

Interactive comment on “Undersizing of Aged African Biomass Burning Aerosol by an Ultra High Sensitivity Aerosol Spectrometer” by Steven G. Howell et al.

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This is a very interesting and well written paper on the UHSAS sizing behavior for absorbing aerosols encountered in an aged biomass burning plume. The topic overlaps with some recent laboratory work that we've been doing at NASA Langley to understand the performance of our Laser Aerosol Spectrometer (LAS) and UHSAS instruments, as well as the airborne measurements of fresh biomass burning plumes conducted during the 2019 FIREX-AQ field campaign. In this paper, Howell et al. use combined electrical mobility and optical particle sizing to study the UHSAS response to biomass burning particles. The main findings are reported as

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- Particles with electrical mobility diameters between 70 and 280 nm are optically sized by the UHSAS, and the singly-charged monodisperse aerosols between 100-200nm diameters show up as two peaks on the UHSAS.
- The peak with 70-100% of the particle number has a mode diameter that is undersized relative to the mobility diameter set point by 0-15% (depending on size), and Mie theory calculations show that particle composition-dependent refractive index changes are unlikely to explain this undersizing.
- The other “anomalous” peak with 5-30% of particles has a mode diameter that is even more significantly undersized by 25-35%, which is consistent with the UHSAS undersizing of laboratory-generated, fullerene soot particles.
- The ORACLES polydisperse size distributions are corrected using a power-law fit to the monodisperse data between 70-280nm that is extrapolated to 600 nm, while Mie theory is used for particles larger than 600 nm.
- The reader is cautioned that “UHSAS data should be treated cautiously whenever the aerosol may absorb infrared light”.

In this short comment, I ask that the authors consider the following:

1. The main peak undersizing of 0-15% (depending on size) is largely consistent with the difference in UHSAS size response between PSL particles and size-classified ammonium sulfate particles, and that the finding reported in the second bullet point above is due to differences in refractive index between the PSL calibration standard and aged, biomass burning aerosols.
2. Can the dual peaks between 100-180 nm be attributed to a stitching error in the transition region between the G3 to G2 gain stages, where the smaller ‘anomalous’ peak is from the G3 detector? While such a stitching error would not bias



the polydisperse size distribution (or indeed even be noticeable in many cases), it may give rise to extra peaks when looking at monodisperse aerosols near the gain stage transition point. What was the G3 gain stage saturation diameter for this instrument during the ORACLES campaigns as well as the subsequent tests?

3. Differences between Mie theory and the actual instrument performance may be substantial between 600-1000 nm as indicated by Figure 3, which may explain the poor performance of the extrapolated correction. The difference between NaCl (refractive index of 1.53+0i) and PSLs is particularly noticeable and seems to exhibit 20% undersizing between 600-1000 nm.
4. Along the lines of my Point 1 and the authors' caution in the final bullet above, it's not clear to me that aged biomass burning particles or other atmospherically-relevant absorbing aerosols far from emissions sources are meaningfully different from non-absorbing aerosols in terms of UHSAS sizing. Instead, these results motivate the need to calibrate the UHSAS with particles of atmospherically-relevant refractive index instead of PSLs.

Discussion:

Figure SC1 is included below to support these considerations, which shows mobility-classified ammonium sulfate (AS) sizing data for our UHSAS. We have used these data in past field campaigns to convert PSL-calibrated size bins to AS-calibrated size bins (e.g., Sawamura et al., 2017). Ammonium sulfate was chosen as the calibrant following Brock et al., 2011, and its real refractive index (1.52) is within the range of 1.52-1.54 identified by Shingler et al. (2016) as being representative of average aerosols encountered during SEAC4RS over a diverse range of air mass types (urban, marine, biogenic, and biomass burning). The

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biomass burning peak sizes are consistent with the ammonium sulfate curve up to about 200nm, after which the curves diverge slightly. It seems reasonable to expect that this might be caused by slight differences in the shape of the internal instrument calibration curves, and I'd suggest that the authors compare their biomass burning curve from Figure 6 to the NaCl curve from Figures 2-3 rather than relying only on Mie Theory calculations as in Figure 7 to rule out the refractive index explanation. Figure 3 shows that the Mie calculations significantly overestimate the expected size response for NaCl at all sizes. The refractive index of NaCl (1.53+0i) is very close to that of AS, and our experiments and those reported by Cai et al. (2008) indicate that the NaCl asphericity has a negligible effect on the UHSAS sizing of DMA-classified aerosols.

