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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Chemical characterization of laboratory-generated tar ball particles" *by* Ádám Tóth et al.

Anonymous Referee #3

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Tóth et al. discuss a detailed analysis of particles generated in the lab that are supposed to mimic atmospheric tar balls. They performed several analyses including elemental analysis, Raman, FTIR, OC/EC, and pyrolysis-gas chromatography-mass spectrometry. From the results, they conclude that their TB surrogates contain a large fraction of elemental carbon, making them more similar to black carbon than to HULIS. I think the paper is nicely written and the analytical methods are sound, and it is worth publication. I have, however, a few concerns that need to be addressed before publication.

General comments:

The main issue I have with the paper is the attempt to extrapolate the findings to the properties of all atmospheric particles including the optical properties of atmospheric

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tar balls. In the literature there are plenty of pieces of evidence that the properties of atmospheric tar balls are variable and therefore, the laboratory particles generated by Tóth and collaborators might be easily representing only a sub-fraction (maybe small?) of what is in the atmosphere, especially considering that there is here no discussion of measured optical properties. I will discuss more this issue in the specific comments next. I would suggest calling these "surrogates" of some TBs, not necessarily all atmospheric TBs.

Specific comments:

Abstract

Line 26: The authors should write "laboratory TBs", instead of "atmospheric TBs", because that what they measured. As mentioned earlier, the issue here is how well these laboratory-generated particles actually represent tar balls generally found in the atmosphere. More on this issue will be discussed next.

Introduction

Lines 32-34: "Since these particles... are able to absorb solar radiation quite efficiently in the visible (Hand et al., 2005; Alexander et al., 2008) and up to the near-IR range (Hoffer et al., 2017)". This ignores an important fraction of the literature that shows much lower absorption properties from atmospheric TBs. Neglecting to mention these works here is biasing the paper toward those studies that showed particles more similar to those discussed here. The authors should acknowledge the fact that there is a wide range in the published values of the imaginary index of refraction for atmospheric TBs and in general a large variability in the TBs properties. See for example, [1-4]. The variability in O/C ratios, for example, is well discussed in the result section on page 4, and the authors clearly acknowledge, there, that different types of TBs might exist in the atmosphere. Therefore, it is reasonable to believe that also the index of refraction values, and therefore, the absorption properties might be quite variable.

Lines 18-20: Similar issue here. Considering the high variability of the properties of atmospheric tar balls, it seems more logical to say here that these laboratory surrogates are similar to some of the TBs studied in the atmosphere but different from others.

Experimental section

Page 2, lines 29-31: “The concentrated aqueous phase of the tarry condensates (wood tars) was nebulised to produce tar droplets which were first exposed to a ’thermal shock’ by passing them through a heated (at 650 °C) quartz tube, then cooled and dried with dry filtered air.” It might be that this ‘thermal shock’ is resulting in TBs that represent well some atmospheric biomass burning smoke particles, but not others. A different “formation” (or transformation?) mechanism has been recently proposed for example by [3]; in that case, a thermal shock is not likely, considering that the TBs abundance increased substantially only far from the flaming region of the plume. This “delayed” formation has been shown in other studies before, as well.

Page 2, line 33: “distorted spheres” this seems in contradiction with the definition of “perfect spheres” discussed in other parts of the paper (e.g., page 1, lines 35-36). It is a bit disturbing that the not perfect sphericity is used as an argument to dismiss the study by Chakrabarty et al. in line 5 of page 2, which is one of those studies that found a week absorption for atmospheric TBs. Please be coherent.

Page 7, lines 1-2: also this high EC content points to the fact that these TBs might be at the high side of the range of absorption properties measured in the atmosphere.

Page 7, lines 2-7: How much would this artifact affect the estimated EC/TC ratio? Conclusions:

Page 7, line 32-33: Because of what mentioned earlier, I find this sentence to be biased toward those studies that found higher absorption and might not represent the large range of optical properties found in atmospheric TBs. I, therefore, suggest that the authors clearly point out this caveat to avoid providing a sense of generality that

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might not be warranted.

Figure 1.: I believe that China et al. (2013) reported only the oxygen content, not the carbon. How did the authors calculate the corresponding values reported in the figure?

References

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4. China, S., C. Mazzoleni, K. Gorkowski, A.C. Aiken, and M.K. Dubey, Morphology and mixing state of individual freshly emitted wildfire carbonaceous particles. Nature Communications, 2013. 4.

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