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Interactive comment on “Properties and evolution of biomass burning organic aerosol from Canadian boreal forest fires” by M. D. Jolley et al.

Anonymous Referee #1

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This paper reports on measurements of organic aerosol (OA) made in biomass burning plumes during the 2011 BORTAS campaign. The authors find that the average $\Delta\text{OA}/\Delta\text{CO}$ was higher in the three transects that sampled fresh smoke than in the aged smoke. The authors posit that this is due to the loss of OA with aging of the smoke, which seems true for comparisons between the three fresh smoke transects (see page 25112) and for comparisons between the ages smoke transects (see page 25104). However, a direct comparison between the fresh and aged smoke is complicated by the fact that the aged smoke came from more flaming combustion than the fresh smoke samples, and so the apparent decrease in OA could be due to just this change in combustion phase. The authors also find that the ratio of f44 (a tracer of oxygenated OA) to f60 (a tracer of levoglucosan OA) generally increase with plume age

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as expected, but only when the fresh and aged smoke transects are considered separately. When the fresh and aged smoke samples are compared, while the f44 value of the aged plumes is higher than the fresh plumes as expected, the f60 value is also higher in the aged plumes, more likely due to the change in combustion phase and characteristics than due to aging which would be expected to steadily reduce f60.

The paper provides new data on boreal biomass burning from a significant field campaign. While the methods and concepts are not new, the analysis appears to have been well done, although the discussion of the analysis and conclusions can use some revision for clarity and accuracy. Overall, this is a solid piece of work and deserves publication after minor revision to address my detailed comments below.

Minor Comments:

P25096, L8-9: I would follow the discussion of the near and far-field ratios with something like “However, the changes in combustion conditions between the near- and far-field samples (smoldering for the near-field, flaming for the far-field) make it unclear whether the apparent decrease is due to aging or the change in combustion efficiency.” Right now, it sounds like you are saying it is clearly due to aging, and that the changing combustion conditions are a minor effect, and I don’t think you have proven that.

P25096, L18-20: I don’t think I understand this sentence. F44 is lower in the fresh smoke than in the aged according to Figure 7, right? If so, I think this sentence is potentially misleading. Do you mean that you would have expected more formation in the far-field plumes than you observed?

P25096, L26-27, and P25097, L1-2. Again, I don’t think you can state that the effect you observed was primarily due to aging and not due to combustion phase given the evidence you present. You need to put both effects on more equal footing in your discussion.

L25097, L13: I think you want the word “emissions” after “(BBOA)”

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L25097, L14-16: This sentence doesn't really say much that I relevant to the paper, so I'd suggest cutting it.

P25097, L21: There are several places you use this "influence of" phrasing, but it sounds awkward to me. Here, you don't mean the influence of SOA is unclear, you mean the rate of formation and loss of OA is unclear, or the importance of SOA is unclear, right?

P25098, L27: I don't see why Figure S1 and S2 are not in the main paper rather than the supplement. They seem pretty important to understanding the conclusions of the paper, so I'd recommend putting them in the main text. More discussion of the source locations and transport pathways o the plumes, including any potential vertical motion during transport, would also be useful.

P25100, L24: I'd say "this size range" rather than "the full size range" – the second makes me think you mean the full size range of the aerosol distribution, not just the part measured by the SMPS.

P25101, L2: Do you mean the excess particle number concentrations as well, or the absolute values?

P25101, L10-12: I think these two sentences would fit better after L2, when the CO and number conc. thresholds have been introduced.

P25101 L16-25: This section was confusing to read. The beginning of this section suggested that your proposed indicators were already well established, but here you are presenting evidence of how they correlate with CH3CN and HCN. If this comparison is a key result o your paper, it should be in the results section, but if the indicators you are using have been used successfully in the past, it's not clear why you need this additional evaluation.

P25102, L12-13: Consider changing to "at the source to up to 5 days after emission."

P25103, L7-8: Concentrations of what in aged plumes? OA? CO? Both?

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P25103, L11: I get 180/50, not a factor of four, from Figure 1a. Either the text is wrong or the figure is cut off and should be fixed.

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P25103, L15: CO is also associated with moldering combustion.

P25103, L27: I'd suggest "analyzed" instead of "relevant" here.

P25104, L2: Is this a typo? How can the campaign average be less than both the fresh and aged samples?

P25104, L22-24: I think you can cut this sentence.

P25104, L25-28: I think you want to add "in the aged plumes" after progressively and end the sentence with ", suggestive of OA losses during aging in these plumes with predominantly flaming smoke."

P25105, Section 3.2: Why don't you discuss the modified combustion efficiency (MCE) of the plumes when you discuss combustion conditions? That should give you a pretty god indication of the change in combustion phase.

P25105, L9-11: You need to make clear here that f44 isn't exclusively produced during smoke aging, but that fresh some also has a significant amount of f44 as well.

P25108, L12: Again, why don't you use MCE here?

P25108, L25: I think you switched your numbers here?

P25109, L5: low CO₂ relative to what?

P25109, L16: I can't see any correlation with f44, but I can see it with f60.

P25109, L24-26: This is true, but almost trivially so. Aren't you just saying that aging is more important for the aged smoke?

P25111, L2: I'd add "during BORTAS" after "plumes" here.

P25111, L16: Missing delta before BC.

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P25111, L19-22: Didn't you say on page 25110 that this boundary layer enhancement was due to biogenics? Doesn't your statement here conflict with that?

P25113, L21-24: I don't think this sentence adds anything, so I'd cut it.

P25114, L14: Could this be due to secondary production of aldehydes and ketones in the smoke plumes?

P25117, L22: I'd say a range of "ages and combustion phases" is more accurate.

P25118, L1: This is not a fair summary of the literature. You do need to point out the many studies that have found significant OA formation in biomass burning plumes here as well.

P25188, L5-7: This last sentence is more of a generically true statement about fires than a conclusion of your paper, so I would cut it.

P25118, L20: I'd say "near the source", not "at the source" here.

P25118, L22: I'd add "near the source" at the end of this sentence on photochemical age.

P25118, L24: I'd say "these aging BB plumes" to make clear again that other have gotten different results.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 14, 25095, 2014.

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