Aircraft measurements of cloud droplet spectral dispersion and implications for indirect aerosol radiative forcing

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[1] Using a large amount of aircraft measurements of cloud droplet size distributions, the relationship between cloud spectral relative dispersion (ε) and cloud droplet number concentration (N_c) is studied. The results indicate that the value of ε varies between 0.2 to 0.8 when the cloud droplet number concentration is low (about 50 cm^{-3}), and converges toward a narrow range of 0.4 to 0.5 when the cloud number concentration is higher. Because the distribution of the cloud droplet size is an important parameter in estimating the first indirect radiative effect of aerosols on the climate system, the uncertainty in the corresponding radiative forcing can be reduced by 10–40% (depending on cloud droplet number density) under high aerosol loading.. This finding is important for improving climate change projections, especially for the regions where aerosol loading is high and continues to increase. **Citation:** Zhao, C., and et al. (2006), Aircraft measurements of cloud droplet spectral dispersion and implications for indirect aerosol radiative forcing, *Geophys. Res. Lett.*, *33*, L16809, doi:10.1029/2006GL026653.

1. Introduction

[2] Aerosols in the atmosphere linked to natural and anthropogenic emissions influence the Earth-Atmosphere system in several distinct ways [*Zhang et al.*, 2004]. For example, they directly or indirectly affect the Earth's radiation budget, and modify cloud and precipitation [*Charlson et al.*, 1987]. Clouds cover (on average) roughly half of the Earth's surface, and affect the energy balance for our planet. The Intergovernmental Panel on Climate Change [*International Panel on Climate Change*, 2001] has repeatedly identified clouds as the largest uncertainty in our current understanding of the climate system. Clouds themselves are composed of individual droplets or ice crystals, essentially all of which started as aerosol particles. A large part of the uncertainty in our ability to describe and model clouds in the climate system is due to the complexity of the processes controlling the interactions between aerosol particles and clouds. Many studies have shown that increasing aerosol concentrations result in an increase of cloud droplet numbers, as well as in a decrease of cloud droplet size and hence in enhanced cloud reflectivity (first indirect aerosol effect identified by *Twomey* [1974, 1991]; *Twomey et al.* [1984]). In this paper, we examine the relationship between cloud droplet number and the relative dispersion of the cloud droplet size distributions. This latter quantity is important in parameterizations of the radiative transfer through clouds, and in estimating the importance of the indirect effect of aerosols on the climate system [*Peng and Lohmann*, 2003].

[3] According to *Stephens* [1984] and *Twomey and Bohren* [1980], the cloud optical thickness τ and single scatter albedo ω_0, two quantities that need to be accurately determined to estimate the first indirect effect of aerosols, can be expressed by

$$\tau \approx \frac{3}{2} W r_e^{-1} \tag{1}$$

$$1 - \omega_0 = 1.7k\, r_e \tag{2}$$

if W represents the liquid water path, r_e the effective radius of cloud droplets, and k the complex part of the refractive index of water. These two equations show that the Twomey effect is strongly dependent upon the effective radius of cloud droplets (r_e). According to *Bower and Choularton* [1992], this effective radius is proportional to the volume weighted mean radius of the cloud droplets r_v and can be expressed as

$$r_e = \beta \left(\frac{3q}{4\pi \rho_w N_c} \right)^{1/3} = \beta r_v \tag{3}$$

where q is the cloud liquid water content, Nc the number concentration of cloud droplets, and ρ_w the water density. The proportionality factor β, also called the scaling factor [*Peng and Lohmann*, 2003], is specified in most climate models as a fixed parameter, whose value is based on observational data over different types of surfaces [e.g., *Lohmann and Roeckner*, 1996]. *Liu and Daum* [2000] have shown, however, that the scaling factor is dependent on the

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size distribution of cloud droplet, and derived the following relationship between β and the relative dispersion of the cloud size distribution ε, a dimensionless parameter that represents the ratio between the standard deviation of the size distribution and the mean droplet radius:

$$\beta = \frac{(1+2\varepsilon^2)^{2/3}}{(1+\varepsilon^2)^{1/3}} \quad (4)$$

Climate models that attempt to quantify the first indirect effect must therefore account for the influence of the cloud droplet spectrum on the scaling factor β. *Liu and Daum* [2002] have suggested that changes in the spectral distribution of cloud droplets resulting from air pollution could reduce significantly the cooling expected from the Twomey effect. In this study, we analyze aircraft measurements of the cloud droplet size distribution and the relationship between cloud droplet number concentration and relative dispersion. Based on these measurements, we discuss the uncertainty associated with the estimate of indirect radiative forcing resulting from the relative dispersion.

