Atmos. Chem. Phys. Discuss., 3, S935–S942, 2003
www.atmos-chem-phys.org/acpd/3/S935/


ACPD
3, S935–S942, 2003

Interactive Comment

# *Interactive comment on* "Physical properties and concentration of aerosol particles over the Amazon tropical forest during background and biomass burning conditions" *by* P. Guyon et al.

**P. Guyon et al.**

Received and published: 1 July 2003

Reply to Anonymous referee #3

http://www.cosis.net/members/journals/df/article.php?a_id=148

1) Abstract single-scattering albedo only lists values from the end of the dry season. They should probably give values from both time periods.

Response: This is not correct. In the abstract we do not only give single-scattering albedo values for the end of the dry season, but for both the first part of the wet season (the cleanest period) and the biomass burning influenced dry season. Single-scattering albedo for the cleanest period of the wet season was 0.97, and a median value of 0.91 was found for the dry season period. All values given in the abstract are actually

Full Screen / Esc
Print Version
Interactive Discussion
Discussion Paper

comparing these two periods (for aerosol number concentrations, aerosol optical depth, scattering and absorption coefficients, and single-scattering albedo).

2) Methods: Index of refraction used to correct PCASP. This relates to the most significant weakness and or uncertainty of the manuscript. The authors are basing a great deal on their knowledge of the index of refraction of smoke particles and values they give are significantly lower than what is typically used (typically 1.5). (This is based on chemistry and AERONET inversions.) This is not to say that the employed values are not correct, but rather the uncertainty in the field needs to be fully explored and their ramifications discussed. These changes strongly influence the reported volume and hence all subsequent size and density parameters that are later derived. A propagation of error assessment must be performed before the paper can be published.

Response: A manuscript has already been published on the method we developed for the retrieval of the refractive index of the particles that were sampled during these two campaigns, including one example of an uncertainty calculation and a summary of the uncertainties:

Guyon, P., O. Boucher, B. Graham, J. Beck, O. L. Mayol-Bracero, G. C. Roberts, W. Maenhaut, P. Artaxo, and M. O. Andreae, Refractive index of aerosol particles over the Amazon tropical forest during LBA-EUSTACH 1999, J. Aerosol Sci., doi:10.1016/S0021-8502(03)00052-1, 2003, in press.

For a more detailed description of the uncertainties related to these calculations, please refer to Chapter V of Pascal Guyon's PhD Thesis, which is available online:

Guyon, P., Chemical and physical properties of Amazonian aerosol particles, PhD thesis, Johannes Gutenberg-Universität Mainz and Université de Paris VII-Denis Diderot, Mainz and Paris, URL: http://ArchiMeD.uni-mainz.de/pub/2003/0035, 2002. These two references were added to the text.

However, referee #3 is comparing the results we obtained for the real part of the re-



ACPD

3, S935–S942, 2003

Interactive Comment

fractive index to those based on chemistry or AERONET inversions, arguing that our values are too low.

First of all, one should remain cautious about drawing conclusions based on comparison of refractive indices of atmospheric aerosols obtained using different techniques. The notion of refractive index is somewhat ill-defined when applied to inhomogeneous particles and can only be expressed as an "equivalent refractive index" specific for the given measurement conditions and set of assumptions made about the properties of the aerosol. Here, the calculated refractive indices can be defined as equivalent refractive indices for spherical, homogeneously internally mixed particles with the same bulk absorption and scattering properties as the actual particles. It should also not be forgotten that concerning AERONET measurements, notably for biomass-burning aerosols in South America, Dubovik et al. (2002) noted that, we cite, "w0 determined from in situ measurement techniques are typically lower than AERONET retrieval valuesĚ The cause for those significant differences from different measurement types (in situ versus remote sensing) is not fully understood and should be the focus of future examination." In the AERONET retrieval, the index of refraction is not independent from the size distribution and single-scattering albedo of the aerosols, which could explain partly the observed differences. However, concerning this issue, the imaginary part of the refractive index has the strongest influence, and the AERONET values are comparable to ours. These authors reported for Brazilian forest burning a value of 1.47 (+/-0.03) - 0.0093 (+/-0.003)i, compared to our value of 1.41 (+/-0.05) - 0.013 (+/-0.005)i. (Reference: Dubovik, O., B. Holben, T. F. Eck, A. Smirnov, Y. J. Kaufman, M. D. King, D. Tanré, and I. Slutsker, Variability of absorption and optical properties of key aerosol types observed in worldwide locations, Journal of the Atmospheric Sciences, 59 (3), 590-608, 2002.)

