Atmos. Chem. Phys. Discuss., 5, S5941–S5947, 2005
www.atmos-chem-phys.org/acpd/5/S5941/
European Geosciences Union

Atmospheric Chemistry and Physics Discussions

ACPD

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Interactive Comment

Interactive comment on "Optical and physical properties of aerosols in the boundary layer and free troposphere over the Amazon Basin during the biomass burning season" *by* D. Chand et al.

D. Chand et al.

Received and published: 28 April 2006

Manuscript: 1680-7375/acpd/2005-5-4373: Optical and physical properties of aerosols in the boundary layer and free troposphere over the Amazon Basin during the biomass burning season by D. Chand , P. Guyon, P. Artaxo, O. Schmid, G. P. Frank, L. V. Rizzo, O. L. Mayol-Bracero, L. V. Gatti, and M. O. Andreae

Reply to reviewer #1.

First of all we thank the reviewer for meticulously going through the manuscript and providing numerous helpful comments. The constructive and valuable comments of the referee have improved the quality of our article.

Abbreviations used: A - Authors R - Reviewer

R. This is an interesting and useful manuscript presenting analysis of measurements of the optical properties of biomass burning aerosols during the SMOCC field campaign in Brazil in 2002. The unique aspect of this study is the presentation of optical properties of smoke aerosol measured at differing altitudes: the surface, boundary layer, and free troposphere. However, these in situ measurements are all made of aerosol downstream of drying units, and therefore may differ somewhat from optical properties under ambient atmospheric conditions. The manuscript would be strengthened if data on ambient relative humidity as a function of altitude were presented in order to help in assessing the possible influence of RH on ambient aerosol hygroscopic growth. Differences in aerosol scattering coefficient relationships with altitude are attributed to aerosol growth by coagulation and condensation, however some of this particle growth may be hygroscopic or as a result of interaction with clouds. There are numerous clouds imbedded in the smoke from mid-September through early October in Rondonia (dates of this investigation) and I suspect that there may be significant influences on the aerosol properties from the cloud-smoke interaction or simply by smoke passing through the high humidity environment of these clouds. Some discussion of this issue should be added to the paper.

A. In this paper we present results/data from the biomass burning period, which represents relatively dry conditions and events of high RH are less important. Also, the hygroscopic growth factors for pyrogenic aerosols are relatively modest (Reid et al., 2005; Rissler et al., 2006). The RH variation in the airborne nephelometer was between 50-80%. For most of the flights, the RH was lower in the FT compared to the BL. The hygroscopic effects can not explain the factor of 2 to 3 increase in Delta(scat)/Delta(CO) that was observed when transitioning from the boundary layer to the free troposphere.

In general, we believe that sampling dry aerosols has both merits and demerits. Since the optical properties of aerosols are dependent on relative humidity (RH), comparison of intensive and most extensive properties of aerosols requires fixed RH conditions.

For practical purposes, to control the RH it is easier to maintain low relative humidity than a higher constant relative humidity when sampling. It is easy to understand and reduce instrumental artifacts under uniform/constant conditions. The data at constant conditions are easier to use by modelers. Also, emission parameters like excess ratio are more appropriate if observed in the dry state instead of humid/ambient condition. To study the hygroscopic effect, we have made observations under ambient RH. These results will be presented in a separate manuscript [Chand et al., MS in preparation] as mentioned in the footnote text (page 4382).

Specific Comments: R. Page 4383 Section 2.3.4: Adding a couple of sentences of explanation regarding the utility and physical significance of the excess ratio would be helpful.

A. We have added more text.

R. Page 4385 line 1: “The average wo, calculated using Eq. (3), is about 0.92 +- 0.02. This value is significantly higher than the observations in earlier campaigns.” It should be stated that this is for dry aerosol and if possible it would be useful to give the range of ambient relative humidity during measurement times. I also suggest that the authors compare their estimates of aerosol single scattering albedo (SSA) obtained from their in situ measurements to remote sensing retrievals made from the AERONET measurements in Amazonia. See Dubovik et al. 2002, Table 2 that gives SSA of about 0.935 (interpolated to 550 nm) for Amazonian forest sites (which includes sites in Rondonia). This comparison is of the SSA retrieved for ambient aerosol and the entire atmospheric column versus dried aerosol at the surface, and therefore the implications of the differences should be discussed, such as an expected higher SSA for ambient conditions due to some hygroscopic growth (Kotchenruther and Hobbs, 1998).

A. We have added the RH range. The SSA text is modified and compared with SSA obtained from AERONET data. Some text is added to address the implications of humid aerosols.