2. Results

[4] The size, number concentration, and distribution of cloud droplets were measured by a Forward Scattering Spectrometer probe (FSSP-100) from Particle Measuring System (PMS), Inc., which covers droplet sizes ranging from 3 to 47μm. The measurements took place at 4 locations in Asia (clean, polluted, and marine regions). The 4 geographical areas representing distinct conditions for aerosol loading are the following: (a) western China with no large anthropogenic aerosol emissions (Qianghai province, 34–37°N, 98–103°E) where 15 flights took place from 2001 to 2004; (b) central eastern China, a heavily polluted area with high emissions of anthropogenic aerosols (Hebei province, 35–40°N, 112–119°E), where 54 flights occurred between 1990 and 1993; (c) East China Sea near Japan, a region characterized by marine aerosol conditions and influenced by downwind transport of aerosol pollutants (28–31°N, 127–131°E) where 23 flights took place during the 2001 to 2003 period; and (d) the heavily polluted Beijing metropolitan area (37–41°N, 113–120°E) with 14 flights in 2004 and 29 flights in 2005.

[5] Figure 1 shows the relationship between cloud droplet number concentration (N_c) and the relative dispersion factor (ε) derived from the measurements. There is a clear indication that at all locations under consideration, the relative dispersion of cloud droplets is characterized by a wide range of values when the number concentration of the cloud droplets is low. With an increase in the concentration, the relative dispersion decreases and converges toward a very narrow range between 0.3 and 0.5. For example, in the clean region of Qinghai, the relative dispersion varies from 0.2 to 0.8 when cloud droplet concentrations are close to 50 cm^{-3}, and converges to about 0.3 to 0.5 when the concentration becomes as large as 400 cm^{-3}. In the marine region (Asian Atmospheric Particulate Environment Change Studies, APEX), the relative dispersion (ε) ranges from 0.2 to 0.8 when the concentration is equal to 50 cm^{-3}, and converges to about 0.4 at a concentration reaching 800 cm^{-3}. In the polluted regions of Hebei, the relative dispersion varies from nearly 0.0 to 0.8 when the cloud droplet concentration is equal to 50 cm^{-3}, and converges to about 0.3 to 0.5 when the concentration reaches 250 cm^{-3}. In the other polluted region (Beijing), the relative dispersion varies between 0.0 and 0.8 for cloud droplet concentrations of 50 cm^{-3}, and converges to about 0.3 to 0.5 for concentrations of 800 cm^{-3}. The convergence of the relative dispersion at high values of N_c is similar at the four different locations. In particular, there is no clear distinction in the value of the converging relative dispersion (ε) between clean and polluted regions.

[6] The cloud droplet size dispersion is jointly determined by aerosol loading and cloud dynamical processes. For similar dynamical conditions, it has been suggested that higher aerosol concentrations lead to an increased dispersion [*Liu and Daum*, 2002]. On the other hand, the dynamical effects (updraft and turbulent processes) may cause a negative dispersion relation [*Liu et al.*, 2006], that is, a decreased dispersion relation with increasing updraft or turbulence. The combined aerosol (positive relation) and dynamical effects (negative relation) could lead to the observations presented here of an overall convergence in the relative dispersion of cloud droplets at high number concentrations. It is also plausible that the presence of elevated aerosol concentrations impacts the cloud dynamics because of cloud invigoration [*Williams et al.*, 2002; *Andreae et al.*, 2004; *Koren et al.*, 2005], which in turn produces a feedback on the microphysical processes. *Ferek et al.* [2002] present observations from the Monterey Area Ship Tracks (MAST) experiment showing a decrease in the concentration of drizzle-sized droplets in a polluted ship track relative to the surrounding unperturbed clouds. The possible explanation for this observation is that the coalescence processes that generated drizzle-sized droplets in the unperturbed clouds were less effective in the polluted ship track. *Lu and Seinfeld* [2006] using three-dimensional large-eddy simulations also found that the relative dispersion decreases with increasing aerosol number concentration.

