

Interactive comment on "Absorptivity of brown carbon in fresh and photo-chemically aged biomass-burning emissions" by R. Saleh et al.

Anonymous Referee #1

Received and published: 13 June 2013

In this paper a unique rather convoluted method to infer the complex component of the biomass burning aerosol refractive index is used. The method makes many assumptions, however, the results seem reasonable when compared to published values (although as noted below, I believe some of the comparisons are incorrect). It would be useful if the authors discuss if the method could be used for ambient aerosol. Example, over what concentrations of smoke would the method work? These measurements were made in what would represent fairly fresh (max 1 hr old) plumes in the ambient atmosphere. One major issue with this paper relates to how useful the data presented really is given the complexity of biomass burning emissions and the rather limited data presented; this is discussed more below. Secondly, it is not clear how the problem is constrained when there is no actual information on the size distribution of the brown

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carbon. The implicit assumption is that the brown carbon distribution is the same as the OA distribution, which there is no evidence for. It is highly possible that the mass concentrations of light absorbing components are very small relative to the POA or SOA. Thus, size distributions and chemical properties of the bulk OA, as measured by an AMS in this case, could be of little value. This limitation is not considered in the uncertainty discussion.

The background and references are not complete. There a number of missing references to papers that have specifically investigated the prevalence of brown carbon from biomass burning, however, more importantly, recent papers have investigated the chemical composition of brown carbon, one specifically focusing on smoke, which likely came out following submission of this work (Desyaterik et al, JGR, 2013). This is relevant to the note above and also to the authors claim that linking light absorption and composition is a future goal (pg 11524 lines 9 -12); others researchers have done it and this should be noted.

Pg 11511 lines 5-10. This seems to imply the mass weighting is due to brown OA and black BC, but the OA is composed of many compounds, all of varying optical characteristics, thus the overall color of the OA depends on the mass weighted optical properties of the various chromophoric OA components and their size distributions. It is more complex than OA to BC ratios.

Pg 11511 line 16-17. Data on atm OA k as a function of size are even more rare, and are what is needed.

Pg 11511, lines 19-23. There are assumptions made that influence the magnitude of k calculated from solvent extracts. Generally, there is no particle size information so the k represents an average for some unknown size distribution, which will depend on the emission and subsequent processing. Furthermore, the k calculation requires a measure of the mass concentration of the absorbers in solution. Clearly not all extracted components absorb light so use of some bulk mass concentration, such as organic

carbon mass, is only representative of that extract (eg, a different extract may have different proportions of organic carbon that are absorbers or non-absorbers). Some studies have used carbon mass, but different measures of mass could be used. This use of a rather arbitrary mass concentration for absorbers is not a significant issue if in the subsequent Mie calculation the same mass concentration is used. However, the reported k values do depend on mass concentration and so comparing k from these bulk measurement is not straight forward and the direct comparisons shown in Fig 6 may be incorrect.

Page 11512, Lines 5-14. The argument is that biomass burning BC gets coated with OA whereas in urban plumes this does not happen? Urban measurements of BC size distributions show rapid shifts to the accumulation mode do not support this.

Page 11516 line 10-12, This statement seems to strong, all that can be said is that the aethalometer and 4-wavelength PAS give similar AAE, not that they are the true values for the ambient aerosol.

A discussion on the sensitivity of the predicted absorption coefficient on AAE would be informative.

Figure 3: The mass distributions would also be of interest since it would provide some insight on coating mass. Also note Fig. 3c typo in legend.

Page 11523. These arguments attempting to explain the wide differences in the various results shown in Fig 6 seem to demonstrate the great limitations when attempting to apply these results to make predictions for ambient smoke plumes. Factors of 10 or so between koa at low wavelengths (300nm), possibly due to different types of burning, as argued here, seem to make the idea of reliable model predictions highly unlikely. The apparent high degree of variability in the optical properties of biomass burning aerosols means that many ambient measurements will be required to link burning type, conditions etc, to refractive index. This means a fairly simple robust method will be required. The authors might want to discuss if their technique could provide this type

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of data.

Page 11524, lines 12-14. It should also be pointed out that there are studies that show very little SOA formation in biomass burning plumes.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 11509, 2013.