



The role of adiabaticity in the aerosol first indirect effect

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[1] Aerosol indirect effects are the most uncertain of the climate forcing mechanisms that have operated through the industrial period. Several studies have demonstrated modifications of cloud properties due to aerosols and corresponding changes in shortwave and longwave radiative fluxes under specific cloud conditions, but some recent studies have indicated that cloud dynamical processes such as entrainment-mixing may be the primary modulator of cloud optical properties in certain situations. For example, day-to-day variations of the cloud drop effective radius (r_e) determined from the ground-based remote sensing at the Southern Great Plains were found to be weakly associated with the variations in aerosol loading as characterized by its light-scattering coefficient at the surface, implying that other processes were impacting the cloud radiative properties. To study these other impacts, we extend a previous study to investigate the role of changes in liquid water path (LWP) and r_e in single layer stratiform clouds that are induced by entrainment-mixing processes and their effects on cloud radiative properties. We quantify the degree of entrainment-mixing in terms of the adiabaticity defined as the ratio of the observed cloud liquid water path to the corresponding adiabatic value. The cloud optical depth is, as expected, governed primarily by LWP, but that adiabaticity is the next most influential factor. In contrast, r_e is found to be equally sensitive to adiabaticity and LWP. In adiabatic clouds the aerosol first indirect effect is clearly observed and related to independent measures of aerosol loading. In sub-adiabatic clouds the aerosol first indirect effect is not readily observed; this may in some circumstances be due to interference from heterogeneous mixing processes that change the droplet number density in a manner that attenuates the effect.

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1. Introduction

[2] Clouds are an important regulator of the Earth's radiation budget. Measurements of the Earth Radiation Budget Experiment [Collins *et al.*, 1994] indicate that small changes in the macrophysical and microphysical properties of clouds have significant effects on climate, and a 5% increase in shortwave cloud forcing would compensate the increase in greenhouse gases between the years 1750–2000 [Ramaswamy *et al.*, 2001]. Meanwhile, substantially increasing aerosols during the industrial period may have affected global climate by altering cloud microphysical and radiative properties, so-called the aerosol indirect radiative forcing, which remains one of the largest uncertainties in

climate modeling and climate change prediction [IPCC, 2007]. The root of this uncertainty is a lack of fundamental understanding of the feedbacks of external forcings on clouds and adequate parameterizations of important processes [Lohmann and Feichter, 2005; Lohmann *et al.*, 2007].

[3] Increases in anthropogenic sources of cloud condensation nuclei (CCN) may increase cloud albedo by increasing the concentration and reducing the size of cloud droplets, in the absence of other mitigating factors [Twomey, 1977; the aerosol first indirect effect], which has strong observational support [Han *et al.*, 1998; Brenguier *et al.*, 2000; Nakajima *et al.*, 2001; Schwartz *et al.*, 2002; Kim *et al.*, 2003; Feingold *et al.*, 2003, 2006]. Additionally, a reduction in the size of cloud droplets could suppress precipitation and result in increased cloud lifetimes (aerosol second indirect effect) [Albrecht, 1989], which has been supported by limited observations [Rosenfeld, 2000; Kaufman *et al.*, 2005; Rosenfeld *et al.*, 2006]. While these mechanisms apply uniformly to the cloud droplet size distribution and uniformly reduce droplet size, recent studies have shown that an increase in aerosol loading leads to an increase in the relative dispersion of the size distribution of cloud droplets as well, and the enhanced dispersion (dispersion effect) acts to diminish the cooling effect associated with

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Table 1. Summary of Primary Instrumentation and Value-Added Products^a

Instrument	Measured Quantities	Comments	Temporal Resolution	References
MFRSR (Multifilter Rotating Shadowband Radiometer)	cloud optical depth (τ_c)	Measures direct and total-horizonal irradiances at 415 nm.	20 s	<i>Min and Harrison</i> [1996]
MWR (Microwave Radiometer)	liquid water path (LWP)	uses microwave brightness temperature	20 s	<i>Liljegren et al.</i> [2001]
Nephelometer	scattering coefficient (σ_{sp})	At 450, 550, 700 nm for the size of aerodynamic diameter less than 1 μm	1 min	<i>Sheridan et al.</i> [2001] http://www.cmdl.noaa.gov/aero/data/
ARSCL (Active Remotely Sensed Cloud Locations)	cloud boundaries	Best estimates from MMCR, Ceilometer and Lidar	10 s	<i>Clothiaux et al.</i> [2000]
BBSS (Balloon-Borne Sounding System)	temperature (T), Relative humidity (RH), wind speed (WS)	Sounding at 6 h intervals (3 h interval for intensive observation period)	10 s	www.arm.gov/docs/instruments/static/bbss.html

^aValue-added products refer to data sets resulting from assimilation and analysis of data from multiple instruments. Data are from www.archive.arm.gov except where indicated.

the aerosol first indirect effect [*Liu and Daum*, 2002; *Rotstajn and Liu*, 2003; *Peng and Lohmann*, 2003]. The dispersion effect has been further confirmed by subsequent theoretical studies [*Liu et al.*, 2006], parcel model simulations [*Yum and Hudson*, 2005; *Peng et al.*, 2007], and field campaigns [*Lu et al.*, 2007; *Daum et al.*, 2007].

[4] Several observational strategies have been used to study aerosol indirect effects. One approach combines remote sensing of cloud radiative properties with in situ characterization of cloud microphysics during intensive, but short-lived, field campaigns [*Radke et al.*, 1989; *Brenguier et al.*, 2000]. An alternative approach uses satellite remote sensing to systematically characterize the aerosol effects on global basis, albeit with a limited spatial resolution [*Han et al.*, 1998; *Nakajima et al.*, 2001; *Breon et al.*, 2002]. Yet another approach uses long-term measurements obtained with ground-based remote sensors to examine the relation of aerosols with cloud microphysics at specific sites [*Feingold et al.*, 2003; *Kim et al.*, 2003; *Garrett et al.*, 2004].

[5] In previous work [*Kim et al.*, 2003], we have found in measurements in north central Oklahoma that the cloud drop effective radius (r_e) in persistent single-layer stratiform clouds on different days was weakly associated with the variation in aerosol loading characterized by its light-scattering coefficient at the surface; this association is supportive of the aerosol first indirect effect, but there was substantial scatter in the observations, which was attributed to unquantified meteorological influences such as drizzle and entrainment. These processes lead to the subadiabatic liquid water content profiles frequently observed in the continental and marine stratus clouds [*Kim et al.*, 2005; *Chin et al.*, 2000; *Miller et al.*, 1998; *Albrecht et al.*, 1990]. Other factors that may have contributed to this scatter involve in the cloud droplet nucleation process, which is dependent on aerosol characteristics (chemical composition and size distribution etc) [*Nenes et al.*, 2002; *Chuang et al.*, 2002; *Breon et al.*, 2002] and the updraft velocity; the latter implies a link to static stability [*Feingold et al.*, 1999; *Kim et al.*, 2003; *Leitch et al.*, 1996]. Recently *Shao and Liu* [2006] demonstrated the strength of the aerosol first indirect effect is about half that estimated by many previous investigators, and attributed the difference to the evaporation associated with entrainment-mixing processes. Especially the LWP is known to play a major role in determining the clouds' radiative forcing [*Dong et al.*, 2002; *Kim et al.*,

2005]. The variation of LWP due to these non-adiabatic processes would induce the uncertainty in estimating aerosol indirect effect.

[6] Ideally, all the controlling factors described above should be considered in order to better understand aerosol indirect effect. The problem is that these mechanisms appear to be dependent upon each other, and accounting for them is impossible with the current understanding of aerosol indirect effect. Therefore in the present study, the subadiabatic character of the clouds, or adiabaticity, is used to characterize the entrainment-mixing processes, and attempts are made to determine the extent to which these properties affect cloud optical properties, apart from the aerosol first indirect effect. For this purpose, we extend the previous one-year study to include 3-years (1999–2001) of data collected in relatively uniform stratus clouds by ground-based remote sensing at the Department of Energy Atmospheric Radiation Measurement (ARM) Southern Great Plain (SGP) site in north central Oklahoma.