3. Implication for the Estimate of Climate Forcing

[7] As shown in Equations 1 to 4, the relative dispersion has an important impact on the estimated indirect radiative forcing (Figure 2). In order to calculate the effect of relative dispersion variation on the first indirect radiative forcing, we consider three different conditions (N_c = 50, 100, and 300 cm^{-3}, respectively) (Table 1). The result shows that, when cloud droplet concentrations are low (50 cm^{-3}), the range in the relative dispersion is very large, and the uncertainties in the estimate of the cloud optical depth and in the absorption coefficient are 29% and 41%, respectively. When cloud droplet concentrations are of the order of 100 cm^{-3}, the range in the relative dispersion is reduced, and the uncertainty in the estimate of the cloud optical depth and absorption coefficient becomes 16% and 19%, respectively. When cloud droplet concentrations are as high as 300 cm^{-3}, the range in the relative dispersion (ε) converges and the uncertainty in the estimate of the cloud optical depth and absorption coefficient is further reduced to 8% and 9%, respectively. The work by *Liu and Daum* [2002] suggested

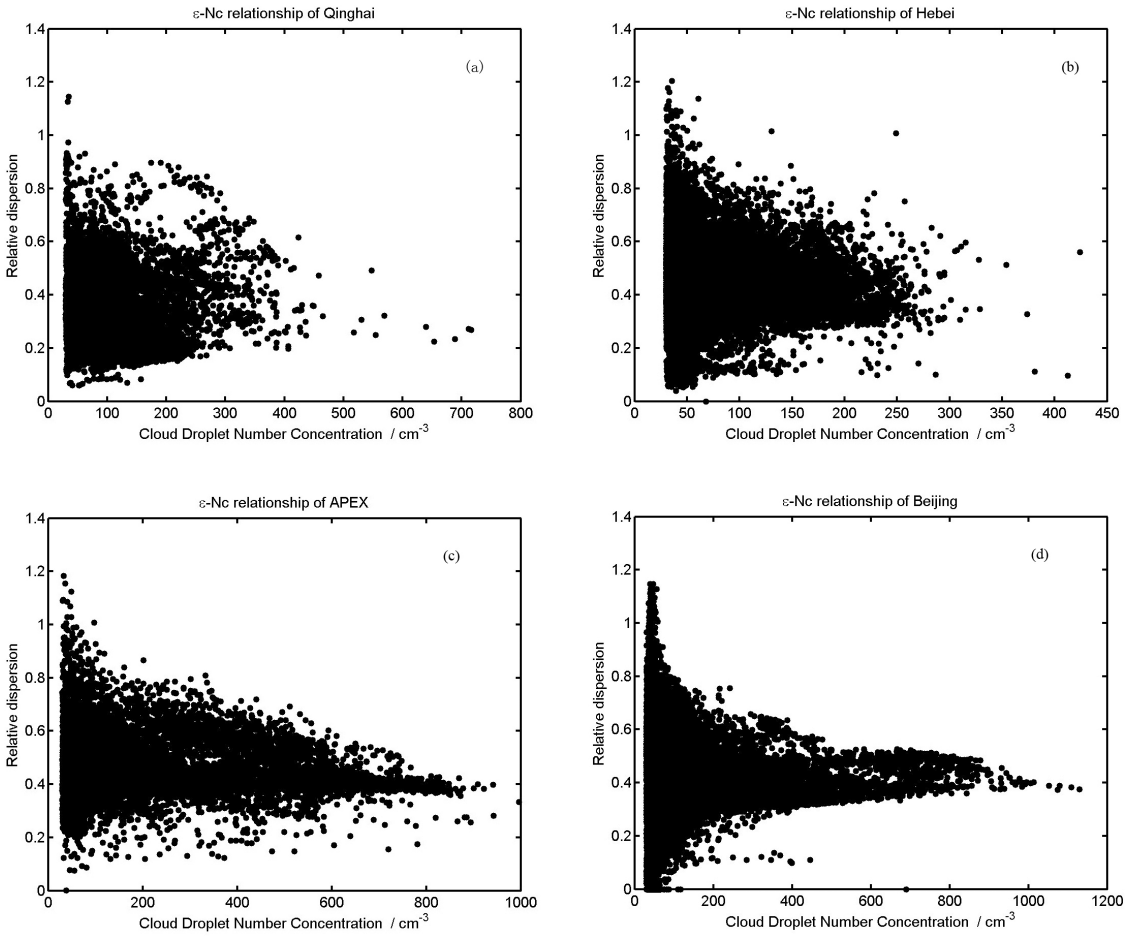

Figure 1. Relationship between the relative dispersion (ε) of cloud droplets and their number concentration at 4 locations (a) in the Qianghai Province during 2001–2004, (b) in the Hebei Province during 1991–1993, (c) over the Yellow Sea of the eastern coast of China during the APEX project, (d) in the Beijing region during 2004–2005. Data corresponding to Nc less than 30 #/cm^3 are not represented.