I find the presence of the 'anomalous' peak very interesting, and I enjoyed reading the authors' well-written discussion of potential artifacts caused by particle absorption and heating (and the limitations of this explanation given theoretical temperature increases). The laboratory measurements clearly show substantial undersizing of the absorbing species, and this is consistent with Kucp et al. (2018) as well as our past laboratory work (Figure SC2), which I've included here in case it helps inform the conclusions related to the difference between fullerene soot and nigrosine dye sizing. That the 'anomalous' particle size curve lines up so well with that for fullerene soot seems like more than a coincidence and would seem to imply the existence of a small, externally-mixed aerosol population in this aged biomass burning plume.

An alternative explanation that I'd like to hear the authors' thoughts on is that the anomalous peak is due to a slight gain stage stitching error at the G3-G2 transition that causes a small fraction of particles to be sized low by the G3 detector up to its saturation limit. These sorts of stitching errors seem to be fairly common in our measurements but are generally inconsequential given

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the noise in a the polydisperse size distributions. However, for a monodisperse aerosol, this might result in the small side peak that would be misinterpreted as an externally-mixed aerosol mode. As an example, I've included the gain stage saturation points for our UHSAS during a recent PSL calibration in Figure SC1, but I'd expect that these sizes would be different for the authors' instrument and calibration during ORACLES. **In sum, can a gain stage stitching error be ruled out as the explanation for the ‘anomalous’ peak?**

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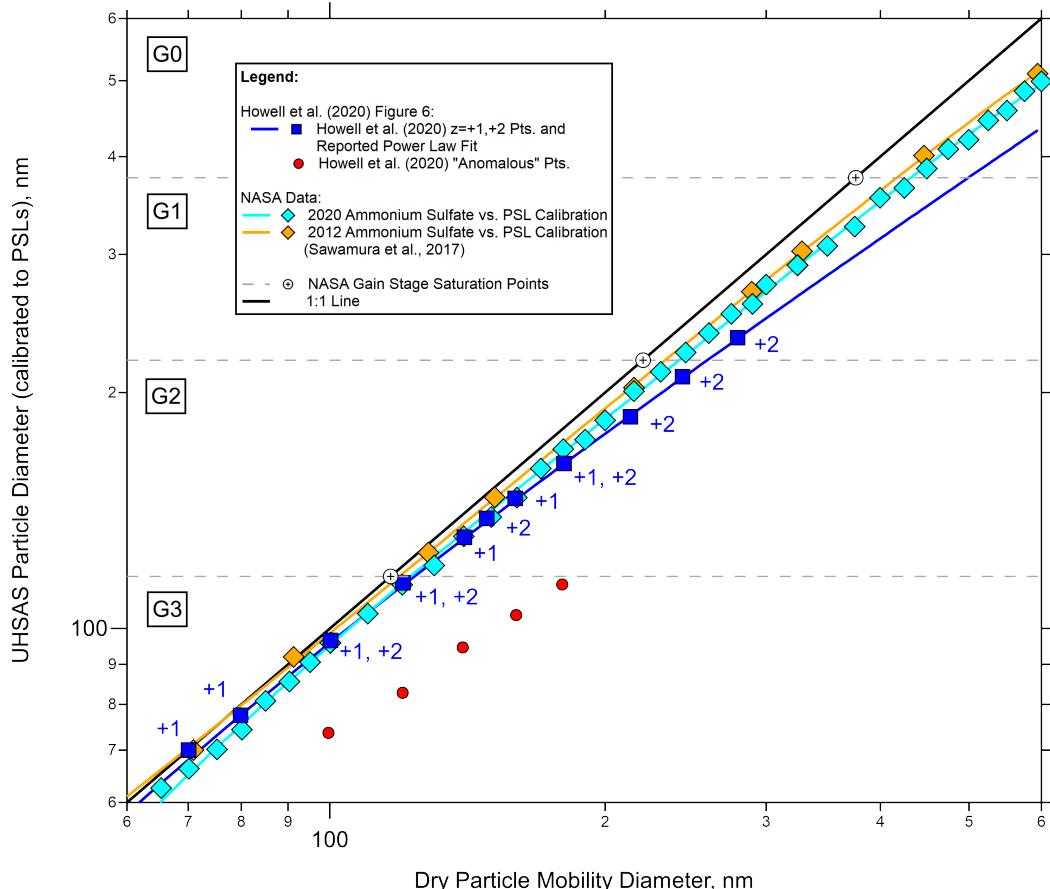
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Fig. 1. UHSAS optical diameters vs. DMA electrical mobility diameters for digitized data pts. in the present paper as well as NASA ammonium sulfate calibrations. Gain stage transition pts. are also shown.

