Response to comments on “Undersizing of Aged African Biomass Burning Aerosol by an Ultra High Sensitivity Aerosol Spectrometer” by J. C. Corbin

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We’d like to thank Dr. Corbin for his review. He clearly took great care and pointed us towards real problems with some of our calculations. It will disappoint him that we are not actually in a position to run significant experiments with the UHSAS, as it needs repair for which we don’t have funds at the moment. We hope to perform the tests he recommended at some point in the near future (or read about someone else doing it). So we don’t have the thorough theoretical plus experimental analysis of the UHSAS that he would like to see. However, we have done much of the reanalysis that he requests and hope this revised version will be regarded as a significant enough contribution to the literature.

The bottom line is that Dr. Corbin was entirely correct in his suspicion of Eq. 1, which applies only in the continuum regime, when the mean free path L is much smaller than particle diameter D_p. While the particles were indeed smaller than L, the non-continuum effects are significant even when $D_p > 20L$. When implementing the transitional scheme of McCoy and Cha (1974) as used in Bambha and Michelsen (2015), it is clear that our previous estimates of particle heating were wild underestimates. There are significant uncertainties, but it is clear that raising rBC particles to volatilization temperatures is exceedingly likely. That resolves many of the issues we had trouble with and changes many of the arguments we made.

I also implemented more realistic optical calculations, though not the Rayleigh-Gans-Debye approximation recommended. Instead, I used Multiple Sphere T-matrix (MSTM v3.0) (Mackowski, 2014; Mackowski and Mishchenko, 2011), which requires similar assumptions about the particles (nonintersecting spheres) but is a numerical solution rather than an approximation, and is more accurate for absorption calculations (Mackowski, 2006; Sorensen et al., 2018). It wound up not making a large difference in particle sizing, but added roughly 10 % to the absorption and hence particle heating. The way MSTM was used is detailed an a new appendix.

In the rest of this document, Dr. Corbin’s comments are in **bold**.

Major concerns

Briefly, my two major concerns are (i) an alternative hypothesis for the field data is internal mixing of soot and organic matter (OM), and (ii) the UHSAS has a similar laser intensity than the SP2. The second concern implies that tarballs should evaporate in the UHSAS (in support of this work’s conclusions), rBC should be vapourized and not detected (in apparent contradiction of the laboratory work), and particle heating is much larger than currently calculated, at least for soot. These two concerns are fleshed out in the following comments, which address the manuscript more directly.

The idea that rBC would not be detected because it was vaporized seems incorrect, because the particle must enter the beam to get heated. Once in the beam it will scatter light and be detected. As the particle progresses father into the beam, two processes compete: scattering gets stronger as the light intensity increases, and scattering is reduced as the particle vaporizes.

1. The evaporation hypothesis has not been unambiguously shown from the laboratory experiments. The authors have not measured absorbing spheres to demonstrate evaporation. It would be relatively simple for the authors to reproduce the experiments of Sedlacek et al.

(2018), using nigrosin. This would let the authors clearly demonstrate what the hypothesized evaporation effects would look like in the UHSAS. Ideally, different laser powers would be used (by varying the pump laser power). Because evaporation in the UHSAS is a strong claim, it should be supported by this direct demonstration.

As mentioned above, we are not able to do that experiment at the moment. It would be a really good idea. However, in light of the new calculations of particle heating, there is very little reason to doubt that evaporation would occur.

2. The laboratory experiments have not shown the response of the UHSAS to realistic soot with a DLCA morphology. This morphology plays a major role in the light scattering properties of atmospheric black carbon (Sorensen et al., 2018). Light scattering by fractal soot aggregates is significantly lower than that of equivalent spheres due to its morphology alone (Mishchenko, 2009's Figure 12). Atmospheric black carbon either has DLCA morphology or is compacted from DLCA by coatings. The fullerene soot sample may at best represent compacted DLCA soot, which is adequate but could be much improved by a simple experiment with a kerosene flame or similar.

