Environ. Res. Lett. 4 (2009) 015002 (8pp) doi:10.1088/1748-9326/4/1/015002

The impact of smoke from forest fires on the spectral dispersion of cloud droplet size distributions in the Amazonian region

J A Martins[1,4] **and M A F Silva Dias**[2,3]

[1] Universidade Tecnológica Federal do Paraná, Londrina, PR, Brazil
[2] Centro de Previsão de Tempo e Estudos Climáticos, Cachoeira Paulista, SP, Brazil
[3] Universidade de São Paulo, São Paulo, SP, Brazil

E-mail: jmartins@utfpr.edu.br

Received 1 August 2008
Accepted for publication 28 November 2008
Published 13 January 2009
Online at stacks.iop.org/ERL/4/015002

Abstract

In this paper, the main microphysical characteristics of clouds developing in polluted and clean conditions in the biomass-burning season of the Amazon region are examined, with special attention to the spectral dispersion of the cloud droplet size distribution and its potential impact on climate modeling applications. The dispersion effect has been shown to alter the climate cooling predicted by the so-called Twomey effect. In biomass-burning polluted conditions, high concentrations of low dispersed cloud droplets are found. Clean conditions revealed an opposite situation. The liquid water content (0.43 ± 0.19 g m^{-3}) is shown to be uncorrelated with the cloud drop number concentration, while the effective radius is found to be very much correlated with the relative dispersion of the size distribution ($R^2 = 0.81$). The results suggest that an increase in cloud condensation nuclei concentration from biomass-burning aerosols may lead to an additional effect caused by a decrease in relative dispersion. Since the dry season in the Amazonian region is vapor limiting, the dispersion effect of cloud droplet size distributions could be substantially larger than in other polluted regions.

Keywords: cloud condensation nuclei, biomass burning, effective radius ratio, relative dispersion, specific cloud water content

1. Introduction

The aerosol first indirect effect predicts a cooling of the Earth's atmosphere since the more numerous and smaller droplets increase the cloud albedo (Lohmann and Lesins 2002, Lohmann and Fleichter 1997). The aerosol forcing may even offset global greenhouse warming to a substantial degree (Charlson *et al* 1992). However, the magnitude of this effect remains uncertain, due in part to the complexity in treating aerosol–cloud interactions (Menon *et al* 2008). Clouds are affected by a set of environmental factors, such as water vapor availability, topography, atmospheric dynamics and stability, land cover, and aerosol concentration and distribution. These factors, together with microphysical variables, act on the strength of the convection process to define the precipitation process as a whole.

Climate models have increasingly used variables related to cloud droplet size distribution in their physical schemes (Lohmann *et al* 2007). Comparison among modeling results reveals that simulations are very sensitive to changes in cloud properties (Dandin *et al* 1997, Rotstayn and Liu 2003). A modeling study performed by Ghan *et al* (2001) showed that approximately half of the indirect radiative forcing is due to changes in droplet radius and the other half to increased cloud liquid water. The cloud droplet effective radius (r_{ef}) is a variable frequently used in climate models. According to Reid *et al* (1999), the cloud droplet effective radius is a key variable because it determines many important radiative properties of

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

a cloud, including the cloud albedo (Pontikis and Hicks 1992, Martin *et al* 1994). Dandin *et al* (1997) performed simulations with changes in parameterization of the effective radius and found that these changes can induce strong differences in the global value of the radiative fluxes. In addition, variations of the relationship between cloud optical thickness and droplet radii may indicate variations in cloud microphysical regimes (Han *et al* 1994).

Liu and Daum (2002) showed that anthropogenic aerosols exert an additional effect on cloud properties derived from changes in the spectral shape of the size distribution of cloud droplets in polluted air. This additional effect can be represented by the spectral dispersion, which plays a significant role in determining cloud radiative fluxes (Liu and Daum 2000a, 2000b, Daum and Liu 2003). The cooling effect on the climate, as predicted by the Twomey effect (Twomey 1977), can be increased or diminished as a consequence of spectral dispersion. An important feature suggested by Shao and Liu (2006) is that the spectral dispersion is also affected by the meteorological properties of clouds, causing uncertainty in the relationship between spectral dispersion and droplet concentration. According to Peng and Lohmann (2003), the comparison between the calculated cloud albedo from microphysical measurements and the aircraft measurements of the cloud albedo confirms the significance of the influence of the cloud droplet spectral shape on cloud properties. When taking into consideration the cloud droplet spectral shape, the calculated values present a better agreement with the independently measured cloud albedo.

The main results of the above-mentioned papers are that it is necessary to include the spectral dispersion in the calculation of the effective radius in global climate models. The dry season in the Amazon region is characterized by an almost permanent smoke plume that has a strong impact on the radiation budget at all levels (Procopio *et al* 2004) and also on the cloud condensation nuclei (CCN) concentration (Martins *et al* 2009b) and physical–chemical properties (Rissler *et al* 2004, 2006). The extremely high biomass-burning aerosol concentration in that region can strongly impact the cloud microphysics and dynamics (Lin *et al* 2006, Martins *et al* 2009a). Undoubtedly, all the other players in the cloud microphysical scenario, e.g., dynamic and thermodynamic environmental patterns, cannot be discarded, since they also have substantial contributions to the precipitation development and climate, influencing the indirect aerosol effect in opposite directions (Koren *et al* 2004, Wang 2005).

