

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

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Response to Review by Anonymous Referee #1

Overall, this paper represents a great deal of work and will become a useful reference to our scientific community. Yet, the following major comments need to be addressed for clarification of the results.

Response: We thank the Reviewer for his/her comments and consider each below.

Minor comments:

1) Eliminate all acronyms in the abstract – FLAME and USFS

Response: These acronyms have been removed or explained.

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2) Line 17 should state (e.g. Ramanathan et al., 2007), page 7470

Response: This has been changed.

3) Between lines 13 and 15 equivalent sign should be replaced by approximate sign, page 7472 ; also page 7474 line 14

Response: This has been changed.

4) The following sentence on page 7475 needs to be clarified: “It is well known that particulate emissions vary considerably between the flaming and smoldering phases of combustion, and Reid et al. (2005) suggested relationships between the combustion conditions and the w of the smoke aerosol.” The phrase “well-known” needs to be supported by references.

Response: A reference to McMeeking et al (McMeeking, et al., 2009) has been added after this sentence.

5) Further needs to be replaced with Furthermore on Page 7476, line 7

Response: This has been changed.

6) The statement on page 7477 regarding the properties of ammonium sulfate (nearly spherical) need to be supported with a reference

Response: A reference to Mikhailov et al. (2009) has been added.

7) Page 7477 – statement by a factor of 0.884 needs to be clarified. 88% different? Or a 12% discrepancy?

Response: See below: we divided by 0.884 to attain agreement.

8) Page 7478 – line 21, nonsphericity statement with regards to size, needs a reference.

Response: References to Decarlo et al. (2004) and Slowik et al. (2004) have been added.

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9) Page 7479 – Need to cite the size distribution instrument used by Hand et al., 2010, was it also the OPC?

Response: Hand et al. (2010) used the same DMPS system used for this work. This has been stated in the manuscript.

10) Page 7481 – need reference or further explanation after – high absorption lead to inaccurate OPC sizing. . .

Response: A reference to Levin et al (2010) who discuss this issue has been added.

11) Page 7481 – need reference on line 15, after 630 nm are present.

Response: 630 nm just refers to the upper end of the DMPS measurements. This has been clarified in the text.

12) Page 7484 – need reference on line 14 after smoke aerosol.

Response: A reference to Reid et al. (2005) has been added.

Major comments:

1) The SSA mean standard deviation is surprising low at 0.007. Are you sure of this calculation? I would definitely suggest the authors look again at this method for calculating the relative uncertainty in SSA measured. This is difficult to review, as there are no plots of SSA over time for the different species.

Response: Please note that the SSA mean standard deviation is not a measure of uncertainty, which is addressed using Equation (1). It represents the consistency of the measured SSA during each chamber burn experiment, after the initial approximately 30-60 min transient during which mixing through the chamber occurred. The weighted average (weighted by the number of SSA values used from that burn) of the standard deviation values of all burns was 0.008. While this number was indeed surprisingly small, the standard deviations for the individual burns did vary between 0.002 to 0.023, with the majority of the values below 0.01. We have modified the text to explain this.

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2) I agree with the previous comment that all Latin species names should be used for clarification. The term needlerush does not have meaning to me.

Response: Latin names have been added to Table 1.

3) The 0.884 correction factor needs a further explanation. Why do you expect different losses of particles in the sampling system for the 2 instruments? Did the instruments have different lengths of sampling lines? You should provide a few calculations pertaining to particle losses with respect to this system. Could there be other reasons for the discrepancy?

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, $r^2 = 0.99$ (we have added this r^2 to the text). The high correlation and constant multiplicative offset suggest a bias. The ~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.

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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 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

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the correction should have been applied to the scattering measurements instead, then the ω_{meas} reported in Table 1 are too low. This would affect our subsequent calculations, 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.

4) In section 3.3, the uncertainty described for Figure 2 should be carefully considered. All calculations were done for the upper and lower limits of the range of refractive index/density pairs as previously reported in the literature. Yet, these values in the literature are NOT absolute and are associated with a certain degree of uncertainty. This uncertainty was not included within this figure. Furthermore, there are no uncertainty bars on any of the points, including the measurements (maybe they are too small, but see comments above). This statement also propagates into Figure 3, where the same methodology is used.

Response: The values we used for the refractive index of light absorbing carbon are the upper and lower limits reported by Bond and Bergstrom (2006) based on varying void fractions within the carbon aerosol. Bond and Bergstrom do not report any additional uncertainty in these numbers; although, there is of course uncertainty in the measurements they are using. We use this range only to show the effects of unknown LAC properties on the refractive index. In most cases, this is small compared to the differences resulting from the different carbon analyzers and methods.

Finally, the comments from Paul Zieger should be considered carefully, as it represents a very good review.

Response: We agree; Dr. Zieger's comments have been addressed.

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