R. Page 4385 lines 9-11: "Observations of higher scattering efficiency for the aerosols at higher altitudes compared with those near ground corroborate our observation of higher wo during the day time." More explanation or elaboration is needed here. Are you suggesting downward convective mixing of higher altitude aerosols (by convective downdrafts) is increasing the single scattering albedo at ground level at mid-day?

A. Text is added to make it clear.

R. Page 4385 lines 22-29: I suggest that you consider comparing mass scattering efficiencies for aged smoke in Amazonia from the literature (4.1 and 4.2 m2/g from Reid et al., 2005, ACP, Part 3 - tables 2 and 5 respectively) rather than just discussing data for young smoke from SCAR-B. Also consider adding some comparisons of your mass absorption efficiency data to those given in Reid et al., 2005, Part 3 (0.50 for tropical forest aged).

A. Comparison is made and the Reid et al 2005a, 2005b references along with mass scattering and absorptions efficiencies are included in text.

R. Page 4386 lines 26-27: You mention two types of aerosol in the BL and FT. I suggest that it would be more accurate to describe these differences as 2 ages (and therefore differing size distributions) of the same aerosol type, which is biomass burning aerosol.

A. Text is added as suggested.

R. Page 4388 lines 26-27: See my comment for Page 4385 lines 9-11 (above). More explanation/elaboration is needed here for the physical mechanism of relating the altitude gradient in single scattering albedo to the diurnal cycle of SSA at the ground.

A. Some text is added to elaborate the comments of page 4385. We are including the diurnal variations in a separate MS under preparation.

R. Page 4389 lines 6-11: A comparison of the Angstrom exponent computed from in

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situ measured light scattering coefficient alone to the AERONET sunphotometer based measurements (from total extinction) is given in the text and Table 3. However, I suggest that you explicitly discuss the differences in the quantities utilized to compute Angstrom exponent, such as the fact that the absorption coefficient is expected to have lesser wavelength dependence than the scattering coefficient. Therefore this fact alone could partly account for in-situ Angstrom exponent (computed from scattering coefficient only) being greater than sunphotometer measured Angstrom exponent (based on total extinction optical depth). Also, the spectral scattering coefficients measured by the nephelometer are for dry aerosol particles and this could result in somewhat higher Angstrom exponents than for aerosols under ambient humidity that may have experienced some hygroscopic growth.

A. We had missed to add this important result! Now we have added it. Regarding the hygroscopic growth, we want to make clear here that there may be little difference in RH in the aircraft nephelometer and ambient aerosols, however, no additional drying unit was applied to dry the aerosols in the aircraft nephelometer unlike the nephelometers at FNS. The drying effect in the aircraft was due to the somewhat higher temperatures (4 - 8 C) inside the aircraft compared to ambient temperatures and the self-heating inside the nephelometer.

Technical Corrections:

R. Page 4376 line 15: Reid et al. 2004 a, b should be changed to Reid et al. 2005 a, b since the final version of this paper is now published in ACP.

A. Added

R. Page 4383 Equation 5: Missing parentheses - log should be of the ratio of AOT at the two wavelengths.

A. Corrected.

R. Page 4383 line 14: Equation 6 is missing.

A. Included

R. Page 4384 line 9: You list September 20 as a rain event at FNS, yet there is no rain event indicated for this date in Figure 2. Either correct this or explain the discrepancy.

A. Corrected.

R. Page 4384 line 19: “..and CO (r2=0.87).” In Figure 3c the r2=0.88. These values should be consistent between the text and figure.

A. Corrected.

R. Page 4385 line 3: “Eˇ pronounced diel variations” should be “Eˇ pronounced diurnal variations”

A. Corrected.

R. Page 4389 lines 17-20: Need to refer to Figure 6 here.

A. Referred

R. Page 4390 line 15: I suggest that you state that the SSA of 0.92 at 540 nm is for dry aerosol particles.

A. Mentioned

R. Page 4391 line 3: The light scattering coefficient should be used here, not the light absorption coefficient (i.e. Fig 6).

A. Corrected

R. Page 4391 line 8: I suggest that it would be preferable to avoid referring to Tables in the conclusions section.

A. The sentence is modified.

R. Page 4400 Table 3: You present a burning period average Angstrom exponent of 2.0 in the text (from the nephelometer data), so why is the period average left blank in

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Table 3?

A. The blanks were mentioned for the period Sep 16-27. The aircraft flights were conducted later than this period. To avoid the confusion, we have modified the title of the bottom row from ‘burning period’ to ‘burning event’.

R. Page 4402 Fig. 2: The subscript ‘a’ should be used in the first line of the caption for the light absorption coefficient.

A. Corrected

R. Page 4404 Fig. 4: “The slopes in the BL and FT are shown by the dashed and long dashed lines.” I suggest that using solid lines for the FT would make the figure much easier to read.

A. Changed

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