Review of acp-2014-720: Aged boreal biomass burning aerosol size distributions from BORTAS 2011 by Sakamoto et al.

General comments:

This discussion paper presents aerosol size distributions originating from biomass burning that have undergone > 1000 km of atmospheric transport. As the authors note in their introduction, understanding the evolution of biomass burning related aerosol size distributions is critical to improve global models used to simulate aerosol effects on climate.

In this reviewer's opinion, the results and discussion sections are very well-written and are in need of minimal improvement. However, I do have numerous suggestions to improve the introduction and methods sections to yield a finalized paper of higher quality. Once these comments are addressed, I recommend this discussion paper for publication in ACP.

Specific comments:

Page 24351, line 27 through page 24352, line 3: Engelhart et al. (2012) investigated CCN activity of fresh and aged BB aerosol during the FLAME-III campaign and found that the κ value for fresh aerosol was occasionally greater than the κ value of aged aerosol.

Page 24352, lines 21-22: I am not so sure that the Hennigan et al. (2011) is the best reference for discussion of the evaporation of semivolatile POA, since this work focuses on photochemical aging. In this reviewer's opinion, there are other references that are more appropriate (Huffman et al., 2009; May et al., 2013).

Page 24353, lines 8-11: Being familiar with the work of McMeeking et al. (2009) and Hennigan et al. (2011), I cannot recall in either of these papers where they discuss size distributions as a function of plume age, combustion phase, or fuel type. I would suggest the following papers as a replacement: (Adler et al., 2011; Capes et al., 2008; Hobbs et al., 2003).

Page 24353, lines 14-15: To the best of my knowledge, the work of Levin et al. (2010) did not utilize a smog chamber, and any aging during this study was due to microphysical processes occurring within the large combustion laboratory at the Fire Sciences Laboratory.

Page 24355, lines 7-8: Do the authors mean that the samples collected on these flights were primarily comprised of biomass burning influenced aerosol? Please re-write for clarity.

Page 24355, line 27-remainder of paragraph: This paragraph is important to describe plume age. However, in this reviewer's opinion, the writing could use some streamlining. For example, the last sentence in the paragraph could be clarified as "The estimated photochemical age of the plumes was calculated by Palmer et al. (2013) to be 1-5 days for b622 and 2-4 days for b623 based on non-methane hydrocarbon analysis (Parrish et al., 2007); these estimated ages may be longer than the backtrajectory estimates due to the influence of background air mixing into the plume."

Page 24356, lines 25-27: Unless I am mistaken, Jolleys et al. (2014) also report AMS data from the Manchester group. This discussion paper appears to have been published online after the authors submitted their paper.

Page 24357, line 19 through page 24358, line 6: Are these thresholds completely arbitrary? The authors establish criteria that plume intercepts are 1.5x greater than background for CO, 2x greater for CH_3CN, 2.5x greater for BC number, and 10x greater for OA mass. Is there a reason for this widespread inconsistency? How would analysis change if everything was set to 5x greater?

Page 24358, lines 16-17: While the authors make a very good argument in Section 3.2 regarding no net evaporation/condensation within their observations, I am struggling to grasp the claim of no evaporation/condensation near the source. For example, Yokelson et al. (2009) observed an increase in the OA emission ratio within the first 1.5 hours of aging, while Akagi et al. (2012) observed a decrease in the OA emission ratio within the first 1.5 hours of aging. In both cases, the emission ratios did appear to reach a steady-state value, which would be consistent with the authors claims in this work. Similarly, for boreal fires, Hecobian et al. (2011) and Cubison et al. (2011) report no net change to OA emission ratios with increases in photochemical age. Perhaps the authors should reframe their argument for no net evaporation/condensation based on the literature summarized here. Further, it may be more appropriate to define their young aerosol size distributions as ~3 hr (or longer), given that net evaporation/condensation is non-zero for shorter timescales in the literature, rather than extrapolating to a value closer to the source and introducing significant uncertainties due to the exclusion of evaporation and condensation.

Page 24358, lines 18: This reviewer would argue that 1 hour should not be considered "fresh", but rather "young", as many microphysical and chemical processes could influence the size distribution during this time. Please consider revising the wording here, and elsewhere in the paper, as appropriate.

Page 24359, lines 6-10: Here, the authors are referring to the dilution timescale within their measurement constraints, correct? Presumably a dilution timescale near the source on the order of 1 hr would appear to be much slower at these distances downwind. Given the vector wind velocities provided in Palmer et al. (2013), the reported timescales seem long for wind speeds on the order of 15 m s^{-1}. This may tie back into the initialization time comment above and may be more appropriately referred to as "apparent dilution timescale", which is only defined over the period after which no net evaporation/condensation occurs.

Page 24363, lines 5-7: What is a reasonable entrainment timescale? Are these typically < 5 hours?

Figure 7: Typically when emission ratios are calculated from scatter plots using ΔOA and ΔCO, the intercept is forced through zero. I would suggest taking this approach here to provide emission ratios that are consistent with the rest of the literature. The values reported in the legend may be of interest to researchers interested in aged emission ratios from boreal fires.

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