The aim of this paper is to compare how different the spectral dispersions of cloud droplet size distributions in clean and polluted conditions are during the biomass-burning season in the Amazonian region. The purpose is to improve our understanding of the indirect aerosol effect and its treatment in climate models. Assuming that the shape of the cloud droplet size distribution constitutes one of the most important players in the biomass-burning scenario, three main variables related to the cloud droplet spectra are analyzed in this paper: effective radius ratio, relative dispersion, and specific cloud water content. The droplet size distribution of clouds developing in a high smoke influenced environment and clean areas will be compared according to these variables. A database with several cloud events for different atmospheric conditions (clean and polluted) obtained from the large-scale biosphere–atmosphere experiment in Amazonia (LBA) will be used. The datasets were compiled from aircraft flyover measurements, taken in the southwestern Amazon region between September and October, 2002.

2. Data description and methodology

2.1. Data acquisition

Aircraft measurements were performed aboard the UECE's (Universidade Estadual do Ceará) Bandeirante EMB 110 plane during the LBA experiment in September and October, 2002. This period was characterized by a transition from a dry condition, with great number of deforestation fires, to a wetter condition with decelerating biomass-burning activity in the southwest of Amazonian region. The aircraft instrumentation included a CCN counter, a forward scattering spectrometer probe (FSSP-100) to measure cloud droplet spectra, and conventional instruments to measure positioning, pressure, temperature, and humidity. Data from this campaign have been analyzed by Andreae *et al* (2004), Freud *et al* (2008). Measurements of CCN properties taken during the campaign were specifically discussed in Martins *et al* (2009b).

The forward scattering spectrometer probe (FSSP) model 100 is an instrument developed by Particle Measuring Systems (PMS Inc., Boulder, CO) for the measurement of cloud droplet size distributions. This instrument is a particle counter that works by measuring the intensity of light that is scattered by the particle when it passes through a light beam. The system makes use of the Mie scattering theory to relate this intensity to the particle size. The size is categorized into one of 30 channels and the information is sent to a data system where the number of particles in each channel is accumulated over a preselected time period. The channels are distributed in the range 2–47 μm. The calibration method involves glass beads with well-controlled sizes and is described in Dye and Baumgardner (1984). The relation between the FSSP-derived (cloud) liquid water content and the liquid water content measured by the hot-wire King probe for the same measurements used in this paper was discussed by Freud *et al* (2008). They found a good agreement between the CSIRO-King and the FSSP instruments regarding the cloud's liquid water content.

The set of flights available (see figure 1 for the location of the flights) includes a large number of flights with different characteristics. There are flights where the aircraft follows a plan transecting regions of different air masses. This type of flight covers regions ranging from smoke plumes of biomass burning in deforested regions to relatively clean environments along the remote areas of rainforest in the western Amazonian region. There are also flights in which a vertical profile of up to 5 km is obtained for a given area, with the aircraft surrounding a specific site. A total of 785 clouds were sampled in 20 different flights. A detailed description of the main characteristics of each flight can be found in Martins *et al* (2009b). The main sites in the region where flights were

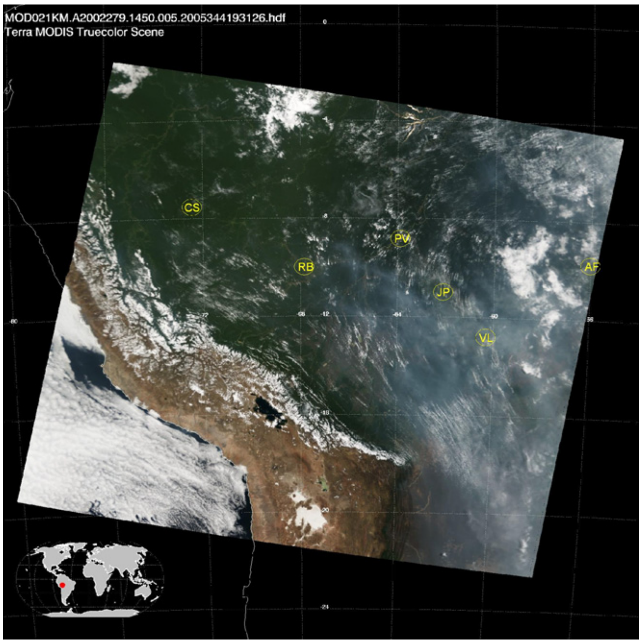

Figure 1. Moderate resolution imaging spectroradiometer (MODIS–TERRA) image showing part of the Amazon rainforest in western Brazil. The symbols represent the main cities in the region where flights were conducted: Cruzeiro do Sul (CS), Rio Branco (RB), Porto Velho (PV), Ji Paraná (JP), Vilhena (VL), and Alta Floresta (AF). Much of the deforestation has occurred in Rondonia and Mato Grosso states, which cover the image in the southeastern region of the Madeira River, which in turn runs in a northeast–southwest direction (see PV to identify the river). Dense smoke covers the south of the Madeira River while the dark green area predominating in the northwest of the image suggests undisturbed clean areas. Image acquired: 06 October 2002, 14:50 UTC (10:50 at JP)—http://modis-atmos.gsfc.nasa.gov.

conducted are those cities identified in figure 1: Cruzeiro do Sul (CS), Rio Branco (RB), Porto Velho (PV), Ji Paraná (JP), Vilhena (VL), and Alta Floresta (AF).

