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 #3

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This paper provides an analysis of BC, OA, CO and OA oxidation state for a 2012 airborne field campaign of biomass burning emissions at Porto Velho Brazil. Data is presented in two parts, a case study of a single smoldering tropical plume and a regional analysis of 9 other regional flights. Some aspects of the paper were nicely put together, such as the observation of the evolution of smoke oxidation state. But the overall purpose of the paper on the evolution of aerosol mass is short on many important details. Trying to sort out mass evolution is quite tricky, especially for an individual plume. Accounting for the temporal evolution of the fire, controlling for combustion efficiency, linking source plumes to the regional haze, all take a great deal of care. One also needs to demonstrate appropriate cross correlation between numerous parameter to ensure an apples to apples comparison to anything about temporal evolution. This is especially true in the present paper where it is clear from the regional survey work that the smoke particle properties show a lot of heterogeneity. While, I think the authors have spent a great deal of time on this paper, I found the narrative unconvincing. I think the paper probably needs significant revisions and resubmitted. At this point I think I can keep my comments to three main themes.

1) The paper references a great deal of “Recent activity” but the whole line of scientific thought on particle evolution came out of the ABLE, ESPRESSO, SCAR-C, SCAR-B missions of the late 80's and 1990s. These studies were much more rigorous than anything that is presented here. Lioussse demonstrated he issues with particle evaporation, and Martins, Reid and Hobbs evaluated secondary production and found in well documented Lagrangian plumes samples significant production. We know the authors of this paper are aware of this work because some of the co authors were actually on these papers. A summary of this work is in the 2005 biomass burning review papers by Reid. The conclusion “secondary production is complicated and varies by fire” is indeed true, but there has been a great deal of work done in the past and even currently (all un referenced) that actually narrows down processes. Reid and Martins points are that secondary production and basic condensation happen very rapidly. Secondary production of sulfate requires cloud processing. Going back to the late 1980s significant and rapid organic acid production has been observed (I think ABLE mission). This paper lacks any concrete linkage to past knowledge to move the field forward.

2) The single case study presented is for a low combustion efficiency plume without any presented evidence that downwind samples are of the same fire characteristics. During the observation of fires in SCAR-C and SCAR B it was found that fire properties change rapidly. Given that the test case had a MCE< 0.8, then black carbon production must have been at a minimum. Perhaps there was some flaming combustion along
the periphery. Therefore the relationship of secondary production to rBC is probably pretty tenuous. At the same time, most of the cases observed of secondary production have been associated with flaming combustion. Smoldering combustion is essentially a surface reaction. So with the limited data provided, I am not sure what to make of this particular test case.

3) Both comments one and two then project onto overall issue of making an apples to apples comparison to evaluate particle evolution. The authors report an MCE value, so there must be CO2 data available. But no time series of MCE is provided, nor even a CO to rBC ratio plot. Rather the reader has to do an eyeball comparison of the two on a log plot. For the regional samples we are presented with a great deal of variability in particle properties (other than well known oxidation with time) but not the types of additional data that other studies have used to sort out what is going on. I think the authors need to spend more time on the data narrative.