Atmos. Meas. Tech. Discuss.,
doi:10.5194/amt-2020-416-RC1, 2020

Atmospheric Measurement Techniques Discussions

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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Received and published: 10 November 2020

Review of "Undersizing of aged African biomass burning aerosol by an ultra high sensitivity aerosol spectrometer" by S. Howell et al.

This manuscript is a well-written and clear analysis of surprising results from an ultra-high sensitivity aerosol spectrometer (UHSAS) found during the ORACLES airborne project. The size distributions measured by this optical particle counter substantially under-predicted directly measured aerosol scattering and size distributions measured by a scanning mobility particle spectrometer. By introducing size selected particles into the UHSAS, the authors found that 10-30% of the particles measured in biomass

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burning plumes downwind of Africa were being undersized. Extensive laboratory investigation led to the conclusion that the UHSAS instrument heats brown carbon or "tarball" particles found in biomass burning plumes, causing partial evaporation and substantial undersizing. The implication is that all UHSAS measurements made in aerosols with a substantial absorbing component must be treated with caution.

Since the UHSAS is widely used in both airborne and ground-based measurements, especially in cases of rapidly changing aerosol conditions where SMPS response is too slow, this manuscript's findings are important. The scientific finding that brown carbon or tarball particles exhibit significant light absorption into the infrared is also of interest to readers.

This manuscript is quite clear, logical, and thorough. The graphs are (mostly) clear, there is a good mix of airborne observations, laboratory experiments, and theoretical analysis, and the data have been made available for public use. This paper is acceptable for publication in AMT with only minor technical edits.

Comments/edits:

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.

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?

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?

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 W/cm^2. This is not very different from the stated (line 204) UHSAS laser intensity of ~5x10^9 W/m^2 (= 0.5x10^6 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.

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.

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 correspond 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.

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?

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.

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.

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

11) The Appendix is very clear and helpful.

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.

Adler et al.: Evidence in biomass burning smoke for a light-absorbing aerosol with properties intermediate between brown and black carbon, Aerosol Sci. Technol., 976-989, https://doi.org/10.1080/02786826.2019.1617832, 2019.

Michelle Stephens, Nelson Turner, and Jon Sandberg, "Particle identification by laser-induced incandescence in a solid-state laser cavity," Appl. Opt. 42, 3726-3736 (2003)

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