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Atmospheric
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Interactive comment on “Spectral absorption of biomass burning aerosol determined from retrieved single scattering albedo during ARCTAS” *by* C. A. Corr et al.

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The manuscript reports the determination of aerosol optical properties in two biomass burning plumes made aboard the NASA DC-8 aircraft during the ARCTAS mission in April and June 2008. Spectrally resolved (350-500nm) aerosol single scattering albedos (SSAs) are determined from actinic flux measurements by using a radiative transfer model. Spectrally resolved aerosol absorption, calculated from the SSAs, is used to determine Angstrom absorption exponents (AAEs).

The data presented build on similar observations of enhanced aerosol absorption in biomass burning plumes reported previously. Although it has been suggested that extended measurements over a wide wavelength range are important in determining the

impacts of absorbing aerosols, little data are available that cover a wide wavelength range due to the measurement difficulties. However, as the authors admit, there are also major difficulties in retrieving aerosol optical properties from actinic flux measurements and therefore there are several important problems with the data interpretation in the present work that need to be addressed.

The spectrally resolved aerosol optical depths from 350 to 550 nm are estimated by using an Angstrom exponent calculated from aerosol absorption measured at 450 and 550 nm by a PSAP on board the aircraft. The authors state that the errors associated with the extrapolation of the AE from 2 visible wavelengths into the UV are likely small (page 13974, ln 16-28). However, the results reported in Table 2 and page 13981 (ln 5-10) show that the AAE results are much different if calculated in the visible versus the UV range. In fact, the authors use this discrepancy as a possible explanation of the very high AAEs reported in this study compared with values reported previously (page 13982). A much more detailed assessment of the errors associated with the extrapolation of the AE from the 450/550 nm pair into the UV needs to be included.

Although the actinic flux measurements extend to 600 nm, SSA values were only calculated to 550 nm. The data sets were also truncated at 500 nm due to unexpectedly low SSA values below this point on Apr 17 (Fig. 3). This was attributed to low AODs. However, it would seem that low AODs would result in a noisier data set not in a systematically low error. Therefore discarding the data below 500 nm seems arbitrary. Since a major goal of the work is to present spectrally resolved aerosol optical properties, the SSA aerosol absorption determinations should be extended to include all the applicable actinic flux data (350 – 600nm) and results should not be discarded simply because they are unexpected.

Aerosol absorption (Fig 5) is calculated from the SSAs and aerosol extinction (Equation 4). How was aerosol extinction calculated? If extinction was calculated by using the AEs from the 2 visible wavelength pairs, (as in page 13974, ln 6-9), how does this bias the results, especially in the UV? The authors state that “the b(abs) values in the UV wavelengths are much higher than those calculate at the visible vavelengths”. . .. How was this determined? Were values simply averaged over the entire range (350-400nm; 400-500nm)? If so how were the sharp bands attributed to interferences treated? The absorbance plots in Fig. 5 seem to have an enhanced absorption appearing as a broad band from 410-455 nm in both events. Is this considered real? How does this affect the calculation of AAEs?

It can only be assumed that the Retrieved AAE reported in Table 2 is calculated across the entire wavelength range of Fig 5 (350-500nm), although this is not explicitly stated. How was this determined? Were all data used exactly as shown in Fig 5? What AAEs are obtained from the data sets if only absorbances at 450 and 550 nm are used - analogous to the PSAP calculations? 470/532 (Table 2)? 470/660 nm (Table 2)? Table 2 should be expanded to include AAEs calculated for different wavelength ranges including those used by other authors (page 13981 - 13982). Standard deviations and goodness of fit (r2) of the logarithmic plots should also be included in the Table.

The retrieved AAE reported for Apr 17 is very high (6.78)!! Values this high have not been reported for whole aerosols. Although AAEs >6 have been reported for water soluble species extracted from biomass burning aerosol samples, these extracts were composed primarily of humic-like substances, which have been estimated to be ∼35% of the whole aerosol and the AAEs for the whole aerosol samples from which they were extracted were much lower (∼3). The retrieved AAE for Jun 29 (3.3) is more in line with those reported previously in both fresh and aged biomass burning plumes. The aerosol absorbances reported for Apr 17 were a factor of 10 lower than on Jun 29 and showed strong high frequency variations in the UV which would not be expected for absorption due to aerosols. The authors attribute this variance to absorption by atmospheric gasses. Whatever the reason, these interferences render the very high retrieved AAE for Apr 17 suspect. If the lines observed in Fig 5 on Apr 17 are indeed absorption from interfering gasses, an attempt should be made to identify and remove them or at the very least report the AAEs +/- one sigma.

The contributions of OA to the absorption profiles were determined by subtracting a λ-1 profile extrapolated from absorbance at 660 nm determined by the PSAP. This assumes no absorbance at 660 by OC. Is this assumption valid? The AAEs reported from the OC absorbance profiles for Apr 17 (Fig 6) are unreasonably high (9.59 for the UV and 7.56 for the visible). What are the standard deviations and r2 for these results? Using this same procedure for the PSAP measurements, what would be the b(abs) for OC at 470? 532?

How were percentages in Tables 3 and 4 calculated? Do the numbers represent averages of the absorbances over the entire wavelength ranges (350-399nm; 400-500nm)? Since the absorbances vary according to the AAEs, averages over a wide wavelength range are meaningless. Tables 3 and 4 should be combined and values reported only for single wavelengths, or narrow wavelength ranges, with standard deviations included. Also, a direct comparison should be added between retrieved data and that calculated from the PSAP measurements, both at the same wavelengths.

An attempt is made to correlate enhanced UV absorption and higher AAEs by calculating the extent of OC oxidation (OSc). However, values reported from +0.3 to -.4 on the 2 events have little meaning without previous results for comparison. A better approach would be to compare AAEs with EC/OC measurements or OA from the AMS.

In the final analysis the data set represents 2 points, one of which is suspect due to low AODs and possible interferences from trace gasses. As such, the data are over interpreted. The paper should be shortened, focusing on the results as an example of obtaining aerosol absorption from actinic flux data with a more in-depth error analysis and detailed comparison to in-situ measurements as a means of validating the results. Omit the attempt to predict the extent of aerosol oxidation (Table 5; section 4.2) or aerosol composition as this is not substantiated by the data as presented.

Additional Comments.

âŮŔ Table 1 is difficult to read and should be expanded for clarity. In-plume and profile means should be listed in separate columns. List ranges instead of deviations, again in separate columns. Add distance of aircraft travel and number of points averaged. It would also be helpful to include any EC/OC results, and measurements of greenhouse gasses expected to be interferences (O3, NO, PANs, CH4).

âŮŘ Be consistent with event titles: e.g. Fig 3 refers to 04/17 while Fig 4 refers to 29 June.

âŮŘ Include PSAP measurements of absorption in Fig 5.

âŮŘ Tables 3 and 4 should be combined and standard deviations added.

âŮŘ Figs 6 and 7 (log axis) are difficult to reconcile with Fig 5 (linear axis). Portions of the data have been omitted without explanation. The AAEs calculated from these data should be accompanied with standard deviation and goodness of fit criteria.

âŮŘ Keep in mind the standard deviations and instrumental accuracies when reporting significant figures. . ..e.g: Table 1 BC = 6974.00 +/- 1688.06. I don't think the PSAP is either accurate or reproducible to 8 significant figures.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 13967, 2012.