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Interactive comment on "Brown carbon absorption in the red and near infrared spectral region" *by* A. Hoffer et al.

Anonymous Referee #1

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In this paper the authors isolate components that comprise tar balls, re-aerosolize them and then measure the light absorption at multiple wavelengths. They find low (but non-trivial) levels of light absorption at high wavelengths. They make extensive use of Absorption Angstrom Exponents (AAEs) to interpret their data and to verify that indeed, according to them, tar balls are making significant contributions to overall absorption in the ambient atmosphere impacted by biomass burning.

The paper is interesting, but I do not believe the authors have presented a convincing argument. In my view the authors over interpret their data and make very strong statements based on weak evidence (eg, sections 3.3 and 3.4 are highly speculative). Yet, there is a simple and direct way to test their idea. They apparently have the technology to do it, since they have reported papers using it in the past. Measure the light absorption spectra, over a wide wavelength range, of the methanol solution containing

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Discussion paper

the dissolved components of tar balls.

The authors delineate tar balls from HULIS and BrC, implying it is a separate aerosol component in smoke. But it is generated by essentially dissolving an isolated product of smoke in methanol and then re-aerosolizing. Many studies, both from smoke generated in the lab and from collected ambient particles, have used methanol as a solvent to determine the light absorption spectra of smoke chromophores. Presumably these experiments/measurements would include chromophores that comprise tar balls. Thus I do not understand why tar balls are exclusive to BrC determined in methanol extracts. This is important because I am not aware of any strong evidence that methanol soluble smoke particles contain a significant number of chromophores that absorb at such high wavelengths (ie based on experiments where collected smoke particles are dissolved in methanol and the light absorption spectra measured). My interpretation is then, based on these observations, that what the authors are measuring is likely a very small component of the overall aerosol absorption spectra.

First, why not measure the light absorption spectra of the tar ball material they generate. Based on the generation method, they have a methanol solution from which they generate the aerosols. The chromophores must absorb at high wavelengths if the aerosols are also expected to. This is a simple and direct way to determine if the hypothesis is correct. It gets away from all the issues with correcting aethalometer data and the use of AAEs to infer absorption over the whole spectrum when only 5 or so wavelengths are measured. (Although it does have other limitations).

Secondly, if the authors are to claim that tar ball absorption in the red and infrared is important in the ambient atm, as noted above, why has no significant absorption been observed at these wavelengths based on filter methanol extracts in fresh and aged plumes? Again, there most be chromophores that absorb at the high wavelengths if this hypothesis is to be supported. These authors could obtain filters loaded with aged biomass burning smoke from their field site (K-puszta station), extract in methanol and measure the absorption spectra. If substantial absorption is observed in the red and infrared, then they have direct proof. If they wish, use Mie theory to estimate the optical effects, based on some assumptions on size and mixing state.

Specific Comments;

The following statement is only partly true: Separating the BrC absorption from BC absorption in field and laboratory studies has relied on the assumption that no other carbonaceous particle type except BC absorbs solar radiation at the wavelength of ∼700 nm or larger (Bahadur et al., 2012; Kirchstetter and Thatcher, 2012; Saleh et al., 2014; Lu et al., 2015). All of the references are based on using optical instruments to measure light absorption. In that case, since total light absorption is measured, it is true that some assumptions have to be made about BC and BrC to separate the two. This statement is not true if one measures the light absorption of chromophores, what the authors refer to as acetone (or methanol) soluble OC.

Pg 6 it states: The absorption coefficient at 880 nm exceeds by 10% that at 470 nm for both wood types, undermining the common assumption that all BrC particles have zero absorption at 880 nm. As noted above, this assumption does not have to be made if one measures light absorption of the aerosol extract. The authors are ignoring much of the literature on BrC.

Regarding Fig. 3, it is claimed on page 8 lines 10 and 11 that the AERONET AAEs can be explained by tar balls and BC. But what are the assumptions about BC AAEs? State them. Given that there can be a range in BC properties (ie, effect of coatings on AAEs, etc), how can the authors make such a strong conclusion? It seems just the uncertainty in BC absorption could explain the AERONET data, no need for tar balls (ie, the AERONET AAEs are in the range that could be complete explained by BC). This needs to be discussed. Overall, I find this section highly speculative and not convincing given the well documented uncertainties in inferring AAEs from AERONET data, combined with uncertainties in BC AAEs.

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