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

Anonymous Referee #2

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The paper describes observations of organic aerosol and precursor VOCs at a rural site in Egbert, Ontario. The authors focus on one event, when they observed a strong correlation between organic aerosol and carbon monoxide, which they attribute to secondary formation of both species from biogenic VOCs. The organic aerosol and CO during the analyzed event were very high: the mass loadings of organic aerosol rose above 15 $\mu\text{g}/\text{m}^3$ and the mixing ratios of CO close to 200 ppbv. Ordinarily such high concentrations are only observed in fairly polluted conditions, but the authors argue -and I cannot fault with their arguments- that that was not the case here. If SOA and CO from biogenic VOCs can be so large, however, it begs the question why others have not made similar observations. Unfortunately, the authors never really address

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that question. They attribute the timing of the event on the temperature dependence of biogenic emissions, but surely there are warmer regions of the atmosphere than Ontario and Quebec. The manuscript is certainly interesting, careful and thought provoking, and should be published after the significant revisions outlined below:

Page 18122, line 27: I guess you mean SESQ emissions were a factor of 0.16 times the monoterpene emissions? The sentence is ambiguous as is.

Page 18123, line 25: This is a different density than mentioned above for the conversion of mass loading into volume (page 18118, line 24). Why?

Page 18125, line: I suggest using “evaluated” instead of “validated”.

Page 18125, lines 5-13: I was confused about this. You mentioned that panel d has significantly less structure than panels a-c, so from the legend I would conclude that going from 4 to 5 factors still improves the quality of the PMF fit. However, the text mentions that 4 is the optimal number.

Page 18127, line 6: Eliminate “with”.

Page 18128, lines 11-13: I cannot see the dashed periods in Fig. 4.

Page 18130: lines 8-11: deCarlo (ACP 2008) observed OA/CO ratios in biomass burning plumes that were much higher than mentioned here. I believe that the variability in biomass burning emissions is too high to rule out a biomass burning origin based on the OA/CO slope.

Page 18131, lines 13-16: Hudman et al. (GRL 2008) calculated secondary CO mixing ratios of 25 ppbv or higher. Hudman et al. used biogenic emissions from MEGAN, which are significantly higher in the eastern U.S. than the emissions according to BEIS used in AURAMS. I suspect that differences in emissions inventories may explain why earlier estimates of secondary CO gave lower mixing ratios (Granier et al., Atmos. Environ. 2000; Kanakidou and Crutzen, Chemosphere 1999). Some discussion of these uncertainties is warranted.

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Page 18131, lines 16-18: The paper by Miller et al. (ACP 2008) should be cited and discussed in this regard.

Page 18131, lines 19-22: Monoterpenes may be the dominant precursor of SOA in this region, but why are they assumed to be the dominant precursor of CO in this analysis? According to Hudman et al. (GRL 2008) and Granier et al. (Atmos. Environ. 2000), isoprene is a much more important precursor of CO than monoterpenes. Assuming that a significant fraction of the CO during the biogenic period came from isoprene oxidation, the analysis would have to explain a much higher SOA/CO ratio from monoterpenes than it currently does in order to be consistent with the measurements.

Page 18132, lines 8-9: A NO_x level of 1 ppbv would actually point to non-negligible anthropogenic or biomass burning emissions, in contrast with the designation of these air masses. I suspect such mixing ratios may be close to the detection limit of the TECO instrument? Some discussion of this is needed in section 2.2.3.

Page 18132, lines 10-14: I guess the $\Delta(\text{CO})/\Delta(\text{alpha-pinene})$ ratio represents the CO formed divided by the alpha-pinene removed? I was expecting this ratio to indicate the instantaneous enhancements as a function of time, in which case I expected this ratio to go to infinity as alpha-pinene gets removed within hours.

Page 18132, lines 15-24: Ng et al. (ACP 2007) determined yields for alpha-pinene in high-NO_x and low-NO_x conditions and this work should be cited here as well. It might also be useful to convert the NO_x emission rate in Fig. 6 to NO_x mixing ratios.

Page 18134, lines 16-18: As argued above, I do not believe that this argument is very convincing: deCarlo et al. (ACP 2008) observed similarly high OA/CO ratios in biomass burning plumes.

Page 18135, lines 19-26: I disagree with that argument. Capes et al. (ACP 2008) showed a tight correlation between OA and CO at various distances from the fire location. Photochemistry changed the particle composition measured by AMS, but did not

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enhance the OA mass loadings relative to CO.

Page 18136, lines 2-15: I am not convinced that the MODIS AOD in Figs. 9, 10a and 10b is unaffected by biomass burning. The authors argue in section 3.3.3 that the enhanced OA measured at the surface was not from biomass burning, and I do not disagree with that analysis. However, the aerosols that determine the AOD can also be in the free troposphere. The authors show the observed fire counts in Fig. 10a, but transport of biomass burning plumes over much larger distances is routinely observed. For example, Fig. 10b averages data from 2001 through 2006, which period includes 2004 when fires in Alaska very significantly affected eastern Canada and the U.S. in June and July (e.g. Pfister et al., GRL 2005; McMillan et al., JGR 2008). The authors must do much more to rule out that their analysis of AOD is not affected by biomass burning.

Page 18136, lines 16-24: The seasonal variation in AOD peaks in June (Fig. 10b). Average temperatures must be higher in July and August, but the average AOD in August is almost half of what it is in June. Thus far, the authors have interpreted their observations solely in terms of the temperature dependence of biogenic emissions, but there must be other factors.

Page 18136, line 25 – Page 18137, line 17: The authors calculate the forcing due to biogenic SOA and the CO₂ formed from biogenic VOCs. This flux of CO₂ is likely inconsequential compared to any imbalance between carbon uptake and respiration. Also, this would not constitute a climate forcing since it is a natural source of aerosol and CO₂. I believe this is the weakest part of the manuscript and is also unnecessary.

Figure 2: The labeling should be changed. The caption identifies the top four panels with mass spectra as panel a and the bottom four panels with time series as panel b. However, the four mass spectra are also labeled a through d.

Figure 7: The caption mentions that SOA is plotted, but the color scale gives OC. Which is right? OA, of course, is typically a factor of 1.6-1.8 times OC.

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