2.2. Calculating the spectral dispersion

To investigate the relationships between biomass-burning aerosols and spectral dispersion of cloud droplets, a combination of field measurements of the cloud droplet effective radius r_{ef} and a scaling factor that relates the liquid water content L and droplet concentration N was made. The r_{ef} is defined as the ratio between the third and the second moment of the droplet size distribution (Hansen and Travis 1974). Size distributions measured by the FSSP-100 probe were used to calculate the r_{ef} according to the following expression (Reid *et al* 1999):

$$r_{\text{ef}} = \sum_{i=1}^{N_c} n_i r_i^3 \Big/ \sum_{i=1}^{N_c} n_i r_i^2 \tag{1}$$

where N_c is the total number of size channels used (30 channels), r_i is the droplet radius, and n_i is the number of droplets in channel i. Cloud droplet size distribution data were sampled at a frequency of 5 Hz. FSSP data also provided the values of N and L.

Most global climate models use a '1/3' power law to parameterize r_{ef} as a function of the cloud liquid water content L and droplet concentration N. The parameterization has been demonstrated both empirically and theoretically by a number of studies (Slingo 1990, Bower and Choularton 1992, Bower *et al* 1994, Dandin *et al* 1997, Liu and Hallett 1997, Liu and Daum 2000a, 2000b). The expression can be written as

$$r_{\text{ef}} = \beta \left(\frac{3}{4\pi\rho_{\text{w}}}\right)\left(\frac{L}{N}\right)^{1/3} \tag{2}$$

where ρ_{w} is the water density and β is a dimensionless parameter, named the effective radius ratio, which depends on the spectral shape of the cloud droplet size distribution. According to Liu and Daum (2000b), the only difference between different power laws lies in the specification of the parameter β.

Analytical distributions are often applied to describe cloud droplet size distributions (Liu *et al* 1995). Using different distribution functions, Liu and Daum (2000a) derived expressions to calculate the parameter β. They compared the expressions to observations and concluded that Weibull or gamma distributions best describe the dependence of the effective radius on the spectral dispersion. As the two expressions perform equally well, we applied the expression corresponding to the gamma distribution. For a gamma distribution, the expression for β takes the form

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

The relative dispersion (ε) of the cloud droplet size distribution is defined as the ratio of the standard deviation to the mean radius of the droplet size distribution and has been calculated using the above-described cloud data.

3. Results and discussion

3.1. Size distributions

The dataset used in this paper involves clouds sampled varying from very clean air to heavily polluted atmospheric conditions. The two flights conducted in clean Amazon conditions at the very far western edge of the Amazon region during 05 September 2002 total 118 cloud events (see table 1) and include measurements performed during both morning and afternoon periods. These flights include measurements of the whole precipitation forming process in a clean atmosphere from weakly organized cloud structure (flight undertaken during the morning) to a vigorous convection period with associated precipitation (flight undertaken during the afternoon). No significant spatial differences can be observed between the two flights. Because of the remote nature of this specially clean condition, the related cloud droplet size distributions are expected to exhibit characteristics of an environment which is free from anthropogenic aerosols.

Flights performed around JP during 23 and 26 September 2002 and the last part of the flights on 06 October and 13 October 2002 were flights in which the clouds were

Table 1. Mean and standard deviations of properties from cloud droplet size distributions measured during the dry season in the Amazon region obtained from the FSSP. Ranges of sampling altitude are also included.

