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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Absorption closure in highly aged biomass burning smoke" *by* Jonathan W. Taylor et al.

Anonymous Referee #3

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The authors have investigated the optical properties of black carbon (BC) and organic carbon from highly aged biomass burning plumes as part of the CLARIFY-2017 field campaign. They measure the mixing state of BC using an SP2, and the optical properties using photoacoustic spectroscopy. They use these measurements to obtain MAC, MAC enhancement, and AAE values for the aged biomass burning aerosol. These measurements are then compared to several different models for calculating biomass burning optical properties. These include coated sphere models, homogeneous grey sphere models, and more complicated aerosol optical models that account for aerosol morphology (semi-empirical models). These measurements also allow for an estimation of the contribution of brown carbon aerosol to overall absorption in aged biomass burning aerosols (10% at 405 nm). The authors conclude that all models are sensitive

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to the choice of refractive index for BC. The authors also conclude that Mie models be implemented with great caution when calculating aerosol optical properties.

Major comments:

1) The authors rely heavily on SP2 measurements for most of their analysis. It would be helpful to comment on potential effects of charring of organics in the SP2 as detailed in Sedlacek et al. 2018 (Aerosol Research Letters 52:15, 1345-1350) and if these would affect any of the measurements detailed here.

2) p 14. line 12-22: The authors describe alternatives to the lensing effect of MAC and mention the possibility of externally mixed intermediate absorbers (IA) affecting total particle absorption and demonstrate that the resulting calculations do not match their observations. If possible, could the authors perform similar calculations for IA internally mixed with BC and show if such a scenario matches the values observed here. An internal mixture of IA and BC would reduce the BC MAC while also reducing the resultant AAE.

3) The main critique I have of this paper is that they provide too little detail on what makes each optical model unique. It is good that they are verifying different optical models with real world data, but one needs to be familiar with the models used for it to make sense why they give different results. I believe a little more explanation is warranted.

4) The semi-empirical models all matched the measured AAE well, and MAC values calculated using Chakrabarty and Heinson method and the Liu method matched the measured values well. MAC enhancement predicted using Liu's method matched MAC enhancement values the closest, but it is unclear why the Chakrabarty MAC enhancement did not, as they are very similar techniques. The authors reason that the enhancement calculated using the Chakrabarty and Liu methods give different results but are similar methods. The authors speculate that this has to do with morphology, did they collect any samples to image the particle morphologies?

5) Overall, the paper is well written but is a bit lengthy. I think perhaps the finer details in sections 4 and 4.1 could be shortened or relegated to the SI.

Other comments:

1. Section 2.2: Are there any limitations or artifacts in the instrumentation that should be mentioned or accounted for?

2. Figure 3: Should error in the MAC of BC as reported by Bond and Bergstrom account for error?

3. p. 6, line 1: Were checks put on the upper limit of the SP2 measurements as high BC concentrations can be underestimated by the instrument, or were concentrations below the upper limit of the SP2 measurement range throughout the campaign?

4. p. 8, line 8: As the OA absorption is calculated by subtracting total absorption by extrapolated BC absorption, the uncertainty propagation would also need to account for uncertainties in BC absorption measurements.

5. Page 8: add details about optical models

6. Page 9: I think the 6 step outline is going to be confusing for some, I would consider rewriting to make it more clear what is a measurement and what is a theoretical calculation

7. Page 10: There is some explanation of the Liu correction that should be moved to an earlier spot in the text

8. p 11. line 24: typo “experimental” written twice

9. p 13. line 21: It would be better to quantify the coating rather than just stating the particles to be thickly coated.

10. p 16. line 11: The line reads as if BC acts as the coating material and I think that is not the intended meaning here. Please edit the sentence to make it clear.

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