We do actually have some lab data of particles generated by a kerosene lamp (Fig. 1), but omitted it from the paper because

1. freshly generated kerosene soot was not representative of the types of aerosols seen in ORACLES;
2. we do not know how much rBC was present in the soot, nor how much organic carbon;
3. we do not know the characteristics of the soot, which was from a hurricane lamp rather than a well-designed soot aerosol generator so spherule diameter and fractal dimension are unknown; and
4. as a consequence, we did not do the DMA→UHSAS tests.

With such poorly known aerosol, the results are hard to interpret, but it is clear that the UHSAS saw a similar overall concentration, so particles were not vaporizing fast enough to shrink below the detection limit. The largest particles were clearly undersized by the UHSAS, and the peak was undersized by about 10 %, representing 50 % less scattering than a PSL sphere with equivalent mobility diameter. That's actually much less undersized than the fullerene soot and Aquadag. The obvious interpretations are that either the kerosene flame actually produced little soot compared to organic matter or that the DLCA soot produced by the lamp scattered less, as you predicted, but did not heat up enough to vaporize, while the more compact Aquadag and fullerene particles absorbed much more light and vaporized. Denser, more compact particles both absorb more heat and diffuse it away less efficiently.

In addition, as you'll see in the new appendix, better scattering calculations do show that total scattering for compact aggregated absorbing spherules is significantly less than Mie calculations suggest. However, the side scatter seen by the UHSAS is actually enhanced. It doesn't cause a large sizing error because of the 6th power dependence of scattering on diameter. Absorption is also enhanced, which is directly related to particle heating.

3. The calibration experiments are also missing a non-absorbing, non-spherical case, which would help to explain whether absorption is really important here, or just morphology. Silica or titania aggregates could be used (Schmoll et al., 2009) and would ideally be generated as DLCA aggregates (Eggersdorfer and Pratsinis, 2013) for comparison with soot. The authors may avoid this suggestion by including both soot and absorbing spheres, however. 4. In the scattering calculations corresponding to the calibrations, the authors should follow the literature to use the relatively simple RDGFA approach (Sorensen et al., 2018) rather than an effective medium approach in approximating soot properties. The combustion literature has long used the RDGFA approach to obtain reasonable results for soot and to show that soot scatters very little light at similar wavelengths (Liu et al., 2019). With the addition of an absorbing spherical particle type, a DLCA soot sample, and optionally a non-absorbing DLCA aggregate, the authors' work would represent a comprehensive study of the UHSAS response. So, these first 4 comments would not only close an important gap in the reasoning here but provide valuable reference data for others.

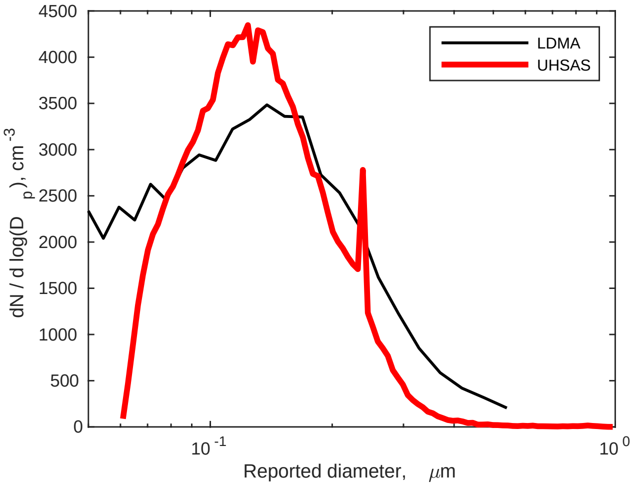

Figure 1: Smoke from a kerosene lamp sampled simultaneously by the LDMA and the UHSAS. This is a 6 minute average. Yes, those are stitching errors at 0.13 and 0.24 µm.

Interesting ideas. As mentioned earlier, we have implemented better scattering calculations, but aren't in a position to run the recommended tests.