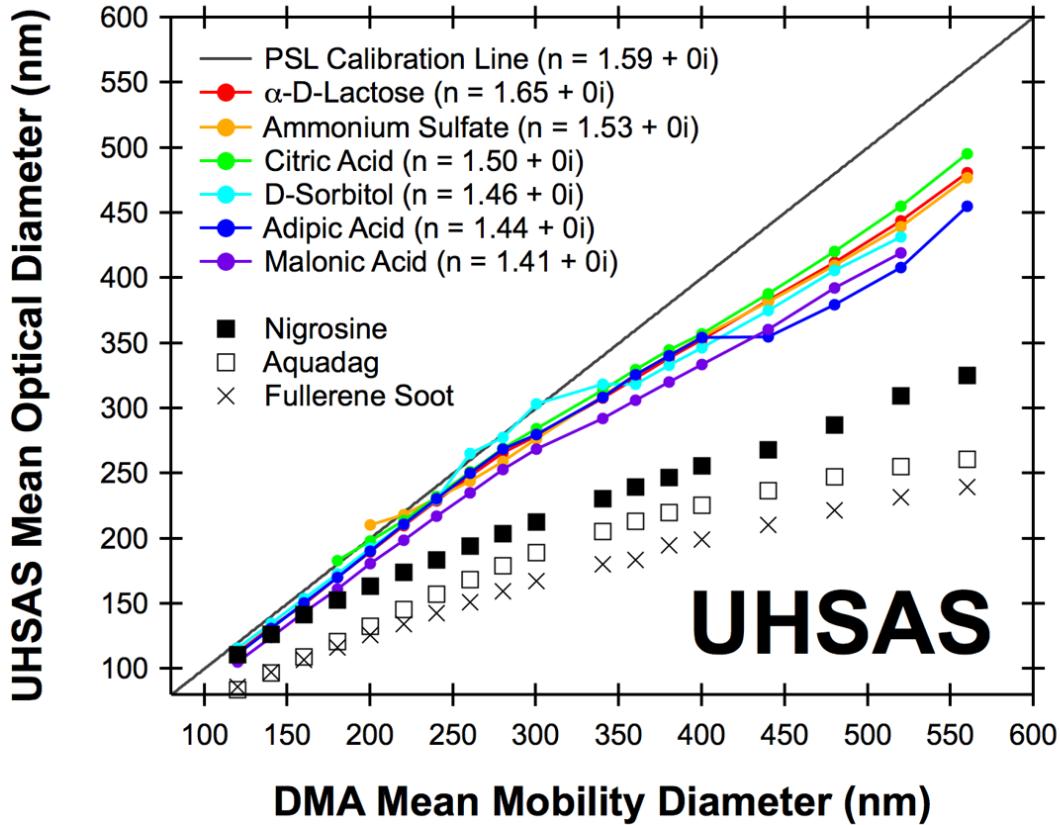


Fig. 2. UHSAS size response to aerosol of different chemical compounds with reported refractive indices from the literature. Reproduced from Zimmerman et al. (2015).

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