Dispersion bias, dispersion effect, and the aerosol–cloud conundrum

Yangang Liu[1,3]**, Peter H Daum**[1]**, Huan Guo**[1] **and Yiran Peng**[2]

[1] Brookhaven National Laboratory, Building 815E, Upton, NY 11973-5000, USA
[2] Canadian Centre for Climate Modeling and Analysis, University of Victoria, 3800 Finnerty Road, Victoria, BC V8P 5C2, Canada

E-mail: lyg@bnl.gov

Abstract
This work examines the influences of relative dispersion (the ratio of the standard deviation to the mean radius of the cloud droplet size distribution) on cloud albedo and cloud radiative forcing, derives an analytical formulation that accounts explicitly for the contribution from droplet concentration and relative dispersion, and presents a new approach to parameterize relative dispersion in climate models. It is shown that inadequate representation of relative dispersion in climate models leads to an overestimation of cloud albedo, resulting in a negative bias of global mean shortwave cloud radiative forcing that can be comparable to the warming caused by doubling CO_2 in magnitude, and that this dispersion bias is likely near its maximum for ambient clouds. Relative dispersion is empirically expressed as a function of the quotient between cloud liquid water content and droplet concentration (i.e., water per droplet), yielding an analytical formulation for the first aerosol indirect effect. Further analysis of the new expression reveals that the dispersion effect not only offsets the cooling from the Twomey effect, but is also proportional to the Twomey effect in magnitude. These results suggest that unrealistic representation of relative dispersion in cloud parameterization in general, and evaluation of aerosol indirect effects in particular, is at least in part responsible for several outstanding puzzles of the aerosol–cloud conundrum: for example, overestimation of cloud radiative cooling by climate models compared to satellite observations; large uncertainty and discrepancy in estimates of the aerosol indirect effect; and the lack of interhemispheric difference in cloud albedo.

1. Introduction

Twomey (1974, 1991) pointed out that an increase in aerosol loading leads to increases in cloud condensation nuclei (CCN) and cloud droplet number concentration (N), which in turn reduces the effective radius (r_e) and thereby enhances the cloud albedo (R) under the condition that other variables (e.g., liquid water content) remain unchanged. Although the N-induced Twomey effect builds on solid physics and ample observational evidence has been reported supporting the effect (e.g., Coakley *et al* 1987, Han *et al* 1998, Breon *et al* 2002,

Feingold *et al* 2003, Kim *et al* 2008, Penner *et al* 2004), the quantitative picture of the first aerosol indirect effect is hazy, and many puzzles remain to be solved (Anderson *et al* 2003, Lohmann and Lesins 2002, Lohmann and Feichter 2005). For example, the estimates of the aerosol indirect effect (AIE) from global climate models (GCMs) still constitute the largest uncertainty among all the known climate forcings (IPCC 2007). The high end of forward GCM AIE estimates tends to be much larger than those inverse calculations constrained by observations (Anderson *et al* 2003). In particular, some GCMs project an AIE cooling that is comparable to the warming from increasing greenhouse gases, which is obviously at odds with the fact of a warming world (Anderson *et al*

---

[3] Author to whom any correspondence should be addressed.

2003, Penner 2004). Another related puzzle is that clouds in the Northern Hemisphere are not much brighter, and the temperature is not much lower, than their counterparts in the Southern Hemisphere, as expected from the substantial interhemispheric difference in anthropogenic sulfur emissions since the industrial revolution (Schwartz 1988, Schwartz puzzle hereafter). The representation of cloud properties in climate models is fraught with outstanding puzzles as well. One of the puzzles was identified in Potter and Cess (2004), which found that, compared to satellite observations, many GCMs significantly overestimate cloud radiative cooling (cloud overcooling hereafter).

All these puzzles mentioned above, collectively referred to as the aerosol–cloud conundrum here, are likely rooted in inadequate understanding and representation of aerosol–cloud interactions and cloud properties, especially in GCMs. Previous studies have been mainly concerned with the influences of liquid water content and droplet concentration, largely ignoring the effect of the spectral shape of the cloud droplet size distribution. However, it is well known that droplet size distributions exhibit a variety of spectral shapes, and relative dispersion (ε), a measure of the relative width of the cloud droplet size distribution, has been demonstrated to substantially affect the parameterization of effective radius (Martin *et al* 1994, Liu and Daum 2000). Furthermore, Liu and Daum (2002) empirically showed that ε in clouds that had been influenced by anthropogenic aerosols was higher than for background clouds that had formed under similar dynamical conditions, and that the enhanced ε exerts a warming effect (dispersion effect) that acts to reduce the cooling of the Twomey effect by 10–80% (Liu and Daum 2002), depending on the relationship between ε and N.

While subsequent GCM studies have reinforced the importance of the dispersion effect (Peng and Lohmann 2003, Rotstayn and Liu 2003), and a firm theoretical basis has been established for the concurrent increases in ε and N when aerosol loading increases through analytical and modeling studies (Yum and Hudson 2005, Liu *et al* 2006a, Peng *et al* 2007), the factors that determine ε and its relationship to N are poorly understood, and quantification of the dispersion effect is still in its infancy (Zhao *et al* 2006, Pawlowska *et al* 2006, Lu *et al* 2007, 2008).

The primary objectives of this paper are to further previous studies to (1) derive an analytical formulation for cloud albedo and cloud radiative forcing that accounts explicitly for the effect of ε, and examine the influences of ε on cloud albedo and cloud radiative forcing, (2) present a new approach to representing ε, (3) explore the important advantages and implications of this new representation for studying AIE, and (4) discuss the potential role of ε in causing the aerosol–cloud conundrum in general.

