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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Optical closure experiments for biomass smoke aerosols" *by* L. E. Mack et al.

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Response to Review by A. Ali Abo Riziq

This article presents very important experiments and interesting results in studying the optical properties of biomass smoke aerosols of different types of fuels. This article undoubtedly is suitable for being published due to the significance of the experiments and the findings that are presented in this paper. However, some major comments have to be addressed.

Response: Thank you for your comments and careful review. Below we address your concerns.

1) Page 7477: The authors discuss the correction factor 0.884 due to particle losses.

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This is not clear; do the losses occur in the sampling system or inside the instruments (PAS and Nephelometer)? If the authors mean that the losses occurred in the sampling system, that correction factor would be reasonable. However, it is not clear how that would affect the size distribution measurements. In addition, if the authors mean that losses occurred in the instruments, this factor seems very high. The authors could test this issue by measuring the particles at the entrance and the exit of the instruments.

Response: All three reviewers asked about this correction factor, and we here summarize our responses to all of these related comments.

The scattering coefficients calculated from Mie theory and measured size distributions were about 12% lower than those measured by the nephelometer (corrected for calibration and truncation errors); however, they were highly correlated, r2 = 0.99 (we have added this r2 to the text). The high correlation and constant multiplicative offset suggest a bias. The $\sim$12% difference is somewhat larger than that found by Anderson et al. (Anderson, et al., 1996) who reported a maximum difference of $\pm$10% between measured and calculated (for ammonium sulfate, from size distribution data) scattering coefficients for the same type of nephelometer. While our 12% discrepancy is not so different from their estimate, and thus from one point of view is consistent with the level of "closure" that might be expected, it is puzzling as to why it appears as a bias in our dataset. Possible sources of bias include the nephelometer calibration, sizing / counting biases, and different particle losses between the two instruments.

It is unlikely that flow errors in the DMA are responsible for this discrepancy, as flows were checked and reset before every experiment and were always within 1% of the target. Further, we have a lot of experience running the sizing rack and using these data for a variety of purposes and have not encountered this large of a bias in size distributions before.

With respect to the nephelometer gas calibrations, these are relatively standard, and Anderson et al. (1996) attribute only $\pm$1% uncertainty in measured scattering to the gas

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calibration. It's not entirely clear to us how this large of a bias could be introduced from this source, but it's possible there were errors in the concentrations used to compute the expected gaseous scattering.

The smoke was sampled from the main combustion chamber through mixing barrels and long lengths of tubing, into a manifold situated within a meter of all of the optical instruments. We ignored any losses up to the manifold, and assumed particles were well-mixed within the manifold. Short lines of quarter-inch conductive tubing that led to each instrument were attached to the manifold. The lengths of the lines were chosen, based on the different instrument sampling flow rates, to ensure the same residence time in each, and we did not attempt separate loss corrections for the tubing connections. We have characterized and accounted for the particle losses for the actual inlet and other tubing connections used in our sizing rack (e.g., they were measured and reported by Hand and Kreidenweis (2002) and are included as part of the alignment procedure). The manifold and conductive tubing connections are not typically part of this sizing system, so there may be some additional, unaccounted-for losses associated with those elements. Unfortunately, we cannot follow the suggestion of A. Ali Abo Riziq to attempt to re-measure losses because the system configuration no longer exists.

In any case, the final result was that we had to divide the Mie-calculated scattering coefficients by 0.884 (or, alternatively, had to multiply the nephelometer data by 0.884) to achieve closure for our ammonium sulfate calibration aerosol. The purpose of a calibration is to establish the best baseline, so we believe it is reasonable and necessary to apply this same correction factor in every case. Nevertheless, Dr. Zieger is correct in pointing out that if particle loss was, in fact, causing this discrepancy then we might expect this factor to change with changing size distribution and particle shape, although we have no way to account for this. We now note this in the text. Further, if the correction should have been applied to the scattering measurements instead, then the ωmeas reported in Table 1 are too low. This would affect our subsequent calcula-

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tions, including the comparison in Figure 3. We mention this in the text, but have not propagated this change as we think it is more likely that the bias is associated with our size distribution measurements.

Finally, we note that the refractive index for dry ammonium sulfate that we used is that reported and applied by Garland et al. (2007) as appropriate at 532 nm; we have corrected our wording in the text and the reference in the Table.

2) Table 2. The imaginary part of the refractive index of the organic carbon is taken as zero. This value needs to be checked since several studies suggested slight absorption for organic carbons from biomass burning.

Response: We agree it is possible that the organic carbon contributes some absorption in the wavelengths addressed in this work. We have added a statement that we have neglected this potential contribution and refer to the recent work by Adler et al. (2010), which nicely addresses this point. We cite their estimate of effective total RI of OC in diesel soot of n=1.519+i0.048. We also mention that Chakrabarty et al. (2010), working with data from the FLAME studies, attributed an imaginary part of the refractive index at 532 nm of 0.0027 and 0.0006 to the OC component of Ponderosa pine duff and Alaskan duff, respectively. This is shown in their Fig. 4b, but the numbers are not explicitly mentioned.

3) Page 7477: The retrieval of the refractive indices: the authors present the routine for retrieving the complex refractive index using the bscat or babs measured by the Nephelometer and the PAS systems, respectively, including the calibration test of ammonium sulfate. In the calibration test, they included the babs measurements to retrieve the complex refractive index. The authors should clarify what the meaning of "including" is (e.g. is it just summing the basb and bscat so that ymeas=babs+bscat?).

Response: During the ammonium sulfate calibration we measured the optical properties using both the nephelometer and the PAS. Because ammonium sulfate is a purely scattering aerosol at this wavelength we would expect measurements of babs = 0 from

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the PAS. However, there is some amount of uncertainty in the measurement of the PAS, and including the actual babs measurements in the complex refractive index will include the PAS uncertainty in the retrieval. We chose this approach to more accurately represent the uncertainty in the retrieved refractive indices, which use both pieces of information.

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