

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

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Response to Review by P. Zieger

We have discussed your publication with pleasure within our literature group seminar (Laboratory of Atmospheric Chemistry (Aerosol Physics Group), Paul Scherrer Institut, Villigen, Switzerland) and will give here a short summary of the major comments and questions that came up during our discussion and hope that they might help to improve your manuscript.

Response: We thank Dr. Zieger and his team for this thoughtful and thorough review. Below we address your specific comments.

1) What are the wavelengths in Tab. 2? Did you neglect the wavelengths dependency

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when calculating ω_0 for the PAS wavelength of 532 nm? This should be mentioned in the table caption.

Response: In calculating and comparing refractive indices, we are assuming that these values exhibit minimal changes over visible wavelengths. The refractive indices we are using are reported at different wavelengths, as follows. The refractive indices taken from the CRC Handbook of Chemistry and Physics (Lide, 2008) are reported at 589 nm. Garland et al. (2007) report the refractive index of ammonium sulfate at a wavelength of 532 nm. The values we are using for LAC (Bond and Bergstrom, 2006), were calculated at 550 nm. Our refractive index for organic carbon is taken from Hand and Kreidenweis (2002) who derived this value from the partial molar refraction data reported by Stelson (1990) for the visible range. It is a commonly-applied value for aerosol OC at visible wavelengths, but is a quantity that should be characterized better (see Schmid et al. (2009)). The CRC handbook also reports refractive indices at 750nm for KCl and NaCl. These are less than those at 589 nm by 0.006 and 0.011 respectively (Lide, 2008). Bond and Bergstrom (2006) report that assuming a constant refractive index for LAC across visible wavelengths is a valid assumption. We are therefore assuming that all of these values are valid at 532 nm. We now make clear in the Table the wavelengths for the various cited values and, in the text, state our assumption neglecting wavelength dependence.

Please note that in making these changes we realized Table 3 was referred to before Table 2. We have therefore switched the order of these tables in the revised manuscript.

2) Lide (2008) is not found in the references.

Response: Thank you; we have added this reference.

3) Page 7472, Line 19 "...the chamber was diluted with clean outside air and prepared for the next experiment.": Did you use only filtered air (particle free) or air from a clean air generator? Was the "clean outside air" used as background?

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Response: The chamber was diluted with ambient outside air between experiments and this was used as the background. There are particle filters on this outside air source at the FSL, but they are not high efficiency.

4) Page 7472, Line 28: Explain the abbreviation SUVA.

Response: SUVA is a registered trademark by DuPont for the gas HFC 134a. We now refer to the gas as HFC 134a as it is more standard.

5) Page 7473, Line 4: The nephelometer integration time of 2 seconds seemed to be very short to us. Did you apply the Anderson and Ogren correction scheme on those 2 second values or on the average?

Response: The Anderson and Ogren correction was applied to the 2 second values.

6) Page 7473, Line 7: Explain the abbreviation CSU.

Response: CSU stands for Colorado State University. This abbreviation has been removed from the manuscript.

7) Page 7475, Line 3: Did you derive Eq. 1 from Gaussian error propagation? Why do you divide the relative error $\Delta\omega_{\text{meas}}$ by ω_{meas} ? We were a bit confused. Therefore we tried it ourselves and came up with the following equation:

[equation not repeated]

where dbscat , dbabs , and $d\omega_{\text{abs}}$ are absolute errors. We tested our equation to yours and came up with definitely larger errors.

Response: You are correct; our equation was incorrect. Thank you for pointing this out. The calculations have been re-done using the correct equation.

8) Page 7475, Line 12-17: There is an inconsistency between the text and Tab. 1: Douglas fir needles and branches (ID 247) were dry not fresh. The number of digits of ω_{meas} is different in the table compared to the text.

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Response: The text is correct; burn ID 247 was dry Douglas fir. Thank you for pointing this out. This has been fixed in the Table. Also, the number of reported digits has been changed in the text to match the Table.