Date	Region	Time (LT) altitude (m)	Clouds	N (cm^{-3})	L (g m^{-3})	r_{ef} (μm)	β	ε
23/09/02	JP	14:07–16:16 1950–3350	12	1284 ± 710	0.63 ± 0.55	4.82 ± 1.09	1.13 ± 0.03	0.38 ± 0.05
24/09/02	JP	14:43–17:04 3000–4800	47	707 ± 485	0.30 ± 0.05	4.47 ± 1.28	1.17 ± 0.04	0.43 ± 0.05
26/09/02	JP	14:02–16:18 1810–2120	13	1182 ± 709	0.27 ± 0.21	3.92 ± 0.52	1.17 ± 0.02	0.43 ± 0.04
27/09/02	JP	14:27–16:03 1910–2300	25	834 ± 567	0.15 ± 0.13	3.70 ± 0.37	1.12 ± 0.05	0.35 ± 0.09
28/09/02	JP	13:37–15:38 1760–4920	91	876 ± 559	0.52 ± 0.56	5.22 ± 1.58	1.17 ± 0.04	0.44 ± 0.06
30/09/02	JP–VL	11:14–12:17 1420–1700	54	783 ± 428	0.19 ± 0.15	4.04 ± 0.50	1.14 ± 0.02	0.39 ± 0.03
30/09/02	VL–JP	13:49–16:40 1630–4900	98	840 ± 511	0.40 ± 0.45	4.74 ± 1.27	1.16 ± 0.04	0.43 ± 0.06
04/10/02	JP–PV	11:06–12:25 1880–4800	38	738 ± 486	0.61 ± 0.56	6.40 ± 1.38	1.20 ± 0.04	0.50 ± 0.06
04/10/02	PV–RB	13:54–15:49 1870–4850	57	811 ± 597	0.54 ± 0.58	5.60 ± 1.31	1.17 ± 0.04	0.45 ± 0.07
04/10/02	RB–CS	16:48–18:36 2680–2770	9	859 ± 207	0.87 ± 0.59	7.10 ± 1.31	1.19 ± 0.04	0.50 ± 0.05
05/10/02	CS	12:13–14:18 1450–4890	80	433 ± 218	0.49 ± 0.53	7.10 ± 2.78	1.23 ± 0.08	0.54 ± 0.12
05/10/02	CS	15:24–17:01 1950–4870	38	412 ± 258	0.43 ± 0.33	8.15 ± 2.61	1.26 ± 0.09	0.59 ± 0.12
06/10/02	CS–RB	11:12–13:15 1990–3180	59	609 ± 349	0.64 ± 0.51	7.39 ± 1.46	1.24 ± 0.06	0.56 ± 0.07
06/10/02	RB–JP	14:19–16:45 2250–3160	42	1033 ± 609	0.58 ± 0.52	5.54 ± 1.17	1.20 ± 0.05	0.47 ± 0.08
08/10/02	JP	14:53–16:30 1290–3970	85	572 ± 335	0.43 ± 0.54	5.70 ± 2.15	1.18 ± 0.04	0.47 ± 0.07
09/10/02	JP	13:32–15:27 1370–4050	95	685 ± 463	0.32 ± 0.49	4.64 ± 1.71	1.16 ± 0.04	0.43 ± 0.06
11/10/02	JP–PV	13:21–14:46 1150–2480	30	679 ± 456	0.17 ± 0.16	4.13 ± 1.14	1.15 ± 0.03	0.42 ± 0.04
12/10/02	JP	11:45–13:38 1300–3300	53	661 ± 504	0.25 ± 0.35	4.53 ± 1.35	1.17 ± 0.04	0.44 ± 0.06
13/10/02	JP	13:20–15:50 1890–2910	19	1073 ± 663	0.36 ± 0.33	4.36 ± 0.98	1.16 ± 0.02	0.42 ± 0.03
14/10/02	JP	19:43–20:17 1870–3910	26	610 ± 444	0.42 ± 0.62	5.38 ± 1.82	1.13 ± 0.04	0.38 ± 0.07

embedded in the smoke layers. Data from these polluted flights accounted for 86 cloud events and exhibited different behavior in comparison to the clean case. There is, however, a common picture between the clean and polluted conditions: convection started during the morning, with cloud streets developing in both areas and evolving during the afternoon to a picture characterized by isolated deep cells. During the beginning of the flights, it was suspected that there was the occurrence of a short time delay in the convection initiation under polluted conditions, possibly because of the shading effect of the aerosols in the surface radiation budget. Since this paper does not focus on this subject, a more solid analysis is necessary to reach any possible conclusion.

Gamma distributions were fitted to the measured droplet size distributions for each cloud crossed and listed in table 1. The histogram in figure 2 shows the corresponding frequency distributions of the shape parameters of gamma distributions fitted in the above-mentioned clean and polluted cases. The shape parameter affects how broad or narrow the gamma distributions are assumed to be. The larger the value of the shape parameter, the more narrowly distributed the spectrum is (Costa *et al* 2000, Gonçalves *et al* 2008).

The minimal value of the root mean square error was chosen as the criterion to select the best fitted gamma distribution for each sampled size distribution. Figure 2 shows the calculated frequencies as percentages of all measurements in each condition. The shape parameter number 10 includes the occurrences associated to all other values larger than 10. The average value associated with the shape parameter found under clean conditions was 2.7 ± 2.2 (mean and standard deviation). Otherwise, under polluted conditions the shape parameter was found to be 4.9 ± 1.9. The results show that the pollution causes a significant narrowing of the cloud droplet size distributions. However, the shape parameter of the gamma function is not able to provide information which can be directly used in climate models. According to the discussion in section 2, this is

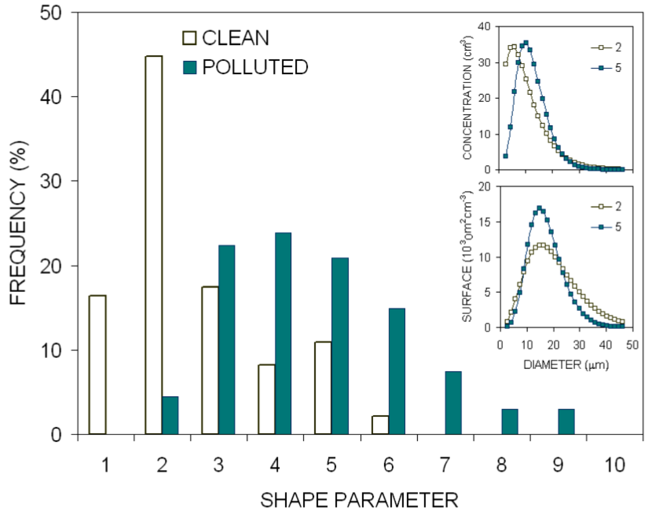

Figure 2. Histogram of the frequency of occurrence of the best fitting shape parameter for two selected regions of study: a clean region in the west side of the Amazon (CS) and a polluted region under strong biomass-burning activity near Rondonia state (JP). Gamma distributions with shape parameters 2 and 5, $L = 0.5$ g m^{-3}, and $N = 300$ cm^{-3} are also included.

