

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 #1 -

1. Even though the primary particle size distributions derived (e.g., Fig. 9a) in this analysis compare with prior studies, it is very likely that chemical processes also had a significant influence on the evolution of the particle size distributions. I do not think one can rule out condensation in the first hours/day of transport simply because net OA formation/evaporation was not observed far downwind of the fires (see comment 2 below, as well). For example, Reid et al. (1998) report for a biomass burning study in the Amazon that “Over a period of 1 to 4 days, coagulation and condensation/gastro-particle conversion probably contributed about equally to the increase in particle sizes”. In the present study, the comparison to prior studies seems to benefit from both the variability observed in prior studies and the variability resulting from differences in the dilution timescale. It is important to at least acknowledge that secondary aerosol formation during the first day of transport also likely influenced the evolution of the size distribution. Some discussion of the uncertainty that condensational processes have on the results is also warranted (e.g., if significant condensation of secondary aerosol species occurred in the first 1-12 hours of transport).

We agree. We've now emphasized the significance of condensational growth during the first hours of aging (when VOC concentrations are highest) by re-framing the discussion of the box model as 'young' plume distributions instead of 'fresh' plume distributions (as suggested by Reviewer #2). The resulting modelling conclusions now detail: "There is therefore an associated uncertainty in the young distributions due to the exclusion of condensational growth from the model, despite evidence of its effects on BB particle sizes especially during the first hours of aging (e.g. Reid et al. 1998)".

2. Since the biomass burning plumes were sampled 1000-1500 km downwind of their source, the conclusion that the $\Delta\text{OA}/\Delta\text{CO}$ ratio did not increase with distance from the fire is not surprising. Significant aging time and chemical processing had likely occurred before encountering the plumes, and SOA formation can occur quite rapidly in fire plumes (e.g., Yokelson et al., 2009). The statement on pg. 24357, line 10-11 implies that this determination has been made for the entire plume transport period –it is important to clarify that this is only applicable to the plume encounters > 1000 km from the source. Thus, I think it is also important to acknowledge that the observations do not preclude OA formation or evaporation happening in the first day of transport when hydroxyl radical and precursor VOC concentrations are likely at their highest levels (e.g., abstract, lines 17-18; pg. 24357, line 10-11; Section 3.2; Conclusion).

We have added to the manuscript to clarify the scope of our conclusions. We found no evidence of organic aerosol formation/loss over the sampling period analyzed, though this does not preclude significant condensational growth in the early stages as has been observed in previous studies. Clarifications have been added to the identified lines.

3. The dilution scheme and timeframes employed seem very reasonable. However, are the authors able to use CO and CO₂ data to infer more about the source emissions? For example – the authors normalize the size distribution in Fig. 5a to get Fig. 5b. Can the same be done for the size distributions in Fig. 9? How do the primary size distributions normalized to CO compare to other published results? The authors derive N₀ values ranging from 62,500-115,000 cm⁻³ – what are these values as inferred emission factors and how do they compare to published particle number EFs (e.g., Janhäll et al., 2010)? It seems this could help to support the representation of dilution rate in the model.

Figure 9a has now been updated to present particle concentrations normalized to values of excess co-emitted CO [ppbv]. The young CO values were extrapolated backwards from median aged CO concentrations, a constant background CO-level of 100 ppb and accounting for dilution rates. We have added the following discussion on the updated figure: “Normalized to estimated freshly emitted excess CO, the young plume number concentrations are 37, 53, and 60 cm^{-3} ppbv⁻¹ (for τ_{dil} =24 hrs, τ_{dil} =36 hrs and τ_{dil} =48 hrs respectively; see Figure 9a). The similar magnitudes of these normalized size-distributions indicate a relatively robust particle/CO ratio regardless of the dilution parameter, though in the absence of knowledge of the source fire fuel densities, a direct comparison to emission factors (kg^{-1}) cannot be made.”

4. For Figure 5: suggest adding a panel ‘c’ that contains normalized size distributions for the plume and background: given the much larger number concentrations in the plumes, it is very hard to compare the two in panel ‘a’.

Figure 5a has now been changed to include two separate y-axes to better distinguish the size distribution features.

5. Pg. 24352, line 29 and following 6 sentences (7 sentences in total): this seems unnecessary.

These sentences are included to give context to the motivation behind studying the microphysical evolution of biomass-burning particles. Though part of this work concentrates purely on the analysis of the experimental BORTAS data, the overarching goal was to investigate how size distributions change with aging.

6. Why was the OA plume threshold set at 10x background, when CO was 1.5x background and CH₃CN was 2x background? This seems arbitrarily high? This seems to significantly reduce the amount of data considered in Flight b622 (Fig. 2) after 18.0 UTC.

We found that the amount of data from the lower-altitude plume were generally insensitive to the exact thresholds for each species within reasonable variations. During the time period you specify, the plume is more limited by the OA threshold than the CH₃CN threshold, and this is due to the aircraft sampling the high-altitude plume that had undergone aerosol wet deposition, and we want to exclude this upper plume since the wet deposition complicates our analysis of the size distribution and aging.

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 (see Section 3.2).”

7. Pg. 24354, lines 16-18: unnecessary.

We removed this sentence.

Technical Corrections:

1. Pg. 24351, line 11: capitalize “Earth’s”

2. Pg. 24357, line 4: suggest changing ‘in this situation’ to ‘for applications to aircraft data’

3. Pg. 24360, line 1: delete ‘a’

These have been corrected.