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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 #1

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This paper describes the analysis and interpretation of biomass burning carbonaceous aerosol emissions over Brazil from an individual plume case study and from regional haze. These emissions are thought to comprise a large fraction of the global aerosol budget and are important in their radiative effects. As these emissions age, there is a potential for changes of organic aerosol mass (OA or OM) relative to the standard inert tracer carbon monoxide (CO) due to evaporation (decrease) and secondary production (increase). The work presented here is from a unique dataset and is particularly relevant to current investigations into how biomass burning emissions are incorporated and

treated in global models. Therefore, the paper is of interest to the readership of ACP. However, the conclusions could be made more strongly if a few more details about the observations were included as described below.

One of the main conclusions is that the observed regional differences in the relationships between OM and refractory black carbon (rBC) with CO are due to the initial emissions from the fire source. However, the data actually presented in this manuscript only partially support this conclusion. The manuscript presents a very clear aging case study of OM from only a single, tropical fire with an initial, low, modified combustion efficiency (MCE) of about 0.79. The ratio of OM to CO for this fire did not change significantly from the source emissions (Figure 3) and is quite similar to the average regional haze OM for the entire study (Figure 8). The near-fire emission paper using the same dataset (Hodgson et al., 2018) included fires from the Cerrado region that had higher average MCE (about 0.94) with roughly half of the OM/CO initially emitted compared to the tropical fires (Table 5 of that paper). The current manuscript shows that the ratio of OM to CO for haze from the same Cerrado region (flight B742) is similar to that observed for the tropical fire case study (Figure 8). While this implies that a Cerrado fire ages from a lower OM to CO ratio to a higher one, this case was not presented as a contrast to the low MCE, tropical fire. It is difficult to see that any regional haze OM to CO ratio differences are directly related to differences in fire source emissions. Hence, this conclusion of the manuscript would be stronger if it included an additional aging case study of a fire with a relatively-high MCE and how the aged aerosols from such a fire transformed into regional haze.

While the analysis of the organic fraction of the biomass burning aerosols was fairly thorough, the analysis of rBC in this paper was surprisingly very limited. It was restricted mostly to coating thickness and some information on delta(rBC)/delta(CO) for the regional haze. rBC (number or mass median?) core sizes were mentioned in the abstract and some of the text, yet no data were presented. Furthermore, the sizes reported here were very different (250-290 nm) from those presented in the Hodgson work (number medians around 100 nm and mass medians around 200 nm). It looks like the mass of rBC in the fire overpass intercepts (and associated background) were significantly higher than in the along plume straight-and-level run (SLR) (Figure 2). There did not seem to be any explanation for this or how it might impact the derivation of a "background" rBC mass for the delta(rBC) calculations. More rBC data (not just in the text but in figures) and analysis should be included in this manuscript (e.g., mass of rBC, number fraction of rBC particles to the total number of particles, rBC-core size, and number fractions of rBC particles with coatings compared to all rBC particles as a function of plume age and similar values for the regional haze). Also, there was no data showing changes in the ratio of rBC to CO with wet scavenging. Such additional information about the rBC-containing carbonaceous aerosols should be included in this manuscript.

Minor comments:

P1 L6 and elsewhere: OA was used here and OM was used in other places. Consider using one or the other.

P1 L17-19: If supported with additional analysis, this last sentence is confusing and should be re-written more clearly.

P4 L17: change "refractory" to "non-refractory"

P5 L32: should "cross-plume" be in front of "SLR" here?

P5 L44 and elsewhere: The numbers for several figures are missing in the text.

P5 L51: Add citation to the Hodgson paper for the 97.1% value.

P6 L25: How does the time to reach background levels (3 hours) compare with calculated entrainment/mixing rates base on the boundary layer heights and the atmospheric (in)stability?

P6 L38-41: What were the delta(rBC)/delta(CO) ratios and rBC core diameters referred

to here? Suggest showing in SI.

P7 L48 and others shortly afterward: "absolute difference and correlation between the mass spectrum and the background" – does this refer to deviation of the slope from one and r-value from a linear least squares correlation obtained by plotting the absolute peak intensities (instead of the "fraction of organic signal" peak intensities) of the top spectrum versus the bottom spectrum in Figure 4? Please clarify.