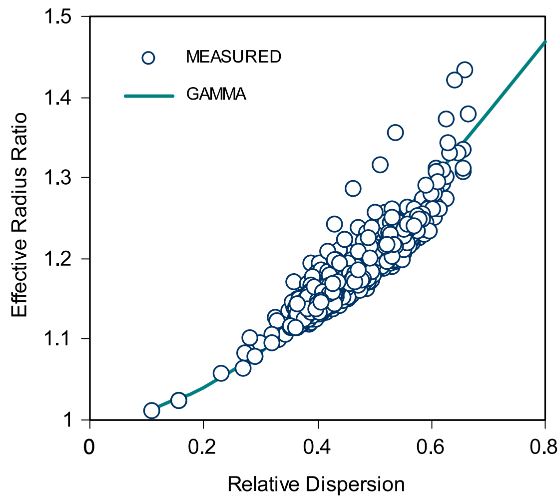

Figure 3. Effective radius ratio as a function of the relative dispersion, calculated from the gamma distribution and from FSSP measurements under clean and polluted atmospheric conditions in the Amazonian region.

done by the parameter β, which links the degree of dispersion of the droplet size distribution and the effective radius.

3.2. Spectral dispersion

Equation (3) shows that β is an increasing function of the relative dispersion ε. The effective radius ratio β as function of the relative dispersion ε, calculated from FSSP measurements under clean and polluted conditions in the Amazonian region, is shown in figure 3, for all 785 sampled clouds. Figure 3 also includes a theoretical curve, predicted according to equation (3). The calculated β from the aircraft measurements under different atmospheric conditions is summarized in table 1. The calculated mean values for β varied from 1.12 to 1.26, with the lowest values generally being associated to polluted conditions. The corresponding mean values for ε varied from 0.38 (polluted) to 0.59 (clean). The r_{ef} was shown to be in general about 50% greater under clean than under polluted conditions, varying from 3.70 to 8.15 μm.

As can be seen in table 1, there is a general decrease of β with the increase in cloud droplet concentration. The relative dispersion ε also decreases in a similar way. On the other hand, both parameters increase with the increase in cloud water content L. This behavior, associated to parameters β and ε, suggests that they are directly related to a variable that relates the cloud droplet concentration and cloud water content, i.e., the specific cloud water content γ (Liu and Daum 2000b), defined as the ratio of the liquid water content to the droplet concentration, i.e.,

$$\gamma = \frac{L}{N}. \tag{4}$$

The effective radius ratio β and relative dispersion ε are shown in figure 4 as a function of the specific cloud water content calculated from FSSP data. Each point represents the mean value associated to the measurements performed in each flight. Note that there are some days with more than one flight, involving morning and afternoon periods. In general, the effective radius ratio and relative dispersion are directly correlated to the specific cloud water content. Both increase with the increase in the specific water content.

Liu *et al* (2006) parameterized the dependence of β on the specific water content by a power law. The result is presented in figure 5 as a comparison to this work. In their parameterized result, the relative dispersion decreases with the increase in the specific water content. This behavior was not found in this paper's results involving data from the biomass-burning season in the Amazon region. In this case, the relative dispersion increases with the specific water content. In fact, as predicted from the idealized model of droplet growth in an adiabatic convective cell (Howell 1949), the relative dispersion (or β) generally decreases when the specific water content increases. The governing equation for the condensational growth of cloud water droplets implies that the droplet radius growth rate is inversely proportional to the droplet radius (see Pruppacher and Klett 1997), so that small droplets grow faster (in radius) than the big ones, narrowing the spectra (Liu and Hallett 1998). However, if the precipitation initiation due to collision–coalescence processes in warm clouds is occurring, the relative dispersion could be increasing with the increase in the specific water content. When the flights are analyzed individually, we observe that clouds sampled in polluted days do not show a clear trend in ε (and also β) as function of the specific water content. Otherwise, the measurements from clean days confirm a very clear increase of β and ε with the increase in specific water content. Figure 6 shows ε as function of γ, confirming the theoretically predicted increase in the spectral dispersion as a consequence of the developing warm rain process.

Liu and Daum (2002) studied the indirect warming effect from dispersion forcing and found a different trend in relation to the present work. According to the authors' results, 11 of the 13 cases show an increase in ε that is concurrent with