We do not have electron micrographs to verify this, but neither the fullerene soot nor the Aquadag used as surrogates for rBC are likely to resemble DLCA aggregates. They are generated in aqueous solution and then the water is evaporated in dried air then any organic materials are either evaporated or charred in the tube furnace. This treatment sort of resembles the processed rBC tested by Bambha and Michelsen (2015) that collapsed their DLCA particles into much more compact shapes.

5. I have requested a direct demonstration of the evaporation hypothesis because I can propose an alternative hypothesis which the authors have not discussed: internal mixing of soot with non-absorbing or slightly absorbing material. Internal mixing is almost inevitable for plumes as old as those studied here (2 days to 2 weeks, Line 309). The authors' laboratory data shows that black-carbon surrogates scatter much less than predicted by equivalent spheres (as expected and noted in the previous comment).

It appears we were not sufficiently clear here, as we certainly never even considered the possibility that particles we sampled in ORACLES were not internally mixed. Indeed, the SP2 data showed thick coatings were the norm. Fig. 11 in the paper showed that the fraction of rBC-containing particles roughly agreed with the fraction of anomalous particles, an indication (though not proof) that the anomalous particles were the ones containing rBC. The main source of our confusion was precisely that they ought to be a mixture that behaved differently than the nearly pure rBC particles tested in the lab.

You do bring to mind a possibility that we did not consider–that the non-anomalous particles also contained rBC, but in quantities too small for the SP2 to detect. Those could heat the particles enough to vaporize relatively volatile materials. We've added that possibility to the discussion.

It can be expected that coated black carbon would behave somewhere in between soot and non-absorbing spheres, as suggested by previous work (Mikhailov et al., 2006). This does not univocally imply a continuous range of UHSAS signals in Figure 7, because particle breakup due to laser heating (Moteki and Kondo, 2007) could cause two UHSAS modes: either coatings evaporate and give a smaller signal, or they fragment and give a larger signal.

In general, fragmentation would not increase scattering signal. If a single particle breaks into 8, for example, then the resulting particles will have half the diameter, but each will scatter $(1/2)^6 = 1/64$ as much, so total scattering drops by a factor of 8. (This is assuming particles smaller than 0.3 µm, so essentially within the Rayleigh scattering regime, and that all particles are in the beam simultaneously.) If they are absorbing particles, they would heat less in the beam so would not evaporate as quickly, but that seems unlikely to overwhelm the inverse relationship between number and scattering.

The Moteki and Kondo (2007) case posits a mixed rBC and non-absorbing OM particle where the two parts split apart, so the non-absorbing part ceases to shrink and continues to scatter as it passes through the beam. If this happened in the UHSAS, the particle would still be undersized relative to the original particle diameter, but by much less than if the OM evaporated. It is certainly possible that this occurred during the inflight DMA→UHSAS tests, but if that was the typical cause of the undersizing of the $z = 1$ and $z = 2$ particles, we would have expected the SP2 to find a much higher fraction of particles containing rBC.

The field data (authors' Figure 7) can therefore be explained simply as a mixture of soot, organic matter (OM), and soot+OM particles:

(a) The soot particles are the anomalous low scatterers with Dmob>100nm, as expected for DLCA aggregates.

Possible, but the SP2 data indicates that most rBC had a thick coating. There does not appear to be a large population of uncoated rBC. It is also unlikely that our very aged soot particles still resemble DLCA aggregates. Electron micrographs in Miller et al. (2021) show much more compact shapes, appearing much more comparable to the processed rBC tested by Bambha and Michelsen (2015) with fractal dimension 2.3–2.4 rather than the 1.78 for DLCA aggregates in Sorensen (2011).

(b) The OM particles are the smallest particles (circles and squares with Dmob<100nm).

Sure.

(c) The soot+OM particles are the larger particles (circles and squares with Dmob>100nm).

But those particles are the most common, and the SP2 indicates that rBC-containing particles are generally only 5 to 18 % of the population. So this can only be the case if the rBC components are too small for the SP2 to detect.