[7] This study is composed of a section that details the data and analysis methods followed by a section that develops an analytical analysis of the relationship between adiabaticity and cloud optical properties under assumption that the clouds are homogeneously mixed. We apply this analytical formalism to data from the ground-based remote sensors with the goal of separating aerosol influences on cloud microphysics from meteorological factors and other cloud dynamics. Finally, we extend the analytical treatment to encompass the details of the mixing process in an effort to explain some of the features of the observed clouds.

2. Data and Methods

[8] The methods and data are similar to those used by *Kim et al.* [2003], except that the analysis period is extended by two years to encompass the period from 1999 to 2001. The primary instruments used in this study at the SGP site (97.48°W, 36.61°N) are in Table 1.

[9] Cloud boundaries are retrieved every 10 s from a combination of data from active remote sensors, mainly a Millimeter-Wave Cloud radar (35 GHz), a micro-pulse lidar, and a ceilometer [*Clothiaux et al.*, 2000]. The height resolutions of radar, lidar, and ceilometer are 45 m, 30 m, and 15 m, respectively. Liquid water path (LWP) is determined by a microwave radiometer (MWR), which measures

time series of column-integrated liquid water based on the microwave emissions of liquid water molecules mainly at 31.4 GHz (vertically pointing; field of view 4.5° half width). The root mean square accuracies for cloud LWP retrievals are about 20 g m⁻² for LWP below 200 g m⁻² and 10% for LWP above 200 g m⁻² [Liljegren *et al.*, 2001].

[10] Cloud optical depth (τ_c) is measured with a Multi-Filter Rotating Shadowband Radiometer (MFRSR) at 415 nm; use of this single MFRSR channel minimizes sensitivity to surface reflectance. Use of the MFRSR limits the analysis to completely overcast conditions because of the wide field-of-view of the instrument (cosine-weighted hemispheric sensor for an effective half width of 120°); however restricting measurements to full overcast also minimizes effects of photon diffusion in clouds. *Min and Harrison* [1996] and *Min et al.* [2001] have developed a family of inversion methods to infer cloud optical properties from MFRSR and MWR. The cloud radiative properties are parameterized in terms of a cloud-average drop effective radius r_e , and total liquid water path, based on Mie theory [Hu and Stamnes, 1993]. In view of the dependence of Mie scattering properties on cloud drop radius, the effective radius is retrieved by an iterative procedure [Min and Harrison, 1996] that accounts for the variation of extinction efficiency with r_e . A sensitivity study illustrates that a 13% uncertainty in observed liquid water path (LWP, 20 g m⁻²) results in 1.5% difference in retrieved cloud optical depth and 12.7% difference in inferred cloud effective radius, on average [Min *et al.*, 2003]. The uncertainty of the LWP measured by the microwave radiometer (MWR) is the major contributor to the uncertainty of retrieved cloud effective radius. In terms of remote sensing, however, the derivative of r_e is independent of LWP.

[11] In the end, five-minute averaged τ_c , LWP, and r_e values are used to characterize the cloud properties, which permits the narrow field of view measurement of LWP to be more compatible with the wide field of view measurement of τ_c by the MFRSR, thereby ameliorating field-of-view issues.

[12] Implicit in this measurement approach is the requirement that the clouds approximate a plane-parallel geometry. Hence we restrict our examination to widespread, low-level, non-precipitating, single-layer liquid water clouds without interference from higher-level clouds. Because we attempt to relate properties of aerosols measured at the surface with those influencing the properties of the cloud, we restrict cloud top height to be a maximum of 2-km above the ground. We also limit our analysis to well-mixed conditions, which are most likely to couple aerosol at the surface to that influencing the cloud [Feingold *et al.*, 2006].

[13] Because measurements of cloud condensation nuclei (CCN), which represent the fraction of all aerosols that may nucleate cloud droplets, were not available, measurements of light scattering coefficient of accumulation-mode aerosols with the aerodynamic diameter of 0.1 to 1.0 μm by integrating nephelometers [Sheridan *et al.*, 2001] are used as a CCN proxy for examination of aerosol influence. The use of light scattering coefficient as a proxy is generally defensible because the fraction of the total aerosol load that serves as CCN is typically found to be a relatively constant fraction of the total aerosol load, which is measured by the integrating nephelometers. Two nephelometers (TSI Model

3563), one dry and one humidified, measure the aerosol light scattering coefficient, σ_{sp} , as a function of relative humidity (RH) at three visible wavelengths (nominally 450, 550, and 700 nm). We use measurements at the 550 nm wavelength and low RH $\leq 40\%$ to represent the light scattering coefficient of the dry aerosol. The TSI nephelometers are preceded by two impactors whose size cut switches every 6 min; a 10- μm impactor removes particles with aerodynamic diameter larger than 10 μm and a 1- μm impactor removes super micron size particles. In this study, we used scattering coefficient of accumulation-mode aerosols only, and thus measurements at 550-nm are available as 1-minute averages for five 6-minute intervals per hour, which are finally interpolated to 5-minute averages for comparisons with the cloud drop effective radius.

3. Effects of Cloud Adiabaticity

3.1. Adiabaticity and the Theoretical Derivation

[14] The propensity of layer clouds to maintain sub-adiabatic integrated liquid water path (W) [Chin *et al.*, 2000; Kim *et al.*, 2005] motivates us to determine analytically the probable impacts of a reduction of liquid water on the cloud optical properties, relative to purely adiabatic clouds that are permitted no mixing or drizzle. In order to examine the effect of entrainment-mixing processes on cloud optical properties, we define the adiabaticity, α , as

$$\alpha = \frac{W}{W_a}. \quad (1)$$

where the subscript “a” in W is referred to the adiabatic value. In addition, to isolate the impacts of entrainment mixing using α as a proxy, we exclude any instances of the following significant drizzle; cloud reflectivity measured by the 35-GHz cloud radar was greater than -15 dBz or the cloud base measured by this radar was lower than the optical cloud base, as measured by a laser ceilometer, which indicates drizzle in the subcloud layer [Kim *et al.*, 2005]. The adiabatic LWP (W_a) can be defined within the model of an adiabatic cloud for which LWC (L) increases linearly with height above the cloud base [Albrecht *et al.*, 1990]. For an isolated cloudy parcel experiencing adiabatic ascent in a shallow layer W_a can be written [Albrecht *et al.*, 1990 and Rogers and Yau, 1989] as

$$W_a = \frac{\bar{\rho}}{2} \Gamma_l \Delta z^2 \quad (2)$$

$$\Gamma_l = \frac{(\varepsilon + q_s)q_s l_v}{R_d T^2} \Gamma_w - \frac{g w_s p}{(p - e_s) R_d T} \quad (3)$$

$$\Gamma_w = -\frac{dT}{dz} = \Gamma_d \left(\frac{1 + l_v/R_d T}{1 + l_v^2 \varepsilon w_s / R_d C_p T^2} \right) \quad (4)$$

where $\Gamma_l = dw_l/dz$ is the vertical variation of the adiabatic liquid water mixing ratio, w_l ; $\bar{\rho}$ is the average air density, $\varepsilon = 0.622$, C_p is the specific heat content at constant pressure, q_s is the saturation mixing ratio of water vapor, and Δz is the cloud physical thickness, l_v is the latent heat of vaporization,

R_d is the specific gas constant for dry air, e_s is the saturation vapor pressure, w_s is saturation mixing ratio of water vapor, and, Γ_d and Γ_w are the dry and moist adiabatic lapse rate, respectively. Γ_l is calculated using the mean temperature T and pressure P at the level of the cloud from the available soundings. Hence W_a can be determined from the vertical profiles of thermodynamic structures and cloud thickness.

