

Interactive comment on “Optical properties of elemental carbon and water-soluble organic carbon in Beijing, China” by Y. Cheng et al.

Anonymous Referee #1

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The manuscript by Cheng et al. attempts to use observations of light absorption, made using a filter-based Aethelometer method, and elemental carbon concentrations, made using a thermal-optical method, to assess how coatings influence the light absorption by EC particles. They also use extracts of WSOC to determine absorption properties of the WSOC. Finally, they attempt to take disparate measurements of absorption and [EC] (reported as the mass absorption coefficient), correct them all to a common basis, and make comparisons between MAC values determined in different regions.

My assessment is that this paper should be rejected. Firstly, it is extremely difficult to follow what they are comparing to what when it comes to the MAC values for EC because so many corrections have been applied AND they have a tendency to intermix discussion of uncorrected and corrected MAC values. Secondly, and most importantly,

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there are some fundamental mis-understandings related to aerosol optical properties and their measurement, which will be detailed below.

1. The authors appear to be conflating intensive and extensive properties. The influence of coatings on MAC is an intensive property, whereas absolute concentrations are extensive properties. It only makes sense to plot e.g. the MAC vs. sulfate if one knows for certain that the [EC] is constant while sulfate varies (thus changing the coating thickness). Here, the requirement to turn correlation into causation has not been met. The authors should look at the recent review of “Time-resolved measurements of black carbon light absorption enhancement in urban and near-urban locations of Southern Ontario, Canada” by “Anonymous Referee #2”, available at <http://www.atmos-chem-phys-discuss.net/10/19939/2010/acpd-10-19939-2010-discussion.html>.

2. Absorption enhancements don't just shut-off at high coating thicknesses. There is a continuum. If the authors want to argue that the low vs. high sulfate dependence is a result of Mie theory and is “consistent with Bond et al. (2006)”, then they need to provide supporting calculations. However, they can't do this because they don't know the fundamental size of their BC particles. Core size has a direct impact on the potential magnitude of the absorption enhancement.

3. Have the authors read Lack et al. (2008) and Cappa et al. (2008)? These are not cited, yet should be, as these works call into question much of the presented analysis related to MACs for EC. Essentially, these works suggest and show that filter based methods are (very likely) incapable of determining absorption enhancements due to inherent biases in filter based methods. The authors should pay particular attention to Figure 4a and 4b in Lack et al. (2008) and how they relate to Figure 3 here. Granted, the Lack et al. and Cappa et al. results were done using a PSAP, not an Aethelometer, but the issues discussed are general to all filter based absorption measurements. In light of these works, the authors need to convince the reader (and reviewer) that their methods are not similarly biased. This will not be an easy task and, in my estimation, will ultimately preclude any analysis of the type presented here.

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4. Why are the authors even presenting uncorrected MAC values? This is confusing/misleading, making the apparent MAC values seem much larger than they actually are. As the authors state, “the unrealistically high values of MAC in Table 1 are mainly due to the artifacts associated with the “filter-based” methods...” If this is true, then these results are meaningless and should not be presented in a table unless the “corrected” values are given right in a column next to them. In other words, Equation 3 should have as part of it a correction for the absorption. Certainly, the uncorrected MAC values should not be presented in the abstract!

5. It might be that the authors have been able to “correct” for differences in [EC] between different TO protocols, but they give no discussion of how different studies may/may not have accounted for biases in the absorption measurements used to determine the MAC. Note that some of the studies listed in Table 1 used an Aethelometer (as in this study), but others apparently derived absorption from the TOT measurement itself. No discussion at all was given as to how these TOT absorbances should/could be corrected. In other words, although the authors may have been able to make the [EC] from different studies “equivalent”, they have in no way made the absorptions “equivalent”, thus invalidating their global analysis of variability in MACs.

6. There are large enough differences in the sources of biomass burning aerosol between the seasons to make the contention that the difference in the MAC for WSOC between the seasons arises from differences in anthropogenic vs. biogenic SOA untenable.

7. Section 3.1: Two fundamental over-(mis-) interpretations here. (1) The authors cannot simultaneously argue for such a large MAC while suggesting that the MAC can be determined from a linear fit to ATN vs. [EC]. The coating thickness may vary for each point, making the single point MAC very different from the average. Further, it is not unreasonable to think that [EC] may be correlated with coating thickness (more emissions = greater potential for coatings). Are the authors arguing that the coating thickness is always the same? (2) Just because there is an intercept close to zero

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does not indicate that EC was the major light-absorbing component, as stated. It is entirely possible (in fact, probable) that [EC] is correlated with brown carbon, and thus as [EC] \rightarrow 0, so would [brown carbon].

8. Section 3.2 (1): Although I see some value in converting different thermal EC measurements to a common basis, what is really needed (if the authors are to truly be quantitative with regards to their MAC results) is a way to put the measurements on an absolute basis. Simply referencing one (potentially wrong) measurement to another (potentially wrong) measurement does not make either correct. By converting measurements to “equivalent” MAC values, are the authors implicitly arguing that the method they used is the best, i.e. most accurate? This would seem to be their argument, since they go on to do quantitative analysis, but they do not state this. Additionally, the variability about any of the conversion factors is so large that such conversion has significant uncertainty. This is partially reflected in the range of values given in Table 1 for “equivalent” MAC values, but it is not clear why only some values have an upper and lower bound while others have only an upper bound.

9. Section 3.2 (2): The authors discuss how brown carbon is important in their region of measurement. Yet they adjust everything to 632 nm (which is seemingly randomly chosen) using an Angstrom exponent of 1. I don't see how this is self-consistent.

10. Table 1: Although I am not convinced that the “charring” and “temperature” protocol corrections are likely to give robust results, the authors should nonetheless include the exact values used for each location in Table 1 so that the reader can understand exactly what correction has been done. From the text alone it is a bit confusing.

11. Equation 12: “f” is not defined. Further, the “f” values that are referenced come from an entirely different study and are almost definitely going to differ depending on the type of aerosol being sampled. Thus, they are not applicable to this study. The authors should be able to use the same methods as Sandradewi et al. (2008) to determine their own “f” values. Further, the R(ATN) factor depends on the current ATN, which is not

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a constant (as implied) and depends on how long the system has been operating and what the current loading is. That the authors give $C \times R(ATN)$ as a constant suggests a fundamental misunderstanding of how the correction is supposed to work.

12. P. 6236, Line 1: I do not see why it matters whether the OC is POC or SOC. The only thing that should matter is how much OC is internally mixed with the EC.

13. P. 6236, Line 7: The relationship between MAC and OC/EC does NOT provide “important information about the extent of secondary organic aerosol (SOA) production.” See point 3 above. Also, there is no Figure 3b in Cheng et al., 2011. Do the authors mean to refer to their own figure in this manuscript?

14. P. 6237, Line 6: The authors state, without proof “the presence of brown carbon would not significantly influence the value of ATN, which is usually measured at wavelengths greater than 500nm.” Brown carbon may indeed absorb at wavelengths above 500 nm. When they say “greater than”, what do they mean? 501 nm? 900 nm? It matters. Also, the authors misunderstand the (potential) impact of brown carbon on the MAC. If brown carbon is not measured as EC, then any absorption at all by brown carbon for a fixed amount of EC will increase the MAC, not decrease it, unless of course brown carbon is identified as EC in the TO method. But if this is the case, then the EC measurement should not be used for this analysis in the first place.

15. I believe that in Equation 3, it should be ΔATN , not ATN.

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