Interactive comment on “An algorithm for retrieving black carbon optical parameters from thermal-optical (OC/EC) instruments” by A. Andersson et al.

Anonymous Referee #1

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1 Summary

This paper suggests a method for inferring absorption by black or “elemental” carbon from standard analyses that detect elemental carbon using heating and laser monitoring. This is an interesting idea and worth pursuing. The paper is well written and organized. However the analysis presented therein exhibits little understanding of either particle optics or the literature on OC/EC analysis. Methods are poorly described and insufficiently examined. I cannot recommend publication of this paper in its present form. Perhaps useful information could come from this investigation, but authors need to understand past literature more deeply and re-evaluate their analysis.
2 Major comments

This manuscript centers on the use of the thermal (Sunset) analyzer to infer both absorption and elemental carbon content. This must be done with a deep understanding of the biases in measurements of both quantities. Authors are either unaware of, or do not cite, the extensive literature describing artifacts in the detection of EC. Many of those artifacts relate to absorption and the expected changes in absorption that occur during heating. These are clearly discussed by Yang and Yu (EST, 36, 5199, 2002). The retention of organic carbon past the EC split point, which could affect the apparent optical properties of inferred EC, is discussed by Subramanian et al (AST, 40, 763, 2006). The differences between reflectance and transmission offer some insight into optics as discussed by Chow et al (EST, 38, 4414, 2004). None of this literature is discussed, and simply citing it will not fix this analysis. All these factors must be considered in the design and interpretation of the measurements.

Authors suggest that the Ram and Sarin comparison of attenuation and EC is the first such effort and that their own investigation of the laser signal is novel. This also betrays a lack of knowledge of past studies. Attenuation has been compared in quite a few studies but it’s often inconclusive and nobody has really relied on it. The point-by-point laser is discussed by Kirchstetter et al (Atmos Env 41, 1874, 2007) and Boparai et al (AST 42, 930, 2008).

3 Specific points

The paper contains several misconceptions discussed below.

p. 1237: “Light is attenuated not only by aerosol absorption, but also scattering” True in the atmosphere, but the statements here suggest that authors do not understand the principle of the integrating plate. Lin 1973 is cited here. I suggest that authors
should read and understand this principle and following work by Clarke regarding integrating plate. Scattering does affect the signal but its contribution to attenuation is much reduced.

p. 1238: “Laser transmission in the OCEC experiment can continue to increase even after the carbon signal has leveled off” What chemical compounds could contribute to this increase? According to the figure it occurs late in the analysis, therefore at a high temperature. Very few compounds absorb light, are stable until this temperature (750C? 900C?), and decompose at this temperature. In fact, I cannot think of any. Ammonium sulfate would have decomposed early in the analysis. Iron oxides are stable but remain on the filter even at the end of the analysis. Brown carbon would have been removed already either by oxidation or volatilization. I can think of one explanation: The optical signal in most analyzers (DRI, Sunset) is rather unstable and sometimes temperature dependent. It is quite possible that this entire investigation examines an artifact of the laser and not any contribution by other aerosol components.

p. 1239: Even though no physical explanation has been offered, the next investigation tries to separate attenuation by carbon and attenuation by the unknown species. This treatment assumes that the two substances could be separated. It does not address the question of whether both substances might evolve from the filter at the same time. How is this assumption justified?

p. 1239: I found this section especially difficult to read and understand. What is a “novel contribution to the transmission?” What does the “response function” tell the analyst? If transmission increases and carbon decreases, shouldn’t the response function be negative? What is a “false positive”? It implies that a “true positive” is being sought, and what is it?

p. 1241: I think authors have misinterpreted the shadowing effect. Although Wein-gartner discussed it for aged aerosols it seems to relate more to total absorption (and scattering) by aerosol on the filter.
p. 1241: “...can partially be controlled by ensuring that the sampling time/air flow rates match the ambient loadings.” I do not understand this at all. What has to be matched? A flow rate does not match a loading. Maybe authors mean that the filter shouldn’t be overloaded.

Inter-site comparisons: Why were these sites chosen? The number of samples for each site is not given. More should be said about this.

p. 1242-1243: The discussion of inter-site differences is not very satisfying (even if we could believe the measurements). Authors admit that no general conclusions are drawn. What are we supposed to do with the data? How are they useful? Most of the discussion emphasizes the large amount of variability between sites but doesn’t provide new insights. It doesn’t even provide confidence about data at any site.

Section 3.4 Comparisons with other filter based methods. The first 3 paragraphs here are not very useful. They are general statements about artifacts and considerations. They don’t help the reader understand how to interpret the results given in the paper. The final paragraph compares MAC given in this paper with MAC given by others, and then suggests that they are comparable if one chooses a new filter correction. This is uncompelling.

The concluding paragraph is overstated. It says that a method was developed and successfully applied. It appears that an algorithm was developed and it did produce numbers, but no criteria for success (understanding, accuracy) were set and none were met. This conclusion also says that novel insights were provided. I regret that I could not find any.