[15] Considering a cloud of depth Δz and combining (1) and (2) yields

$$W = \alpha W_a = \frac{1}{2} \bar{\rho} \alpha \bar{\Gamma}_l \Delta z^2. \quad (5)$$

and substitution of (5) into the definition of effective radius of cloud droplets [Hansen and Travis, 1974] yields

$$\bar{r}_e = \frac{3}{4} \frac{\bar{\rho}}{\rho_l} \frac{\bar{\Gamma}_l \alpha \Delta z^2}{\tau_c}. \quad (6)$$

where ρ_l is the average liquid water density.

[16] The effective radius, which is a key property of a cloud in characterizing the optical properties of the cloud [Hansen and Travis, 1974], can thus be retrieved by ground based remote sensors within the underlying assumptions that the radiation extinction efficiency, $Q_e(r, z) \sim 2$ (large particle extinction limit) and that the scattering is conservative. The over bar indicates that r_e is a vertically integrated quantity for the entire cloud. Since W can be measured with a multichannel MWR and τ_c with a MFRSR, \bar{r}_e can be accordingly derived during the daytime in single layer liquid clouds.

[17] Introducing N_{cd} , the number concentration of cloud drops at a given height within a cloud z , $N_{cd}(z) = \int n(r, z) dr$, we note that if $N_{cd}(z)$ is a constant independent of z , $N_{cd}(z) \approx N_{cd}$, then $\tau_c = 2\pi N_{cd} \int \int r^2 dr dz$, so information about the cloud droplet spectrum is required to proceed.

[18] In order to represent this spectrum, we assume that the cloud drop size distribution can be represented by the k -th moment of a lognormal distribution given by $\langle r^k \rangle = r_m^k \exp(k^2 \bar{\sigma}_r^2 / 2)$, where k is the desired moment, and $\bar{\sigma}_r$ is a measure of the relative dispersion of cloud droplet size distribution, the standard deviation of the logarithm of r , which is assumed to be constant for simplicity in this study. With these substitutions, the cloud optical depth is given by

$$\tau_c \cong 2\pi N_{cd} \bar{r}_m^2 \exp(2\bar{\sigma}_r^2) \Delta z, \quad (7)$$

where \bar{r}_m is the mean of droplet mode radius. Combining the LWP (W) expressed in terms of the third moment of the cloud droplet size distribution and a moment generation function of \bar{r}_m leads to

$$W = \frac{4\pi}{3} \rho_l \bar{r}_m^3 N_{cd} \exp\left(\frac{9}{2} \bar{\sigma}_r^2\right) \Delta z, \quad (8)$$

and equating (5) and (8), and solving for \bar{r}_m , yields

$$\bar{r}_m = \left[\frac{3}{8\pi} \frac{\bar{\rho}}{\rho_l} \frac{\bar{\Gamma}_l \alpha \Delta z}{N_{cd} \exp(9\bar{\sigma}_r^2/2)} \right]^{1/3}. \quad (9)$$

Substituting this expression into (7) produces

$$\tau_c = 2\pi \left(\frac{3}{8\pi} \frac{\bar{\rho}}{\rho_l} \right)^{2/3} \left[\frac{\bar{\Gamma}_l^2 N_{cd} \alpha^2 \Delta z^5}{\exp(3\bar{\sigma}_r^2)} \right]^{1/3} \quad (10)$$

and upon substitution into (6) we obtain

$$\bar{r}_e = \exp(\bar{\sigma}_r^2) \left[\frac{A \alpha \bar{\Gamma}_l \Delta z}{N_{cd}} \right]^{1/3} \quad (11)$$

where A is a constant, which is given by

$$A = \frac{3}{8\pi} \frac{\bar{\rho}}{\rho_l}. \quad (12)$$

[19] The above derivation is based on assumption of the homogeneous mixing in that cloud properties through the entire cloud are assumed to be uniformly impacted by the reduction in liquid water path denoted by α . As we will note later, this assumption has important implications in the interpretation of the observations.

[20] Examination of the ARM data archive for three years (1999–2001) yielded fourteen analysis days for which the screening criteria were satisfied for 2 h or more such that the cloud should be widespread, low-level, non-precipitating, single-layer liquid water only without interference from higher-level clouds and also located below 2-km above the ground with the well-mixed condition. Comparisons of the observed LWP with the adiabatic LWP are made for the 6 cases during which there were vertical soundings that permit the calculation of W_a by (2). Comparisons (Figure 1) reveal that most of the cases were subadiabatic except for 1999/03/23, which was nearly adiabatic. The subadiabatic cases have W/W_a ranging from 0.34 to 0.51; these values are somewhat lower than the mixing parameter ($\alpha = 0.6$ – 0.7) used by Boers and Mitchell [1994] in a maritime cloud system. The vertical variation of the adiabatic liquid water mixing ratio with height, Γ_l , ranged from 1.32 to 1.93 g kg⁻¹ km⁻¹ with an average value of 1.63 g kg⁻¹ km⁻¹; this value is somewhat lower than that (2.10 g kg⁻¹ km⁻¹) reported in marine boundary layer clouds off the Southern California coast [Albrecht et al., 1990].

3.2. Sensitivity of r_e and τ_c to Adiabaticity

[21] In order to investigate the dependence of r_e on α (Figure 2a), we classify the data by cloud thickness with the separation of 200–1000 m into thin and thick cloud regimes by its mode value of 600 m. These data show an increase in r_e with an increase in α , and segregation by cloud thickness. According to (11), if the mixing is homogenous we expect changes in r_e as a response to changes in α , Δz , or σ_r . Note that the relationship of r_e to α is not as significant as that of r_e to Δz (Table 2). We cannot evaluate the relationship between r_e and σ_r because we have no means of directly measuring σ_r .

[22] To quantify the sensitivity of (11), we take its logarithm and its derivative, which yields

$$\frac{\delta \bar{r}_e}{\bar{r}_e} = \delta \bar{\sigma}_r^2 + \frac{1}{3} \frac{\delta \alpha}{\alpha} + \frac{1}{3} \frac{\delta \Delta z}{\Delta z} - \frac{1}{3} \frac{\delta N_{cd}}{N_{cd}} \quad (13)$$