2. Dispersion bias and cloud overcooling

This section is an extension of our previous work on r_e (Liu and Hallett 1997, Liu and Daum 2000) to investigate the influences of ε on R and cloud radiative forcing and present an analytical formulation for R and cloud radiative forcing that

accounts explicitly for ε. It has been shown that r_e can be generally parameterized via the following equation (Liu and Hallett 1997, Liu and Daum 2000):

$$r_e = \left(\frac{3}{4\pi\rho_w}\right)^{1/3} \beta \left(\frac{L}{N}\right)^{1/3}, \qquad (1a)$$

where L is the liquid water content, and ρ_w is the water density. Instead of being a constant as commonly assumed in GCMs (e.g., $\beta = 1$ for the monodisperse droplet size distribution), the effective radius ratio β is a dimensionless parameter that depends on the spectral shape of the cloud droplet size distribution. Note that Martin *et al* (1994) used a slightly different but equivalent way to measure the effect of the spectral shape such that

$$r_e = \left(\frac{3}{4\pi\rho_w}\right)^{1/3} k^{-1/3} \left(\frac{L}{N}\right)^{1/3}, \qquad (1b)$$

where the parameter k is equal to β^{-3}. The parameter β is used throughout this paper. For nonprecipitating clouds, there is an increasing body of evidence that β is an increasing function of ε that can be well described by (Liu and Hallett 1997, Liu and Daum 2000, 2002, Lu *et al* 2007, Daum *et al* 2008)

$$\beta = \frac{\left(1 + 2\varepsilon^2\right)^{2/3}}{\left(1 + \varepsilon^2\right)^{1/3}}, \qquad (2)$$

where ε represents the relative dispersion of the cloud droplet size distribution.

The cloud optical depth τ is related to r_e by

$$\tau = \frac{3H}{2\rho_w} \frac{L}{r_e}, \qquad (3)$$

where H is the cloud thickness. Under the two-stream approximation of a nonabsorbing, homogeneous, plane-parallel cloud, the cloud-top albedo R is given by (Bohren 1987)

$$R = \frac{(1-g)\tau}{2 + (1-g)\tau}, \qquad (4)$$

where g is the asymmetry parameter and considered a constant in this study.

It is expected from the above four equations that a smaller ε leads to a smaller r_e, but larger τ and R, all other variables remaining the same. To further quantify the influence of ε on R, the difference between the real R and that for the corresponding reference cloud with a monodisperse cloud droplet size distribution ($\varepsilon = 0$ and $\beta = 1$, hereafter monodisperse cloud), ΔR, is examined. It can shown by combining the above four equations that ΔR is given by

$$\Delta R = R - R_0 = -\left[\frac{(\beta - 1)(1 - R_0) R_0}{R_0 + (1 - R_0)\beta}\right], \qquad (5)$$

where R_0 denotes the cloud albedo of the corresponding monodisperse cloud. Because clouds generally have polydisperse droplet size distributions with $\varepsilon > 0$ and $\beta > 1$, the minus sign in front of the square bracket indicates that the monodisperse cloud assumption generally overestimates R.

The overestimation of cloud albedo in turn translates to an overestimation of global mean shortwave cloud radiative forcing. Charlson *et al* (1992) provided an expression for the perturbation in the global mean shortwave cloud radiative forcing that would result from a change in R of the marine stratus and stratocumulus clouds (their equations from (9) to (11)). It can be shown similarly that the difference between the cloud radiative forcing including ε and that of the corresponding monodisperse cloud is given by

$$\Delta F = \frac{0.8}{4} A S \left[\frac{(\beta - 1)(1 - R_0) R_0}{R_0 + (1 - R_0)\beta} \right], \qquad (6)$$

where A is the fraction of the globe that is covered by marine stratiform clouds and S is the solar constant. Again, because ambient clouds generally have $\varepsilon > 0$ and $\beta > 1$, $\Delta F > 0$, which suggests that the monodisperse cloud assumption overestimates the cooling from shortwave cloud radiative forcing. Because the overestimates of cloud albedo and shortwave radiative forcing resulting from the monodisperse cloud assumption are systematic, we call them dispersion bias collectively to distinguish it from the concept of dispersion effect reserved for the special dispersion bias that is caused by changes in pre-cloud aerosol properties.