(d) The ratio of Dopt/Dmob (y axis of Figure 7) may decrease with increasing Dmob because larger soot particles have larger shape factors (Sorensen,2011), or because larger soot particles have larger internal coupling parameters (Sorensen et al., 2018), or both. I made a rough calculation of the latter effect and it appears to be smaller than the former.

A second internal-mixing hypothesis replaces soot with tarballs above.

Yes, it may well be that tarballs behave very similarly to rBC.

The current laboratory experiments on soot surrogates (Aquadag and fullerene soot) actually support this internal-mixing hypothesis more than the brown carbon evaporation hypothesis. These surrogates anyway probably scatter more light than soot since their structure is more compact, and since Aquadag is made up of larger graphite flakes. The authors may perhaps consider my alternative as a sub-set of their evaporation hypothesis, or disprove it using their thermal denuder data.

It seems simpler to assume that there are only two major particle types: particles that contain rBC and thus absorb lots of light, heat up, and shrink considerably; and particles that are weakly absorbing, heat up less and lose only a relatively volatile fraction of their mass. The absorbing species in the latter particles could be BrC, tarballs, aged tarballs that are no longer recognizable as such, the intermediate BrC found by Adler et al. (2019), or even tiny amounts of rBC. This scheme agrees with the SP2 data indicating that the fraction of particles containing rBC is roughly comparable to the anomalous particle population, the thick coatings seen by the SP2, and the lab calibration of rBC surrogates.

6. In the context of the previous comments, I question the value of a "correction" to the UHSAS. If the surprising signals represent real physical phenomena, and the UHSAS is working correctly, why "correct" the data? Section 3.2 could in- stead follow the tone of Section 3.1, and focus on the prediction of the UHSAS response from fundamental particle properties. Since the particle properties are not known exactly, the properties (morphology effects on scattering, morphology effects on Dmob, refractive index and – if justified – evaporated volume fraction) required to explain the observations can be discussed. If the authors' answer is that a correction is valuable to predict volumetric size distributions and total light scattering, then please modify the manuscript to emphasize this.

To a large extent, I agree that a correction is futile–it utterly fails to account for the anomalously undersized particles and it's not likely to generalize to other projects.

However, the whole point of using the UHSAS in ORACLES was to rapidly measure particle size distributions through the entire range of important CCN diameters and to make detailed measurements of the optical properties of the aerosols. As it turns out, we got a lesson in the limitations of the UHSAS, and that became the theme of this paper. Nevertheless, we'd like to make the UHSAS data as useful as possible. We hope it's clear that the correction is not universally useful.

7. The manuscript has cited relevant SP2 work but there are a few points where the SP2 literature should be used to constrain the UHSAS predictions.

(a) The intensity of the SP2 laser has been reported as 1.7E+05 Wcm-2, 6.5E+05 W cm-2, and 4.05E+05 W cm-2 by Schwarz et al., 2006, Moteki and Kondo 2007, and Bambha and Michelsen 2015, respectively. Cai et al. (2008) reported the UHSAS laser intensity as 5.1E+05 W m-2. These are all similar to the 5.1E+09 W m-2 (5.1E+05 W cm-2) reported here. The SP2 and UHSAS wavelengths are about the same. So, the behaviour of a given particle in the SP2 can be extrapolated to the UHSAS. (While thinking about this I consulted Figure 1 of Corbin and Gysel-Beer 2019, which shows the behaviour of various particle types in the SP2 laser.) Any particles which vapourize (or carbonize) in the SP2 must vapourize in the UHSAS. Therefore, Equation 1 and Figure 4 cannot be correct. The SP2 routinely observes soot particles down to about 80 nm from their incandescence at >3000 K, yet Equation 1 apparently predicts only 1000 K at steady state for 100 nm soot. Moreover, this size is an overestimation for aggregate particles like fullerene soot. The assumptions behind Equation 1 must be flawed, at least for non-spherical particles. Bambha and Michelsen (2015) performed more detailed calculations than Cai et al. (2008); probably too detailed for this manuscript. So it becomes even more important to perform UHSAS calibration experiments with DLCA soot.