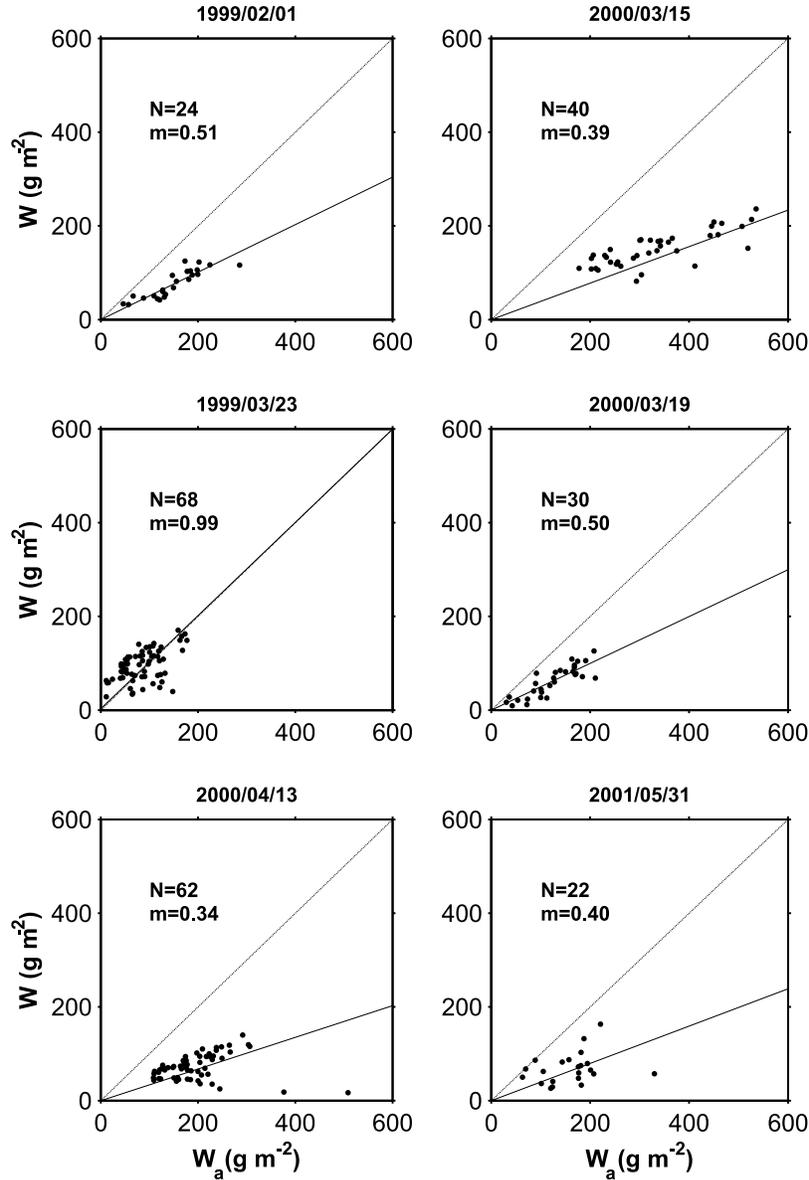


Figure 1. Comparisons of the observed LWP with the adiabatic LWP for the analysis period on 6 days. Solid line denotes the regression line of the observed vs. the adiabatic LWP. N denotes the number of data and m is the slope of the regression.

[23] Equation (13) assumes a homogeneous mixing and a uniformly change of droplet size through the whole droplet spectrum. If we assume $\bar{\sigma}_r$, Δz , and N_{cd} to be constant, a relative change in r_e is proportional to a relative change in α with the slope of $1/3$. The dashed reference line (Figure 2a) denotes r_e for $N_{cd} = 288 \text{ cm}^{-3}$ and $\bar{\sigma}_r = 0.35$, values representing average values in continental clouds by Miles *et al.* [2000], Γ_l of $1.63 \text{ g kg}^{-1} \text{ km}^{-1}$, an average value obtained from 6 soundings of 14 candidates of this study, and the cloud thickness of 500 m, which is the average cloud thickness. Neither the thick or thin cloud subgroups appear to possess the requisite $1/3$ slope suggested by (11), though the thick clouds appear to more closely approximate the analytical formulation. We also note a lack of slope in

the thin cloud regime, implying that r_e could be subject to variability in $\bar{\sigma}_r^2$.

[24] We examine the dependence of r_e and Δz in more detail by classifying the data into different bins of α : 0.1–0.8 (subadiabatic regime; black dots in Figure 2b) and 0.8–1.2 (adiabatic regime; gray dots in Figure 2b). A reference line denotes r_e for the same conditions listed above, but for $\alpha = 0.7$. The sensitivity of r_e to Δz seems slightly stronger, as indicated by the closer proximity to the predicted $1/3$ slope. The stronger association of r_e to Δz is shown in the adiabatic regime (slope = 0.42 and square of Pearson correlation coefficient $R^2 = 0.48$, in Table 2), which is comforting because r_e increases systematically in an adiabatic ascent. This relationship of r_e to Δz could be due to physical connections, since the r_e represents column aver-

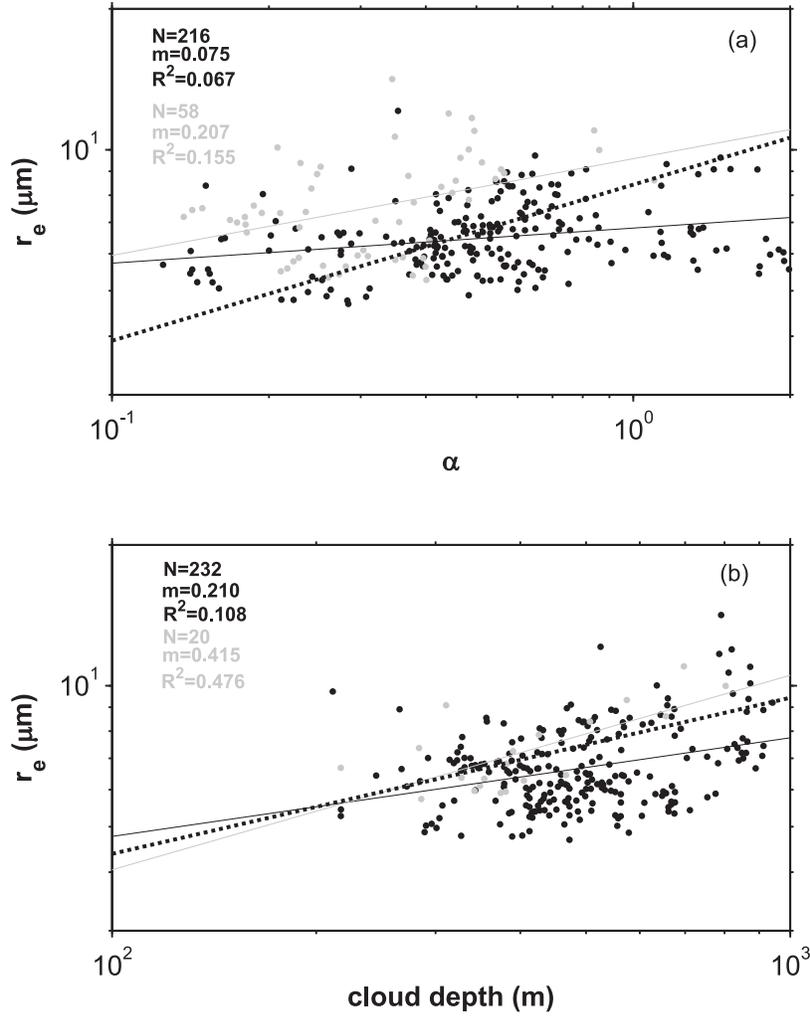


Figure 2. (a) Effective radii of cloud droplets as a function of α with the different cloud height; the dot in black color belongs to the thin cloud whose thickness ranges between 200 and 600 m, whereas the dot in gray color to the thick cloud between 600 and 1000 m. Similarly (b) r_e as a function of the cloud thickness with the different adiabaticity; the black dot belongs to subadiabatic cloud and the gray dot to adiabatic cloud. The subadiabatic cloud denotes α of 0.1 – 0.8 and the adiabatic cloud indicates α of 0.8–1.2. The thick dashed line of each figure indicates r_e for the indicated N of 288 cm^{-3} , Γ_l of $1.63 \text{ g kg}^{-1} \text{ km}^{-1}$, and the cloud thickness of 500 m and α of 0.7, respectively.

aged effective radius and the derivative of r_e is independent of LWP in terms of remote sensing. We conclude from this analysis that r_e is weakly associated with α , a proxy for mixing and only dependent upon cloud thickness in spite of the assumption of constant droplet dispersion. The poor correlation of r_e with α suggests that the homogeneous mixing is not the major mechanism of entrainment-mixing processes in these clouds.

