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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.

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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 #2.

First of all we thank the reviewer for carefully reading our manuscript and comments.

Abbreviations used: A - Authors R - Reviewer

R. Overall review: In this manuscript the authors present regressions of light scattering, absorption, CO and TEOM estimated mass at a surface site and on an aircraft for the LBA campaign. While I empathize with the authors on what they are trying to do and express, the paper is in a state where the findings are not easy to apply. Key information on the vertical profile of aerosol particles and CO is totally absent and we are left with a series of non-interpretable regressions. The two regressions with real meaning, the construct of the mass scattering and absorption efficiencies, are not well executed on a number of levels and most likely are biased. Most significantly, the TEOM is well known to underestimate organics species, particularly smoke. I suspect that their derived values of the mass scattering/absorption efficiencies are biased high by 10-30%. Gravimetry using samples off of the exact pipe from the nephelometer and PSAP is the only reasonable way for the mass efficiency measurements to be constructed.

A. We have added text to make the results more clear. Figure 7 contains vertical profiles of both particle number concentration and scattering coefficients. Simultaneous observations of TEOM and gravimetric techniques are used to validate the TEOM data. Text is added to show the magnitude of underestimation of organics by TEOM measurements (about 8%).

R. While I have many criticisms, I see real promise in this data if presented properly. Although I do not knwo if the author's can make all of the changes in a timely manner. Specific issues are listed below.

Abstract and throughout: The usage of "Boundary layer (BL)" and other planetary boundary layer terms throughout the manuscript needs to be more specific. Authors should be specific as to what they mean, Planetary Boundary Layer (PBL), Surface Layer, Convective Boundary Layer (CBL), beneath the trade inversion, etc. It is not at all clear from the manuscript what the boundary layer dynamics they are referring too are. A plot of an ideal boundary layer structure would be very helpful.

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Interactive Comment

A. We have added text and explained the boundary layer in MS. A plot has been published in a companion paper (Rissler et al., 2006).

R. Page 4377 line 8, Section 2.1, Line 5. “see level” should be “sea level”

A. Corrected

R. Section 2.2 Use of “Dry air (RH<40%)” is not really dry. You need to get down below 30% to be considered dry. If you look at the collective works of Tang, and consider that the background RH for the region is fairly high, then particles will no doubt be on the upper hysteresis curve. Hence, there can be tightly bound water on the order of 10 to 20% of particle mass.

A. Yes, we agree that ideally the dry air should be much below down 30 % to get the dry aerosols. But due to practical difficulties we could best achieve the RH about 25 - 40 % only. The observed hygroscopic growth in scattering coefficient at higher RH (30 - 50 %) is <10 % for the sampled submicron aerosols (<1.5 u m). The water uptake by pyrogenic aerosols is much smaller than by the inorganic salts considered by Tang and coworkers. In measurements made during the same campaign, Rissler et al. (2006) have shown that water uptake is only about 9% at RH=90%. To avoid any confusion, now we have changed the words ‘dry aerosols’ to ‘dried aerosols’ throughout the MS.

Tang, I. N., and Munkelwitz, H. R.: Water activities, densities, and refractive indices of aqueous sulfates and sodium nitrate droplets of atmospheric importance, Journal of Geophysical Research, 1995.

R. Section 2.2. It is fairly unclear as to which instruments have which cut-points. Similarly, instrumentation is presented in a hodge-podge manner and is difficult to follow. A table would be very helpful. Authors mention the SMPS but do not give any data. Why? It may help their case on a number of discussed topics.

A. Table (#2) is added to show the size cut-points. Here, the main objective of using SMPS data only from aircraft flights is to see if there is any change in the sizes

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of aerosols in vertical profiles in BL and FT (Figure 5). Detailed size distributions data/results at surface site FNS are discussed in Rissler et al., 2006 (now reference is included).

R. Section 2.2 Use of the TEOM. While TEOMs are considered "usable by the EPA" in reality they tend to have serious biases, specifically with regards to organic aerosol particles such as smoke. By heating to 50oC, often semi-volatile organics are driven off. If drying for light scattering is done by perma tube, and for the TEOM by significant heating, then no doubt any measurement of the mass scattering efficiency will be biased high, at least 15-30%, and maybe more. Please see "Long-Term Field Characterization of Tapered Element Oscillating Microbalance and Modified Tapered Element Oscillating Microbalance Samplers in Urban and Rural New York State Locations, by Schwab et al., AWMA 2004" This problem has been known for some time, but this is the most recent discussion. I suspect that the very high mass scattering efficiencies that are found here are a result of this bias.

A. Yes we agree that TEOM measurements operated at 50 C may be biased for the semi-volatile (organic) aerosol. Nevertheless, the magnitude of this bias depends on the ambient temperature and chemical composition of sampled aerosols. At a tropical site like FNS, the higher ambient temperature and fraction of submicron size aerosols ($<$1.5 u m) produce less effect of volatilization in organic compounds. We have validated/compared the TEOM results with gravimetrical observations; the difference between the two instruments was found to be about 8%. Now we have included this information in the MS.

R. Section 2.3.1 Why did you make corrections to the radiance research nephelometer and not the TSI? Besides, as shown by Anderson lon-lambertian light source errors are much larger than truncation to begin with. The use of a RR neph at the surface and the TSI on the airborne platform constitutes a study design issue for the paper's primary premise- comparing surface and airborne particle properties. How did the instruments compare in fly-bys?

