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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Characterization of a large biogenic secondary organic aerosol event from eastern Canadian forests" *by* J. G. Slowik et al.

Anonymous Referee #1

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This paper reports unique observations on the formation of secondary organic aerosol apparently from biogenic VOCs. The authors have presented an extensive analysis based on measurements, modeling, and satellite data. A number of broad questions may need more discussion or clarification. These relate to:

1)How regionally important are the biogenic SOA events

2)Is the model really doing a reasonable job if it only predicts the magnitude of the aerosol produced when averaged over a significant time period, but cannot predict with much skill the temporal evolution of the event or diurnal tends.

3)The extensive use of PMF analysis on AMS spectra in the data interpretation even

though specific factors used to identify sources or aerosol properties are shown to have very weak correlations with independent tracer data.

General Comments:

It is claimed that high T (and the resulting enhanced BVOC emissions) likely produced the observed biogenic SOA event that produced 15 to 20 ug per m3 of organic aerosol. It is also asserted that this process is likely a large source for aerosol over the whole region, even in more urban locations to the south (see Conclusions). However, during the duration of the study period (roughly 5/12 to 6/15) there were many periods of similar high T, yet throughout this whole period there was no other evidence for large biogenic SOA contributions at the measurement site. This seems odd if the process really is generally widespread and of sufficient range to significantly impact urban areas. Thus more than T is important. Is the argument that most of the biogenic SOA is occurring significantly to the north of the site, and it is only occasionally transported south? I think it would also be of interest to know if the model agrees with the observations and predicts less of a biogenic SOA influence at that site during the whole study (not just select number of days, as shown in Fig 7)?

In the Abstract and Conclusions it is stated that a regional model approximately predicts the event timing and accurately predicts the aerosol loading. This is summarized in Fig 9. Given the very different shapes in the temporal profiles of the model prediction and observations (Fig 9), is it meaningful to claim that the model is doing a good job in predicting SOA mass? For example, peak SOA is observed slightly after midday on 6/13/2007 and peak predicted SOA occurs very early in the morning (looks like near sunrise) on 6/14/2007. (It would be interesting to see more of the temporal profile past 1200 am 6/15/2007 where model and observations appear to significantly diverge). Thus in essence, averaged over some significant time interval (say 24hrs) the magnitudes agree. All the diurnal properties or processes are then lost and maybe the agreement is just fortuitous. For example, in Fig 9, the OOA tends to have daily peaks from noon to late afternoon on 6/12, 6/13, and 6/14, as expected since SOA formation

is strongly diurnal. The model shows none of this structure.

Some Specific Comments:

Pg 18118 line 7, give uncertainty in Eb (0.6 plus/minus what)? I suspect there is some level of uncertainty in the SMPS measurement, converting an SMPS number distribution to volume, and then using measured composition to convert to mass. Also, does calculating the AMS average collection efficiency for one period really prove that the method was quantitative throughout the whole study for all the different aerosol chemical components? This is a critical question since the authors claim that the measured OOA agrees with the predicted SOA from monoterpenes. Some estimate of the measurement uncertainty should be given when these types of statements are made.

Pg 18118 line 25, it is not clear how the difference in the upper size limits of SMPS and AMS are reconciled for the comparison.

A significant portion of this paper is devoted to AMS spectra interpretation by factor analysis. A problem with factor analysis is over interpretation – something that this analysis is in my view approaching. In order to provide some confidence in the analysis the factors are compared to various independent tracers, however, often the correlations are really very poor. Eg., Pg 18126 lines 15 and on, the correlations between F1 (HOA) and NOx, and F1 and Benzene are low (less than 50% of the variation can be explained), moreover, F2 (BBOA) correlations (r2) with K, levoglucosan (both inferred from AMS data), and independently measured acetonitrile, etc are very low ($\sim$ 0.3) – any insights why? Is this typical? Based on this data, labeling these as anthropogenic or biomass burning seems questionable. This raises concern over how well PMF uniquely identifies the OOA-1 and OOA-2 parameters. I think it might be worth discussing if the main conclusions reached in this study are sensitive to the PMF analysis results. My impression is that they are not.

Page 18128, lines 6-8. Can it really be stated that a large fraction of OOA-2 is formed within half a day give that: MVK+MACR to OOA-2 r2 is roughly 0.6 and the MVK+MACR

vs OOA-1 (aged OA according to this paper) r2 is 0.2. I would characterize it as: some fraction, larger than that found in OOA-1, is possibly associated with freshly formed aerosol.

Page 18128, line 11 – don't know what is meant by dashed backgrounds of Fig 4, are they missing?

Page 18129, lines 13, 14, What is the variability given for OOA-2 fractions (eg, 65 percent plus minus 10 percent). Is this the standard deviation?

Page 18133 lines 15 -20 and Fig 9. The authors do make convincing arguments that the predicted SOA from monoterpenes is in reasonable agreement with observations (ignoring for the moment that the timing of the event is off). But does that really prove that it is correct? I would use the word consistent. In my view this leads to overreaching arguments later in the paper regarding possible influence of biomass burning. The authors believe that because the predicted SOA is of similar magnitude as observed OOA that proves no biomass burning influence. Given the uncertainty in SOA yields, AMS measurement uncertainty, PMF analysis, etc this argument should be qualified.

Page 18136, line 4. Given the ranges in AOD shown in Fig 9, it could be argued that the AOD data do not show the large increase in SOA (one could almost draw a flat line through the AOD vs time data.

Page 18136, Line 10; it takes some imagination to infer from this graph that areas to the north were also from biogenic SOA. Maybe the scale should be changed to focus more on the region of interest.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 18113, 2009.