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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 #2

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General Comments:

Tóth et al. describes the use of multiple analytical techniques to study the organic composition of laboratory generated tar ball particles and compared them to field collected tar balls and other carbonaceous particle types including HULIS and soot particles. From these analyses, the conclude that laboratory generated tar balls are similar to some types of field tar balls based on the O:C and H:C ratios, but have much lower O:C than other possibly aged tar ball samples. Additionally, they conclude that their laboratory generated TBs are more closely related to BC than HULIS based on the O:C ratios. Overall the paper gives a good description of the laboratory generated TBs and a compelling case that they have properties between HULIS and soot. This being said, there are numerous areas that need to be addressed further in this paper to make

the comparison between their TBs and BrC/BC species. Specifically, there is some discussion about the comparison between species types, but it is lacking in some sections and the comparison data is absent in multiple figures and tables that would lead to a more polished manuscript. The IR data is of insufficient quality to make the claims in the paper and leads to questions about the quality of the Raman spectroscopy due to the low signal observed in the IR spectra for the wood tar samples. Overall the paper has a good analysis of the laboratory generated tar balls, but there seems to be some missing information that needs to be addressed.

Specific Comments:

Table 1: Please define in caption what the parenthesis represent What is the error in the individual CHNSO measurements?

Pg. 2 Line 25: "with a view to locate TBs in the light-absorbing carbon continuum" There is no analysis of the optical properties as is indicated by the last sentence of the introduction, the purpose of the paper needs to be clarified here

Pg. 2 Line 32-34: There is a description of the shape of these particles, but no actual TEM images. Please include TEM images and clarification of perfect vs. distorted spheres. An analysis of the shape factors (roundness etc.) could be used here to quantify the sphericicty

Pg. 3 Line 2: How is it calculate that 44% of the mass is collected with this stage? And what is the overall size distribution?

Pg. 4. Lines10-13: It is not stated how they believe this transformation of change in the O/C and H/C is accomplished? Could it be purely that the low volatility organics/water are driven off and what other factors could be occurring?

Pg. 4 Line35: I am not sure what "mean carbon to mass conversion factor" is telling me, perhaps showing how it is calculated would help.

Figure 1: "TB- Black locust" etc. should be labeled "laboratory generated TB" or similar

In the legend it should be noted what technique was used for analysis (e.g. EDS, CHNO, etc)

Pg. 5 Lines 7-10: The broad region around 3400cm-1 usually indicates that there is water present along with the sharp peak at 1643. Looking at Figure 2, these account for some of the peaks present in both spectra. This data indicates that there is possibly still quite a bit of water present which would possibly skew the results of the O:C analysis as well.

Figure 2: The absorption on the wood tar samples is < 0.2 a.u., which indicates a significantly lower sample loading compared to tar balls and is also a very noisy spectra below 2000 cm-1. The IR spectra needs to be improved to make any definitive statements about the carbon speciation of the wood tar and the possible presence of water needs to be addressed and corrected for.

Figure 2: It would be nice to have a comparison spectra of HULIS and soot that shows the similarities and differences since they are compared in this manuscript.

Pg. 5 line 30: Throughout the paper the “laboratory generated tar balls” becomes “tar balls” which refers to a specific natural source which this paper is showing similarities to. Clarification throughout the paper is needed as to which is being discussed.

Pg. 5. Line 34: “do not contain carboxyl groups” This is misleading based on the IR analysis, it would be better to say that they are not detected in the IR analysis

Pg. 6 Line 3: “All three types of wood tar were Raman inactive” is not substantiated because of the low noisy signal in the IR spectra demonstrating low loading of the wood tar compared to the laboratory tar balls.

Pg. 6 Lines 2-15: This needs to be more descriptive in comparison to soot as well as HULIS

Pg. 6 Line 16: Since there is already a lot of comparison between the laboratory generated tar balls and the tar starting material, why not compare at least one of these

in this section as well.

Pg. 7 Line 7: I was left wondering here how the EC/TC compared to HULIS and soot using the same method

Pg. 7 line 12: The similarity to the savanna fire data is valid for O/C only since H/C was not calculated for the savanna fires.

P7. 7 Lines 27-30: “Our results. . .combustion aerosol” this paper only shows the similarity between the laboratory generated tar balls and atmospheric tar balls, there is no data to confirm a mechanism of formation of tar balls

P7. Lines32-33: “In harmony. . .global radiation budget” please add citations to the studies on optical properties of TBs here. The main purpose of this paper was to describe the chemical composition of laboratory tar balls and the similarity to other carbonaceous particles, but there is no discussion throughout the manuscript on how they are important for light absorption (though indeed they are!).

Table 2: For the * samples (e.g. 2,4 dimethylfuran) that are only in the laboratory generated TBs, they should be excluded since it is misleading on first read through

Technical Corrections:

Pg. 2 Lines 9-16: TEM-EDS/SEM-EDS/ETEM/ESEM should all be combined into a single EDS since that is the technique used to analyze the composition

Pg. 2 Line 28: Should “chops” be “chips”?

Pg. 6 Line 7-8. “The peak fitting. . . software” should be moved to the experimental

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