However, the major difference between AERONET retrieval and our method is that we derived the refractive index of aerosol particles that were measured close to the ground at near-ambient conditions of temperature and relative humidity, whereas the



ACPD

3, S935–S942, 2003

Interactive Comment

AERONET inversion is integrating the whole aerosol column. In our calculations, the water content of the aerosols certainly contributed to a decrease in the value of the refractive index towards that of water (1.33 - 0i) since we were confronted with high humidities most of the time, whereas AERONET would depend on humidity and aerosol concentration profiles. This is not something new, and similar observations of a decrease in the index of refraction of aerosol particles with decreasing measurement height (from aircraft measurements) and/or increasing relative humidity are available in the literature, e.g.:

- Redemann, J., Turco, R. P., Liou, K. N., Russell, P. B., Bergstrom, R. W., Schmid, B., Livingston, J. M., Hobbs, P. V., Hartley, W. S., Ismail, S., Ferrare, R. A., & Browell, E. V. (2000). Retrieving the vertical structure of the effective aerosol complex index of refraction from a combination of aerosol in situ and remote sensing measurements during TARFOX. Journal of Geophysical Research-Atmospheres, 105, 9949.

OR:

- von Hoyningen-Huene, W., Schmidt, T., Schienbein, S., Kee, C. A., & Tick, L. J. (1999). Climate-relevant aerosol parameters of South-East-Asian forest fire haze. Atmospheric Environment, 33, 3183.

The same remark is applicable to refractive indices retrieved from the chemical composition of the particles, as the latter technique often does not include the water content of the particles.

Given the fact that this discussion is available in details from the above-mentioned publications, we limited ourselves to specify the conditions at which the measurements were made in Section 2.3, where the refractive indices are first mentioned.

3) Section 3.2, end of second paragraph: The authors may want to be careful about trying to pull two modes out of the PCASP. PCASP bin sizing is not optimal for typical accumulation volume distributions and particle index of refraction most likely varies

between particles. There have never been any bimodal size distributions of smoke published (that I know of) from mobility analysers. As these are better instruments for the task I suspect artefact in the PCASP.

Response: Correct, and we are actually very cautious about it. We do mention in the text (page 1382) that this second mode for accumulation particles has been typically observed with PCASP instruments previously, but usually not with a differential mobility particle sizer, citing as an example Reid and Hobbs (1998), who measured similar type of aerosols as ours with both instruments. Moreover, we mention twice in the text that this bimodal accumulation mode could be a product of some artifact in the PCASP data Finally, when fitting the accumulation mode of these size distributions (and particularly the volume/size distributions) with a lognormal equation, we do apply a unique equation to this mode, because we think it has "a more physical meaning". However, this does not mean that the observed size distributions from the PCASP are fundamentally wrong, and we are as confident in our data as one can be when using this instrument, since the instrument was working without problem during the campaign, was calibrated by the manufacturer prior to the campaign, and data were corrected for the refractive index of the particles using the software provided by the manufacturer.

It is also interesting to note that the inverted MOUDI mass/size distribution from this sampling period also shows a shoulder for this mode. Nevertheless, bimodal size distributions of submicron smoke aerosol particles from mobility analyzers have been published previously, with the first mode typically peaking at particles diameter $<$0.1 mm, i.e. smaller than what was observed in the present study. For a recent publication, see for example: Hedberg, E., A. Kristensson, M. Ohlsson, C. Johansson, P. A. Johansson, E. Swietlicki, V. Vesely, U. Wideqvist, and R. Westerholm, Chemical and physical characterization of emissions from birch wood combustion in a wood stove, Atmos. Environ., 36 (30), 4823-4837, 2002.

