

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

Anonymous Referee #1

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General comments:

This study, “Chemical characterization of laboratory-generated tar ball particles”, by Tóth et al., generated tar balls and characterized their chemical properties using several unique instruments. They compared tar balls with other similar carbonaceous particles (BC and HULIS) and discussed how they are different or similar. Tar ball particles are important and abundant in the atmosphere but are not well known. Thus, the characterization of tar balls is significant in this field. The manuscript is clearly written with enough data. I feel more discussion is useful regarding the similarity of the atmospheric and laboratory-generated tar balls, in addition to their morphology and O/C molar ratio. Overall, I think this manuscript includes important results on the atmospheric science.

Specific comments:

1. Page 2 line 18-20: The morphological and structural characteristics and the elemental composition of the TB particles generated by this experimental system were highly similar to those of atmospheric TBs.

Reviewer comment: Please discuss more about how they are similar to each other. Their similarity is a critical point to use the laboratory-generated tar ball data to the atmospheric tar balls. Although some such discussion may have been done in their previous studies, I think some discussion is useful here. Also, the tar ball formation process in this laboratory experiment may be different from that observed in other studies from wild fire (e.g., Posfai et al., 2004; Adachi and Buseck, 2010). They found more tar balls in aged smoke than young smoke. The laboratory-generated tar balls are emitted as primary particles and can be detected directly from the fresh smoke, suggesting that no tar ball number change as aged(?). More explanation will be useful here how they are similar to or different from ambient tar balls regarding their formation process.

2. Page 2 line 32-34: The shapes of the TB particles (investigated by TEM) generated from European turkey oak were mostly distorted spheres.

Reviewer comment: Tar balls are defined as “TBs can be unambiguously identified by electron microscopy as perfectly spherical amorphous particles externally mixed in relatively fresh biomass burning plumes” in Page 1 line 35-36 or “in this paper we use the term exclusively to refer to combustion particles that share all the key characteristics that were described above” in page 2 line 5-6. However, “distorted sphere” contradicts “perfectly spherical amorphous particles.” Clear definition of tar ball will be needed here, if they include those are not perfectly spherical. TEM images of European turkey oak tar ball may be useful as well as those from other sources.

3. Page 3 line 11.

Reviewer comment: Please spell out “TCD.”

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4. Page 4 Line 10-13. It should be noted that wood tars (starting material for TB generation) exhibited significantly higher O/C and H/C molar ratios (0.182 and 1.215, respectively), which strongly suggests that the 'thermal shock' employed during TB generation (as described in Hoffer et al. (2016)) has markedly increased the degree of aromatisation (Francioso et al., 2011).

Reviewer comment: Is it possible that water (H₂O) is included in the wood tar to increase these molar ratios?

5. Page 4 line 17-19: It can be clearly seen that the average O/C molar ratio of our laboratory-generated TB particles is very similar to that of atmospheric TBs examined by Pósfai et al. (2004), whereas it is lower than those obtained by some other authors (Tivanski et al., 2007; Chakrabarty et al., 2010; China et al., 2013).

Reviewer comment: Using microscopy technique, O and C may be also from the substrate that supports tar balls in addition to the particles themselves. Please explain how such substrate effects were considered in this study and others, because the O/C ratio is important data to compare the laboratory-generated tar balls and those from ambient.

6. Page 5 line 28-30: On the other hand, HULIS spectra contain characteristic features (a broad band at 3400–2400 cm⁻¹ and a band of C=O at ~1700 cm⁻¹) suggesting the presence of carboxyl groups, whereas these bands cannot be found in the TB spectra.

Reviewer comment: This result seems to be different from that by Tivanski et al (2007), who found more carboxylic carbonyls functional groups in tar ball and concluded that tar balls are similar to HULIS. It is better to have some discussion.

7. Page 7 line 30-33: In this regard, the combination of all analytical results presents an array of supporting chemical evidence that spherical atmospheric TBs with C/O molar ratio around 10 are closer to BC in many of their properties than to weakly absorbing HULIS. In harmony with the findings of several independent studies on the

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optical properties of TBs, the present results imply that TBs are indeed quite strongly light-absorbing aerosol particles and likely play an important role in the global radiation budget.

Reviewer comment: This conclusion regarding the optical properties of tar ball is too strong as the most discussion in this study focuses on their chemical properties but not on the optical properties. More discussion regarding the optical properties will be needed to conclude their optical properties. RF-IR result may be useful to this discussion (similarity of C=C?).

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