

Interactive comment on “Biomass-burning derived particles from a wide variety of fuels: Part 1: Properties of primary particles” by Crystal D. McClure et al.

Anonymous Referee #2

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The authors have investigated optical, physical, and chemical properties of primary aerosol particles generated from laboratory biomass burning of various fuels under different combustion conditions at the Missoula Fire Science Laboratory. The measured properties were correlated with bulk properties such as the MCE and OA/BC ratios. The authors concluded that intensive optical properties of aerosols are well parametrized using OA/BC mass ratios. The authors also observe negligible coating-induced black carbon absorption enhancements and little relationship between bulk OA chemical properties and OA/BC mass ratios. This is a dense manuscript; the results are presented in a coherent and logical manner and most figures are easy to follow. That said, I recommend major revision of this manuscript—my concerns and comments

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are provided below.

Major comments:

1) First major concern is the source of black carbon (BC) constituting the Class 6 particles. Per table 1, the fuels burnt constitute duffs, peat, dung - all of which have been found to smolder (low MCE values) and produce tar-balls or spherical brown carbon (BrC) aerosol with negligible/no BC, very high SSA, and AAE >6 in the 405-532 nm. For example: Chakrabarty et al. ((2010), ACP 10, 6363) observed and reported no BC from duff burning at Missoula FSL. More recently, peat collected from Alaska and Indonesia were burned in a Missoula FSL-replica chamber (Sumlin et al. 2017 and 2018 series of papers) and negligible BC was found. These fuels have been only observed to smolder (low-temperature fires) both in the lab as well in field. Consequently, the particle formation mechanism is distinct in these fires, meaning soot (BC) formation is not supported.

2) Could charring of organics by the SP2 and/or SP-AMS be responsible for enhanced rBC concentration erroneously showing up in particle classes, especially in Class 6? This needs to be addressed. Sedlacek et al. (Aerosol Research Letters 52:15, 1345-1350) convincingly showed that initially near-IR transparent low-volatility compounds (fulvic and humic acid) particles at room temperature undergo chemical transformations as temperature is increased in a heated tube, creating new near-IR absorption transitions. They also say that this phenomenon enable SP2-induced charring of organic aerosol including tar-balls (akin to Class 6 particles in this article). Sedlacek et al. observed around 5-10% mass loading of rBC in case of fresh OA/tar balls resulting from SP2-induced charring through near-IR light absorption. The reviewer is suspicious that the authors erroneously report rBC concentration corresponding to Class 6 particles due to this phenomenon and then draw their conclusions. Please provide substantive proofs that no soot photometer induced artifact is involved during the experiments, especially for Class 6 particles. If no evidence can be provided, please remove Class 6 particles from all plots which have [OA]/[BC] as the x-axis.

3) The authors purport negligible absorption enhancement at 781 nm for Rcoat values as large as 10 based on results from Figure 4c. My concern with this assertion is that the axes in these graphs are extremely skewed which can misrepresent the actual MACBC enhancements. The average Eabs for Rcoat less than 10 is mentioned to be close to 1.2, but theoretical Eabs for longer wavelengths at these coating values are not expected to exceed 2 regardless (see Chakrabarty and Heinson, Phys. Rev. Lett., 2018). I believe that if the axes were not disproportionately skewed due to the extremely large MACBC values at the large OA/BC mass ratios (corresponding to Class 6 particles which in turn are due to the very small BC concentrations rather than large OA concentrations) we would be able to discern larger coating-induced absorption enhancements even at 781 nm. The conclusion that there is negligible coating-induced absorption enhancement based on visual comparison with a skewed axis is in my opinion is highly misleading. I notice that there are points in Fig 4c which have MACBC values larger than 10 which in turn would correspond to Eabs close to 2 which is significant in terms of absorption enhancements.

4) The authors claim that the increased MACBC at 781 nm is due to OA absorption and not coating-induced. They need to cite relevant literature which demonstrates significant BrC absorption at longer wavelengths to back up this assertion.

5) In Figure 4, it does not make sense to include points for MACBC where the contribution of BrC to total absorption is much larger than that of BC. So, removing all points for Class 6 OA would make the plots in Figure 4 more informative. The BC concentration in Class 6 is very likely an artifact. The absorption enhancement at longer wavelengths have a weaker dependence on coating thickness than at shorter wavelengths as observed by Pokhrel et al. (2017) cited in the manuscript, but it is still significant. I am unconvinced of the insignificance of coating-induced BC light absorption enhancement asserted by the manuscript or at least from the results as they have been presented right now.

6) Title should bear the word “laboratory” since the observed results might not be appli-

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cable to real world fires. For e.g., something like: “Laboratory-based biomass burning particles from a wide variety of fuels: Part 1: Properties of primary particles”

7) The authors provide no explanation (beyond a hand waving argument) to back the statement “The contribution of coating-induced enhancements (i.e. lensing effects) to absorption by black carbon are shown to be negligible for all conditions”. Lensing or focusing of light to the core could also be possible with weakly light-absorbing coating materials such as brown carbon with low imaginary index of refraction. Such a coating would facilitate lensing in addition to itself absorbing. One convincing way to declare that “no lensing” takes place is by looking at the internal field strength of a brown-carbon coated BC aggregate (see methodology in Chakrabarty and Heinson, Phys. Rev. Lett, 2018). I would like to see such a rigorous analysis performed (DDA or T-Matrix and not Mie-based core-shell) by the authors on a few BC aggregates coated with BrC vs non-refractory materials and convincing the reviewer and the community if indeed the “no lensing” claim is valid. If the authors cannot perform such an analysis, then I suggest that they remove all statements from the abstract and the main text regarding “negligible coating-induced enhancements (lensing effects)”. Instead, rephrase or replace the sentences with “brown carbon-coated BC particles yield absorption enhancements of x and y values. . .”

Minor comments:

L68: change vary to vary Add units for MACBC at all relevant plots. L164: When the authors say that the relationship between SSA and OA/BC is consistent with those of Pokhrel et al. could they expand on whether the corresponding fits parameters in the two studies match as well and if they do not then possible reasons for the mismatch. L169: change Table S1 to Table S2 L330: change Eqn. 7 to Eqn. 6

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