9) Page 7477 Line 7 to 24: The correction of all measured size distributions by a factor of 0.884 seems to be quite a critical thing to us. What was the correlation coefficient of the measured to calculated scattering coefficients? How can you be sure that this factor (which you attribute to losses) is similar for all fuels which will probably have a totally different size distribution? Apart from impaction losses, we would expect diffusion losses, but they are less critical because those small particles don't contribute much to scattering. Did you also consider an ill-defined flow within the DMPS for these differences? You could also interpolate the refractive index of ammonium sulfate to the exact wavelength used. Is this factor valid for all three wavelengths of the nephelometer?

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 $\sim 12\%$ 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

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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 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 neces-

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sary 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 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.

10) Page 7480 Line 15: The error bars you refer to at this point are missing in Fig. 2 (or are they realistically so small of being noticed?). See comment above about the error calculations.

Response: Error bars have been added; they are now evident in Figure 2 (see attached).

11) Tab. 1: We thought it might be helpful if you would add the exact Latin scientific name (species and genus) to the individual fuel type.

Response: The Latin names for each fuel have been added to Table 1.

12) When you discuss the differences in the DMPS and OPC due to shape effects, you should extend this discussion to the shape effect of aerosol particles on the nephelometer measurement itself (and how you possibly correct for them).

You are correct that the shape will affect the nephelometer response. We do not consider any correction for this in the nephelometer measurements. We now explicitly state this in the paper.

Interactive
Comment

13) The single scattering albedo ω_0 was measured and presented for 21 samples. Why is the refractive index retrieved and discussed only for 6 species?

Response: We had the best size distribution data for these six cases; indeed, size distributions for most of the other cases had very large uncertainties. Since, as discussed in the paper, errors in the size distribution will lead to large errors in retrieved refractive index, we only picked those cases we were most confident in to attempt the retrieval. Even so, size distribution errors proved to be the largest source of uncertainty in the retrieved refractive index values.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 7469, 2010.

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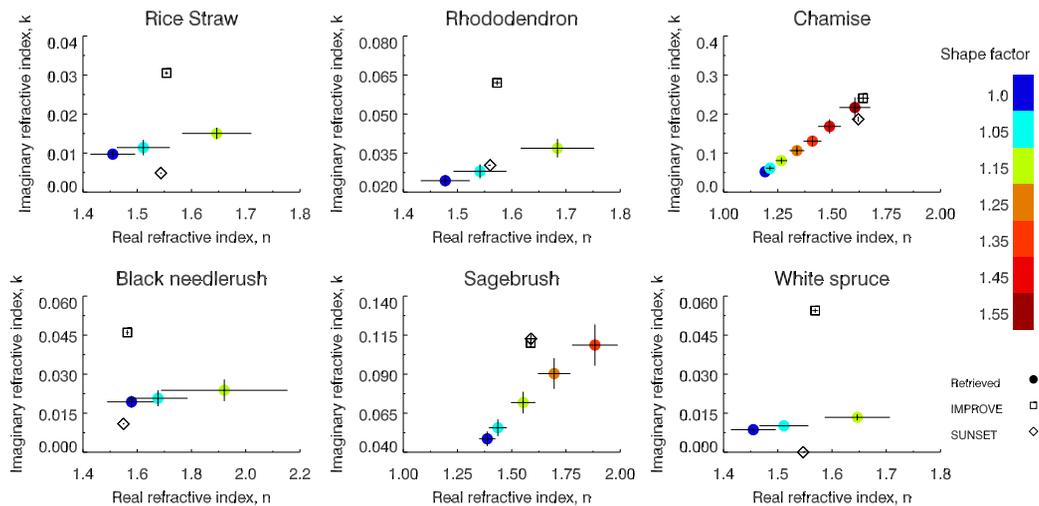
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Comment

Fig. 1.

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