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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Detection of tar brown carbon with the single particle soot photometer (SP2)" *by* Joel C. Corbin and Martin Gysel-Beer

Anonymous Referee #1

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The current manuscript builds upon recent findings that the SP2 can induce some NIR light absorbing organic aerosol particles (commonly referred to as brown carbon or BrC) to char and subsequently be detected as refractory black carbon in the SP2. Specifically, the present manuscript examines tar brown carbon particles (referred to as "tar brC") that are produced by a marine engine running on heavy fuel oil. Using both the time-dependent scattering and incandescence channels of the SP2, the authors report that tar BrC annealed by the SP2 laser, and then detected as "rBC", can be distinguished from primary emission rBC from incomplete combustion. This new analysis methodology provides a framework that will expand the utility of the SP2 to include the subclass of BrC particles that absorb at the lasing wavelength of the SP2 (1064 nm). The methodology and analysis presented are sound and the paper is writ-

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Discussion paper

ten well. Publication is certainly warranted, once the authors address a concern on material definition and clarify a nomenclature issue as highlighted below.

With respect to material definition, a core statement that the authors make, and one that traces back to the lead author's 2019 Climate and Atmospheric Science paper, is that tar ball material is insoluble (e.g., cited on page 2, line 27 and again page 3, line 89-90 in present manuscript). The authors are encouraged to look take a second look at the closing section in Hand et al., 2005 where these authors state (page 12; ". . ..ESEM experiments of particle hygroscopicity indicate that tar balls observed during YACS are water soluble at high relative humidity (RH > 83%), albeit to a much lower degree than inorganic salt particles and with no distinct deliquescence point." Additionally, this reviewer wishes to draw attention to the recent work of Li et al., (ACP, 19, 139-163, 2019) where the presence of both polar and non-polar TBs (e.g., water soluble and non-soluble components) are reported on and discussed. The observation that wildfire TBs are water soluble and that some laboratory-generated TBs can be as well, suggest that care should be exercised with the application of generalized labels such as that used in the present manuscript. While the tar brC particles investigated in the present study may very well be completely insoluble, the reported presence of water-soluble TBs suggests that there are in indeed two classes of "tar-like" particles and further suggests that some reconciling is in order. The fuel source for a marine engine is very different from that available for wildfires - not to mention the emission sensitivity to burn conditions in the latter - so perhaps we should not be too surprised that there could be some fundamental differences of specific particle types within this subclass of brown carbon particles. The authors need to address this and adjust their material definition argument accordingly. If indeed biomass burning tar particles are different from fossil fuel tar particles, how generalizable are the conclusions about tar brC behavior in the SP2 laser?

This reviewer is confused by the presence of a non-incandescing distribution for the "soot BC case" (blue line) in Figure 2. In section 3.2, the authors reference a "pure-BC

case" as a "control" to help identify the unique features of the tar particles, yet in the figure, they cite "soot BC case". It is not clear whether the authors are referencing two different "BC" controls or are using two labels for the same material. My concern is that a "pure-BC case" should be composed of solely rBC particles that would all incandesce in the SP2 and that would necessarily lead to a non-Gaussian scattering signal distribution for in Figure 2. Additionally, the traces presented in Figure 2 are presumably for non-incandescing particles, which, again, for a pure-BC case, no pure scattering signals should be present. In short, clarification is in order. I believe the solution is straightforward enough. As this reviewer understands the manuscript text, the "soot BC case" are those particles generated when the marine engine is running under a specific operation condition and hence serves as a control on the particle-type produced and believes that their reference to "pure-BC case" is the same material. If correct, please clarify this nomenclature and stick to one label for your control material.

Other specific issues:

Page 1, lines 5 -6. The authors write ". . ..tar brC results in unique SP2 signals due to a combination of complete or partial evaporation, with no or very little incandescence. Approximately 70% of tar particles incandesced." The juxtapositioning of these two sentences is very awkward. The first seems to say that there is very little to no incandescence while the follow on sentence says that 70% of the particles incandesced. Which is it? Please reword.

Page 4, line 95, 96. The authors are encourage to examine (and possibly reference, if they think it is relevant) the recent paper by Li et al., (ACP, 19, 139-163 (2019)) where the optical properties of laboratory-generated TBs were investigated and refractive indices derived.

Page 6, line 183-185. The $\sim$3000K refractory requirement is dictated by the bandpass filters used in the commercial-grade SP2. A different set of bandpass filters would enable a lower temperature blackbody to be detected. (Admittedly, this reviewer is

being a bit pedantic here, but still. . ..)

Page 18, Figure 4. While the authors discuss how different thickly-coated rBC particles would look cast on a similar plot as shown for two cases in Figure 4, this reviewer wonders whether it would be useful to have a third panel on this figure for thickly-coated particles. As a picture is worth a thousand words, so too is a plot worth a thousand arguments and thus might prove a useful visual comparison for those readers who are not well acquainted with the SP2 mixing state analysis and data products. This is certainly not critical, but rather a suggestion, and thus at the authors discretion.

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