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Interactive comment on "Deriving Brown Carbon from Multi-Wavelength Absorption Measurements: Method and Application to AERONET and Surface Observations" *by* Xuan Wang et al.

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Review of "Deriving Brown Carbon from Multi-Wavelength Absorption Measurements: Method and Application to AERONET and Surface Observations" by Xuan Wang, Colette L. Heald, Arthur J. Sedlacek, Suzane S. de Sa, Scot T. Martin, M. Lizabeth Alexander, Thomas B. Watson, Allison C. Aiken, Stephen R. Springston and Paulo Artaxo

This paper uses empirical methods to derive organic carbon absorption from optical and in situ measurements. The authors apply their results to hundreds of AERONET sites and 10 field sites. Unfortunately, their methodology is not sound, and their results are not consistent with what is known about BrC.

MAIN POINTS

The authors are merely adding a new twist to the AAE method pioneered by Chung (2012) and Bahadur (2012) (although they did not acknowledge the similarity of their work to these previous works). The authors determine the AAE in two spectral regions via Eq 1, and then define the nonlinearity of the AAE as the wavelength dependence of the AAE (WDA) in their Eq 2. Their WDA is presented as a ratio of two exponentials, so mathematically it is equivalent to

WDA = exp(AAE_440/870 - AAE_675-870).

Later, the authors use ln(WDA) in their Eq 4, so they are really just using the difference of AAE measured over two spectral regions to characterize the BC and BrC absorption at 440 nm. This is very similar to Bahadur (2012)!

The authors go on to compute WDA for black carbon (BC) using a set of lognormal distributions (p4, lines 22-31), and provide a range of WDA values for their computations as the grey shaded area in their Figure 2. The median of this range of values is the black line, which represents the WDA_0 of Eq 4. Thus, the authors are using theoretical BC AAE calculations as a baseline AAE; differences from this baseline are attributed to BrC. Particle size effects on AAE are not considered, although extreme dust cases are later omitted from the analysis (EAE < 1 and AAE > 1.5).

There are many problems with this. First of all, the description of the parameters used for their Mie calculations does not include enough details for a reader to understand and duplicate the shaded region of figure 2. For instance, how thick are the coatings when the absorption enhancement factor is 2? What are the optical properties of the coatings? How does varying the optical properties of the coatings affect the size of the shaded area? Which size distributions represent the baseline? Are these 20nm particles, or 300 nm particles? Do the baseline particles have coatings? How thick? Which AERONET sites are falling inside of the shaded region of Fig 2? Urban OC is non-absorbing, so the urban aerosols should fall inside of the shaded region, and the biomass burning aerosols should fall outside of the shaded regions. Looking at Figs

3&4, I don't believe that this is the case.

How come the shaded area of Figure 2 does not include the point (1,1), which would represent AAE = 1 for the entire wavelength range? Some of the particles sizes for their calculations are small enough (20 nm GMD) to reach the theoretical limit for small absorbing particles.

Why aren't any coarse mode aerosols included in the computations of the shaded region? There is always a coarse mode in the AERONET retrievals, and this will affect AAE. Sure, the authors eliminate aerosols dominated by dust in Section 3.1 by filtering for EAE < 1, but aerosol systems with 1 < EAE < 2 are mixtures of fine and coarse particles. Thus, there is dust in much (most?) of their analysis.

How well is the AERONET AAOD known? How does the uncertainty in AAOD at each wavelength affect the uncertainty in the AAE in the two spectral regions that you are using? How do the AAE uncertainties affect the BrC AAOD retrieval?

Beyond the theoretical issues with this approach, though, the results do not seem to back up this approach. Figure 3 shows regional maps of seasonal mean BrC AAOD. The biomass burning sites in S. America never show elevated BrC AAOD, and the biomass burning seasons are not detectable over Africa.The Asian sites seem to have elevated BrC in all seasons except for Summer.

Figure 4 provides seasonal mean fractions of (BrC AAOD) / (total AAOD), and those results are even more ambiguous. The colors on these maps are all very random, with dark reds co-mingled with light blues; one would expect neighboring AERONET sites to exhibit similar colors if they were representing similar source regions. The randomness of the colors on these maps, though, seem to indicate noise.

OTHER IMPORTANT POINTS:

Page 3, line 6: AERONET provides *retrievals*, not measurements.

Page 3, line 23-25: Chung (2012) and Bahadur (2012) do not assume AAE = 1 for

BC, as stated here. The authors need to be more careful when attributing concepts to others.

Page 5, line 8: Bahadur (2012) has nothing to do with the SSA screening conditions of AERONET. Cite Dubovik (JGR, 2000), instead.

PAge 5, line 16: The authors state: "... and cannot be estimated using our method without additional information about the size and coating state of the particles." However, you have information about size in the AERONET retrievals. Why not use them?

Page 5, equations 3&4: These equations should be reversed. WDA_0 is used to compute BC AAE_440/870, which is used to compute BC abs(440), right? So presently, Eq 4 gets used before Eq3.

Page 6, line 18: The authors state: "In general, BrC-AAOD is smaller than 0.005 at most sites but larger in Asia and occasionally in northern Africa and South America." BrC-AAOD < 0.005 in general? That sounds like a small number. Does the AERONET retrieval really have the sensitivity to quantify this parameter, then? AERONET only claims an accuracy of $\sim$0.01 for AOD; the accuracy of AAOD is certainly no better. So all of the BrC-AAODs that you are retrieving seem to be well below AERONET's accuracy threshold.

Page 6, lines 24-26: The authors are quoting numbers for seasonal variability, but they do not specify seasons or sites. The colors on the maps do not obviously correspond to the numbers quoted here. The authors should at least give these "special" sites a different shape, so that the reader can understand which sites are the "African biomass burning sites." Also, the biomass burning season on the Sahel is not July-October. It starts in November and ends in the Spring.

Page 6, line 31: The authors state: "In winter the BrC AAOD contributions in North America (15%), Europe (17%) and East Asia (23%) are larger than in summer (10%, 13% and 14%)." Why should BrC AAOD fractions be higher in the winter in North Amer-

ica? Few Americans use biofuel, and the biomass burning season is in the summer. Also, higher winter BrC AAOD fractions are not obvious from Fig 4.

Equation 6: The authors assume that all OC are BrC for Equation 6. This is not a valid assumption.

PAge 12, line 10: The authors state “the correlations between BrC-AAE and BrC absorption contributions are only slight to moderate at these sites.” They should still provide the correlations to the readers, though.

PAge 12, lines 22-23. The authors state that OA-MAC is correlated with BC/OC mass ratio, but they do not help the reader to understand why this should be the case (anywhere in the article). Thus, for a given OC mass, the OA-MAC is linearly related to BC mass? This sounds like BC is contaminating their OC-MAC retrieval (rather than proof that their retrieval is working).

PAge 13, lines 26-28 and Figs 10b, 10c: Authors find that the OA-MAC for the non-biomass plumes (Fig 10b) are much much higher than for the BB plumes (Fig 10c). WHY? OA-MAC for urban aerosols should be negligible.

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