Liu et al. (2015) present aircraft measurements of light-absorbing carbonaceous aerosols, specifically black carbon (BC) and brown carbon (BrC). Through a combination of online optical and speciation measurements and offline spectrophotometric absorption measurements of extracts, they attempt to deduce the relative contribution of BC and BrC to atmospheric light absorption. They then use the results in a radiative transfer model to calculate the radiative effect. The measurements are interesting and timely, and seem to be of high quality. However, there are a few major issues that need to be addressed before the paper is suitable for publication in ACP.

Major comment 1:

The conversion of spectrophotometric absorbance measurements to aerosol "absorption coefficients" needs to be done more rigorously. Also, the method and terminology are poorly described.

1.1. It is not clear in the paper how the authors convert the bulk absorbance measurements to aerosol absorption coefficients. I had to go to their previous work (Liu et al., 2013) to get an idea. This needs to be described again, maybe in the SI, or at least a direct reference should be provided. Since they use data from both bulk and aerosol measurements, the authors need to be very clear on what they mean by "absorption coefficient" because in the former it's an intensive property related to the imaginary part of the refractive index and in the latter it's an extensive property (the total aerosol absorption cross-section per unit volume of air) – both having dimension L^{-1}. Without referring to the earlier work (Liu et al., 2013), one could get the impression that the authors confuse the two "absorption coefficient" definitions in their analysis. Incidentally, in their nomenclature (Table 1) they express the units as (M/m), which I assume should be 1/Mm (1/mega-meters).

1.2. How are the aerosol mass concentrations obtained for calculation of H2O_Abs and TOT_Abs?

1.3. Using a single conversion factor of 2 for all data points is not convincing. Ideally, the authors could explicitly calculate the conversion factor for each data point using Mie theory based on the measured size distribution. They need to at least quantify the uncertainty associated with the constant conversation factor and/or show that their conclusions hold in light of this uncertainty. The Mie calculations shown in Figure 1 below suggest that this uncertainty is rather larger. The conversion factor (MAC / [alpha/rho]) calculated at 365 nm varies considerably with particle size and the imaginary part of the refractive index (k).

1.4. The authors acknowledge the uncertainty associated with their mixing state assumption (they assume BrC and BC are externally mixed), but they can do more to quantify the uncertainty. They can perform calculations (at least for some of the data points) using the internally mixed assumption and account for absorption enhancement by lensing as a bounding case. The core-shell sizes can be assumed based on OA/BC ratios obtained from AMS/SP2 measurements. This can be done either explicitly using Mie theory, or the approximation given by Bond et al. (2006).

Major comment 2:

The discussion of the spectrophotometric absorbance measurements is not adequate. The authors discard the data at wavelengths longer than 450 nm, where Figure 4 shows absorbance leveling out. The authors attribute this to the "chemical nature of the chromophores" which I take to mean that there exist chromophores that absorb significantly at wavelengths longer than 450 nm. If this is true, then the data should not be discarded! When they calculate AAE values based on data at wavelengths smaller than 450 nm, then they use this AAE to extrapolate to longer wavelengths, this would introduce major bias.

The authors need to think more about the shape of absorbance curves in Figure 4 and how to interpret the measured absorption at wavelengths > 450 nm.

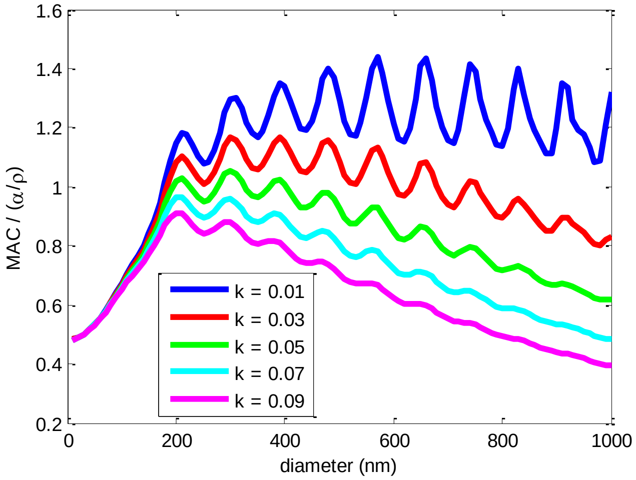

Figure 1: conversion factor as a function of particle size and for different imaginary part of the refractive index (k) values.