[25] The derivative of the cloud optical depth (10) can be expanded with the partial derivative of α , Δz , and N as,

$$\frac{\delta\tau_c}{\tau_c} \propto \delta\bar{\sigma}_r^{-2} + \frac{1}{3} \frac{\delta N_{cd}}{N_{cd}} + \frac{2}{3} \frac{\delta\alpha}{\alpha} + \frac{5}{3} \frac{\delta\Delta z}{\Delta z}. \quad (14)$$

[26] This equation specifies that the relative change in Δz , or equivalently LWP, is the major contributor to the

relative change in τ_c , which is also supported by the observations from the ground-based remote sensors (Figure 3). Here the reference line denotes the same conditions listed above, but with $\Delta z = 500 \text{ m}$ rather than

Table 2. Summary of Data Numbers, the Slope, and Correlation Coefficient of r_e Versus α (Upper) and r_e Versus Δz (Lower) for Each Δz Bin and α Bin, Respectively^a

	r_e versus α		r_e versus Δz	
	Δz bins		α bins	
Effective Radius	Thin	Thick	Subadiabatic	Adiabatic
N (data number)	216	58	232	20
Slope	0.07	0.21	0.21	0.42
R^2	0.07	0.16	0.11	0.48

^aThe cloud thickness of 200–600 m belongs to the thin regime, and 600–1000 m to thick regime. The subadiabatic cloud denotes α of 0.1–0.8 and the adiabatic cloud indicates α of 0.8–1.2.

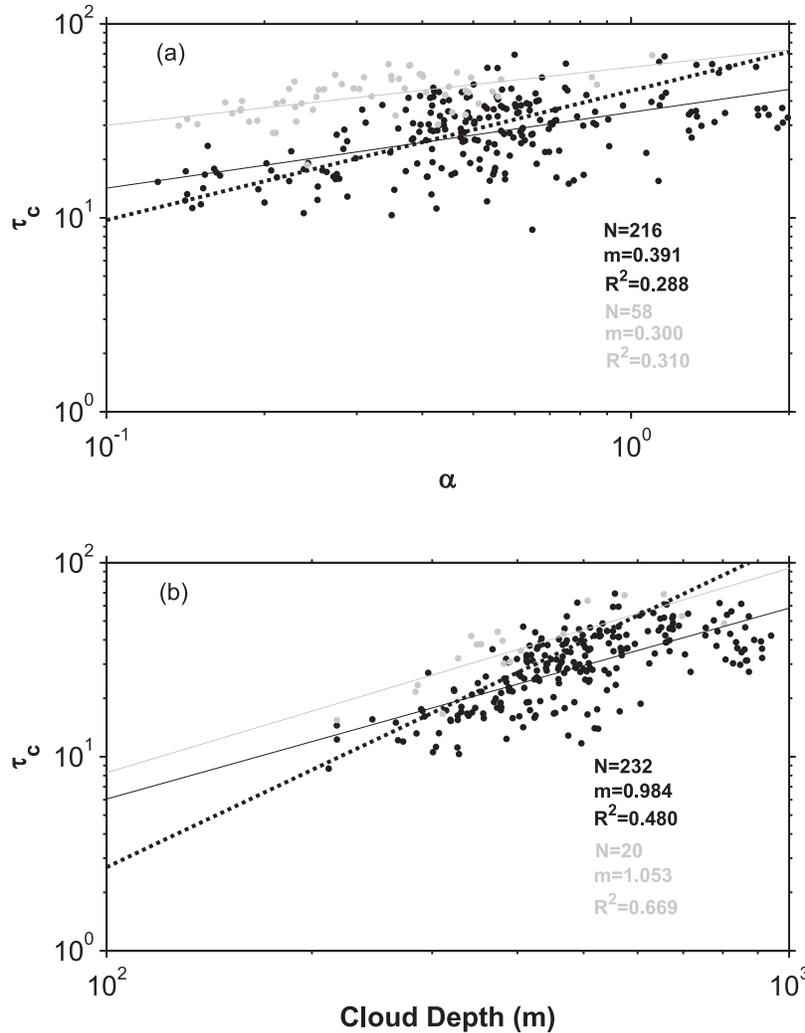


Figure 3. The same as in Figure 2 except for cloud optical depth instead of r_e .

a fixed value of α . There is a notable increase in τ_c with an increase in α and distinct segregation in the two cloud thickness populations, characteristics that are predicted by (10).

[27] We plotted τ_c against Δz to evaluate the sensitivity of τ_c to changes in Δz for different values of α (Figure 3b). The sensitivity of τ_c to Δz is greater than that of τ_c to α , as seen by noting the similarity in the slopes for different α bins. Similar to Figure 2 where we find that r_e is more significantly correlated with Δz than with α , we also find the most significant correlation between τ_c and Δz . Note that the sensitivity of τ_c to Δz and α is 5 fold and 2 fold greater, respectively, than that of r_e to Δz and α (equations (13) and (14) and Table 3).

[28] This formula and supportive observations illustrate the potential linkages with various physical parameters and especially the strong dependence of cloud optical depth upon cloud thickness, or equivalently, LWP. In addition, the derived formula shows that adiabaticity effect has twice that of the cloud droplet number concentration in modulating increases in the cloud optical depth, and, as a consequence, the albedo with the assumption of a homogeneous mixing and a uniform change of droplet size. The observation strongly suggests that mixing processes may overwhelm

the reduction in cloud droplet size dictated by the nucleation processes that underlie the theory of the aerosol first indirect effect.

4. Aerosol First Indirect Effects and Adiabaticity

[29] The observed cloud optical properties (r_e and τ_c) were related to the cloud structural parameters α , a proxy for the entrainment-mixing process, and Δz , a proxy for

Table 3. Summary of Data Numbers, the Slope, and Correlation Coefficient of τ_c Versus α (Upper) and τ_c Versus Δz (Lower) for Each Δz Bin and α Bin, Respectively^{a,b}

Optical Depth	τ_c Versus α		τ_c Versus Δz	
	Δz bins		α bins	
	Thin	Thick	Subadiabatic	Adiabatic
N (data number)	216	58	232	20
Slope	0.39	0.30	0.98	1.05
R ²	0.29	0.31	0.48	0.67

^aThe cloud thickness of 200–600 m belongs to the thin regime, and 600–1000 m to thick regime.

^bThe subadiabatic cloud denotes α of 0.1–0.8 and the adiabatic cloud indicates α of 0.8–1.2.

LWP in the previous section. In this section, we examine the relationship between r_e and aerosol loading, thereby enabling us to evaluate the extent to which aerosols are changing r_e , and whether its magnitude rivals that of the structural parameters examined above.

4.1. Relationship of r_e to Aerosol Load

[30] The aerosol light scattering coefficient σ_{sp} has been used as a proxy for the aerosol loading, and ultimately, the CCN concentration in the previous studies [Kim *et al.*, 2003; Garrett *et al.*, 2004]. The mixing state of the boundary layer must be considered to insure that the surface aerosol observations are representative of those that are influencing the cloud droplet nucleation properties of the clouds. Feingold *et al.* [2006] suggested that the optimal period for the use of this proxy is in daytime well-mixed cases, which we use here.

[31] The aerosol indirect effect (IE) has been defined as the derivative of the logarithm of cloud droplet radius with respect to the logarithm of the aerosol light scattering [Feingold *et al.*, 2003];

$$IE = -d \log r_e / d \log \sigma_{sp} \quad (15)$$

Strictly speaking, the IE must be defined in terms of τ_c rather than r_e , but (15) focuses on the reduction in droplet size and assumes that CCN have no impact on LWP. While there is scant evidence to support the dependence in LWP on CCN, dependence cannot be summarily dismissed in certain cloud conditions. The IE value emphasizes relative rather than absolute sensitivities, which is useful in reproducing trends without regard to the measurement biases [Garrett *et al.*, 2004; Feingold *et al.*, 2006]. It should be noted that the true manifestation of the aerosol first indirect effect rests upon the nucleation of observed aerosols into cloud droplets, which is better described by the CCN activity spectrum. In lieu of this measurement, equation (15) not only assumes that there is a relationship between σ_{sp} and CCN, but that the updraft conditions that exist in cloud nucleate a constant fraction of CCN. While this condition cannot be insured in the current analysis, it is expected that many of the observed cases occur in similar conditions.

[32] The relationships between r_e and σ_{sp} in the logarithm scale are shown in Figure 4 which indicates a general decrease in r_e with the increase of σ_{sp} , which, as expected, is consistent with Kim *et al.* [2003] since the data used in the original analysis are a subset of those presented here. For the data set as a whole the value of IE is 0.15 ($R^2 = 0.28$) (Figure 4a), which is a slightly better correlation than 0.13 ($R^2 = 0.24$), which was found by Kim *et al.* [2003].