P8 L44 and after: Were there any flights where there was a clear distinction in delta(rBC)/delta(CO) before and after raining? This would be interesting to explore in light of the assertions made here about air mass history and fire burning conditions.

P9 L15 and earlier: The fraction of rBC particles that were coated is discussed in this paper, but the actual fraction is not shown in this paper. Should be included somewhere.

P9 L22-23: Should include the rBC core-size data somewhere.

P10 L43 & 44: "limited net enhancement of OA" seems to be inconsistent with "net OA production". Please clarify.

P10 L54-56: This last sentence was confusing; suggest revising. Perhaps add a second statement (if observed) about how the rBC mass was useful for distinguishing differences in air mass history for two similar instances of OA/CO with the same f44.

P11 L34-35: This statement is made without mentioning the actual fractions of coated rBC particles in the different locations studied.

P11 L54: consider adding to the end of this sentence "to essentially background values."

Figure 1: It was unclear how the flight track matched up with the map. Suggest enlarging the map to show where the fire was.

Figure 2: The values may be easier to obtain/visualize directly from the plots if they had

horizontal lines across them. It would be good to see where the background CO, OM, and rBC values were chosen, especially for the along-plume SLR. Consider having plots of OM vs CO, rBC vs CO, and Inorganics vs CO in the SI. Add the fire altitude to the caption. Consider expanding the altitude axis and including the ground level height.

Figure 3: This could be split into three separate figures: one of the top three plots, one of f44, f43, and f60 vs. plume age (instead of Figure 5), and one of the rBC data (delta(rBC)/delta(CO), rBC median number core size, fraction of rBC particles coated, and rBC coating thickness) vs. plume age. All three of these should have "background" values on the right hand side plus both horizontal and vertical grid lines for reference. Consider adding data from the cross-plume intersect points too.

Figure 4: The background mass loading from "regional aerosol haze away from the main fire plume study region" is 4.1 here, but in Figure 7 the average background mass loading for this flight was about 10. Why were they so different?

Figure 5: It is not clear why the dashed lines are important. How would these plots look if only mixing was occurring? Suggest omitting this figure or relegating to SI.

Figure 6: Suggest enlarging the map showing only the portions of the flight tracks where the regional haze data were obtained and labeling the states with regional haze flight data. For the enlarged map it would be helpful to have an indication of scale.

Figure 7: Consider adding the location information from Table 1 to the caption, or at least mention which two flights were not only over Rondonia. Maybe point out the flight numbers on the map in Figure 6?

Figure 8: It is difficult to distinguish variation in delta(rBC)/delta(CO) because the points are outlined in black. Suggest including the average value for each delta(OM)/delta(CO) value shown. Does B737 show the opposite trend (higher delta(OM)/delta(CO) with lower delta(rBC)/delta(CO)) than flights B734 and B746 (with lower delta(OM)/delta(CO) for lower delta(rBC)/delta(CO))? It's difficult to see either

trend for flight B739 – are both those slopes for relatively low delta(rBC)/delta(CO)? Is there another indicator that could be used to distinguish low delta(rBC)/delta(CO) arising from precipitation rather than burn conditions? Perhaps the B737 flight was more/less impacted by precipitation than the other flights? Also, consider a table in the SI of background OM and CO (and rBC) for each correlation shown.

Figure 9: How do low delta(CO) values contribute to the uncertainty in the high delta(OM)/delta(CO) ratios?

Figure 12: Suggest adding tick marks on the x-axis for the flights in the top and middle rows. Also consider coloring each bar with the average delta(rBC)/delta(CO) ratios for that coating thickness bin.

Table 1: (L) must mean Airport. What does Phase P1 or P2 mean?

References

Hodgson, A. K., Morgan, W. T., O'Shea, S., Bauguitte, S., Allan, J. D., Darbyshire, E., Flynn, M. J., Liu, D., Lee, J., Johnson, B., Haywood, J. M., Longo, K. M., Artaxo, P. E., and Coe, H.: Near-field emission profiling of tropical forest and Cerrado fires in Brazil during SAMBBA 2012, Atmos. Chem. Phys., 18, 5619-5638, https://doi.org/10.5194/acp-18-5619-2018, 2018.

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