Major comment 3:

The radiative transfer calculations.

3.1. The authors state that they include scattering aerosols. What types, and how were their concentrations and vertical profiles obtained? The same for gases.

3.2. Also, they do the calculations with and without BrC and find that BrC reduces the overall cooling by 20% (from -24.84 to -19.33). A couple of points: 1) is that the overall effect of BrC or is it the effect of "brownness"? In other words, when they do the calculations without BrC do they take the OA out or keep it but assuming it's non-absorbing? 2) This 20% is only meaningful when compared to the BC effect. What is the difference in cooling with and without BC? If it is much larger than the 20% effect of BrC (which still needs to be better defined), then the BrC effect cannot be considered significant.

3.3. The aerosol radiative effect numbers (-20'ish W/m2) seem to have a very large magnitude, at least compared to the global average aerosol DRF in say the IPCC report (close to -0.5 W/m2). How do the authors' numbers compare to other studies that calculate regional aerosol DRF over the US?

3.4. The chart to extrapolate surface measurements to TOA effect of BrC is not well-supported. The authors need to provide evidence/arguments that the vertical profile measurements done in this study (thus the chart) can be generalized.

Minor comments

1) The authors state uncertainty bounds in measurements at several points in the manuscript without mentioning how these were estimated (e.g. p. 5965 line 21 and line 28, and many other places..)

2) PSAP measurements: The authors rightly mention that the PSAP is notorious for artifacts, however their statement about uncertainty (20%) is not convincing. The adjustment of data needs to be shown explicitly (maybe in SI) for at least one or more representative cases to show the extent of correction applied. For example, is the correction wavelength-dependent?

Specific comments:

Abstract line 21: "BrC is an important component of.." should be something like "BrC is an important contributor to.."

p. 5962 line 6: there are many more studies that have investigated light absorbing OA – add e.g. to the reference list.

Paragraph starting p. 5962 line 26: the authors dismiss online optical instruments as a means to quantify BrC absorption, stating mixing with BC as a major issue. It's true that mixing with BC poses difficulties, but there have been attempts to decouple the effects of BC absorption (including enhancement due to internal mixing – lensing) and BrC absorption by integrating measurements with Mie theory calculations (e.g. Lack et al., 2012; Saleh et al., 2014). These methods should be acknowledged. Then they present offline measurements of OA extracts as the better alternative, which can be argued, but is not necessarily the case. These too suffer from issues, the most obvious being extraction efficiency. This should also be acknowledged.

p. 5970 line 23: it's not clear how the conclusion that "water-soluble faction in the background troposphere could be more strongly related to primary emissions and possibly linked to aged biomass burning" follows from the data.

p. 5970 line 26: I don't see a reason to jump to the conclusion that there used to be BrC but got photobleached. The more straightforward conclusion is that there was not much BrC to begin with.

p. 5972 line 5-10: the difference between PSAP_abs 365 and 660 50% mark is too small to support the conclusion that the authors make on the relative wavelength dependence of BrC vs BC absorption. In fact, from Figure 3, the 50% mark for 365 is closer to 4.2 km (the authors say 4.5 km in the text).

Reword section 3.5 title: BrC is a component of light absorbing aerosols.

Reword section 3.6 title: you don't compare BC to BrC, but compare certain properties of the two (mass, absorption, etc.)

p. 5974 line 25: it's not clear how nitrate (you mean inorganic nitrates?) would impact the data.

Reword title of section 3.7: you can't compare to PSAP – it is an instrument not a measured quantity.

References

Bond, T. C., G. Habib, and R. W. Bergstrom (2006), Limitations in the enhancement of visible light absorption due to mixing state, *J. Geophys. Res.*, *111*(D20), 1–13, doi:10.1029/2006JD007315.

Lack, D. A., J. M. Langridge, R. Bahreini, C. D. Cappa, and A. M. Middlebrook (2012), Brown carbon and internal mixing in biomass burning particles, *Proc. Natl. Acad. Sci. U. S. A.*, *109*(37), 14802–14807.

Saleh, R. et al. (2014), Brownness of organics in aerosols from biomass burning linked to their black carbon content, Nat. Geosci., doi:10.1038/NGEO2220.