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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Determination of OM/OC ratios and specific attenuation coefficients (SAC) in ambient fine PM at a rural site in southern Ontario: implications for emission sources, particle aging, and radiative forcing" *by* T. W. Chan et al.

Anonymous Referee #1

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General comments

The manuscript presents carbonaceous aerosol measurements made by a filter-based thermal optical analysis method and an aerosol mass spectrometer at Egbert, Canada in 2005 and 2007. The paper focuses on interpreting the ratios of organic mass measured by the AMS to organic carbon measured on the filters. It also compares light absorption measurements with POC measured on the filters and found that the ab-

sorption efficiency appeared to decrease with increasing age as represented by POC content. The majority of the paper is well written and the methods and analysis method are described with an appropriate amount of detail. I believe the manuscript is suitable for publication in ACP, but I have two general areas of criticism with the manuscript.

First, there isn't a lot to support including the 2005 results with the 2007 data, other than completeness. The 2005 data is not discussed in as much detail as the 2007 data and the major conclusions of the manuscript don't really hinge on the inclusion of the 2005 data. If the 2005 data are really important, it would be nice to see a bigger discussion of the similarities and differences between the measurements during the two campaigns and what they might mean. At the moment the 2005 results are added almost as an afterthought, and I think the manuscript would benefit if they were given a bit more attention or otherwise omitted.

Second, in my opinion the case made for using POC as a proxy for aerosol age in this case is not strong enough to show a clear relationship between SAC and aerosol aging. As I am sure the authors are aware, POC determination in thermal optical analysis methods is not straight-forward, and relies on assumptions about the optical properties of the POC to accurately quantify it. I discuss this issue in more detail in my specific comments, but wanted to highlight it here, given the somewhat unusual (though physically plausible) observation of decreasing SAC with increasing aerosol age.

Specific comments

Page 14319, line 9: Is there a better word than "easier" to use here? Please state the specific characteristic you are describing here, e.g., is it simply that POC evolves in the first EC stage of the analysis?

Page 14319-14320: Might be helpful to have a table summarizing previous OM/OC measurements.

Page 14321, lines 1-2: Slightly confusing. Are the authors referring to light absorption measurements, or light absorption itself? In either case, it would be helpful to have a few sentences discussing how organic mass increases light absorption, either via mixing or addition of light absorbing material.

Page 14322, lines 4-6: A map showing the location of the site and major emission sources nearby would be useful.

Page 14325, lines 1-9: I have a minor issue with the assertion that just because CE of 1 was found using the same instrument in the other studies cited by the authors, it is valid to apply a CE of 1 in this study. The manuscript implies that ammonium sulphate mass fractions were < 50% during the 2005 study. Were they the same in 2007? Do the recent Matthew et al. (2008) and Crosier et al. (2007) methods for estimating CE support a CE of one? Finally, it would be useful to have a time series of the CE applied in the 2007 in an appendix or supplementary data somewhere. Why was the CE applied to filter samples?

Page 14326, line 11: It would be useful to have the wind data shown either in the main manuscript or in the appendix. The authors mention back trajectories, but do not show the results. They may wish to consider examining air mass sources using some kind of backward trajectory statistical analysis approach such as that employed by Ashbaugh et al., A residence time probability analysis of sulfur concentrations at Grand Canyon National Park, Atmospheric Environment 19, 1263-1270, 1985.

Page 14326, line 22: Was there any reason for the choice of 40% as the selection criteria?

Page 14332, line 7: If direct comparisons between OM/OCtot values measured by different AMS instruments are not meaningful, what does this mean for the applicability of the results on a broader context? How can they make the comparisons to other studies in the following paragraphs if they can't do so for their own measurements? This statement apparently undercuts their entire approach.

Page 14332, line 25: It would be more convincing if they compared POC with the independent oxygen mass measurements from the AMS rather than with the OM-OCtot results that depends partly on accurate determination of POC.

Page 14333, lines 1-3: I think the authors should add a caveat stating that their parameters may not be valid for POC measured by different OC/EC protocols (e.g., NIOSH 5040, IMPROVE_A) that may have different sensitivities to OM oxygen content.

Page 14334, line 11: Why not use the toluene-benzene relationship described previously? If POC is determined incorrectly (e.g., the method over- or under-estimates the actual amount of pyrolysis occurring during the analysis), then the EC measurement would be off by the same magnitude, affecting the SAC determination. Samples with high POC may simply represent an underestimate of EC and corresponding overestimate of SAC. If the behavior of SAC really depends on air mass aging, than we should see some kind of relationship with the toluene-benzene ratio if the absorption and EC measurements are truly accurate. This could explain the negative relationship between POC and SAC and is worth including in the discussion that follows. If nothing else the authors could show the relative mass fractions of EC and POC and give an estimate of the sensitivity of the SAC calculation to the accuracy of the POC measurement. If POC is a tiny fraction of EC than I would have a lot more faith in the use of POC as a proxy for aging. Finally, the argument could be better supported by other estimates of the air mass age, such as using the backward trajectory travel time between likely source regions and the measurements or examining the mz43 and mz44 relationships to total organics in the AMS data.

Page 14336, lines 18-21: Agreed, but the results here hinge on POC's ability to serve as a proxy for aerosol aging, which I don't think has been established very well.

Figures

Figure 3: Remove arrows.

Figures 7 and 8: Legend is a little confusing. I assume the points are scaled by concentration, but the legend makes it seem like they are grouped into three concentrations groups. If they are scaled I think it is better to just show two examples of the symbol size at each concentration.

Technical corrections

Page 14322, line 7: change “regions of the south” to “regions to the south”

Page 14322, lines 12 and 24: change “last” to “lasted”

Page 14323, line 5: delete “as”

Page 14333, line 19: poor wording, “The magnitude of SAC reflects the ability of absorbing light by EC.”

Page 14336, line 6: missing “et al.” in the reference

Page 14336, line 12: change “hypothetic” to “hypothetical”

Page 14336, line 17: see above

Page 14337, line 11: a nitpick, but change “[OM-OC]” to “(OM-OC)”

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 14315, 2009.