Your argument is precisely correct. I hope we have sufficiently addressed it. (Except as noted above, while calibration with DLCA soot would be interesting, it is probably irrelevant for either the lab or the ORACLES aerosol.)

(b) Sedlacek et al. (2018) showed that nigrosin (brown carbon / tarball surrogate) absorbs the SP2 laser. Corbin and Gysel-Beer (2019) reported SP2 time-resolve scattering cross-sections for the evaporation of tar brown carbon (TB) from heavy-fuel oil that was similar to Alexander et al. (2008)'s tarballs Their scattering cross-sections for TB actually look very similar to the "anomalous" particles reported here. In contrast, their scattering cross-sections for rBC actually show that the rBC evaporates before reaching the centre of the laser. So, how can the UHSAS see rBC? Presumably, the software uses the peak signal for all particles. This would correspond to the peak laser intensity for non-evaporating particles, but would occur before the peak for evaporating particles. This difference in incident laser intensity would result in an undersizing of evaporating particles, in addition to their actual change in volume.

(I have presumed that the software uses the peak signal because the SP2 would have to do the same if it did not have a "split" detector.) This is an important issue which will be implicitly addressed by the addition of soot particles in the calibrations.

Yes, the UHSAS has peak detectors only, not the fast data acquisition that allows the SP2 to actually trace the peaks. Therefore, if we have a particle that is a mixture of rBC and more volatile material, scattering would be double-humped, as in Laborde et al. (2012) Fig. 1(c). But we have no way of knowing which peak the UHSAS detected. The same might well be true for tarballs, if coated with more volatile material. With BrC, presumably the heating would be less intense so there might only be one peak (unless the particles char and generate rBC, as nigrosin does in an SP2). So the odds are that any particle with significant absorption will be undersized, whether it is vaporizing sulfates, organic matter, tarballs, or rBC.

Minor comments

While reading the manuscript carefully a number of minor comments arose, which I list here.

1. I appreciated the original structure of the introduction, but please add a short goals paragraph at the end.

Okay

Please also consider moving parts of Section 1.3 to Methods.

It makes sense to move the absorption calculations to Methods.

2. Please add an Appendix section where the prediction of nephelometer signals from

UHSAS signals is explicitly described. I can imagine what was done but it should be spelt out.

Sure

3. In Section 1.2 of the introduction, the authors' use of light-absorbing carbon (LAC) terminology could be refined. The section should cite Petzold et al. (2013) in its first paragraph, which is a review of the topics discussed there.

I've changed the terminology to be consistent with Petzold et al. (2013).

In the next paragraph, change boiling to incandescence (line 79) since rBC actually sublimates.

I've changed the wording.

Please reword the statement that BC is a hypothetical material that includes graphitic soot nanospheres and amorphous C, citing either the definitions given by Petzold et al. (2013), Bond and Bergstrom (2006). The word hypothetical suggests that soot is not of a consistent composition, which may mislead readers (Michelsen et al. 2020).

I agree that "hypothetical" is misleading, as it implies that BC is not real. I meant to convey the ambiguity in the definition of BC (as is mentioned by Petzold et al. (2013)).

Michelsen et al. (2020) is a nice summary, but appears to be focused largely on soot derived from hydrocarbons, not biomass burning. "soot" is defined as "carbonaceous particles formed during the incomplete combustion or pyrolysis of hydrocarbons", neglecting the carbohydrates and other organic material that dominate biomass. I'm sure the processes generating soot from open biomass fires are largely similar to laboratory combustion of hydrocarbons, but there may be some added complexity.

Please also reconsider the statement that quantitatively connecting the amount of carbon with the light absorption of LAC remains a challenge. The issue is not that connecting the two is challenging, but that the range of light-absorbing compounds formed by carbon is vast. These LAC compounds include BrC, black carbon, and amorphous carbon in tarballs (a recent summary is given in Corbin et al., 2019).

