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Atmospheric Chemistry and Physics Discussions

Interactive comment on “Highly-controlled, reproducible measurements of aerosol emissions from African biomass combustion” *by* Sophie L. Haslett et al.

Anonymous Referee #3

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This paper reports organic aerosol (OA) and refractory black carbon (rBC) emissions along with gas phase carbon dioxide (CO2) and carbon monoxide (CO) emissions from controlled laboratory experiments where the same amount and type of fuel was burned with varying applied heat and flow rates of air through the sample. The results are relevant to the rapidly growing body of work on aerosol emissions from biomass burning. A major finding of this work is that there are distinct types of aerosols from each of the three different phases of the controlled fires: predominantly less-oxidized OA during the initial pyrolysis phase (Phase 1, prior to ignition with no emissions of CO2 or CO), almost entirely rBC during the flaming phase (Phase 2, after ignition with a relatively high modified combustion efficiency or MCE), and a mixture of mostly more-oxidized OA with a small amount of rBC during the smoldering phase (Phase 3 after ignition with a relatively low MCE). The relative amounts of aerosols between the three phases were generally consistent between experiments with the same heat+flowrate conditions. The results of these laboratory experiments were compared with other laboratory and field measurements.

The manuscript is fairly clear to understand, but some of the analysis and conclusions are confusing given the types of experiments conducted. This paper is important to many readers of Atmospheric Chemistry and Physics and should be published after some clarifications are made to the manuscript. The key areas needing revisions are described below.

Overall Comments:

1) The manuscript is lacking a conceptual picture of the processes examined in these experiments, and interpreting the results depends on such a foundation. For this study, the heat applied to the sample (high heat = H or low heat = h) and the flow rate across the sample (high flow = F or low flow = f) were varied for 8 experiments. How should the results vary with these changing experimental conditions? It is likely dependent on the applied heat and flow rate in different ways for the different phases.

2) Prior to ignition, it is expected that the amount of heat applied and sample flow rate will affect the amount of vapors up to their flashpoint (ignition) as well as the amount available for subsequent re-condensation as OA. Higher heat applied increases the vaporization rate of semi-volatile species from the sample. Higher/lower flow rate would then decrease/increase the concentration just prior to ignition. rBC is not expected to be formed during the pyrolysis phase since there is no combustion occurring (no CO2 or CO). What would happen if an ignition source was not there? Since this is an important phase for producing OA and is not a combustion phase, how might the fuel type be relevant for the emissions of this phase? What kind of differences might be expected between wood used as cooking or heating fuel versus wildfires? A little more

discussion on this subject should be included in the paper.

3) After ignition, how does the applied heat compare with the heat produced by combustion for the flaming and smoldering phases? Applied heat may be irrelevant to the results if the combustion processes generate more heat than is applied. Perhaps the only affect from varying the applied heat is related to how quickly semi-volatile species evaporate from the sample (higher heat causes them to evolve faster than the lower heat)? As for the pyrolysis phase, how might the heating environment differ between real cooking/heating fires and wildfires compared to these laboratory studies?

4) It was a bit confusing that the graphs with the data as a function of time are not indicative of how much fuel was lost during each phase, which was important for the emission factor calculations. Although using the fuel lost for each phase in the calculation was mentioned in the methods section (and shown in the mass loss plots), it should be repeated again in the text of the emissions section. The other piece of information to include in that section was whether or not the aerosol concentrations were dilution corrected.

5) A short discussion on some (potentially large) sampling issues should be included, especially the effect of dilution on the OA measurements and losses of the fluffy, fractal BC-dominated particles.

6) It is interesting that the organic spectra from the pyrolysis phase contain more alkene and aromatic peaks whereas the smoldering spectra contain more oxygenated peaks. It is quite remarkable that these two types of spectra were also observed in the ambient London data. The dominance of m/z 60 in the smoldering spectra is somewhat surprising since it should be from levoglucosan which is a pyrolysis product from cellulose that forms without oxidation. The rest of the spectra indicate that the OA from this phase is oxidized compared to the OA from the pyrolysis phase. What does this indicate about how m/z 60 is formed?

Minor Comments:

Page 5, line 133: change Appendix 1 to Supplementary Material.

Page 6, Figure 2: The text says that the wood was sanded down and the photo looks like it does not have any bark. Is that the case? Does this fuel have any residual latex that can influence the emissions?

Page 7, lines 197-198: Add further description of the “small flame” that piloted ignition.

Page 8, lines 203-207: All four pieces of wood extinguished at different times. Did they all ignite at the same time?

Page 12, lines 292-295: Could you add the average MCE’s from these studies to support these statements?

Page 14, line 337: Should probably be “alkenes, alkanes, cycloalkenes” instead of “alkanes, alkenes, cycloalkanes.” Could also mention here that some “saturated hydrocarbon” peaks (e.g., m/z 43) could contain an oxygen.

Page 18, lines 433-435: Where are the qualitative potassium measurements that are referenced here? Maybe include in the SM?

Page 19, line 444: Could mention that Zhou et al. study was a summertime wildfire and the Brito et al. study was of open biomass burning (in contrast to the Young et al study of wintertime heating fires).

Section 3.2 on Nitrogen: Since the organic mass is high, there could be some organic interferences at m/z 30 and 46 that are currently not subtracted in this analysis for organic nitrate. Considering the uncertainties in this calculation, how much of the total mass could have been inorganic nitrate? The next to the last sentence seems to indicate that the ratio of (NO+ plus NO2+) (presumably organic nitrate) to organic mass is 0.29:1. That is quite high. What about other inorganic constituents? There needs to be a conclusion to tie this back to the beginning statements on hygroscopicity.

Table 2: What do the errors indicate? Might want to note in the caption that the emis-

sion factors are for the measured mass loss during that phase and a “weighted” average is shown in Figure 7 (although this starts to become clear with Figure 8).

Page 21, line 512: Consider different wording than “both factors.”

Page 23, line 555-556: Consider revising “if a flame reached a high intensity and rate of mass loss sooner. . .” since this phrase is confusing.

Page 24, lines 566-567: The pyrolysis phase as defined in this paper cannot be parameterized with MCE because CO2 and CO are not produced prior to ignition. Consider rephrasing this sentence.

Page 24, line 571. Consider replacing “this third phase” with “the pyrolysis phase.”

For all figures: Examine the positons of the text boxes, arrows, and shaded regions to make sure nothing is obscured and all are placed correctly. Consider using three different colors consistently for the three phases, including the bars/shaded regions for Phase 1 and 3 and data points in Figures 6 and 7 (suggest the three colors used for Figure 8).

Figures 3 and S1.1-1.8: Make the CO and MCE traces darker. Make all of the OA traces solid lines (some are dashed).

Figure 4: Change OA and rBC to lines which shows the clear presence/absence of each species instead of filling to zero. Consider omitting the scattering volume from these plots, since they do not appear clearly in the figure nor provide additional information. Why are there multiple Phase 2 regions?

Figure 8 (and related figures in SM): Change x-axis to “mass loss rate”, also the associated text in the manuscript should say “rate.”

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