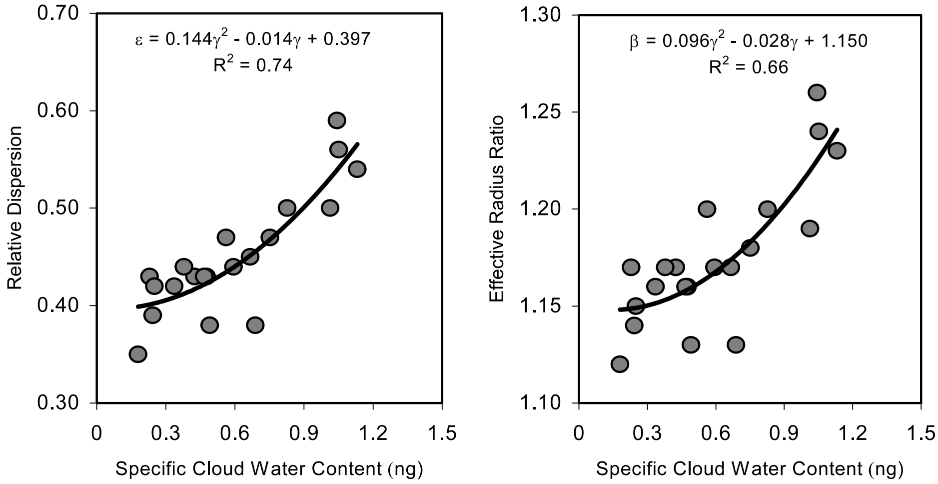

Figure 4. Relative dispersion and effective radius ratio as a function of the specific cloud water content, calculated from FSSP measurements. Each point represents a flight average during the dry season in the Amazonian region. The polynomial functions represent the best fit.

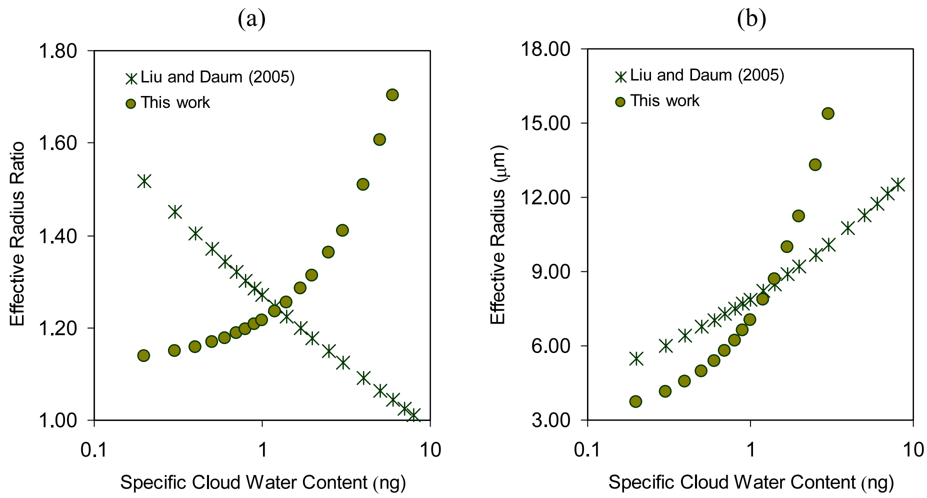

Figure 5. Effective radius ratio (a) and effective radius (b) as a function of the specific cloud water content calculated from parameterizations from Daum and Liu (2005) and this work.

the effect caused on the effective radius by the increase in N, with negligible change to slight decreases in the other two. Lu and Seinfeld (2006), using a three-dimensional large-eddy simulation of marine stratocumulus, found that the relative dispersion decreases with increasing aerosol number concentration (for aerosol number concentrations less than about 1000 cm^{-3}), which is in agreement with our results, but for a different environment and cloud structure.

Liu and Daum (2002) support anthropogenic aerosols having a more complex chemical composition and a broader size distribution than marine aerosols, and that the more numerous small droplets formed in a polluted cloud compete for water vapor and broaden the droplet size distribution when compared to clean clouds that have fewer droplets and less competition. We might conceptualize the suggested explanation as a chemical-limiting effect and suppose that some particles are more efficient than others in growing by vapor condensation. We may also assume that the increase in the spectral dispersion is observed due to the presence of background marine aerosols, since they can grow at a rate faster than anthropogenic aerosols.

Lu and Seinfeld (2006) suggested a different mechanism to explain the dispersion effect found in their numerical results. Note that, in this case, the effect is opposite to the one found by Liu and Daum (2002). The smaller droplets resulting from higher aerosol number concentrations lead to less spectral broadening by suppressed collision and coalescence processes and more spectral narrowing by droplet condensational growth. In this case, a collision–condensational-limiting effect is a conceptual explanation.

3.3. The clean–polluted contrast

If all the cloud droplets were of the same size, monodisperse, then the β parameter would be equal to unity. In real size distributions, the broader the size distribution, the larger is the β parameter, i.e., β is a measurement of the degree of dispersion associated to the size distributions. Although there

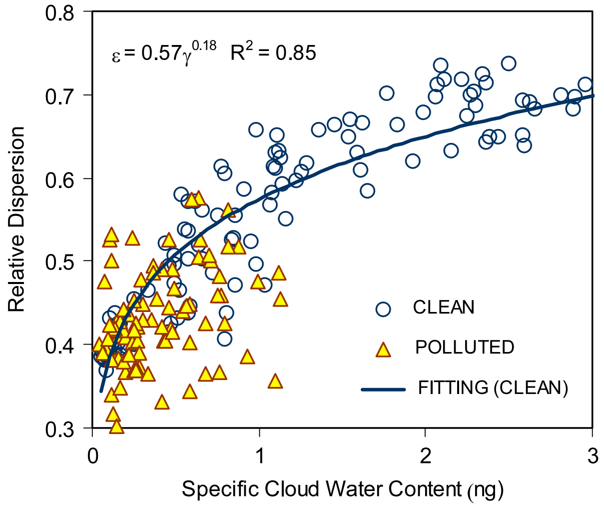

Figure 6. Relative dispersion as a function of the specific cloud water content, calculated from FSSP measurements. Each point represents a cloud average during the dry season under CLEAN and POLLUTED conditions in the Amazonian region.

are uncertainties as to whether the relative dispersion ε is correlated with the cloud droplet number concentration, the results found in this work confirm a relationship between the two variables, with the more dispersed (broader) cloud droplet size distributions being obtained under clean conditions (low cloud droplet number concentration).

