure 4 in the corresponding dashed lines. Contrary to ξ_N, the assumption of a linear N_a-M_a relationship overestimates the indirect aerosol effect in the sublinear regime ($\delta<1$), but underestimates the indirect aerosol effect in the superlinear regime ($\delta>1$). In practice, aerosol index, aerosol optical depth or scattering/extinction coefficient has been used, instead of N_a or M_a (Nakajima et al. 2001; Kim et al. 2003; Feingold et al. 2003). The wide range of variation as shown in Figure 3 suggests that the large uncertainty in current remote sensing of indirect aerosol effects may reflect the definition ambiguity associated with a nonlinear N_a-M_a relationship.

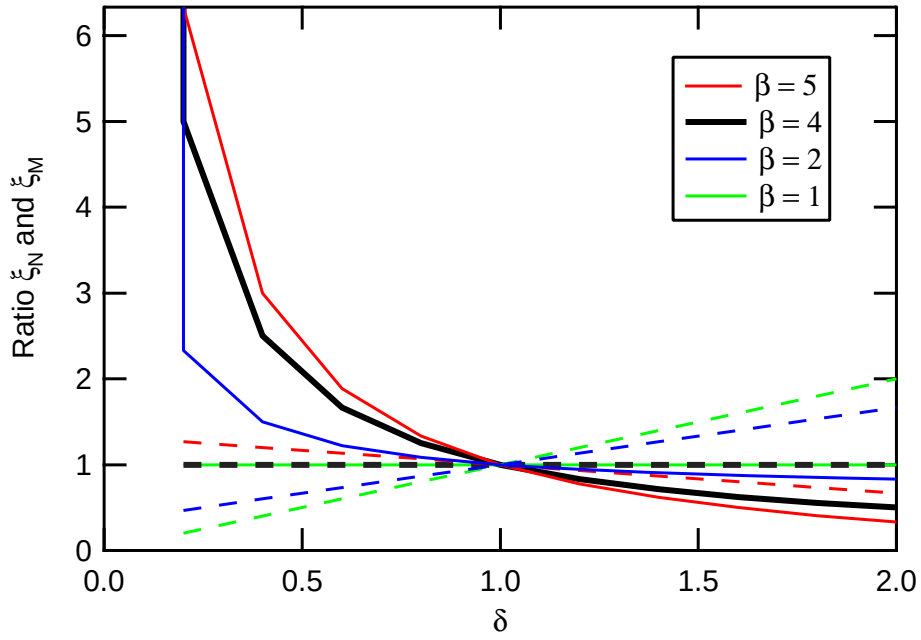

Figure 4. The ratio of the indirect aerosol effect as a function of δ. The solid and dashed curves are for the number-based and mass-based definitions, respectively. Different colors denote values of β.

Concluding Remarks

A theoretical formulation is presented that relates CCN spectra, cloud droplet number concentration to the three commonly used moments of the aerosol size distribution: aerosol number concentration, aerosol mass loading and aerosol relative dispersion by various power-law relationships. The newly derived power expressions are then applied to examining issues regarding indirect aerosol effects. It is shown that the relationships between the aerosol number concentration, aerosol mass loading and aerosol relative dispersion are critically important for quantifying indirect aerosol effects, and that part of the uncertainty in estimates of indirect aerosol effects is due to the use of either aerosol number concentration or mass loading as the sole aerosol variable in parameterization of the cloud droplet number concentration. It is also shown that if the relationship between aerosol number concentration and aerosol mass loading is nonlinear, the remote sensing definition of the first indirect aerosol effect that has been commonly used in various studies is ambiguous, causing additional uncertainties. The results highlight the importance and need to simultaneously measure and analyze N_a, M_a, ε_a, and their mutual relationships.

It is noted in passing that despite the wide use and simplicity of the various power-law relationships derived from the power-law aerosol size distribution, ambient aerosols often do not follow the power-law size distributions as assumed in this paper. This is especially true when the size range over which aerosol particles can activate into cloud droplets is sufficiently large (another reason for not ignoring the size truncation t). For example, field and laboratory measurements have shown that a more realistic

CCN spectrum is not linear in log-log coordinates as implied with a single power-law CCN spectrum, but has a concave curvature, i.e., k decreases with increasing S (Hudson 1984; Ji and Shaw 1998; Yum and Hudson 2001). The leveling-off of CCN spectra is indicative of non-power-law aerosol size distributions. The issue of non-power-law aerosol size distributions will be addressed in the future.

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