Equations (5) and (6), together with equation (2), further indicate that the magnitudes of the ε-induced ΔR and ΔF depend on both ε and R_0. To illustrate this dependence and to make the proceeding arguments quantitative, figure 1 depicts ΔR (left axis) and minus ΔF (right axis) as a function of R_0 for different values of ε. As expected, both the magnitudes of ΔR and ΔF increase monotonically with increasing ε, and the magnitude of ΔF can reach up to 10 W m^{-2}, depending on the values of ε and R_0. Even for a typical cloud with $\varepsilon = 0.3$ and $R_0 = 0.5$, the overestimation of cloud albedo and cloud forcing are 0.02 and 1.68 W m^{-2}, respectively, which are comparable to the climate forcing caused by doubling CO_2 in magnitude but opposite in sign. The ε-induced ΔR and ΔF are not evenly distributed over R_0. Instead, as R_0 changes from 0 to 1, their magnitudes first increase with increasing R_0, peak at some value denoted by R_0^*, and then decrease with further increasing R_0. This quadratic behavior arises from the fact that, for a given ε, both ΔR and ΔF are determined by the product of R_0 and the corresponding monodisperse co-albedo $(1 - R_0)$. Optically thin clouds have very small R_0 but a large co-albedo, while optically thick clouds have very small co-albedo but large R_0, leading to small dispersion biases at both ends of R_0 and maximum dispersion biases at R_0^*. It should be emphasized that R_0^* is not highly sensitive to ε over the range of ε that is typically observed in ambient clouds (from 0.1 to 1; Liu and Daum 2000, 2002, Zhao *et al* 2006). The maximum values of ΔR and ΔF, $(\Delta R)^*$ and $(\Delta F)^*$, occur near $R_0^* \sim 0.5$ regardless of the specific value of ε. Furthermore, the variation of ΔR and ΔF is rather flat over the typical range of cloud albedo ($R_0 \sim 0.3$–0.7). It is interesting to note that the dependence of the maximum ΔR and ΔF on R_0 is similar to that of the Twomey effect, which has been known to be most sensitive at $R_0 = 0.5$, and the cloud susceptibility is rather flat over R_0 from ~ 0.3 to 0.7 (Twomey 1991, Charlson *et al*
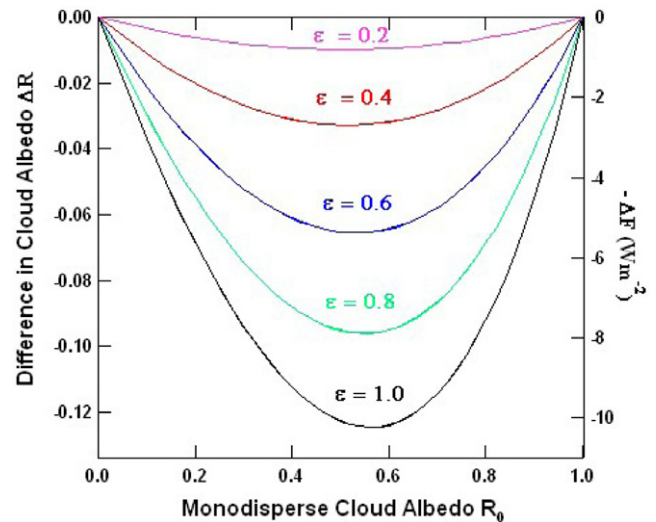

Figure 1. Difference in the cloud-top albedo (ΔR, left axis) and the negative difference (ΔF) in the global mean cloud radiative forcing as a function of the cloud-top albedo for a monodisperse cloud (R_0) at different values of relative dispersion (ε). In the calculation, the values of the global mean fraction of low stratiform clouds (A) and the solar constant (S) were set to the same values as used in Charlson *et al* (1992), i.e., $A = 0.3$, and $S = 1370$ W m^{-2}.

1992). This coincidence has important implications for aerosol indirect effects (see section 3 for more discussion).

The insensitivity of R_0^* to ε, and the dependence of $(\Delta R)^*$ and $(\Delta F)^*$ on ε can be further investigated by establishing and examining the analytical formulation for R_0^*, $(\Delta R)^*$, and $(\Delta F)^*$. By setting the derivatives of equations (5) and (6) with respect to R_0 equal to 0, we obtain the following equations:

$$R_0^* = \frac{\beta - \sqrt{\beta}}{\beta - 1}. \qquad (7)$$

$$(\Delta R)^* = \frac{\left(1 - \sqrt{\beta}\right)}{\left(1 + \sqrt{\beta}\right)}, \qquad (8)$$

$$(\Delta F)^* = \frac{0.8}{4} A S \frac{\left(\sqrt{\beta} - 1\right)}{\left(1 + \sqrt{\beta}\right)}. \qquad (9)$$

Coupled with equation (2), the above equations analytically quantify the dependence of the peak quantities on ε. As shown in figure 2, R_0^* only varies by about 12%, from about 0.5 to 0.56, when ε changes from 0.1 to 1. This confirms the preceding result that R_0^* is not highly sensitive to the variation of ε over the likely range from 0.1 to 1, and $R_0^* = 0.5$ serves as a good approximation.

Figure 3 shows the dependence of $(\Delta R)^*$ and $-(\Delta F)^*$ on ε calculated from equations (8) and (9), respectively. It is evident that the magnitudes of both $(\Delta R)^*$ and $(\Delta F)^*$ increase as ε increases, which is consistent with the general trend of variations of (ΔR) and (ΔF) with ε. Together with the fact that the dependence of ΔR and ΔF on R_0 is relatively flat over the typical cloud albedo, this consistency suggests that the set of equations (7)–(9) can be used as an analytical approximation to quantify the ε-induced biases in cloud albedo and cloud radiative forcing, which has the nice feature of being dependent
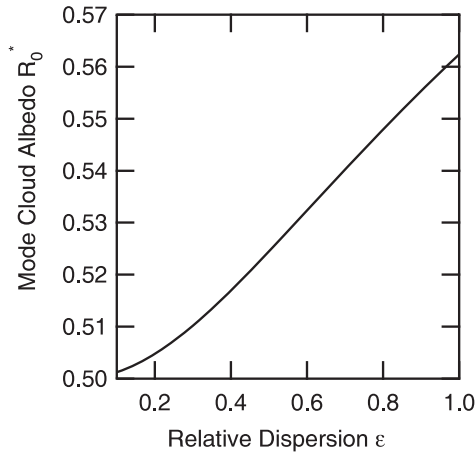
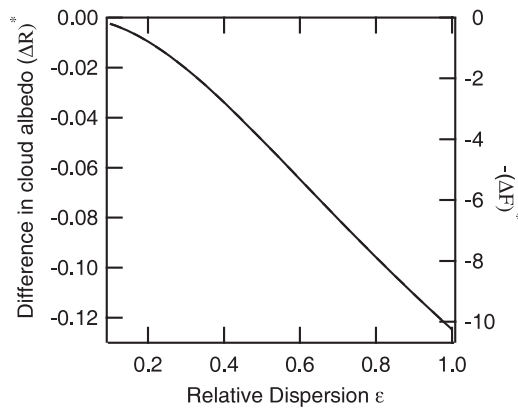**Figure 2.** Dependence of R_0^* on the relative dispersion ε.