[33] The physics that underlie the first indirect effect dictate that the IE comparison must be made between clouds having the same liquid water content because r_e is also a function of LWP [Twomey, 1977; Garrett *et al.*, 2004; Feingold *et al.*, 2006]. Accordingly, the data are divided into 5 bins of LWP = 10–50, 50–100, 100–150, 150–200, and >200 g m⁻², respectively. Values of IE range from 0.04–0.17 (Figure 4b and Table 4), indicating a decrease in IE with an increase in LWP and better correlation with the lower LWP, though the correlations are, in general, poor ($R^2 = 0.02\sim 0.17$). Despite the poor correlations, these values are also compatible with the previous works using

ground-based remote sensors; 0.02–0.16 by Feingold *et al.* [2003], and 0.13–0.19 by Garrett *et al.* [2004].

[34] The IE values and adiabaticity (α) with 5 bins of LWP are demonstrated in Figure 5, respectively. The IE has relatively larger value around 50–150 g m⁻² bins, which is also coincident with larger value of adiabaticity, except for the bin of LWP less than 50 g m⁻², where the measurement confidence is lowest. It is interesting to note that the higher the LWP, the more subadiabatic clouds are with the lowest bin of LWP excluded. The thicker clouds of high LWP tend to contain subadiabatic LWP and lower IE values, partly due to larger interacting surface area for the entrainment-mixing processes, which could eventually damp aerosol first indirect effect with an increasing LWP. Observational evidence that thicker clouds tend to exhibit stronger entrainment-mixing has also been reported from in situ aircraft measurements [e.g., You and Liu, 1995].

[35] The correlations between r_e and σ_{sp} are, in general, poor, despite the more extensive data set considered here. A natural question to pose is the extent to which the influence of aerosol loading on r_e might be dependent on the cloud structural parameters. In other words, are their specific structural configurations that permit cloud-aerosol interactions to dominate?

4.2. Aerosols Indirect Effect With the Change of Adiabaticity

[36] To compute the adiabaticity α , the averaged value (1.63 g kg⁻¹ km⁻¹) of the adiabatic lapse rates (Γ_l) of LWC obtained from the six soundings was applied in the 14 cases (Figure 6). The calculation of α strongly depends on Δz because the adiabatic LWP is proportional to square of cloud thickness, and linear proportional to Γ_l as seen in (6). In addition, the observation indicated that the relative variation of Δz is from 200 m to 1000 m greater than Γ_l ranging from 1.3 to 1.9 g kg⁻¹ km⁻¹. Accordingly, the sensitive of the adiabatic LWP to Δz is more than 7 times larger than that to Γ_l . Therefore the current method using the averaged value of Γ_l is thought to introduce very little uncertainty in calculating α .

[37] Similar to the LWP bins of section 4.1, α of 0.1 to 1.2 is divided into 2 classes: a subadiabatic regime (0.1–0.8) and an adiabatic regime (0.8–1.2). The thick cloud cases (600 to 1000 m) are not shown here because they are all generally subadiabatic (95%) with α ranging from 0.1 to 0.8 (Table 5) and data points are very few (the number of points is 3 only) for the adiabatic clouds. Therefore the cloud thickness over which both adiabatic and subadiabatic clouds exist is 200 to 600 m, but only 8% of these clouds were found to be adiabatic (Table 5). Poor correlations of r_e and σ_{sp} are indicated in the subadiabatic clouds and a negative slope (–0.17) with the significant correlation ($R^2 = 0.53$) of r_e and σ_{sp} in the adiabatic cases. While the number of cases analyzed here is still small, r_e appears to have virtually no correlation with the σ_{sp} in the subadiabatic clouds associated with turbulent entrainment-mixing processes.

[38] These results suggest that entrainment-mixing processes have substantial effects on the optical properties of these subadiabatic clouds which constitute a large fraction of the clouds that we observed. The predilection of the clouds at SGP to be subadiabatic due to entrainment-

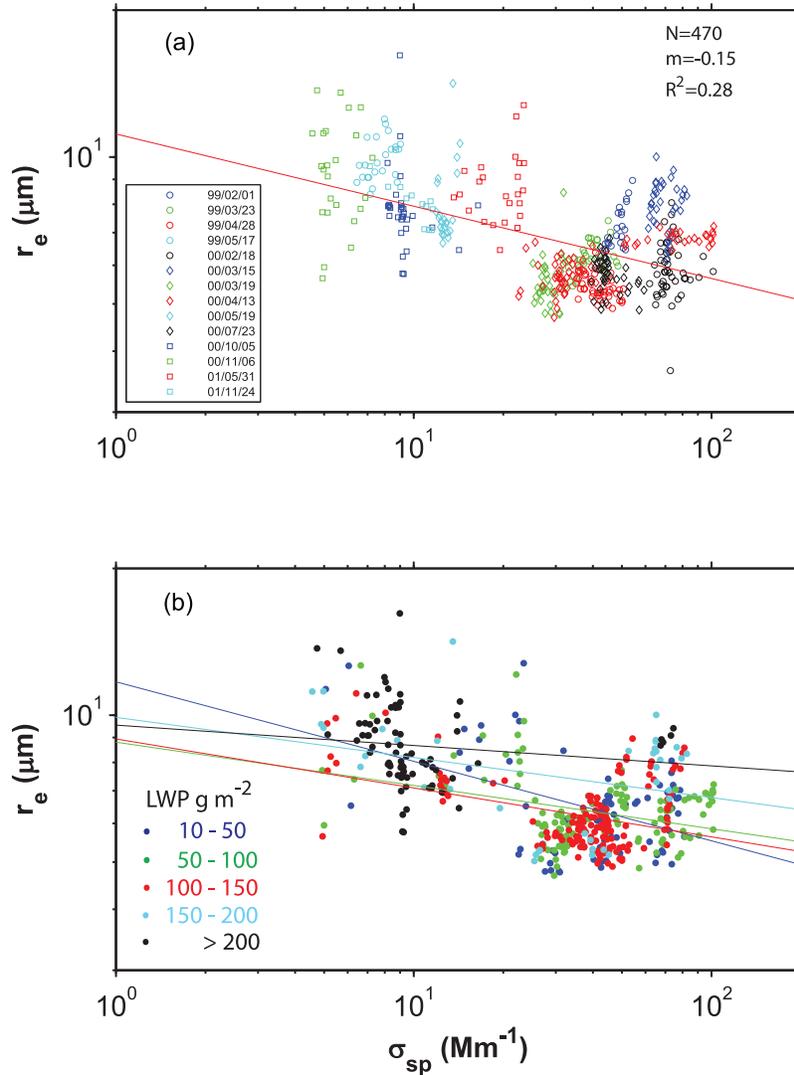


Figure 4. (a) Scatterplot of 5-min average r_e vs. light scattering coefficient (σ_{sp}) for submicrometer aerosol at 550 nm. Data for individual days are distinguished by color and symbol. (b) Scatterplot of r_e versus σ_{sp} with the different LWP classes; The blue color indicates LWP of 10–50, the green 50–100, the red 100–150, the cyan 150–200, and the black larger than 200 g m^{-2} . The slope of each solid line is a value of IE .

mixing could partly explain why it is hard to detect the aerosol indirect effect, as we will later demonstrate analytically. It is noteworthy that the reasons could partly be observational uncertainty [Kim *et al.*, 2003] or an instrument collocation problem [Feingold *et al.*, 2006] even though we tried to reduce these uncertainty and problem through the strict criteria and careful data screening processes.

