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Interactive comment on “Observations and analysis of organic aerosol evolution in some prescribed fire smoke plumes” by A. A. May et al.

Anonymous Referee #1

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Overview: I find this manuscript to be well written and logically organized. The manuscript describes particle chemical compositional changes measured downwind of two prescribed fire smoke plumes in SE US over 1.5 and 5 hours, respectively. As these measurements are difficult to obtain and there are very few such measurements reported in current literature, this manuscript is both timely and appropriate material for ACP. The manuscript should be published with attention paid to the following minor issues.

- 1.) IE's given have units of “ions/molecule”.
- 2.) The paragraph about using no gas-phase correction in the AMS data analysis for CO₂ is incorrect and misleading. It needs to be removed or rewritten. The standard

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Discussion Paper



AMS analysis directly incorporates a standard CO₂ correction, as it is an important correction, in the fragmentation tables. Please see Allan et al., 2004. It is possible that the intent of the authors was to suggest that differences in gas phase CO₂ concentrations in and out of plume were insignificant, or that the constant pressure inlet reduced gas phase CO₂ concentrations below relevant signal levels, both of which might be true. However, since the authors present total OA, f₄₄, and O:C measurements, all of which can be dramatically impacted by incorrect gas-phase CO₂ corrections, the authors need to clear this issue up.

3.) In order to limit the size of this manuscript, Section 2.2.4 should be removed, unless the data is directly used in the manuscript, which I cannot seem to find.

4.) In addition to (3) above, the discussion of Lagrangian or non-Lagrangian could be removed, as again while the data points are duly marked and the description and intent is clear, the differences in L or non-L data points appear to be never discussed or utilized in any way to suggest the differences are important. If the authors' decide to keep this in, then it would be important to at least describe how they differ or why they do not differ.

5.) One must assume that the significance level assumed in the manuscript for the statistical tests is 0.05. It should be included.

6.) Page 1968 line 22 appears to have the incorrect trend stated, which should read “f₆₀ (Fig. 4c) is significantly lower downwind than at the source. . .”

7.) Page 1970 line 10 “decrease” should be “increase”.

8.) The discussion of O:C and H:C in the same paragraph ends by comparing the trends in changing O:C and H:C downwind with the same trends that were demonstrated for thermal denuded OA. What is left unstated is that these same trends are also true for SOA formation. Can refer to Kroll et al., 2011. Thus, by chemical changes alone, this connection is a bit misleading. It needs to be paired with the decreasing OA

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loadings to suggest that dilution/evaporation may dominate. Part of the issue here is that the H:C typically goes down with increasing oxidation, as more H's are lost than C's, an explanation that was not included by the authors when attempting to describe why the H:C does not go up with dilution of higher H:C background aerosol.

9.) Given the decrease in measured OA over the short time frames, it is definitely tempting to implicate dilution/evaporation over photochemical oxidation, though photochemical oxidation processes may also reduce the amount of OA in time. However, as the authors' note, it is not necessarily clear how much of a role photochemical oxidation may have affected the observations. The authors give due time and effort to model the OA measurements as if dilution/evaporation was the only significant process in section 3.2. However, previous work by (some of) these authors reported observations of photochemical activity for these same biomass burning plumes (page 1967 lines 4-6). Why did the authors not try to at least quantify potential photochemical oxidation effects, especially if they might be predicted to be small?

10.) Page 1972 line 14. "with decreasing plume-integrated COA" should probably read "with decreasing total measured (not background subtracted) COA".

11.) Page 1973 lines 21-24. The authors switch from the discussion of how dilution/evaporation may dominate the biomass burning particle processes during downwind advection and appear to make a more concrete conclusion here and only here in the conclusions, suggesting that not only does the dilution-driven evaporation dominate over photochemical oxidation, but it happens in the first hour, after which the "OA in the plume reaches an equilibrium state with the background in our observations." It is not clear where this additional information is presented in the results and discussion sections. On page 1967 at the end of section 3.0, the authors note that after 1.5 h from emission, no statistically-significant detectable change was observed in NEMR_OA for either of the two downwind burns. The authors' should clearly discuss this statement, the associated uncertainties, and the underlying assumptions prior to the conclusions section.

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 1953, 2015.

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