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Atmospheric Measurement Techniques Discussions

Interactive comment on "Towards the development of standard reference materials for soot measurements – Part 1: Tailored graphitized soot" *by* O. Popovicheva et al.

Anonymous Referee #2

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This manuscript describes techniques used to measure elemental carbon (EC) and organic carbon (OC) mass and absorption coefficient and uncertainties of these techniques, with a focus on the thermal-optical analysis method for OC and EC mass, and a focus on the filter-based and photoacoustic methods for absorption coefficient. It proposes the use of reference materials for reducing measurement uncertainties, and then describes the development of a standard reference material (SRM) and some of its chemical and physical properties. The authors indicate that a subsequent manuscript – currently in preparation – will present more data on how thermal-optical analysis methods can be calibrated with their SRM.

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Interactive Discussion
Discussion Paper

Without intending to minimize the careful work of the authors in preparing their SRM, I must say that I am not convinced that this manuscript should be published. I don't see much value in this manuscript without the proposed follow-on work. While this manuscript does describe well-enough their approach to coating soot with organics, this alone is not of great value, in my opinion. The excitement lies, as acknowledged by the authors, in using the SRM to reduce uncertainty in measurements of OC/EC mass and absorption coefficient, and ultimately uncertainty in our understanding of air pollution health impacts and climate change. This manuscript doesn't begin to take that step, and more importantly, I am not convinced that the SRM that the authors have developed will be able to reduce measurement uncertainty.

The main uncertainty in thermal-optical analysis of OC/EC lies in the complexity of ambient OC, which is refractory and prone to forming char when heated in the thermal-optical analyzer, as summarized by the manuscript and the studies cited. The OC/EC ratio of ambient particles ranges from approximately 50% to >95%. The OC is partly refractory and possibly light-absorbing, especially when the OC is secondary-organic aerosol or from biomass burning, and, therefore, it turns to EC-like char during thermal-optical analysis. Since the OC»EC in ambient particles, the EC-like char can be much greater in mass than the EC originally present in the particle sample, leading the large uncertainty in quantification of EC. That, in brief, is why it is difficult to measure EC. My trouble with this manuscript is that the authors do not make a convincing argument – they really don't explain it at all – exactly how their SRM will tackle the problems of the thermal-optical analysis method. Since their SRM has OC/EC ratios that are mostly <1% and has relatively simple OC composition that is similar to products of fossil-fuel combustion – rather than the more chemically complex, refractory, and prone to forming char OC of secondary-organic aerosol and biomass smoke – it is true that their SRM is unlike ambient carbon-containing aerosol in composition and it is likely true that their SRM will not challenge thermal-optical analysis methods as they need to be challenged to reduce measurement uncertainty. Their simplifying step of choosing off-the-shelf OC compounds is the main problem, in my opinion (and frankly, I'm not

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sure there is any way to get around this problem b/c it is the complexity of ambient OC that prevents us from putting it in a bottle and using it to coat reference soots). Second, the OC coverage is more than an order of magnitude lower than in ambient aerosols. The author's have chosen an important topic, but the major flaw of this manuscript is that the benefit of the developed SRM is, in my opinion, greatly overstated and certainly unsupported by its content.

In the end, it is not clear to me that the SRM will do more than help to strictly calibrate thermal-optical analysis instruments, i.e., confirm that they quantitatively count carbon atoms. The SRM is not needed for this b/c these instruments are already suitably calibrated with sucrose or glucose samples that are easily prepared in the laboratory.

It is not clear what is the form of the SRM: is it dry powder, dry powder on a filter, powder suspended in a solvent? Similarly, the manuscript does not explain how the SRM was applied to filters for thermal-optical analysis or suspended for size-distribution measurement, but it should clearly explain these.

The manuscript concludes with a statement that the next manuscript will discuss how the SRM will be used to calibrate the thermal-optical analysis instrument, but this has already been shown in the current manuscript, the results are shown in Figure 7. This current manuscript, in my opinion, should tackle the real issue – the difficult problem of separating correctly OC and EC in ambient samples (as already noted above).

A much more minor concern, since the manuscript really focuses on thermal-optical analysis, is that the manuscript doesn't apply the SRM to methods of measuring absorption. The discussion of these absorption methods in the introduction could, therefore, be deleted.

I found the Methodology section somewhat cumbersome to read. Perhaps partly b/c I was unable to easy follow the progression of terminology: technical carbon, GTS, ECRM, SRM, modifier, probe molecules, and partly b/c of the style: it seems like some of this section should be in the introduction (such as the first sentence and the entire

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second paragraph of the Methodology section).

I didn't find Figure 2 very helpful.

What is the proof for the statement made in the caption of Figure 4 – that the apparent smaller sizes are due to changes in particle diffusivity and shape?

Interactive comment on Atmos. Meas. Tech. Discuss., 3, 1743, 2010.