Figure 3. Dependence of $(\Delta R)^*$ and $-(\Delta F)^*$ on the relative dispersion ε.

on $\beta(\varepsilon)$ only. Therefore, the generalized equation for R can be approximately expressed as

$$R \approx \frac{\left(1 - \sqrt{\beta}\right)}{\left(1 + \sqrt{\beta}\right)} + R_0. \tag{10}$$

As mentioned in section 1, many GCMs tend to overestimate the cooling of the shortwave cloud radiative forcing compared to satellite observations, and deciphering the puzzle of the cloud overcooling has been a major challenge (Potter and Cess 2004). The above analysis of the dispersion bias, together with equation (10), suggests that assuming unrealistically narrow cloud droplet size distributions (too small β, explicitly or implicitly) may be one reason for this outstanding problem.

3. Dispersion effect and AIE haziness

Treating the effective radius ratio β as a constant had been a common assumption implicit in the evaluation of the indirect aerosol effect as well, until very recently, when ε (or β) was shown to increase with increasing aerosol loading, and it was shown that the enhanced ε leads to a warming effect that acts to substantially offset the cooling of the Twomey effect by 10
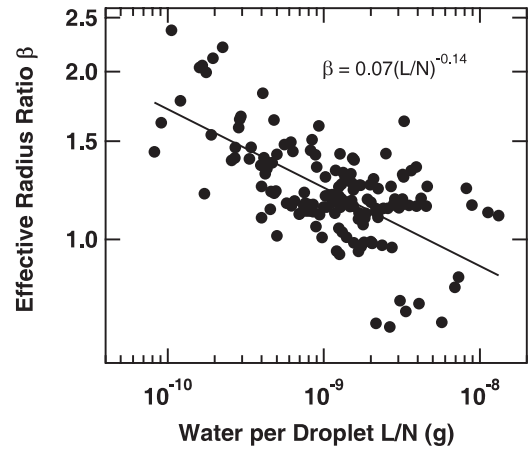

Figure 4. Relationship between the effective radius ratio β and the average water per droplet, L/N. The data are from several projects, including ARM 1997 Spring and Fall IOPs (Liu and Daum 2000), ARM 2000 IOP (Dong and Mace 2003), NARE, RACE, FIRE-ACE (Peng *et al* 2002), and MASE (Daum *et al* 2008). Each point in this figure represents a flight or horizontal leg average.

to 80%, depending on the $\beta(\varepsilon)$–N relationship (Liu and Daum 2002, Rotstayn and Liu 2003, Peng and Lohmann 2003). It is noted that some studies consider the dispersion effect in part by assigning $k = 0.67$ and 0.80 to continental and maritime air masses, respectively, based on Martin *et al* (1994). Virtually all existing estimates of the dispersion effect were based on some empirical $\beta(\varepsilon)$–N relationships, which suffer from large uncertainty. Subsequent theoretical analysis (Liu *et al* 2006a) and parcel model studies (Yum and Hudson 2005, Peng *et al* 2007) have further demonstrated that, in addition to aerosol properties, ε, N, and the β–N relationship also depend on cloud updraft, and that, unlike the aerosol effect, variation in cloud dynamics leads to a negative correlation between ε and N. It is anticipated that the large uncertainty in the empirical β–N relationships likely stems from the neglected differences in cloud dynamics. Another serious deficiency of using the β–N relationship to parameterize the dispersion effect is the implicit assumption that the data used to derive the empirical relationship have the same L. Therefore, a new parameterization of $\beta(\varepsilon)$ that helps reduce these deficiencies is desirable.

In an effort to improve the parameterization of r_e for stratocumulus clouds, Wood (2000) empirically demonstrated that β is better parameterized in terms of the mean volume radius than by using N alone. This empirical finding is supported by the theoretical analysis by Liu *et al* (2006a), which reveals that cloud updraft should have much less impact on the relationship of $\beta(\varepsilon)$ to the mean radius than on the β–N relationship. According to these studies, we examined the dependence of β on L/N (i.e., the average water per droplet), using the data from several projects that cover a variety of environmental and dynamical conditions (figure 4). It is evident from this figure that β generally decreases when L/N increases, and that the dependence can be described by

$$\beta = a_\beta \left(\frac{L}{N}\right)^{-b_\beta}, \tag{11}$$

where $a_\beta = 0.07$, $b_\beta = 0.14$, and L and N are in cgs units. Equation (11) embodies the fact that the cloud droplet size distribution narrows as droplets grow according to the adiabatic condensation theory, and accounts for the variation of L to some extent. The ability to account for the variation in L is a tremendous advantage of representing the dispersion effect with equation (11) over the $\beta(\varepsilon)$–N relationship in view of the difficulty in choosing data with the same L from which to develop an empirical relationship. The dispersion effect is reflected in equation (11) by the fact that an increased N leads to an increased $\beta(\varepsilon)$. Note that the scatter in the figure may be due to turbulent entrainment-mixing effects, which likely lead to concurrent increases of $\beta(\varepsilon)$ and L/N or broadening toward large sizes (Daum *et al* 2008).