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A. We have all the supporting/extensive measurements at ground site (FNS), but it was not possible to have all these measurements in aircraft. Using these supporting observations at FNS, we corrected the Radiance Research (RR) nephelometer data. Since truncated angles are higher in RR nephelometer compared to TSI, we feel that the correction in the RR neph is required and useful. Using the comparison in the fly-by times, the TSI nephelometer is higher by 5-15% compared to the RR neph. This difference is likely attributable to different nature and number density of aerosols at higher heights, higher relative humidity and higher cut-points in the TSI nephelometer. Now, in addition to the truncation correction, we have corrected the dried aerosols nephelometer data at FNS for non-Lambertian effects. Inter-comparison results during fly-bys are included in MS.

R. Section 2.3.2 The authors argument with the Bond corrections is reasonable, but not well executed. First, at the very least the authors should state the difference in photoacoustic and Bond corrections, especially of they are going to present data to three significant figures. Referencing a paper in preparation in this context does not help us reviewers. Also, Pat Arnott's instrument is not a primary standard either.

A. We have added text to emphasize the differences between the Bond correction (laboratory-based; using extinction cell combined with nephelometer as reference absorption device) and our correction (calibration with ambient aerosol in the field using the PAS as reference device). In addition, we referenced the Schmid et al. (2005) paper that describes in detail the procedure and the results of the field calibration. We agree that Pat Arnott's instrument (PAS) is not a primary standard for aerosol absorption; however, no such claim was made in the manuscript. We only emphasized that in contrast to filter-based methods, the PAS measures light absorption of aerosols in their suspended state and the PAS can be calibrated in the field (using NO2 as calibration gas).

R. Section 3.1 Discussion of Figure 3. Clearly there are two populations in this figure, with a slop changing at Ÿ120 ug m-3. These should be analyzed separately.

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A. The number of point (hours) in the second population is less than 1% of the total data. So we decided to focus the main results on the first population. We have added some text to mention it in the MS.

R. Section 3.1. Comparison to SCAR-B. The comparison is not really complete. First the SCAR-B measurements were made at ŸPM4 as opposed to the modified PM1.5 done here. This would in itself make a 20% change. Also in the SCAR-B campaign comparisons are made between aged and source optical properties. Aged values equivalent to PM1.5 would be Ÿ4.1 m2 g-1, compared to the 2.7 to 3.6 m2 g-1 given here. Besides, if you have a value for mass scattering efficiency that is higher than every other measurement in existence for all other places in the world, you may want to try and justify it. If the TEOM is off by 20%, well within its uncertainty for smoke particles, and an almost certain bias here, the measurements are in agreement. As for absorption, in their review paper Reid et al admits that the original integrating plate values for SCAR-B are biased low. But, even here the values do not quite reach the values presented here, even with the extinction cell. One thing that the authors may want to consider is that the particle concentrations for this study are higher than those in SCAR-B. This no doubt effects the evolution process that converges to larger particle size. What does the SMPS data suggest?

A. We have modified the text in MS.

R. Figure 4. This is almost unreadable and impossible to follow. Such regressions offer very little context. How about a few vertical profiles? How are the authors defining the PBL? Thermodynamic soundings. “data for different flights is shown in different colors”? A key on the figure would be most helpful. Even so, in this context all that can be said is that the regressions are different at different levels and flights- no surprise there. This data needs to be analyzed in the larger context of large-scale meteorology. Reid et al., [1998] found the development of the surface mixed layer and the top of the CBL/trade inversions to be very complicated. Given the significant number of flights the authors should be in a position to add to this.

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A. Now the text is made clearer. The current article presents the optical properties of haze (plumes + background air) from biomass burning activities over the Amazon basin. The aim of the regressions in figure 4 is to show the different mode/population of aerosols in BL and FT. So far, such observations are not reported to our knowledge. In addition to the meteorology, we believe the physical processes (e.g. coagulation and ageing) play important role in deciding the optical properties of these aerosols. We have added some text to discuss it. A further discussion of the aerosol properties in the meteorological context is available in the companion paper by Rissler et al. (2006). Also, more integrative papers, that put the detailed results of the SMOCC campaign in a large context are in preparation. Such an analysis would be beyond the scope of the present paper.

R. Section 3.2. I think a scatter plot of aircraft values of light scattering, CN, and CO, versus surface measurements at overpass is an absolute necessity. This is especially true considering the relationship between ground and airborne measurements is the point of the study.

A. Inter-comparison between the observations at FNS and aircraft was made during day time using the seven fly-by flights at a height of 150 - 600 m above FNS. Since the fly-by was on a time scale of 1-3 minutes, we have averaged the observed parameters for that interval at both platforms and inter-compared the values. Text is added in the MS to discuss it. However, it is important to note that for the discussion of the vertical structure of the optical aerosol properties we rely on correlations of bscat and CN (or CO), i.e., constant instrument offsets are irrelevant, since we are only interested in the respective slopes. These slopes are given in Figures 3 and 4, where we find good agreement for bscat/CO for FNS (0.43 Mm-1 ppb-1) and aircraft (for BL: 0.38-0.40 Mm-1 ppb-1). We also show that the correlation between bscat and CN is poor at FNS whereas it shows much better correlation for airborne measurements. In addition, for the discussion of the different optical properties of aerosols in BL and FT (e.g., Figure 4), we rely entirely on aircraft measurements, i.e., these results are not affected by

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potential differences in instrument response at FNS and in the aircraft.

R. Section 3.3. The development of the OSH is totally nonsensical, especially considering you have aircraft data! What are the vertical profiles of bscat? What do those tell you?

A. OSH is an additional piece of information. Here we use surface (bscat and AOT) data to get an additional result (an approximate optical height). This OSH is consistent with the BL height and the height where a sharp transition (1200-2000 m) in Delta(bscat)/Delta(CN) was observed. A figure with vertical profiles of bscat and CN has been added (Figure 7) and the results are compared with OSH. OSH may be useful for meteorology, modeling and radiative studies over the Amazon basin. Now we have added more text to support this result. We feel it should be included in the MS.

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