

“Physical and chemical properties of black carbon and organic matter from different sources using aerodynamic aerosol classification” by Dawei Hu et al. describes a comprehensive set of measurements from a well-designed and rather clever set of experiments. I believe the measurements are important for the community (especially the BC morphology results) and I look forward to the subsequent paper on optical properties. However, the manuscript is in need of some attention to detail, and in some cases, significant revisions and potential re-analysis of data. I believe this manuscript will be suitable for publication in ACP after these questions and comments are addressed.

Major Questions and Comments

Given the scope of the manuscript, the title does not adequately capture the science and I believe a more focused title will help steer readers towards this work when conducting literature searches. With the current title, I would believe that the authors aerosolized a variety of substances and passed them through an AAC, when in reality, the work is geared towards important combustion aerosols and a variety of instrumentation. Consider revising the title to reflect that.

Lines 29-31 of the abstract say “Here we present insights into the physical and chemical properties of the aerosols, with optical properties being presented in subsequent publications” and lines 131-132 of the introduction say “The characterisation and parameterisation of the optical properties of the particles will be the subject of a future publication” yet the introduction seems almost entirely focused on refractive index retrievals. It isn’t until line 108 that the authors begin to make the case for morphology and mixing state as a prerequisite for understanding RI. If the optical properties are the subject of a future publication, then the bulk of this introduction belongs in whatever forthcoming paper the authors are preparing, not here. I suggest re-writing the introduction to focus on the literature surrounding the physical and chemical properties that form the basis for the results presented in this manuscript.

The authors claim that their experimental setup enables them to derive refractive index with “much lower uncertainty than has been achieved previously.” How is this quantified? How does this particular experimental setup achieve this? What is the current state-of-the-art in RI uncertainty? Finally, how can any statement of this nature be made when this manuscript does not report measurement uncertainties?

The SP2 has been thoroughly described in the literature. Is all of section 2.1.3 necessary? At this stage in the maturation of the SP2, I believe it is more constructive to discuss the artifacts and other measurement caveats of the SP2, for example, Sedlacek et al. (2018). How were these charring artifacts handled in data analysis analysis?

The SMPS has an even longer history of prior publication. Section 2.1.6 can be significantly shortened to include only the operational parameters such as scan time and flow rates, and the resulting bounds of the size distribution you are able to measure. Once this has been described, the manuscript would benefit from a discussion on the uncertainties introduced by the SMPS. Finally,

please describe the inversion algorithm used. Is it the typical inversion algorithm that comes with the SMPS software, or is it custom? If it is custom, it should be discussed and cited.

Why was 180 °C chosen for the thermal denuder? The optimal internal temperature for a thermal denuder is the subject of ongoing research, yet the consensus is that it must be chosen based on some optimization of experimental parameters. In this case, I would guess that this optimization corresponds to complete volatilization of coating materials so that BC alone can be studied. However, it is never explicitly stated what the reasons are for choosing 180 °C, nor is the method by which the authors arrived at this temperature. For example, Sumlin et al. (2018) describes an approach using four different TD temperatures in an attempt to volatilize different mass fractions of organic matter. Please discuss the approach to choosing 180 °C.

Furthermore on the subject of the TD, the authors state that the inner diameter of the heating zone is 0.15 meters. This is rather large, and it is almost certain that there is a temperature gradient across the radius. The temperature along the central axis may be several degrees colder than at the walls, and aerosol travelling at different points along the radius will experience varying degrees of thermal processing. Was this temperature gradient measured? How does this gradient affect the analysis?

Finally, on the subject of the TD, the authors state that the residence time in the heating section was approximately 31 seconds. Given the measurements provided for both the inlet and outlet (0.037 m ID tube) and the heating section (0.15 m ID) it seems that the authors did not account for fluid velocity change within the heating section. If one considers only 1 liter min⁻¹ flow through the 0.037 m ID tube, across the 0.51 m heating section (assuming steady and conservative flow), one arrives at 32.9 s, which is “approximately” 31 s as stated in the manuscript. However, that assumes that fluid velocity is constant throughout the TD – it is not, since the ID in the heating section is different than the inlet and outlet. The mean residence time in the heating section is likely somewhere on the order of nine minutes. Given this, and my previous comment, I am unsure that conclusions from the TD experiment are represented accurately. It is likely that this data can be re-processed, or at least re-interpreted, but the data may have been handled according to an incomplete understanding of the experimental setup. This must be corrected or addressed in the manuscript.

It seems that all measurements are reported without uncertainties. The authors should report uncertainties or justify their exclusion.

Finally, please make sure all references are properly formatted and include DOI numbers, if possible.

Minor questions and comments

The name “Manchester Aerosol Chamber” is unfortunate, since it shares an acronym (MAC) with a commonly studied aerosol optical parameter, the mass absorption cross-section. Unless this acronym has been used for your chamber multiple times in other publications, I would suggest you consider renaming it.

Several parts of the Experimental Methods section contain results, especially section 2.3.4. I suggest re-writing these sections to focus on the methods, and thoroughly discuss the results in section 3.

The headings for sections 2.1.1, 2.1.2, 2.1.3, 2.1.5 and 2.1.6 should be spelled out, for consistency with other 2.1.X section headings.

The description about the AAC in section 2.2.1 is overly subjective. Furthermore, the AAC only produces a monodisperse aerosol in the sense of aerodynamic diameter. This is not “truly” monodisperse, since there are other measures of aerosol diameter that run contrary to this qualifier.

Line 278: The authors may consider stating the material that the space blanket (mylar, I’m guessing) is made from so that readers unfamiliar with the term will know what was used.

Line 281: How was the illumination evaluated?

Line 357: Can the authors comment on what effect resin content might have on the resulting aerosols?

Line 366-367: How was the “sufficient concentration” to ignite the pyrolysate determined?

Line 380: Change “exemplar” to “example”.

Line 404: Change “300 s of the ignition” to “300 s after the ignition.”

Line 430: I suggest specifying the NO₂ came from a “compressed gas cylinder”. Also, what was the concentration of NO₂, and what was the balance gas?

Line 432: Change “desirable” to “desired”.

Lines 451-454: I suspect that the increase in ozone concentration is attributed to the absorption of UV photons by O₂ in the chamber – this is a common method to produce O₃ from O₂. I cannot find any references in the literature on O₃ production due to UV absorption by cresol. If the authors can provide a reference for O₃ production by cresol photochemistry, please do so.

Line 464: Change “desirable” to “desired”. What was the desired concentration?

Line 470: “particulate” is an adjective. Change to “particles”.

Line 471-472: What size did the particles stabilize to?

The caption on Figure 1 refers readers to the main text to thoroughly understand what is meant by the different groupings of instruments, however, the first reference to the figure in section 2 does not clarify the groupings whatsoever. The figure is therefore unclear: were different groupings of instruments used during different experiments? Were they grouped in this way during data analysis to calculate specific parameters? This needs to be clarified both in the figure and its caption, and in the main text.

References

Sedlacek et al., Formation of refractory black carbon by SP2-induced charring of organic aerosol, *Aerosol Sci. & Tech*, 52:12, 1345-1350, 10.1080/02786826.2018.1531107, 2018.

Sumlin et al., Density and Homogeneous Internal Composition of Primary Brown Carbon Aerosol, *Environ. Sci. Technol.* 2018, 52, 7, 3982-3989, 10.1021/acs.est.8b00093, 2018.