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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Transformation and aging of biomass burning carbonaceous aerosol over tropical South America from aircraft in-situ measurements during SAMBBA" *by* William T. Morgan et al.

Anonymous Referee #2

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The present manuscript is a highly-focused piece of work that examines chemical and microphysical property changes of biomass burning aerosol emissions that were sampled during the SAMBBA field campaign in Brazil. By and large the manuscript is written reasonably clear (explicit areas where more clarity is needed are highlighted below). As biomass burning (BB) events represent a major source of particulate matter injected into the atmosphere, it is indeed important to characterize the emissions and to understand how the chemical, microphysical, and optical properties transform as the plume ages so that models can, in turn, improve upon there fidelity. Using the

analysis tool of excess mixing ratios, the authors report on a negligible net increases in organic aerosol (OA) even though there is concomitant changes in chemical properties - namely the oxidation state of the OA. This finding is inline with that reported by others and the authors put forth the same argument as that put forth by others that the loss of primary organic aerosol (POA) due to plume dilution is offset by the production of secondary organic aerosol (SOA), hence the negligible change in net OA loading. The other reported findings centers on refractory black carbon and its mixing state. In summary, the present manuscript adds important findings to a growing body of data on biomass burn-generated carbonaceous aerosol production and evolution and as such should be published. That said, to this reviewer at least, this manuscript has a feel of being incomplete in some of its analyses and in other areas left this reviewer wondering why other datasets were not part of this manuscript. This feeling could be simply that the authors are parsing out various subject matter for other manuscripts (e.g., optical properties). If this is indeed the case, the authors should be explicit about that. Once this authors address this ad some other top-level items listed below along with several specific items (e.g., figure numbering and clarification text) the manuscript should be acceptable for publication.

While this manuscript is highly-focused, as highlighted above, it seems that the authors miss the opportunity to role in other data sets that could, potentially, elucidate all that is going on in these complex emissions plumes. For example, the authors report that there are changes in the OA chemical properties and that they observe that the rBC coating thickness changes as a function of plume age, yet authors do not report on, nor reference any parallel paper, that looks at aerosol size distribution - arguably one of the most fundamental of measurements in our business. Doing so could tell us something about the roll of coagulation near the fire source as well as informing us about how the distribution evolves. Does the mode stay constant or grow?As hinted at above, perhaps the authors have the intention of publishing a separate aerosol size distribution-centric paper. If this is indeed the case they should make reference it, as the analysis of these dataset would easily complement what is being learned from

the AMS. The absence of this dataset is even more puzzling given that the authors report on geometric mass mean diameter for rBC - along with estimates of the coating thicknesses. Why present microphysical properties of only one species (rBC) and not the primary particulate species (OA)?

Similarly, the authors say nothing about aerosol light scattering or light absorption. How does the scattering Angstrom exponent evolve with plume age in the near field? Does the mass scattering efficiency track what is observed with the AMS? In their rBC coating thickness analysis, the authors assume that the coating this transparent (coating refractive index of 1.5 +0i). What is the basis for this assumption? BB events are a known source of brown carbon. What does the absorption Angstrom exponent suggest? And, of course, what is the SSA doing in these first few hours where a lot of chemistry appears to be going on? As with the size distribution, perhaps the authors will report on this in a separate manuscript. It just seems to this reviewer that the absence of any reference to either the optical or size distribution datasets misses any opportunity to better examine what is going on.

In their examination of regional BB haze, the authors use the AMS tracer f44 and the ratio of rBC to CO. As the author state, rBC and CO are relative inert tracers that are strongly controlled by initial conditions, and that the ratio can provide some information about the influence of precipitation. But my question to the authors, especially when examining regional haze, is how do you know what the initial ratio was at the various sources that are contributing to the haze? Are all fires assumed to exhibit the same burning phase conditions (e.g., flaming versus smoldering)? Figure 7 indicates that the mass fraction of rBC ranged from 2% to 12.4%. Are the authors saying that the burn conditions are the same for these two bounding conditions? Here is where I would have expected some discussion of modified combustion efficiency (MCE) which might help answer this by telling us something about the initial burn conditions. Under active flaming conditions little CO is produced and more rBC while under smoldering conditions, more CO is produced and little rBC. Could this explain the variability observed in

rBC mass fraction contributions or is the variability driven by differences in source fuel or subsequent cloud processing (e.g., rBC loss through precipitation)? While not listed in this paper, Darbyshire reports that a CO2 analyzer was deployed on the FAAM and thus is presumably available to use along with the CO data set, to estimate the MCE. It might b interesting to see if any of the variability in rBC mass fraction contributions can be explained by MCE. And if so, maybe the MCE can, in turn, improve the robustness of the rBC/CO ratio analysis. Finally, the authors might want to check out Collier et al., (figure 4; EST, 50, 8613, 2016) who reported on the relationship between OA production and MCE - where OA production was favored under smoldering conditions.

The authors state that the fire used in their case study was likely a natural fire and one that is “highly-smoldering”. Do we expect a highly-smoldering fire to generate a 12.4% mass fraction of rBC?

The manuscript seems heavy on the figures (12 figures). The authors are encouraged to try to reduce this number.

Specific comments

Abstract: page 1, Line 32: Please add “mean mass diameter” in front of “250- 290 nm”.

Page 4. Line 11: Please add figure number. (“1”)

Page 4, Line 22: Please add figure number (“1”). Also, not clear what comes after “and”

Page 4, Line 30: Please add figure number (“2”)

Page 4, Line 35: Please add figure number (“3”)

Page 5, Line 4-5: How do the authors explain a slight decrease in rBC core diameter? This is a curious finding and this reviewer cannot help but wonder if the reported decrease is due to a measurement artifact as this was reported for the “case” study where the plume lifecycle could be well constrained (i.e., no cloud processing of coated

rBC particles that could selectively wash out larger diameter rBC-containing particles). What were the highest number concentrations encountered near the source? Back-of-the-envelope calculations assuming a size mode of 125 nm rBC particle and 1.5 ug/m3 suggests < 1000 particles/cc which should be low enough not to suffer from particle coincidence. Again, this is a very curious finding to simply close out a paragraph with, with no follow on statement or discussion.

Page 5, Line 7: Please add figure number (“4”)

Page 5, Line 23: Please add figure number (“5”) - Are both figures 4 and 5 necessary?

Page 5, Line 33: What is the useful “aging” range of the f44 marker as a tracer of age?

Page 6, Line 23: Please add Figure number (“5”)

Page 6, Lines 34-39 and page 21, Figure 8. As discussed above, perhaps examining the MCE might provide some useful insights. The delta rBC/delta CO scale in Figure 8 nominally ranges from $\sim$0.0025 to $\sim$ 0.02. Is this variability driven by cloud processing (i.e., precipitation) or MCE.

Page 25, Figure 12: Again, not to harp on the MCE theme, but it might prove interesting to examine whether the variability in coating thickness is driven by processing or MCE.

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