Furthermore, this new parameterization of $\beta(\varepsilon)$ leads to an analytical formulation for the first aerosol indirect effect. Substitution of equation (11) into equation (1) leads to a generalized power-law expression for r_e,

$$r_e = a_e \left(\frac{L}{N} \right)^{b_e}, \qquad (12a)$$

$$a_e = \left(\frac{3}{4\pi\rho} \right)^{1/3} a_\beta, \qquad (12b)$$

$$b_e = \tfrac{1}{3} - b_\beta. \qquad (12c)$$

A value of $b_e < 1/3$ due to $b_\beta > 0$ indicates that consideration of $\beta(\varepsilon)$ results in a weaker dependence of r_e on L/N, which in turn leads to a smaller indirect aerosol effect. This point becomes more evident from the relative measure of the first indirect aerosol effect that have been widely used in remote sensing of the aerosol indirect effect (Feingold *et al* 2003, Kim *et al* 2008):

$$I = -\frac{\mathrm{d}\ln r_e}{\mathrm{d}\ln N_a}, \qquad (13)$$

where N_a is the number concentration of pre-cloud aerosols. Assuming $N = a_a N_a^{b_a}$, we have

$$I = b_e b_a = \left(\frac{1 - 3b_\beta}{3} \right) b_a = I_0 + I_\varepsilon, \qquad (14a)$$

$$I_0 = 1/3 b_a, \qquad (14b)$$

$$I_\varepsilon = -b_\beta b_a = -3b_\beta I_0, \qquad (14c)$$

where I_0 and I_ε represent the cooling Twomey effect associated with enhanced N, and the warming dispersion effect associated with enhanced ε, respectively.

The set of equations (14a)–(14c) has important implications for studying aerosol indirect effects. First, equation (14a) clearly reveals that the first aerosol indirect effect is an algebraic sum of the two opposing effects: the cooling Twomey effect and the warming dispersion effect. A constant $b_\beta = 0.14$ indicates that the dispersion effect offsets the Twomey effect by 42%. This reinforces the suggestion made by Liu and Daum (2002) that we should avoid the common practice of using the Twomey effect and the first aerosol indirect effect interchangeably. Second, the first aerosol indirect effect either detected by

remote sensing or inversely inferred from other observations reflects the combined signals of both the Twomey effect and the dispersion effect, whereas most forward GCM estimates only consider the Twomey effect. This conceptual inconsistency is at least responsible for part of the discrepancies between forward GCM estimates and those inverse calculations (Anderson *et al* 2003). Equation (14c) further indicates that consideration of the dispersion effect should also significantly reduce the absolute uncertainty in estimates of the first indirect aerosol effect because its magnitude increases proportionally with the Twomey effect, suggesting more reduction by the dispersion effect for a larger Twomey effect.

4. Implications for the Schwartz puzzle

In 1987, Charlson, Lovelock, Andreae, and Warren linked the Twomey effect to the production of dimethylsulphide (DMS) by marine microorganisms and proposed the well-known CLAW hypothesis, which states that an increase in the marine DMS in a warmer environment leads to increases of sulfate particles, CCN, and N in the marine atmosphere, thus enhancing cloud albedo and providing a negative feedback on the Earth's temperature. Schwartz quickly pointed out that if the CLAW hypothesis is correct, one would expect to see brighter clouds as well as more cooling in the Northern Hemisphere relative to the less polluted Southern Hemisphere because global anthropogenic emissions of sulfur dioxide are twice those from marine plankton, and predominantly in the Northern Hemisphere. However, Schwartz demonstrated that neither a larger cloud albedo nor a cooler trend in the Northern Hemisphere was borne out by the data examined. Differences between cloud amounts and liquid water contents in the two hemispheres have been proposed to mask the effect from the hemispheric difference in N (Slingo 1988, Schwartz *et al* 2002).

Noting that ε has never been in the argument of the Schwartz puzzle, here we propose that the overlooked hemispheric difference in ε may be a microphysical reason for the Schwartz puzzle. Based on the same physical principle for the dispersion effect (e.g., equation (11)), more anthropogenic aerosols in the Northern Hemisphere lead to both a larger N and a larger $\beta(\varepsilon)$ in the Northern Hemisphere compared to those in the Southern Hemisphere. Consideration of the hemispheric difference in ε is expected to reduce the hemispheric difference in R as expected from the hemispheric difference in N alone, thereby (at least partially) reconciling the Schwartz puzzle.

Consideration of the hemispheric difference in ε strengthens another explanation for the Schwartz puzzle proposed by Twomey (1991) as well. Twomey related the Schwartz puzzle to the fact that clouds in the Northern Hemisphere are less sensitive to perturbations in N than their counterparts in the Southern Hemisphere. Briefly, Twomey introduced the concept of cloud susceptibility, and derived the following expression for it:

$$\left(\frac{\Delta R}{\Delta N} \right)_0 = \frac{R(1 - R)}{3N}. \qquad (15)$$

Note that in the derivation of equation (15), Twomey implicitly utilized the monodisperse cloud assumption. The subscript 0 denotes this fact. Later studies (Ackerman *et al* 2000) demonstrated that equation (15) holds as long as β does not change with N. It is evident from equation (15) that, relative to the Southern Hemisphere, the Northern Hemisphere exhibits weaker cloud susceptibility because of a larger N. Two recent studies provided some observational evidence from satellite remote sensing by showing that cloud susceptibility is indeed higher over ocean than over land (Platnick and Oreopoulos 2008, Oreopoulos and Platnick 2008).