The results presented show that the largest β parameters are the best fittings for clean conditions and the smallest β parameters are the best fittings for polluted conditions. According to these results (table 1), the liquid water content (0.43 ± 0.19 g m^{-3}) is practically independent of the cloud droplet number concentration. Otherwise, the effective radius was found to be very well correlated with the relative dispersion ($R^2 = 0.81$), as can be seen by figure 3. Equations (2) and (3) show that, under constant L, the effective radius (and also cloud reflectivity) is decreased (increased) by the increase in N. However, a decrease in relative dispersion ε, as caused by the biomass-burning aerosols, increases the effect caused on effective radius. Therefore, in a biomass-burning context, the dispersion effect acts to decrease (increase) the effective radius (and potentially, the reflectivity).

The results of our study show that an increase in CCN concentration from biomass-burning aerosols leads to a decrease in the spectral dispersion. According to equation (2), this impacts directly on the effective radius, increasing the cloud reflectivity. Therefore, during the Amazon dry season, the aerosol effect on spectral dispersion enhances the climate cooling predicted by the Twomey effect. Two important aspects need to be mentioned in comparison to the Liu and Daum (2002) results. First, since the dry season in the Amazon region is also vapor limiting, the dispersion effect could be substantially larger than in other continental polluted regions (low vapor availability, no chemical advantages for the potentially more hygroscopic CCN). Second, the background regime is continental, so there is not a chemical-limiting effect as in the low aerosol concentration regime (maritime regime) influenced by anthropogenic aerosols. In a maritime regime, where stratocumulus clouds typically predominate, the number of cloud droplets is controlled by the number of CCN, with less influence of water vapor availability, dynamical or large-scale effects.

4. Conclusions

Cloud droplet spectra data from both clean and polluted by biomass-burning conditions in the Amazon region were studied in this work. We analyzed results concerning the first measurements involving cloud number size distributions during the dry season in the Amazon region. In agreement with the literature, the results show that cloud droplet spectra are sensitive to the atmospheric aerosol loading. Clean conditions were found to be characterized by a low concentration of cloud droplets. Otherwise, under polluted conditions, high concentrations of cloud droplets with smaller effective radius were observed. The liquid water content during the polluted period was similar to that in the clean one, but the effective radius was substantially smaller.

The analysis of spectral properties suggested that the additional source of biomass-burning aerosols impacts the cloud reflectivity in a different way, as suggested in previous studies concerning the impact of pollution in maritime regimes. The cloud droplet size distributions during the polluted period were found to be narrower than during the clean one. A low relative dispersion means that the particles are all approximately the same size, enhancing the effect caused by the increase in number of cloud droplets (first indirect effect).

The results presented in this study constitute useful information for climate model simulations. Further studies are necessary in this direction to reduce the uncertainties associated to these models. This study involved spectral dispersion of cloud droplet size distributions under dry air conditions and high biomass-burning aerosol concentrations. The two phenomenologies (dry air and biomass-burning aerosol) are not dissociated in the context of the Amazonian region. They may also be acting in order to drive the cloud-precipitation process, with important consequences on the hydrological cycle and surface radiative balance.

Acknowledgments

This research was developed in the framework of the large-scale biosphere–atmosphere experiment in Amazonia (LBA). It was supported by Fundação de Amparo à Pesquisa do Estado de São Paulo (FAPESP) and Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq).

References

Andreae M O, Rosenfeld D, Artaxo P, Costa A A, Frank G P, Longo K M and Silva Dias M A F 2004 Smoking rain clouds over the Amazon *Science* **303** 1337–42

Bower K N and Choularton T W 1992 A parameterization of the effective radius of ice-free clouds for use in global climate models *Atmos. Res.* **27** 305–39

Bower K N, Choularton T W, Latham J, Nelson J, Baker M B and Jensen J 1994 A parameterization of warm clouds for use in atmospheric general circulation models *J. Atmos. Sci.* **51** 2722–32

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

Costa A A, Oliveira C J, Oliveira J C P and Sampaio A J C 2000 Microphysical observations of warm cumulus clouds in Ceará, Brazil *Atmos. Res.* **54** 167–99

Dandin P, Pontikis C and Hicks E 1997 Sensitivity of a GCM to changes in the droplet effective radius parameterization *Geophys. Res. Lett.* **24** 437–40

Daum P and Liu Y 2003 Dispersion of cloud droplet size distributions, cloud parameterizations, and indirect aerosol effects *Proc. 13th ARM Science Team Mtg*

Daum P and Liu Y 2005 Generalized expressions for effective radius, cloud radiative properties, and their application to studies of the first indirect aerosol effect *Proc. 15th ARM Science Team Mtg*

Dye J E and Baumgardner D 1984 Evaluation of the forward scattering spectrometer probe. Part I: electronic and optical studies *J. Atmos. Ocean. Technol.* **1** 329–44

