Carbonaceous Aerosol AAE inferred from in-situ aerosol measurements at the Gosan ABC super site, and the implications for brown carbon aerosol

Chung et al. ACPD 2012

The authors present a detailed study of optical properties of carbonaceous aerosols as monitored over a long-term study conducted in Jeju Island, South Korea. Using co-located mass and absorption measurements, the authors offer an estimate of absorption due to organic carbon (referred to in the manuscript as Brown Carbon Spheres), finding it to be approximately equal to absorption due to Black Carbon. The authors also offer estimates of absorption cross section for black and organic carbon, which may contribute to future studies of absorption by aerosol species. Although the subject matter described is important in terms of current scientific interest, in my opinion the manuscript as written is not suitable for publication without significant improvements.

Structural Concerns

1. In my opinion the Authors do not provide adequate context for their work, particularly in the abstract and introduction sections. For example – they seek to calculate the AAE and MAC, but do not explain why these properties may matter. Further, their most significant result (i.e. OC absorption is 45% of total Carbon absorption) needs to be better related to applied climate science – how does this compare to what present studies and models are suggesting or assuming, for instance. How would this number impact our current understanding of aerosol forcing? These questions need to be answered.
2. The manuscript is very poorly organized. For example, Figure 9 and Table 3 are referred to out of order on Page 4510. Similarly, Section 4, which describes errors and uncertainties in the measurement, should precede Section 3, which describes analysis of the measurements. Sections 5 and 6 could be possibly combined.
3. In my opinion, the text of the manuscript needs to be copy edited by a native English speaker to conform to acceptable standards of scientific publications. For example, lines 5-12 on

page 4510 are completely extraneous. Other examples of problem text are -

   a. "When a particle is coated during aging the entire particle is surrounded by a coating shell", 4509-20
   b. "Alexander (2008) identified different kind of BrC particle", 4511-5
   c. "We use BrC to refer to absorbing organic material, while BrC aerosol is used as such" 4511-20
   d. Lines 17-20 on Page 4512 are poorly written.

4. I also find that the authors have not adequately cited their assertions or statements in the text. Again, a few examples –
   a. "Coating amplifies BC MAC", 4510-13
   b. "There are plenty of ambient measurement studies…", 4510-25
   c. "We expect Aethalometer data to be particularly erroneous during precipitation events", 4513-20

The examples listed here do not constitute an exhaustive list. While some of these concerns may seem trivial, taken together, they severely compromise the quality of the article and therefore I cannot recommend it for publication without significant rewrites.

Major Scientific Concerns

1. Absorption Angstrom Exponent (Section 2.2). The authors determine the AAE by regressing a single curve onto their 7-band absorption measurement. While this satisfies the classical formulation, recently it has been shown [see for example, Flowers et al. 2010 (ACP) and Moosmuller et al. 2011 (ACP)] that the AAE itself is wavelength dependent. Further, the non-linear features of the AAE, i.e. the deviation from the standard value of "1.0" are more pronounced for OC like aerosols at short wavelengths. By regressing a single curve onto their measurements, it is my opinion that the authors are emphasizing the long-wavelength (i.e. flatter) part of the curve, which may explain their very low value of OC AAE.
2. Filtering out dust-influenced events (Section 3). The authors eliminate measurements that could potentially contain a large dust mass (and correspondingly absorption) by tracking the PM10/PM2.5 ratio, working on the premise that dust is typically

found in the coarse mode. While I find this reasoning to be sound, it is problematic that the authors do not justify their threshold value of 1.6. What is this based upon? As best as I can see, only Figure 4 actually illustrates measured values of this ratio, presented as monthly values. Based on this figure alone, only measurements for November and June are valid – therefore this figure is clearly inadequate, and worse, misleading. The authors need to provide a concise visualization of the entire data set (perhaps as a probability distribution function), and justify why 1.6 was chosen as a cutoff.

3. Somewhat minor – as written, Equation 2 is incorrect as the intrinsic OC is being subtracted from both the BC and OC.
4. Assumption that OC constitutes 20 % of the BC mass – again, this assumption needs to be justified in some fashion. The authors find that this does not impact their OA MAC (understandably, since OC concentrations are typically very high in Asian outflow), but it does significantly change their estimate for the BC MAC – this in turn impacts the authors core conclusion that OC absorption is roughly 45% of total carbonaceous absorption. At the very least, the authors need to provide a range of uncertainty here.

Minor Points –

1. In addition to combustion, biogenic and marine sources are significant contributors to organic aerosol - see Russell et al, 2011 (PNAS).
2. In general I find the figure labels and legends to be small and hard to read. This is particularly true for Figure 3.