In studying cloud susceptibility using ship track data, Ackerman *et al* (2000) generalized equation (15) to include the effects of both N and ε such that

$$\frac{\Delta R}{\Delta N} = \frac{R\,(1 - R)}{3N}\left(1 - 3\frac{\partial(\ln\beta)}{\partial(\ln N)}\right). \qquad (16)$$

They found by examining the relationship between $\Delta\ln(\beta)$ and $\Delta\ln(N)$ that on average ship track emissions enhance β and further reduce the cloud susceptibility. Large scatter in the relationship between $\Delta\ln(\beta)$ and $\Delta\ln(N)$ was also reported in this study. By use of the new equation (11), equation (16) can be further simplified as

$$\frac{\Delta R}{\Delta N} = \frac{R\,(1 - R)}{3N}\left(1 - 3b_\beta\right). \qquad (17)$$

Equation (17) indicates that consideration of the dispersion effect ($b_\beta > 0$) reduces the cloud susceptibility further, which is consistent with the finding reported in Ackerman *et al* (2000). Equation (17) also indicates that the absolute measure of the first aerosol indirect effect is an algebraic sum of two competing effects, the Twomey effect and the dispersion effect, as well.

It is noteworthy that the explanation of the Schwartz puzzle only requires a larger ε in the Northern Hemisphere. The physical processes leading to a larger ε are not limited to a larger aerosol loading. For example, a stronger turbulent entrainment-mixing in the clouds of the Northern Hemisphere would result in a larger ε as well (Telford 1996, Liu *et al* 2002).

5. Concluding remarks

Developing ways to represent liquid water content (Kessler 1969, Sundqvist 1978) and cloud droplet number concentration (Ghan *et al* 1997, Lohmann *et al* 1999) has dominated the effort to improve the capability of climate models to quantify cloud–climate interactions and aerosol indirect effects. The influence of how water is distributed among cloud droplets has been largely marginalized. Motivated by the aerosol–cloud conundrum that is still baffling the climate (change) community, this paper focuses on the relative dispersion, examining its influences on cloud albedo, cloud radiative forcing, and the first aerosol indirect effect, and seeking a new approach for parameterizing it in climate models.

It is first shown that using a unrealistically small relative dispersion, such as assuming a monodisperse cloud, can lead to significant overestimation of cloud albedo by climate models, resulting in a negative bias of global mean estimates of shortwave cloud radiative forcing that can be comparable to the warming caused by doubling the CO_2 concentration. This result suggests that the dispersion bias may be one reason for the overestimation of shortwave radiative cooling in many climate models. The analysis also shows that the maximum dispersion bias occurs over a narrow range of cloud albedo from 0.5 to 0.56. An analytical formulation is presented that expresses the maximum dispersion bias and the peak cloud albedo as a function of relative dispersion. Cloud albedo and cloud radiative forcing are shown to be an algebraic sum of the contributions from a monodisperse cloud and from the relative dispersion.

It is then shown that the effective radius ratio β can be represented in terms of the ratio of the cloud liquid water content to the droplet concentration (i.e., water per droplet). This new representation not only relaxes the stringent requirement of grouping clouds with the same liquid water content and cloud dynamics in studies of the dispersion effect, but also leads to a new analytical formulation for the first aerosol indirect effect. This new formulation reveals that the first aerosol indirect effect is an algebraic sum of the conventional Twomey effect and the dispersion effect, and that the dispersion effect is proportional to the Twomey effect in magnitude. The proportional changes of the Twomey and dispersion effects offer a plausible explanation for the large discrepancy between the forward estimates and inverse calculations of the aerosol indirect effects.

The results of dispersion bias and the dispersion effect are also applied to explain the Schwartz puzzle. It is argued that an interhemispheric difference in relative dispersion may be a plausible reason why clouds in the Northern Hemisphere are not much brighter, and the temperature is not much lower, than their counterparts in the Southern Hemisphere, as expected from the substantial interhemispheric difference in anthropogenic sulfur emissions since the industrial revolution.

Compared to the effective radius, the spectral shape of the cloud droplet size distribution has been considered to be of marginal importance in computing cloud radiative properties (Hansen and Travis 1974). Taken together, the triad of dispersion bias, dispersion effect, and their plausible relationships to the Schwartz puzzle clearly challenge this conventional view; relative dispersion can exert a significant impact on cloud radiative properties by affecting the parameterization of effective radius itself.

Additional points are worth noting. First, unlike in the climate community, where the importance of the spectral shape of the cloud droplet size distribution has just started to be appreciated, understanding and quantifying the spectral shape of the cloud droplet size distribution has long been a central issue of cloud physics. To some extent, this contrast seems ironic in view of the well-recognized importance of cloud processes in shaping the climate. Furthermore, this work is only concerned with issues regarding nonprecipitating clouds. It is anticipated that the influence of relative dispersion becomes particularly important in understanding and quantification of precipitation and the second aerosol indirect effect because the importance of spectral dispersion in rain formation cannot be overemphasized. The interplays

