Response to comments on "Undersizing of Aged African Biomass Burning Aerosol by an Ultra High Sensitivity Aerosol Spectrometer" by Charles Brock

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We'd like to thank Charles Brock for his review. His comments point out ways to make the paper clearer, and identified the single biggest weakness–that the UHSAS laser intensity is actually comparable to the SP2, so incandescence and particle vaporization are likely to happen. It turns out that the heat transfer calculation we used to demonstrate that incandescence was unlikely was in fact inappropriate, as it assumed that particles were much larger than the mean free path of air molecules. This realization had substantially changes our conclusions.

Comments from Charles Brock

1. *Line 66. You may want to cite Kupc et al. (2018) here. Kupc et al. describe modification and calibration of a UHSAS for airborne use, and is one of only two (now three) papers discussing the performance of the UHSAS.*

   I'm sorry to say I had missed that paper. It's a good one! It's now cited.

2. *Line 106. You mention later (line 139) that a "grab" sampler was used by the SMPS. Why do you need a "vast" and apparently homogeneous plume to do the size-resolved analysis? Doesn't the grab sample eliminate the need for homogeneity?*

   That is a good point–this kind of in-flight calibration can be done at a small scale, but there are a couple of limitations. Most obviously, the grab sampler only has enough volume for 3 scans, so multiple grabs were necessary to characterize the UHSAS response. More importantly, if aerosol composition changes at small scales, then one cannot assume that a calibration at one location is valid elsewhere in the plume. The consistency of our results over many samples and suggest that they are generally valid for this large plume.

3. *Figures 2-5. These figures are well laid out, but they use very similar colors and line types. The colors for the graphite, Aquadag, and NaCl are too similar, as are the sulfate types. I'm not color-vision-impaired, but I know many who are, and these figures would be really tough to read. Can you use more distinctive colors and different line types (dotted, dashed, dash-dotted, etc.) to better distinguish the curves?*

   I have tried to make those figures clearer, altering line types and choosing colors that shouldn't be confusing to the most common types of colorblindness. Part of the difficulty is that the symbols are overloaded–the colors identify test materials and the shapes indicate the number of charges. In my opinion, the number of charges information information is worthwhile, but it could be excluded, allowing the symbol shapes to clearly identify the test materials.

4. *Line 236. You state that the SP2 has a "much more powerful IR laser". Stephens et al., (2003), who describe the prototype for the SP2, quote a laser intensity of* $10^6\,\mathrm{W\,cm^{-2}}$*. This is not very different from the stated (line 204) UHSAS laser intensity of* $\sim 5\times10^9\,\mathrm{W\,m^{-2}}$ *(*$= 0.5\times10^6\,\mathrm{W\,cm^{-2}}$*). Thus I*

would not be surprised at all if the UHSAS is able to at least partially incandesce BC particles, leading to a mis-sizing.

This turns out to be an extremely important point. The SP2 laser system is so much more elaborate that it hadn't occurred to me that the energy density is comparable. As it turns out, the equations I used for thermal conduction to the air, following Cai et al. (2008), which assume particles are large enough to be in the continuum regime, indicate that the SP2 would be unable to heat small rBC particles to incandescence. That's clearly wrong! It turns out that applying a transitional regime model, as in Bambha and Michelsen (2015), makes a huge difference. It is now clear that incandescence and vaporization is a very real problem in the UHSAS. The paper now reflects that.

5. *Line 357. The corrected UHSAS data are not "an order of magnitude too high near* 1 μm*" for the free troposphere case (Fig. 8f). It's less than a factor of 2.*

   Good point. All of the corrected UHSAS volume distributions are high, but only the MBL example is high by nearly an order of magnitude. I've rephrased it to "the corrected UHSAS is considerably higher than the APS between 0.6 and 1 μm"

6. *Line 373. Here you say that it's not clear which refractive index to use to calculate scattering for comparison with the TSI nephelometer. Each bin of the UHSAS corresponds to a certain amount of scattering into the detection volume. If you quantify how much scattering each bin represents, which you are effectively doing by calibrating with a monodisperse aerosol, you should just use the same calibrant refractive index to calculate total scattering. In effect, you are just summing up the scattering represented by each bin, getting the total scattering. Of course, this ignores the difference between hemispherically integrated scattering vs. the narrower viewing angles of the UHSAS detection optics, but it is a very good first approximation to just use the refractive index of the calibrant (in this case, PSL). You can investigate the magnitude of the error due to the scattering geometry using Mie calculations.*

   That's what we've normally done in the past and in fact what we have done here. However, the situation is made more complicated by the fact that the refractive indices at the UHSAS wavelength are different than those in the visible, so the accuracy of the results are affected by the relative change in refractive index of the sample particles and the calibration spheres. I've explored that a bit in Fig 1.

7. *Line 371. Can you use the rBC number fraction from the SP2 to estimate the number of anomalously undersized particles, and boost the number in the main mode by this fraction to compensate?*

   Certainly one could do that, but that would require making assumptions about exactly which size particles to move.

8. *Line 384. The obvious explanation is that the coatings are not volatile at 400 C. This is consistent with Adler et al., 2019, who found coatings on biomass burning particles that did not evaporate (at lower temperature) but that were not incandescent in the SP2.*

   The new heat loss calculations indicate that even a pretty small amount of absorption by rBC can heat up particles sufficiently to volatilize almost anything organic, so this isn't really an issue any more. I expect that the coating simply takes long enough to volatilize that the peak scattering occurs at the initial peak as shown in Fig 1c of Laborde et al. (2012) rather than at the second peak.

