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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Formation and evolution of Tar Balls from Northwestern US wildfires" *by* Arthur J. Sedlacek III et al.

Anonymous Referee #1

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This manuscript characterizes the microphysical properties of tar balls sampled during the BBoP campaign. This is an important finding and deserves to be published. I would recommend publication of this manuscript after mandatory revision. Below are my major comments:

1) In the abstract, the sentence "Brown carbon is a poorly characterized mixture that includes tar balls (TBs)" conveys a very vague meaning and needs to be edited or removed. I suggest not to mention the word "brown carbon" here. Just defining tar balls should suffice.

2) I like that this paper finally tackles the question of how tar balls are actually made. I like the terminology "processed primary particles". This needs to be mentioned in the abstract. A sentence or two should also be added to the abstract on how these

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particles differ in composition and optical properties from those generated using heat-shock treatment in the laboratory by Hoffer et al.

3) The authors determine aging using the NOX/NOY metric, which is fine. However, by parameterizing aging using this metric will make it difficult to place their findings within the accepted context of the atmospheric chemistry community. I recommend a Van Krevelen (VK) diagram if they do have the necessary data from the AMS. I think adding a VK plot would enable better comparison of this dataset with other studies.

4) If the authors have optical measurements and size distribution data available, why didn't they just inverse Mie calculations to derive complex m rather than perform forward calculations of literature data? The consensus is that m varies according to fuel type and burn conditions, and using a single value for a given fuel is an incomplete basis for comparison. For example, assuming that all Alaskan duff particles have m=1.75-0.002i is neither reasonable nor rigorous.

5) Continuing the discussion of complex m, Sumlin et al. (2017 and 2018) have shown that m associated with the organic fraction of carbonaceous aerosol varies widely as a function of burn conditions and photochemical age. This work further emphasizes that it is preferable to do inverse Mie calculations rather than rely on previously reported values.

Sumlin, B. J.; Pandey, A.; Walker, M. J.; Pattison, R. S.; Williams, B. J.; Chakrabarty, R. K., Atmospheric Photooxidation Diminishes Light Absorption by Primary Brown Carbon Aerosol from Biomass Burning. Environ. Sci. Tech. Let. 2017, 4 (12), 540-545.

Sumlin, B. J.; Heinson, Y. W.; Shetty, N.; Pandey, A.; Pattison, R. S.; Baker, S.; Hao, W. M.; Chakrabarty, R. K., UV-Vis-IR spectral complex refractive indices and optical properties of brown carbon aerosol from biomass burning. J. Quant. Spectrosc. Radiat. Transfer 2018, 206, 392-398.

6) Figure 5 shows TEM images of aggregates of tar balls. This is very interesting.

Chakrabarty et al. (2016; ACP) have also observed aggregates of spheres from Alaskan Peat combustion. A paragraph is warranted on how the optical parameters (SSA etc.) would differ if aggregate morphology is accounted for in the calculations. Would schemes such as Rayleigh-Debye-Gans be appropriate?

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