

We'd like to thank the reviewers for their time and insights. Our revised paper has benefited from their critiques. Our responses to individual comments are below:

Reviewer #2 -

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.

The line has been updated to acknowledge the kappa sensitivity to age is dependent on fuel type, "...though this range may be slightly larger or smaller for fresh biomass-burning particles due to these particles being initially more hydrophobic/hydrophilic (depending on fuel type) than typical ambient aerosol (Carrico et al., 2010; Engelhart et al., 2012; Petters and Kreidenweis, 2007)"

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 semi-volatile 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.

These references have all been updated to reflect these works.

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.

This has been changed to: "...[the two flights] also contained the majority of the biomass-burning aerosol sampled during the 14-flight campaign." These flights contain the most concentrated and longest duration of BB sampling during BORTAS.

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."

The ending of the paragraph in question has been simplified to: "The estimated photochemical age of the plumes, calculated by Palmer et al. (2013) (by non-methane hydrocarbon analysis; Parrish et al. (2007)), were 1-5 days for b622 and 2-4 days for b623. These estimates may be longer than the physical transport ages due to the entrainment of background air (which is more photochemically aged) into the plumes."

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.

This reference has been added to the list of papers discussing the Manchester group AMS BORTAS data.

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 CH₃CN, 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?

The thresholds were chosen subjectively (i.e. we did not choose something like 2 standard deviations above background based on the background variability), but it was not completely arbitrary as we needed to increase the thresholds for the particles so we could exclude the high-altitude plumes that had undergone particle wet deposition. The CO and acetonitrile thresholds are capable of excluding the most data (since their peak values are relatively closer to their background values), while given that this paper concentrates on particle characteristics, the OA and BC thresholds are designed to exclude those sampling periods with few particles (e.g. severe rainout events where particle concentration is abnormally low). The 1.5x and 2x thresholds for CO and CH₃CN are sufficiently high to ensure in-plume sampling without excluding significant amounts of suspected BB data. The OA and BC thresholds are even higher since the plume is so concentrated relative to their respective backgrounds, and the upper plume still contained OA and BC above background yet we didn't want to include this upper plume. Changing the thresholds of OA or BC within (2x-15x) or (1.5x-3x) respectively does not significantly change the amount of lower-altitude-plume data.

We have added the following text to the discussion of the thresholds, “These thresholds for particles are higher relative to background than CO and CH₃CN because we wanted to exclude a higher-elevation plume that had undergone aerosol wet deposition (will be described later).”

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.

Our work shows that the OA emission ratio is unchanging (or below significance) across the observed times in the plume evolution (roughly 1-2 days). We agree that we have no evidence of early plume processing. This has been clarified in Page 24358, lines 18-19, in the abstract, and the conclusions, to emphasize that our lack of ER trend doesn't preclude OA prod/loss earlier or later in the aging process. The use of the coagulation-only model has been re-framed to modeling 'young'-plume size distributions with a discussion on the uncertainties associated with condensational growth in the early aging process.

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.

This recommendation has been followed, with 'young' plumes being defined as ~ 1-3 hrs old. From Results 3.2: “The plume size distributions modeled here are very sensitive to microphysical processes directly after emission. Very close to the source, rapid dilution and condensation (due to cooling) may occur, which are not captured by the coagulation/dilution model we have developed. Thus the modeled plumes are better categorized as 'young' rather than freshly emitted.”

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.

The reviewer is correct that the dilution timescale of plumes is shorter when the plume is young and narrow when it takes less time to mix background air into the plume. Our estimates represent the mean timescale over the first 48 hours of Gaussian plume expansion calculated for near neutral stability classes. We have edited a sentence in the text: “The 36 hr dilution timescale was calculated as the mean timescale for dilution from Gaussian plume equations with an initial plume width of 10 km in a neutral stability environment (Klug, 1969) (note, however, that expansion occurs at faster timescales early in the plume, and this timescale slows with time).”

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

Entrainment and dilution for young/fresh plumes is rapid (possibly < 5 hours), but this depends greatly on the initial width of the plume, which varies greatly for wildfires. An appropriate mean entrainment timescale to this work, given the very high concentrations within the BB plumes at these ages, is most likely much greater than 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.

Figures 7b and 8 have been updated with enhancement ratio intercepts forced through zero. The enhancement ratios now span from 0.09-0.17 $\mu\text{g m}^{-3} \text{ ppb}^{-1}$.