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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Light absorption properties of laboratory generated tar ball particles" *by* A. Hoffer et al.

Anonymous Referee #3

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General Comments:

This is a nice short paper describing an important laboratory study aimed at characterizing 'tar balls' (TBs), which the authors contend are an important component of the atmospheric aerosol, especially in biomass burning emissions. The authors argue that determining the optical properties of atmospheric tar balls is quite difficult because they are co-emitted with many other types of particles. They have developed a method whereby they can produce particles in the laboratory that microscopically closely resemble TBs in their physical and chemical properties. These particles were then produced in quantities sufficient for the determination of their aerosol optical properties (e.g., light scattering, light absorption, wavelength dependence of absorption, etc.). If we can assume that the laboratory-generated particles are actually similar in

their optical properties to atmospheric TBs, then the optical properties of an important component of the atmospheric aerosol have been determined. This information would probably be of use to modelers of global and biomass burning aerosols.

This study was well designed and focused. The authors are very knowledgeable in their respective fields and have extensive experience in the components of this study. The referenced literature was for the most part sufficient. English grammar and usage are excellent. The methods were solid and produced results that should be considered robust. The results are new and quite relevant given the amount of biomass burning aerosols. My only real criticism is that the scope of the research is quite narrow. While interesting to know, do the light absorption properties of laboratory-generated TBs really matter that much? We are already capable of measuring the optical properties of real-world atmospheric (including biomass burning) aerosols. So do we need to know the specific properties of one component of these aerosols? It can be argued that this is important for modeling biomass burning aerosols, so I am willing to accept this as a driving force behind this study.

Overall, I would rate this study as 'very good', and would recommend publication in ACP pending the minor revisions discussed below.

Specific Comments:

Pg. 16217, Line 1-2: Rephrase first sentence. A possibility is... 'Tar balls (TBs) are abundant in the global atmosphere and represent a particle type that is strongly emitted from biomass burning.'

Pg. 16217, Line 4-6: 'TBs ... can withstand the high-energy electron beam of the TEM.' The only way these can be observed in the TEM is when they are illuminated by the high-energy electron beam and have been exposed to local beam heating and high vacuum. How do you know that the TBs have not changed in the TEM before you observe them? The authors state that morphological changes in the electron beam are not observed, but what if transformations or loss of volatile materials have happened before the particle was observed in the TEM. Was a cold-stage used to minimize beam heating? How can the authors be sure that volatile components do not leave the particle in the high-vacuum TEM environment. Have previous studies looked at these possibilities? If so, references should be provided.

Pg. 16218, Line 3: '…absorption Angstrom exponent…'. Please define the acronym 'AAE' here which is used later on the page.

Pg. 16219, Line 6-8: 'By generating pure TB particles in the laboratory… we have directly measured the optical properties of TBs…'. You have not actually measured TB particles, which are atmospheric particles. You have measured laboratory-generated particles which are very similar microscopically to TBs. Assuming these also have similar optical properties, then you have constrained the optical properties of TBs.

Pg. 16220, Line 7: 'A PM1 cyclone (SCC2.229)…'. Please list manufacturer.

Pg. 16220, Line 14-16: '… the raw light absorption … data were corrected according to Bond et al. (1999)…'. The Bond et al. (1999) corrections were determined using a single wavelength PSAP instrument. Since the CLAP instrument was developed based on the PSAP (it is basically a multiple-spot PSAP) and the same filter and similar optics and detection methods are used, the Bond et al. (1999) correction scheme is used there also. There are two important things that must be accounted for when using the Bond et al. (1999) corrections for a CLAP instrument. One is a fundamental error in the filter area measurement of the original PSAP spot used in the original Bond study. This affects the loading correction. The other is how to handle measurements made at different wavelengths. Both of these are discussed in detail in the Comment by J. Ogren (Aerosol Sci Technol., 44:589-591, 2010). This work should be referenced, as it is likely (from the mention in the Acknowledgements section) that the data for this study were processed using the NOAA data processing utilities, which include the Ogren modifications to the Bond et al. (1999) correction scheme.

Pg. 16222, Line 27: Define 'ns-soot'.

Pg. 16223, Line 12-21: 'The uncertainty of the measurements of Particle Soot Absorption Photometer (PSAP) whose measurement principle is very similar to that of the CLAP is 20–30% (Bond, 1999). It was demonstrated that the presence of organic compounds (secondary organic aerosol, SOA) causes positive bias and enhances the uncertainty of the PSAP (Cappa et al., 2008; Lack et al., 2008). This effect has to be considered in the case of particles generated from tar which contain condensable organic compounds as well. Based on the above, if we consider that the CLAP overestimated the absorption of nigrosin by 25% and the scattering is also overestimated by 25 %, we obtain a refractive index of 1.65–0.29i and 1.77–0.27i for nigrosin at wavelengths of 550 and 652 nm, respectively.' How specifically did the authors conclude that 'the CLAP overestimated the absorption of nigrosin by 25%'? The Bond et al. (1999) study suggests an uncertainty in the PSAP measurements of 20-30%, but this could be in either direction (positive or negative). The Cappa et al. and Lack et al. papers show a positive bias in the light absorption measurement in the presence of significant amounts of SOA, but this effect is quite variable and depends on filter loading (i.e., transmittance). How do the authors know that there are lots of SOA particles on the CLAP filter leading to this bias? And even if there are, how do they arrive at the +25% value? This seems a bit arbitrary to me. Please explain your reasoning here in more detail.

Pg. 16224, Line 1: Please mention the wavelengths or wavelength range that the measurements in Table 1 represent. Also, please mention this in the Table 1 caption. Tables should be able to be interpreted on their own merit.

Pg. 16224, Line 8: '… mass absorption coefficients…'. Do the authors mean MAE (mass absorption efficiencies) which appear to be what is listed in column 4 of Table 1. If these are the same, please use consistent naming in table and text.

Pg. 16224, Line 9: '… These values are similar to…'. A roughly factor of two difference in MAC between TB and BC seems like a pretty big difference to me, but that depends on what you are comparing the difference to. It is small compared to the difference in MAC between TB and HULIS, which I guess is the point. These TB-like particles appear to be much closer to BC than HULIS in optical properties.

Pg. 16225, last paragraph of section 5: Figure 3 is discussed in this paragraph but is never called out in the text.

Pg. 16226, first half of the Conclusions section: Much of this discussion is about the importance of TBs in the atmosphere. This should have been discussed in the Introduction so that the reader knows why this study was performed in the first place. Typically new references are not presented in the Conclusions section. The Conclusions section should highlight the major findings of the study, and give numerical values or ranges for the important measurements. My recommendation is that the Conclusions section be re-written to present and summarize the major findings of the study without all of the background material on TBs in the atmosphere.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 16215, 2015.