Freud E, Rosenfeld D, Andreae M O, Costa A A and Artaxo P 2008 Robust relations between CCN and the vertical evolution of cloud drop size distribution in deep convective clouds *Atmos. Chem. Phys.* **8** 1661–75

Ghan S J, Easter R C, Chapman E G, Abdul-Razzak H, Zhang Y, Leung L R, Laulainen N S, Saylor R D and Zaveri R A 2001 A physically based estimate of radiative forcing by anthropogenic sulfate aerosol *J. Geophys. Res.* **106** 5279–93

Gonçalves F L T, Martins J A and Silva Dias M A F 2008 Shape parameter analysis using cloud spectra and gamma functions in the numerical modeling RAMS during LBA project at Amazonian region, Brazil *Atmos. Res.* **89** 1–11

Han Q, Rossow W B and Lacis A A 1994 Near-global survey of effective droplet radii in liquid water clouds using ISCCP data *J. Clim.* **7** 465–97

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

Howell W E 1949 The growth of cloud drops in uniformly cooled air *J. Meteorol.* **6** 134–49

Koren I, Kaufman Y J, Remer L A and Martins J V 2004 Measurements of the effect of Amazon smoke on inhibition of cloud formation *Science* **303** 1342–5

Lin J C, Matsui T, Pielke R A Sr and Kummerow C 2006 Effects of biomass burning-derived aerosols on precipitation and clouds in the Amazon basin: a satellite-based empirical study *J. Geophys. Res.* **111** D19204

Liu Y and Daum P H 2000a 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 2000b Which size distribution function to use in parameterization of effective radius *Proc. 13th Int. Conf. on Clouds and Precipitation* pp 586–9

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

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

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

Liu Y and Hallett J 1998 On size distributions of cloud droplets growing by condensation: a new conceptual model *J. Atmos. Sci.* **55** 527–36

Liu Y, You L, Yang W and Liu F 1995 On the size distribution of cloud droplets *Atmos. Res.* **35** 201–16

Lohmann U and Fleichter J 1997 Impact of sulfate aerosols on albedo and lifetime of clouds: a sensitivity study with the ECHAM 4 GCM *J. Geophys. Res.* **102** 13685–700

Lohmann U and Lesins G 2002 Stronger constraints on the anthropogenic indirect aerosol effect *Science* **298** 1012–5

Lohmann U, Quaas J, Kinne S and Feichter J 2007 Different approaches for constraining global climate models of the anthropogenic indirect aerosol effect *Bull. Am. Meteorol. Soc.* **88** 243–9

Lu M L and Seinfeld J H 2006 Effect of aerosol number concentration on cloud droplet dispersion: a large-eddy simulation study and implications for aerosol indirect forcing *J. Geophys. Res.* **111** D02207

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

Martins J A, Silva Dias M A F and Gonçalves F L T 2009a Impact of biomass burning aerosols on precipitation in the Amazon: a modeling case study *J. Geophys. Res.* doi:10.1029/2007JD009587

Martins J A *et al* 2009b Cloud condensation nuclei from biomass burning during the Amazonian dry-to-wet transition season *Meteorol. Atmos. Phys.* at press

Menon S, Unger N, Koch D, Francis J, Garrett T, Sednev I, Shindell D and Streets D 2008 Aerosol climate effects and air quality impacts from 1980 to 2003 *Environ. Res. Lett.* **3** 024004

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

Pontikis C and Hicks E M 1992 Contribution to the droplet effective radius parameterization *Geophys. Res. Lett.* **19** 2227–30

Procopio A S, Artaxo P, Kaufman Y J, Remer L A, Schafer J S and Holben B N 2004 Multiyear analysis of Amazonian burning smoke radiative forcing of climate *Geophys. Res. Lett.* **31** L03108

Pruppacher H R and Klett J D 1997 *Microphysics of Clouds and Precipitation* 2nd edn (Norwell, MA: Kluwer Academic)

Reid J S, Hobbs P V, Rangno A L and Hegg D A 1999 Relationships between cloud droplet effective radius, liquid water content, and droplet concentration for warm clouds in Brazil embedded in biomass smoke *J. Geophys. Res.* **104** 6145–53

Rissler J, Swietlicki E, Zhou J, Roberts G, Andreae M O, Gatti L V and Artaxo P 2004 Physical properties of the submicrometer aerosol over the Amazon rain forest during the wet-to-dry season transition—comparison of modeled and measured CCN concentrations *Atmos. Chem. Phys.* **4** 2119–43

Rissler J, Vestin A, Swietlicki E, Fisch G, Zhou J, Artaxo P and Andreae M O 2006 Size distribution and hygroscopic properties of aerosol particles from dry-season biomass burning in Amazonia *Atmos. Chem. Phys.* **6** 471–91

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

Shao H and Liu G 2006 Influence of mixing on evaluation of the aerosol first indirect effect *Geophys. Res. Lett.* **33** L14809

Slingo A 1990 Sensitivity of the Earth's radiation budget to changes in low clouds *Nature* **343** 49–51

Twomey S 1977 Influence of pollution on shortwave albedo of clouds *J. Atmos. Sci.* **34** 1149–52

Wang C 2005 A modeling study of the response of tropical deep convection to the increase of cloud condensation nuclei concentration: 1. Dynamics and microphysics *J. Geophys. Res.* **110** D21211