between hydrological and radiative processes (Ramanthan *et al* 2001) further reinforce the need to account for spectral shape as accurately as possible. Relative dispersion affects the asymmetry factor *g* as well (Iorga and Stefan 2005). Second, despite the surge of interest in the context of dispersion effect since Liu and Daum (2002), research activities regarding relative dispersion have been limited in scientific scope and to a few research groups. There is a need for concerted effort to integrate multiple studies over a global scale. Especially lacking is a way to remotely measure relative dispersion that can provide long-term and large-area coverage. Platnick and Oreopoulos (2008) and Oreopoulos and Platnick (2008) explored the idea of using satellites to globally retrieve cloud susceptibility. Although a constant β was assumed in these studies, according to equation (17), a similar approach may be used to retrieve the dispersion parameter b_β from space (e.g., figure 9 of Platnick and Oreopoulos (2008)). Third, despite its marked importance, relative dispersion should not be considered as a panacea to remedy all the symptoms of the aerosol–cloud conundrum. It is known that cloud albedo is affected by many other factors: for example, cloud inhomogeneity (Cahalan *et al* 1994) and macroscopic cloud properties (e.g., liquid water path and cloud cover; Potter and Cess 2004). Nevertheless, there may be some connections between spectral shape and these factors as well, and such relationships are worth pursuing in the future. For example, the variability in liquid water path has been suggested as a major mask to the Schwartz puzzle (Schwartz *et al* 2002). Although this argument appears to make sense, it brings up a new and deeper puzzle: other things being equal, a smaller cloud albedo in the Northern Hemisphere would require a lower liquid water path, which is at odds with the argument of the second aerosol indirect effect outlined in Albrecht (1989) that a higher droplet concentration in the Northern Hemisphere would lead to a weaker precipitation and a higher liquid water path. Even fewer studies have been performed to investigate the impact of relative dispersion on precipitation and the second aerosol indirect effect (e.g., Liu and Daum 2004, Liu *et al* 2006b, Rotstayn and Liu 2005). Obviously, much remains to be learned regarding the aerosol–cloud conundrum. A more comprehensive reexamination of the Schwartz puzzle to take advantage of the progress in related areas such as satellite retrievals since Schwartz (1988) seems useful. Fourth, it should be noted that, in addition to parameterizing relative dispersion for use in bulk microphysical schemes as this work does, there exists some effort to consider the whole cloud droplet size distribution using sectional microphysics (Jacobson 2002, 2003). Despite its advantages, coupling a detailed microphysical model to climate models does not appear to be practical in the near future in view of the formidable computational cost involved. Instead, multi-moment schemes are expected to occupy the center stage in improving cloud representation in climate models. Finally, until now, the Twomey effect has been used synonymously with the first aerosol indirect effect. The existence and importance of the dispersion effect calls for the end of this practice.

References

Ackerman A S *et al* 2000 Effects of aerosols on cloud albedo: evaluation of Twomey's parameterization of cloud susceptibility using measurements of ship tracks *J. Atmos. Sci.* **57** 2684–95

Albrecht B A 1989 Aerosols microphysics, and fractional cloudiness *Science* **245** 1227–30

Anderson T L, Charlson R J, Schwartz S E, Knutti R, Boucher O, Rodhe H and Heintzenberg J 2003 Climate forcing by aerosols—a hazy picture *Science* **300** 1103–4

Bohren C F 1987 Multiple scattering of light and some of its observable consequences *Am. J. Phys.* **55** 524–33

Breon F-M, Tanre D and Generoso S 2002 Aerosol effect on cloud droplet size monitored from satellite *Science* **295** 834–8

Cahalan R F, Ridgway W, Wiscombe W J, Bell T L and Snider J B 1994 The albedo of fractal stratocumulus clouds *J. Atmos. Sci.* **51** 2434–55

Charlson R J, Schwartz S E, Hales J M, Cess R D, Coakley J A Jr, Hansen J E and Hofmann D J 1992 Climate forcing by anthropogenic aerosols *Science* **255** 423–30

Coakley J A, Bernstein J R and Durkee P A 1987 Effect of ship track effluents on cloud reflection *Science* **237** 1020–1

Daum P H, Liu Y, McGraw R L, Lee Y, Wang J, Senum G, Miller M and Hudson J G 2008 Microphysical properties of stratus/stratocumulus clouds during the 2005 marine stratus/stratocumulus experiment (MASE) *J. Geophys. Res.* submitted

Dong X and Mace G G 2003 Profiles of low-level stratus cloud microphysics deduced from ground-based measurement *J. Atmos. Ocean. Technol.* **20** 42–52

Feingold G, Eberhard W L, Veron D E and Previdi M 2003 First measurements of the Twomey indirect effect using ground-based remote sensors *Geophys. Res. Lett.* **30** 1287

Ghan S J, Leung L R, Easter R C and Abdul-Razzak H 1997 Prediction of cloud droplet number in a general circulation model *J. Geophys. Res.* **102** 21777–94

Han Q, Rossow W B, Chou J and Welch R M 1998 Global variation of column droplet concentration in low-level clouds *Geophys. Res. Lett.* **25** 1419–22

Hansen J E and Travis L D 1974 Light scattering in planetary atmospheres *Space Sci. Rev.* **16** 527–610

Intergovernmental Panel on Climate Change (IPCC) 2007: Summary for Policymakers by Working Group 1 http://www.ipcc.ch

Iorga G and Stefan S 2005 Effects of the atmospheric aerosol on the optical properties of cloud *Rom. Rep. Phys.* **57** 426–35

Jacobson M Z 2002 Control of fossil-fuel particulate black carbon and organic matter, possibly the most effective method of slowing global warming *J. Geophys. Res.* **107** 4410

Jacobson M Z 2003 Development of mixed-phase clouds from multiple aerosol size distributions and the effect of the clouds on aerosol removal *J. Geophys. Res.* **108** 4245

Kessler E 1969 *On the Distribution and Continuity of Water Substance in Atmospheric Circulation* (*Meteorol. Monogr.* vol 10) (Boston, MA: American Meteorological Society) p 84

Kim B, Miller M A, Schwartz S E, Liu Y and Min Q 2008 Importance of adiabaticity in evaluating aerosol indirect effect derived from ground-based remote sensing *J. Geophys. Res.* **113** D05210