9. *Fig. 9. Have you modified the UHSAS flow system as in Kupc et al.? We found that both of our UHSAS instruments leaked through the seals around the detectors, downstream of the detection region, reducing the sample flow even though the exhaust flow (which is the nominal flow measurement) was constant. This produced an altitude-dependent flow bias (Brock et al. https://doi.org/10.5194/acp-11-2423-2011), although it works in the direction opposite the trend seen here. Droplet Measurement Technologies has repaired the leak in our UHSAS, and they had a jig and setup to do this, implying that this is a common problem that they have had to fix in the past.*

   No. Unfortunately, our field project was over before the paper was published. We actually did notice a small discrepancy between inlet and outlet flow rates but failed to find the leak. It was small enough that we did not notice an altitude dependence, although we looked for it.

10. *Fig. 11. Change y-axis label to "Fraction of Particles".*

    That's not actually what the Y axis is. The x-axis is the fraction of rBC-containing particles. The y-axis is the number of sample periods that had that fraction of rBC-containing particles normalized by the total number of samples each year. The sample periods were rather arbitrarily chosen as LDMA scan periods, which were 85 s long. I've clarified that in the text.

11. *The Appendix is very clear and helpful.*

    Thanks! I hope the two more appendices are useful as well.

12. *Please check over the references for consistency with Copernicus formatting guidelines. For example, Clarke and Ellis et al. have capitalized titles, journal names are not consistent, etc. This is a consequence of EndNote-type software, which ALWAYS needs manual checking and correction.*

    Yes, something always slips through. I'm actually using the rather outdated LaTeX and BibTeX style files supplied by Copernicus, so EndNote can't be blamed this time.

References

Bambha, R. P. and H. A. Michelsen (2015). "Effects of aggregate morphology and size on laser-induced incandescence and scattering from black carbon (mature soot)". *J. Aerosol Sci.* 88, pp. 159–181. DOI: 10.1016/j.jaerosci.2015.06.006.

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Laborde, M., P. Mertes, P. Zieger, J. Dommen, U. Baltensperger, and M. Gysel (2012). "Sensitivity of the Single Particle Soot Photometer to different black carbon types". *Atmos. Meas. Tech.* 5.5, pp. 1031–1043. DOI: 10.5194/amt-5-1031-2012.

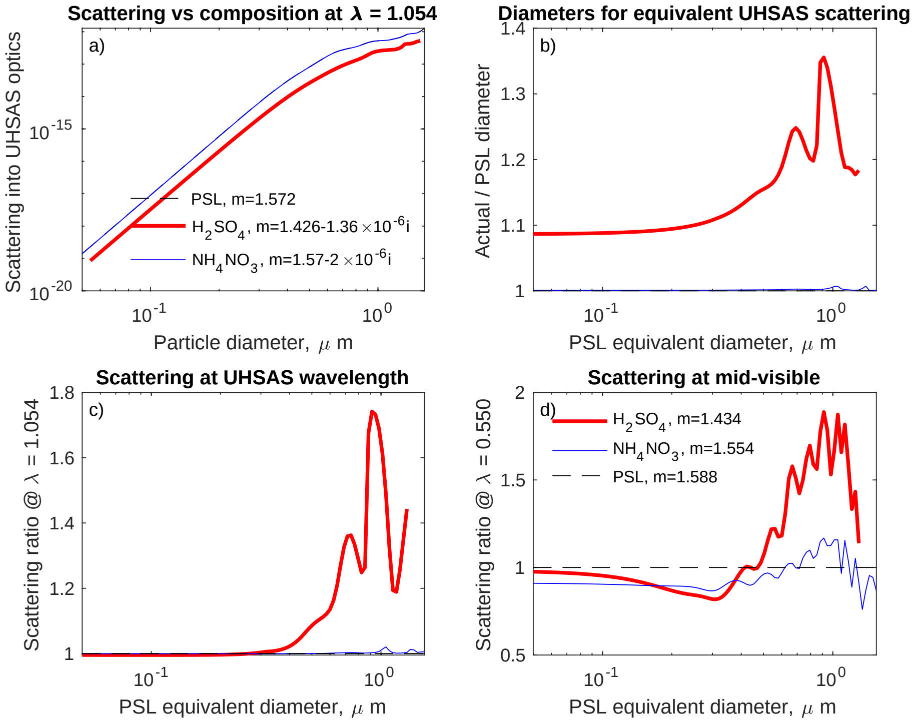

Figure 1: Effect of refractive index on calculated scattering. Panel a) shows the power scattered into the UHSAS optics for 3 materials. The PSL line is almost precisely on top of the NH_4NO_3 since the refractive indexes are almost identical. I then determined how big the NH_4NO_3 and H_2SO_4 particles would have to be to be classified as a PSL particle of a given size. Panel b) shows the ratio of those sizes. When calculating scattering per particle at the UHSAS wavelength (panel c), one sees good agreement up to 0.4 µm for both salts, but then the large size of the H_2SO_4 particles does a lot of forward scattering that isn't seen by the UHSAS. Nevertheless, one would expect very good scattering closure for the accumulation mode. However, when calculating scattering in the mid-visible, the refractive indices of the materials change in different ways, yielding much poorer agreement, something like a 10% underestimate for the accumulation mode. NH_4NO_3 refractive indices for visible light are averages of the 3 axes of orthorhombic crystals (the stable form from $-16\,^\circ$C to $31.2\,^\circ$C) from Bowen (1926) and for infrared light are from Cai et al. (2008).