As you say, the atmosphere has a plethora of varieties of LAC, generally all mixed together. Each has different optical characteristics that are not necessarily well known and are affected by the mixing state. Instruments that actually measure C cannot unambiguously differentiate between types of LAC. Instruments that measure optical properties cannot tell us how much C is present. On top of that is the difficulty that sampling techniques tend to alter the environment around the particles, changing their physical and chemical properties. Petzold et al. (2013) discuss the difficulty of reconciling chemical and optical measurements. There seem to be some challenges left!

When the authors mention amorphous C, are they referring to the degree of graphitization (Michelsen et al., 2020) of the sample? The term amorphous C is often used for a specific carbon material, rather than amorphous domains within soot. Please reword to clarify.

We do not know the precise chemical form of the ambient BC during ORACLES or even of the fullerene soot used in the lab tests. Michelsen et al. (2020) advocate a very restrictive definition of amorphous carbon that may not qualify as BC and is rather at odds with the usage of the term in older literature (e.g the description of fullerene soot in Gysel, Laborde, et al. 2011) that appears to include anything lacking long-range order, including glassy carbon, turbostratic carbon, polycrystalline carbon and perhaps carbon onions. Perhaps it will be useful in the future to refine the definitions that way.

4. Please add a short description of the UHSAS to Methods. How are particle focussed into the beam, at what flow rate, etc.

Done.

5. Please mention the particle counter after the DMA in Methods. Of course, a DMA by itself does not provide size distributions, so I assume a CPC was used. Similarly, at line 126, please clarify that it is not the DMA but the CPC which cannot tell the difference between charges (especially important since this work uses a UHSAS to tell the difference).

I've mentioned the CPCs and reworded the bit about doubly and triply charged particles somewhat.

6. Line 147 mentions an empirical correction to a valve – please clarify if this correction was applied to CPC, UHSAS, or both. Please consider adding a graph to the supplement and/or stating here the magnitude of the correction.

Clarified with the relevant equation and a slightly better explanation.

7. Line 151 and surrounding, please mention what RH the sample would have had without desiccation or what the maximum dewpoint would have been (I assume it is very low given the altitude).

It varies, but I've included the range.

8. Line 161 please briefly mention the reason why the fraction is trivial, especially since the detector is missing from Figure 1. I am assuming that the reason is the small collection angle of the detector optics.

I wish I could be quantitative here! I do not actually know the dimensions of either the avalanche photodiode or the PIN photodiode, as I have not opened up the optical chamber and I have found no mention of the sizes in the UHSAS documentation or in scientific literature. The photodiodes are roughly 5 cm from the particle beam. If I assume they are 1 cm in diameter (an overestimate, I suspect), then they subtend a solid angle of 0.031 sr, which is less than 1.2 % of the 2.65 sr collected by the Mangin optics.

9. Figure 2 please change soot to ambient soot with a citation to Moteki et al (2010) in the legend. I originally misunderstood it as fullerene soot. Please also change graphite to graphite sphere (or similar) for clarity, since Aquadag is also graphite.

Figure 2 has changed in response to another review. I think it is clearer.

10. Line 180 please provide a citation for this description of fullerene soot. Please change to the RDG approximation here.

The soot description is now referenced. As mentioned, I used MSTM rather than RGD.

11. Line 191 "poor statistics"... how exactly was the statistical analysis done? Was the mode or median of the distributions used?

Neither, actually. We used the particle by particle mode of the UHSAS, which reports the peak mV for each individual particle detected. We bin the values, plot a histogram, and report the most frequently occurring bin. I suppose that's akin to a mode. Description added.

12. Line 192 please change "than expected" to "than predicted for equivalent spheres" or similar.

Changed, but to "scatter far less light than the calculations suggest".

13. Table 1, what is "amorphous C" here? I did not see this term used by Moteki et al. 2010. They did use the term "non-graphitic" to describe samples with a lower degree of graphitization. I suggest omitting this entirely as the optical properties of soot (one row above) will be similar. This comment relates to my general comment on LAC terminology above.