It should also be kept in mind that we were measuring a mixture of young and aged plumes of burning and smoldering phases, which are characterized by different size distributions.

Most of the above discussion was already included in the original text. We have added a small section on measurements of bimodal accumulation particles from mobility analyzers.

4) Index of refraction uncertainty in density calculation. This part of the analysis is a bit misleading. First by RAW PCASP I assume they mean calibration bead index of refraction of 1.59 (which is not in common use). It is noteworthy that the Radke/Stith derivation was also from a comparison of OPC data to filter mass. Hence the current estimate may be similar to Radke/Stith because there is a method bias. What can be concluded from this analysis is that the density derivation is highly uncertain and may be better left to mobility analysers than OPCs (which is much more closely related to physical size).

Response: This is correct and is also the reason why, in order not to mislead the reader with data which may not be accurate (the volume being sensitive to the power of 3 to the correction for the index of refraction applied to the size bins of the PCASP), we give density values that were derived using both the corrected and the uncorrected PCASP data. We also provide the reader with a potential explanation for the large spread in the observed values. Therefore, we do point out in the text the large uncertainties associated with these values, and take this opportunity to draw attention to the large discrepancies between the values reported in the literature on particle density (most model study are still using the Radke/Stith value of 1 g cm-3). The density of particles is a parameter widely used in aerosol science, and there are many assumptions made about it. Therefore, we conclude this paragraph with the following sentence: "Too little information on the density of this type of aerosol is available to date, resulting in large uncertainties in aerosol model calculations."

We have now added to the text the comments of the referee that our estimate may be



ACPD

3, S935–S942, 2003

similar to that of Radke/Stith because we used the same method, and there may be a method bias.

5) Page 18: The y intercepts for figure 5 regression are important. One should not simply force a regression through zero because it seems physical. The change in y with x is what is important. Leaving the y intercept in can be useful in detecting bias.

Response: In our case the intercept doesn't have a physical meaning; however, it is true that it can help detecting bias. Therefore, we note here the value of the regression obtained while not forcing the intercept through zero. With MOUDI sampled mass concentrations expressed in mg m-3 (y-intercept), and the PCASP cumulated volume concentration in mm3 m-3 (x-intercept) (identical to that presented in Figure 7), we obtained:

- For the LBA-EUSTACH 2 (n = 27):

- with uncorrected PCASP: y = 1.31x (+/-0.06) + 1.88 (+/-1.31), r2 = 0.95

- with corrected PCASP: y = 0.59x (+/-0.03) + 2.79 (+/-1.27), r2 = 0.95

- For the LBA-EUSTACH 1 (n = 5):

- with uncorrected PCASP: y = 3.25x (+/-0.16) + 0.04 (+/-0.20), r2 = 0.99

- with corrected PCASP: y = 1.88x (+/-0.10) + 0.01 (+/-0.21), r2 = 0.99

The values given in brackets are the standard errors associated with the least square fit.

It can be seen that overall the intercept is small compared to the range of values observed during each season.

For this reason, and because the above additional regression analysis would not have contributed to a better understanding of the data, the text was not modified here.

6) Discussion on density, page 19. Again, this density measurement hinges on using

the correct index of refraction. This is a pretty big spread in values discussed.

Response: The spread is indeed large (see comments above), and this is why we limited our comparison of these results with those from the other campaign.

7) Single scattering albedo discussion is relatively complete and error free.

Response: No comment needed

8) Overall, a relatively complete and well-written paper although not, perhaps, a major advance. But, well documented findings in one of the larger data-void regions of the world. Nevertheless, one should not read too much into PMS probe data.

Response: OPCs are widely used instruments in aerosol science. We think it is valid and useful to point out the pros and cons of using such an instrument by, for instance, regressing the results obtained from an OPC on those obtained with another instrument (here a MOUDI), in order to obtain a "simple" parameter (the particle density) and discussing the values computed.

---

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 1367, 2003.