[39] Box plots of cloud optical properties (Figure 7; LWP, τ_c and r_e) for the subadiabatic and adiabatic clouds indicate consistently higher values of LWP and τ_c for adiabatic clouds. Conversely, r_e shows no sensitivity to adiabaticity. Note that the notches represent a robust estimate of the uncertainty about the medians for box-to-box comparison. The boxes representing LWP and τ_c , whose notches do not overlap, indicate that the medians of the subadiabatic cases differ from the adiabatic ones at the 5% significance level,

whereas boxes of r_e overlap each other. The subadiabatic clouds at the SGP site generally contain less cloud water, on the average, than they would have if they were adiabatic for the thin cloud with cloud thickness of 200–600 m, which is attributable to entrainment-mixing process. Accordingly the cloud optical depth, which is mostly a function of LWP, is

Table 4. Values of IE , Correlation Coefficient (R^2), and Data Number (N) With the Different Classes of LWP^a

LWP (g m^{-2})	10~50	50~100	100~150	150~200	>200
IE^a	0.17	0.09	0.10	0.08	0.04
R^2	0.17	0.12	0.15	0.08	0.02
N	92	126	155	44	77

^a IE is defined as $-d \log r_e / d \log \sigma_{sp}$ and σ_{sp} indicates light scattering coefficient for submicron aerosol.

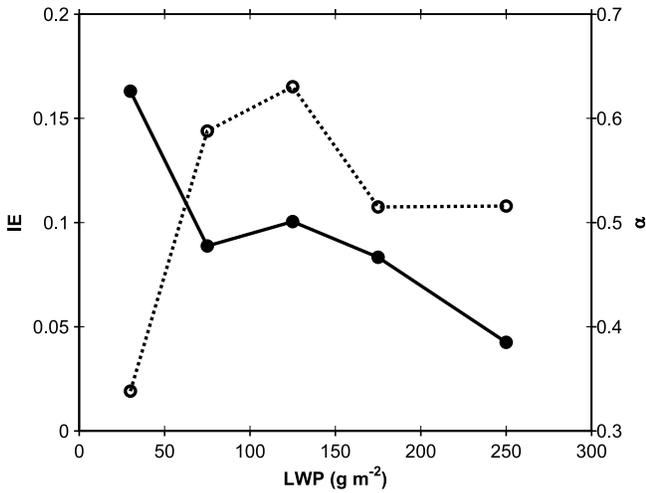


Figure 5. IE values (solid line) and adiabaticity (dotted line) according to the change of LWP.

understandably decreased when there is a reduction of LWP. Meanwhile, the cloud droplets effective radius could either increase or decrease, depending on homogeneous and heterogeneous mixing conditions, as explained in the following section 4.3, which would lead to no significant difference of r_e between the adiabatic and subadiabatic clouds.

4.3. Further Discussion on Entrainment-Mixing Effects

[40] Our simple expression based on the homogeneous mixing assumption accounts only for the effect of entrainment-mixing process on the liquid water content (L). The implied sensitivity of the clouds at the SGP to entrainment-mixing motivates us to analytically explore how the specific details of the entrainment-mixing process may impact the efficacy of the aerosol first indirect effect and our ability to observe it. Without loss of generality, sensitivity to any entrainment-mixing mechanisms can be illustrated by writing (11) as

$$r_e = \left(\frac{3}{4\pi\rho_l} \right)^{1/3} \beta \left(\frac{L}{N_{cd}} \right)^{1/3} \quad (16)$$

where β is a dimensionless parameter depending on the relative dispersion of the droplet spectrum [Liu and Daum, 2002]. For an adiabatic cloud, we have

$$r_{ea} = \left(\frac{3}{4\pi\rho_l} \right)^{1/3} \beta_a \left(\frac{L_a}{N_{cd,a}} \right)^{1/3} \quad (17)$$

where the subscript a denotes adiabatic conditions. The effects of mixing on L , N_{cd} , and β may be represented as

$$\alpha_x = \frac{x}{x_a} \quad (18)$$

where x represents the variable that is being considered. Substitution of (18) into (16) yields

$$r_e = \alpha_\beta \left(\frac{\alpha_L}{\alpha_N} \right)^{1/3} r_{ea} \quad (19)$$

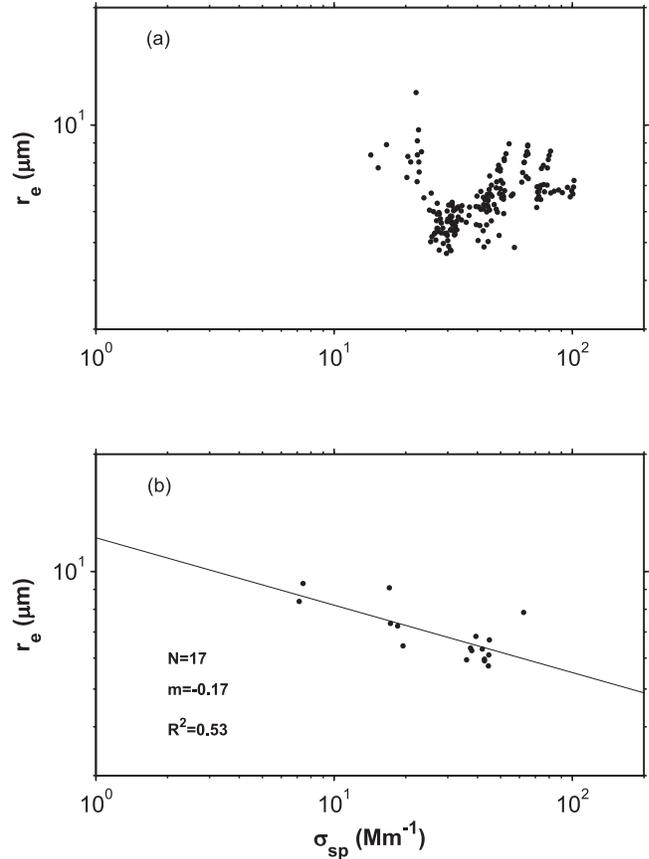


Figure 6. Scatterplot of r_e versus σ_{sp} for (a) the subadiabatic regime ($0.1 < \alpha < 0.8$) and (b) the adiabatic regime ($0.8 < \alpha < 1.2$) with the cloud thickness of 200~600 m.

which relates adiabatic to non-adiabatic conditions. According to (19), the cloud optical properties may respond in several ways depending on the nature of the entrainment-mixing process. Four extremes are evident in (19):

[41] (i) Extreme homogeneous mixing occurs if the mixing timescale is much faster than the timescale of evaporation so that all droplets are exposed to the same relative humidity. In this case, $\alpha_N = 1$, because the mixing process does not change N_{cd} , but evaporation reduces droplet sizes. This is the case considered in (11) and in the analysis above. Taking logarithms produces $\ln r_e = \ln(\alpha_\beta) + \ln r_{ea} + \frac{1}{3} \ln \alpha_L$, which is the equation of the line displayed in Figures 2 and 3.

[42] (ii) Extreme inhomogeneous mixing dictates that all droplets are exposed to different relative humidity and a

Table 5. Frequencies of the Subadiabatic, Adiabatic and Superadiabatic Clouds for Each Thin and Thick Cloud Regime^a

Frequency	Thin Cloud Regime			Thick Cloud Regime		
	Sub.	Adiab.	Super.	Sub.	Adiab.	Super.
Number	177	17	22	55	3	0
(%)	(82%)	(8%)	(10%)	(95%)	(5%)	

^aThe cloud thickness of 200–600 m belongs to the thin regime, and 600–1000 m to thick regime. The subadiabatic (Sub.) cloud denotes α of 0.1–0.8, the adiabatic (Adiab.) cloud α of 0.8–1.2, and the superadiabatic (Super.) cloud α of 1.2–2.0.