It is the fullerene soot, which is ≥ 90 % "amorphous carbon" according to Gysel, Laborde, et al. (2011). I think it is somewhat misleading to call it fullerene soot, because fullerenes are only a minor component and the major species controlling the optical properties ought to be emphasized. But I can see that it's clearer to use the name in common use.

14. Table 1, please indicate either how these values were extrapolated from measurements at other wavelengths, or the measurement wavelength if they were not.

We did no extrapolation except as already documented in the footnotes. All other values are either directly from the sources cited at wavelengths no more than a few tens of nanometers from 1054 or are interpolated from graphs in the sources cited. (Though it is not clear to me how Cai et al. (2008) determined m for NH_4NO_3; the sources cited there are not particularly close to 1054 nm. In any case, we have no calibration data using NH_4NO_3 and no longer show it in the heating calculation figure, so it has been eliminated.)

15. Table 1, please change 'variable' to the values used in order to convey more precise information.

The whole point was that the effective refractive index changed with particle diameter. But, since I'm no longer using the Mie calculations for those materials, they are no longer in the table.

16. Line 124, I do not agree that Gysel et al. (2011) concluded that Aquadag particles are 13% unknown composition after denuding at 450 degrees C, and I do not see the statement in that work. These particles would be 100% rBC by definition, but may have a different SP2 response to other forms of rBC. In other words, this statement is illogical since the SP2 is calibrated to the total mass of denuded particles. Perhaps the authors are referring here to

the EC content of Aquadag; this excludes oxygen and other atoms, so is smaller than rBC mass.

Gysel, Laborde, et al. (2011) state that

> TC content accounts for ∼83 % of the total gravimetrically determined mass, and consequently EC makes up ∼76 % of the total mass. Tests with thermodenuding Aquadag particles at 400 °C before selecting them by mass resulted in ∼15 % higher SP2 signal amplitude per particle mass compared to non-thermodenuded Aquadag.

So an undenuded 10 fg particle would have 7.6 fg of EC. A particle that passed through the denuder and had a mass of 10 fg would had 15 % more EC, or 8.7 fg. That's 13 % of the mass unaccounted for. It does not cause problems with SP2 calibrations because it's accounted for in the effective density calculation.

17. Figure 6 should not show the extrapolation if the discussion describes it as "completely inappropriate", please harmonize.

Changed.

18. Line 250 please change "kernel function" to "transfer function" according to convention.

"Kernel function" is hardly rare in the DMA literature (e.g. Gysel, McFiggans, et al. 2009; Talukdar and Swihart 2003), but that's usually from an inversion perspective. I'll admit that "transfer function" is easier to understand.

19. Line 303 please report the wavelengths which the AAE was calculated from.

Sure. It was 470 to 660 nm.

20. Throughout the manuscript, error bars were generally missing and not discussed, please add them or an overall comment.

Okay.

21. Line 204 please mention 1/e2 as the measure of beam diameter (if correct).

Okay.

22. Line 382 much lower temperatures than 400 C, are enough to evaporate most coatings. e.g. https://doi.org/10.1016/j.jhazmat.2011.12.061

Offhand, I'd agree that that's long been known. I cited Clarke (1991) for sulfates and Ellis and Novakov (1982) for organics vaporizing below 400 °C, but not there and I was not explicit. It's not actually obvious that Maruf Hossain et al. (2012) established that for ORACLES aerosol. The fuel and the aging were far different, and the ORACLES plume had modified combustion efficiencies (MCEs) $CO/(CO + CO_2) > 0.97$ (article in preparation), indicating efficient flaming combustion (Collier et al., 2016). Flaming combustion is where Maruf Hossain et al. (2012) found that their maximum temperature (250 °C) only evaporated 20% of the volume of 0.116 µm particles.

It is amusing to note that another reviewer cited Adler et al. (2019) to suggest exactly the opposite–that the coating might not be volatile at 400 °C. I hope the new discussion about this is clearer.

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