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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aged boreal biomass burning aerosol size distributions from BORTAS 2011" *by* K. M. Sakamoto et al.

Anonymous Referee #1

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General Comments:

This paper presents analyses aimed at characterizing the evolution of the particle size distribution for biomass burning emissions undergoing long-range transport. The boreal fire plumes studied here were encountered during the BORTAS-B aircraft campaign after being transported 1000-1500 km (and 1-2 days) from their source. The authors compute a primary aerosol size distribution by using an aerosol microphysics model with the assumption that coagulation and dilution of background air are the only processes that affect the particle size distribution. Overall, the analysis is novel, the paper is well written, and several findings stand out as significant. Specifically: 1) the recommendation that climate models use aged biomass burning size distributions because coagulation alters the size distribution rapidly, and 2) the high concentrations of

small particles that cannot be explained by the dilution of background air. I therefore recommend the manuscript for publication in ACP after the following comments are addressed.

Specific Comments:

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/gas-to-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).

2. Since the biomass burning plumes were sampled 1000-1500 km downwind of their source, the conclusion that the $\Delta OA/\Delta CO$ 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).

3. The dilution scheme and timeframes employed seem very reasonable. However, are the authors able to use CO and CO_2 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 N_0 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.

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'.

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

6. Why was the OA plume threshold set at 10x background, when CO was 1.5x background and CH_3CN 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.

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

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'

References:

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