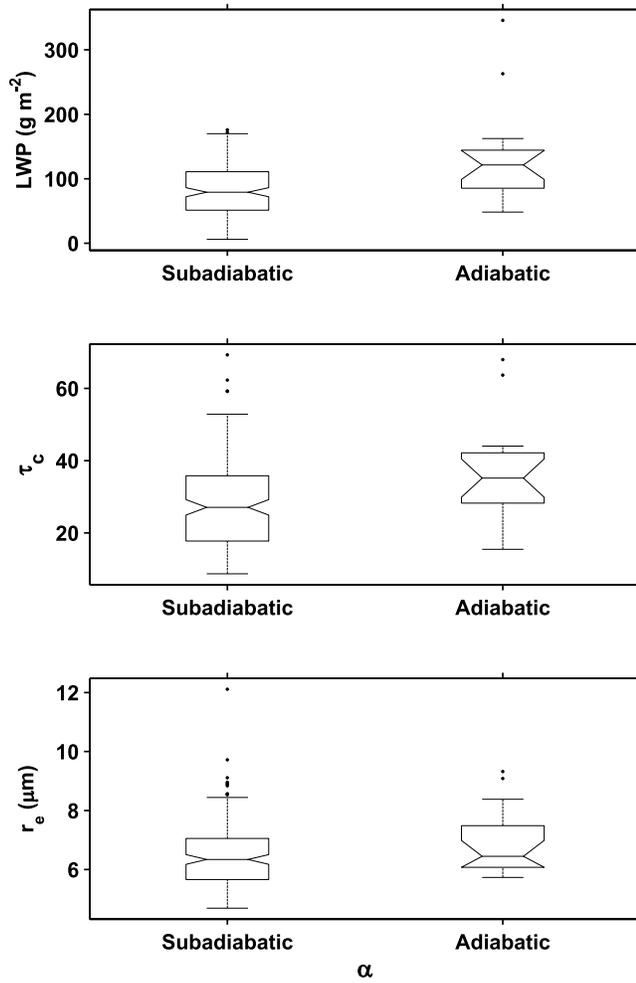


Figure 7. Box plots of LWP, cloud optical depth and r_e for the subadiabatic and adiabatic clouds for the thin cloud with cloud thickness of 200~600 m. The lower, middle and upper lines of the box are the 25th, 50th (median) and 75th percentiles of each variable, and an outlier is a value that is more than 1.5 times the interquartile range away from the top or bottom of the box. The notches represent a robust estimate of the uncertainty about the medians.

constant portion of droplets of all sizes are totally evaporated. In this case, $\alpha_N = \alpha_L$, and we can write $r_e = \alpha_\beta r_{ea}$, which implies that effective radius is basically independent of α_L .

[43] (iii) Secondary activation is diagnosed when $\alpha_N > \alpha_L$, so $c = \alpha_L/\alpha_N$ decreases with decreasing α_L , or stronger mixing brings in more newly activated droplets. In this case, $r_e = \alpha_\beta (c)^{1/3} r_{ea}$, we expect a steeper slope in the decrease in r_e as the cloud becomes more subadiabatic (α_L decreases).

[44] (iv) Enhanced growth occurs when some cloud droplets grow at the expense of others. In this situation, $\alpha_N < \alpha_L$, which implies that c increases with decreasing α_L , so stronger mixing results in less but bigger droplets. The same mathematical relationship as in the above (iii) applies, but there is an increase in the effective radius with a decrease in α_L .

[45] The possibilities suggested by the above extremes as applied to (19) are summarized in Table 6, which suggests that the nature of the mixing process may either attenuate or amplify the aerosol indirect effect. Thus when the observed clouds are sub-adiabatic, the details of the mixing process may be important in determining the radiative impact of the clouds and may, in some circumstances, be the controlling factor.

[46] In summary, it is more likely that the aerosol first indirect effect will be observed in unabated form in adiabatic clouds because they are free of processes such as drizzle and entrainment that may alter the cloud droplet spectrum after the nucleation process occurs. Adiabatic clouds are the exception rather than the rule in real continental clouds and our results suggest that diabatic processes may possess many configurations, particularly when the mixing is heterogeneous.

5. Conclusions

[47] The impacts of mixing on the optical properties of continental stratus clouds were examined in the context of the aerosol indirect effect using ground-based remote sensing at SGP for several cases that occurred in 1999–2001. Adiabaticity was used as a proxy for mixing processes. The results suggested, as expected, that there is a strong dependence of cloud optical depth upon cloud thickness, (a surrogate for LWP) as already emphasized by *Kim et al.* [2003]. The theoretical derivation showed that the impact of adiabaticity is twice that of the cloud number concentration in determining the cloud optical depth for clouds with the assumption of a homogeneous mixing and a uniform change of droplet size. The observed adiabaticity with the change of LWP illustrated that the thicker clouds of high LWP tend to

Table 6. Potential Response of Cloud Droplet Effective Radius to Homogeneous and Heterogeneous Mixing Processes

	Homogeneous Mixing	Heterogeneous Mixing/ETEM ^a		
		Extreme Case	Secondary Activation	Enhanced Growth
Underlying mechanism	faster mixing	uniform evaporation	nucleation	coalescence
α_N and α_L	$\alpha_N = 1$	$\alpha_N = \alpha_L$	$\alpha_N > \alpha_L$	$\alpha_N < \alpha_L$
Mixing function	mixing does not change N_{cd} but reduce the sizes	mixing changes L & N_{cd} proportionally	stronger mixing results in more droplets	stronger mixing results in less but bigger droplets
Response of r_e	depending on α_β and α_L	r_e independent of α_L	r_e decreases with decreasing α_L	r_e increases with decreasing α_L
Formula	$r_e = \alpha_\beta r_{ea} (\alpha_L)^{1/3}$	$r_e = \alpha_\beta r_{ea}$	$r_e = \alpha_\beta \left(\frac{\alpha_L}{\alpha_N}\right)^{1/3} r_{ea}$	$r_e = \alpha_\beta \left(\frac{\alpha_L}{\alpha_N}\right)^{1/3} r_{ea}$
AIE Effect ^b	less AIE effect	no change	more AIE effect	less AIE effect

^aETEM means entity type entrainment mixing proposed by *Telford* [1996].

^bAIE indicates aerosol indirect effect.

contain subadiabatic LWP with accordingly lower α values, corresponding to lower IE , which could eventually damp aerosol first indirect effect with an increasing LWP. A stronger association ($IE = -0.17$ and $R^2 = 0.53$) between cloud droplet effective radius and aerosol light scattering coefficients, a proxy for aerosol concentration and size, was found in adiabatic clouds. Poor correlations between the two variables were indicated in the subadiabatic clouds that are observed most frequently at SGP and other continental locations.

[48] We were able to observe characteristics consistent with the aerosol first indirect effect in adiabatic clouds, while being unable to detect these characteristics in subadiabatic clouds. This difference is likely due to processes other than the aerosol first indirect effect modulating cloud optical properties in subadiabatic clouds. While we recognize that potential limitations in our measurements, particularly in the subadiabatic cases, our selection criteria are significantly stringent that these limitations have been minimized. We also demonstrated analytically that the details of the mixing process might confound detection of the aerosol indirect effect in subadiabatic clouds because homogeneous and heterogeneous mixing apparently produces different microphysical responses.

[49] This study is limited to a continental site where dynamic range of r_e is almost less than $10 \mu\text{m}$. Including the observation data of other remote sites should broaden this range and could improve our understanding of the various manifestations of mixing and its interplay with the aerosol first indirect effect. We note that information of the vertical profile of thermodynamic state is crucial in understanding the cloud microphysics in association with the aerosol indirect effect and that this study was hampered by the sparse frequency of vertical soundings rather than the selection of cloudy periods. Therefore more frequent balloon-borne soundings or alternative strategies to measure the thermodynamic profile are needed to better understand the influence of adiabaticity and meteorology on cloud drop size, so that these impacts can be sorted from those that are true manifestations of the aerosol first indirect effect. Last the quantitative analysis of drizzle and entrainment effects on adiabaticity and its association with the aerosol indirect effect is recommended for future study.

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