Liu Y and Daum P H 2000 Spectral dispersion of cloud droplet size distributions and the parameterization of cloud droplet effective radius *Geophys. Res. Lett.* **27** 1903–6

Liu Y and Daum P H 2002 Indirect warming effect from dispersion forcing *Nature* **419** 580–1

Liu Y and Daum P H 2004 Parameterization of the autoconversion process. Part I: analytical formulation of the Kessler-type parameterizations *J. Atmos. Sci.* **61** 1539–48

Liu Y, Daum P H and Hallett J 2002 A generalized systems theory for the effect of varying fluctuation in cloud droplet size distributions *J. Atmos. Sci.* **59** 2279–89

Liu Y, Daum P H and Yum S 2006a Analytical expression for the relative dispersion of the cloud droplet size distribution *Geophys. Res. Lett.* **33** L02810

Liu Y, Daum P H, McGraw R and Miller M 2006b Generalized threshold function accounting for effect of relative dispersion on threshold behavior of autoconversion process *Geophys. Res. Lett.* **33** L11804

Liu Y and Hallett J 1997 The '1/3' power-law between effective radius and liquid water content *Q. J. R. Meteorol. Soc.* **123** 1789–95

Lohmann U and Feichter J 2005 Global indirect aerosol effects: a review *Atmos. Chem. Phys.* **5** 715–37

Lohmann U, Feichter J, Chuang C C and Penner J E 1999 Predicting the number of cloud droplets in the ECHAM GCM *J. Geophys. Res.* **104** 9169–98

Lohmann U and Lesins G 2002 Stronger constraints on the anthropogenic indirect aerosol effect *Science* **298** 1012–6

Lu M *et al* 2007 The marine stratus/stratocumulus experiment (MASE): aerosol-cloud relationship in marine stratocumulus *J. Geophys. Res.* **112** D10209

Lu M *et al* 2008 Aerosol-cloud relationships in continental shallow cumulus *J. Geophys. Res.* **113** D15201

Martin G M, Johnson D W and Spice A 1994 The measurement and parameterization of effective radius of droplets in the warm stratocumulus clouds *J. Atmos. Sci.* **51** 1823–42

Oreopoulos L and Platnick S 2008 Radiative susceptibility of cloudy atmospheres to droplet number perturbations: 2. Global analysis from MODIS *J. Geophys. Res.* **113** D14S21

Pawlowska H, Grabowski W W and Brienguier J-L 2006 Observations of the width of cloud droplet spectra in stratocumulus *Geophys. Res. Lett.* **33** L19810

Penner J E 2004 The cloud conundrum *Nature* **432** 962–3

Penner J E, Dong X and Chen Y 2004 Observational evidence of a change in radiative forcing due to the indirect aerosol effect *Nature* **427** 231–4

Peng Y and Lohmann U 2003 Sensitivity study of the spectral dispersion of the cloud droplet size distribution on the indirect aerosol effect *Geophys. Res. Lett.* **30** 1507

Peng Y, Lohmann U, Leaitch R, Banic C and Couture M 2002 The cloud albedo–cloud droplet effective radius relationship for clean and polluted clouds from RACE and FIRE.ACE *J. Geophys. Res.* **107** 4106

Peng Y, Lohmann U, Leaitch R and Kulmala M 2007 An investigation into the aerosol dispersion effect through the activation process in marine stratus clouds *J. Geophys. Res.* **112** D11117

Platnick S and Oreopoulos L 2008 Radiative susceptibility of cloudy atmospheres to droplet number perturbations: 1. Theoretical analysis and examples from MODIS *J. Geophys. Res.* **113** D14S20

Potter G L and Cess R D 2004 Testing the impact of clouds on the radiation budgets of 19 atmospheric general circulation models *J. Geophys. Res.* **109** D02106

Ramanthan V, Crutzen P J, Kiehl J T and Rosenfeld D 2001 Aerosols, climate, and the hydrological cycle *Science* **294** 2119–24

Rotstayn L D and Liu Y 2003 Sensitivity of the first indirect aerosol effect to an increase of cloud droplet spectral dispersion with droplet number concentration *J. Clim.* **16** 3476–81

Rotstayn L D and Liu Y 2005 A smaller global estimate of the second indirect aerosol effect *Geophys. Res. Lett.* **32** L05708

Schwartz S E 1988 Are global cloud albedo and climate controlled by marine phytoplankton? *Nature* **336** 441–5

Schwartz S E, Harshvardhan and Benkovitz C M 2002 Influence of anthropogenic aerosol on cloud optical depth shown in satellite measurements and chemical transport modeling *Proc. Natl Acad. Sci.* **99** 1784–9

Slingo T 1988 Can plankton control climate? *Nature* **336** 421

Sundqvist H 1978 A parameterization scheme for non-convective condensation including prediction of cloud water content *Q. J. R. Meteorol. Soc.* **104** 677–90

Telford J W 1996 Clouds with turbulence; the role of entrainment *Atmos. Res.* **40** 261–82

Twomey S 1974 Pollution and the planetary albedo *Atmos. Environ.* **8** 1251–6

Twomey S 1991 Aerosols, clouds and radiation *Atmos. Environ.* A **25** 2435–42

Wood R 2000 Parameterization of the effect of drizzle upon the droplet effective radius in stratocumulus clouds *Q. J. R. Meteorol. Soc.* **126** 3309–24

Yum S and Hudson J G 2005 Adiabatic predictions and observations of cloud droplet spectral broadness *Atmos. Res.* **73** 203–23

Zhao C *et al* 2006 Aircraft measurements of cloud droplet spectral dispersion and implications for indirect aerosol radiative forcing *Geophys. Res. Lett.* **33** L16809