that the net aerosol effect (i.e., excluding the dynamical processes) lead to an increasing dispersion relation. We have demonstrated in this work that the combined aerosol and dynamical processes result in a convergence in the dispersion relation at high cloud droplet concentrations. Furthermore, cloud invigoration [*Williams et al.*, 2002; *Andreae et al.*, 2004; *Koren et al.*, 2005] by polluted aerosols may produce an additional negative feedback on the dispersion relation. Numerous studies have documented an increase in the number concentration of cloud droplets because of high anthropogenic aerosol concentrations [*Twomey*, 1974; *Charlson et al.*, 1992]. Because ε is an important parameter in estimating the indirect radiative effects of aerosols, the present result implies that under high aerosol conditions the uncertainty in the indirect radiative forcing of aerosol is significantly reduced. This finding can be important in improving our understanding of climate change, especially in the regions where aerosol loading is high and continues to increase.

4. Summary

[8] A large number of aircraft measurements of cloud droplet size distributions performed at several locations (clean, polluted, and marine regions) in Asia were used to analyze the relationship between cloud droplet number concentration (N_c) and the relative dispersion. The results show that the range in the relative dispersion of cloud droplet is large (0.2 to 0.8) when the number concentration

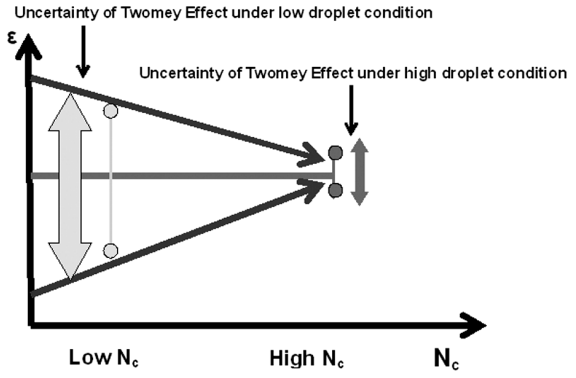

Figure 2. A schematic description of the measured relationship between the dispersion of cloud droplets and their number concentrations, showing that under low N_c conditions, the uncertainty in the quantitative estimate of the "Twomey Effect" is substantially larger than that under high N_c conditions.

Table 1. Estimation of the Impact of Relative Dispersion on Cloud Radiative Properties[a]

Cloud Droplet Number Density, cm^{-3}	Relative Dispersion, ε	$\frac{r_{e1}}{r_{e0}}$	$\frac{\tau_1}{\tau_0}$	$\frac{(1-\varpi_0)_1}{(1-\varpi_0)_0}$
50	0.2∼0.8	1.41	0.71	1.41
100	0.3∼0.6	1.19	0.84	1.19
300	0.35∼0.5	1.09	0.92	1.09

[a]Subscript 1 refers to the upper limit and 0 to the lower limit of the relative dispersion (ε).

of cloud droplets is low. When the number concentration increases, the relative dispersion of cloud droplet converges toward a narrow range of 0.3 – 0.5. This convergence is similar in clean and polluted regions. Because the first indirect radiative forcing increases with the relative dispersion, the wide range of the relative dispersion for low cloud droplet concentrations produces a large uncertainty in the estimation of the indirect radiative forcing. However, for high cloud droplet concentrations, the uncertainty in the estimation of indirect radiative forcing is significantly reduced due to the convergence of the relative dispersion. This finding from the analysis of the aircraft measurements is important for our ability to model indirect radiative forcing, especially for regions where the aerosol pollutants are high and continue to increase. In these regions, the uncertainty in the calculation of the "Twomey Effect" could be significantly reduced, leading to a better quantitative estimate of the indirect radiative forcing of aerosols on climate.

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