Response to Referee #2

We thank the Referee for their interest in our work and the timely and useful comments, which have improved the paper. In the text below we reproduce each comment followed by our response and an exact description of any changes in the revised paper. (At the end of this response we append a short list of minor voluntary corrections/updates that don't affect any conclusions.)

Anonymous Referee #2, This is a very important and generally well-written manuscript reporting on characterization of gaseous and particulate emission from the laboratory burning of a multitude of Wildland fuels. However, comparisons to results from previous laboratory studies, especially for aerosol emissions and their optical properties are largely missing and errors are not quantified in many figures. This manuscript is appropriate for ACO and should be published after these shortcoming have been corrected and the comments below have been taken into account.

1. P2,L33, 37: Replace the technobabble "lab" with "laboratory" here and elsewhere.

We replaced "lab" with "laboratory everywhere except for in a few hyphenated usages to prevent clumsy long words.

2. Introduction: The work presented here needs to be put into the context of the earlier laboratory studies of aerosol emissions and optical properties including the FLAME study, also conducted at the FSL in Missoula, MT; references to earlier laboratory studies and comparison of results are completely missing. For example, the fact that emissions from the combustion of duffs have a very high AAE (P11, L32) has been reported from a previous FLAME study (Chakrabarty et al., 2010). References and comparisons of emissions from peat and rice straw combustion are also missing.

There have been hundreds of papers describing previous laboratory BB studies at the FSL, Max Planck Institute, India, and elsewhere dating back to at least 1991 and we've added text to the introduction on P2, L37 before "However":

"For these reasons, numerous laboratory studies have been crucial to advance our understanding of biomass burning emissions (e.g. Lobert et al., 1991; Yokelson et al., 1996; Lewis et al., 2008; McMeeking et al., 2009; etc)."

References added:

Lobert, J. M., D. H. Scharffe, W. M. Hao, T. A. Kuhlbusch, R. Seuwen, P. Warneck, and P. J. Crutzen.: Experimental evaluation of biomass burning emissions: Nitrogen and carbon containing compounds, in Global Biomass Burning: Atmospheric, Climatic, and Biospheric Implications, edited by J. S. Levine, MIT Press, Cambridge, Mass., 1991.

McMeeking, G. R., Kreidenweis, S. M., Baker, S., Carrico, C. M., Chow, J. C., Collet Jr., J. L., Hao, W. M., Holden, A. S., Kirchstetter, T. W., Malm, W. C., Moosmüller, H., Sullivan, A. P., and Wold, C. E.: Emissions of trace gases and aerosols during the open combustion of biomass in the laboratory, J. Geophys. Res., 114, D19210, doi:10.1029/2009JD011836, 2009.

However, to our knowledge this study is the first to focus specifically on simulation of wildfires. Thus, we agree it makes sense to add a comparison to the FLAME duff-combustion results the Referee recommends since it is an overlapping fuel with our study. At P11, L44 we have added the following:

"We can compare our duff results to previous measurements of optical properties of duff-fire aerosol by Chakrabarty et al (2010). These authors identified tarballs as a major BrC species produced by duff combustion and they measured an AAE of 4.2 (405 and 532 nm wavelength pair) for a Ponderosa Pine duff sample from MT. Including their other duff sample (AK feather moss duff), they obtained a study-average duff-combustion AAE of 5.3. We measured AAE on two much larger burns (~4 times more fuel mass, Fires # 12 and 26) in Engelmann Spruce duff, with different wavelengths, and at much lower MCE (0.843 ± 0.036 versus ~0.91). We obtained a study-average duff combustion AAE of 7.13 (0.057). Both studies observed a high AAE for duff combustion. Their lower AAE values could be related to different wavelengths used, the possibility of some BrC abs at 532 nm (Bluvshtein et al., 2017), the different duff type, and/or their higher MCE, which they attributed to sampling some flaming combustion during the ignition process. The AAE calculated from our AAE versus MCE fit (for all fuels) at their MCE of 0.91 is relatively closer to their value."

New references:

Bluvshtein, N., P. Lin, J. M. Flores, L. Segev, Y. Minon, E. Tas, G. Snyder, C. Weagle, S. S. Brown, A. Laskin, and Y. Rudich, Broadband optical properties of biomass-burning aerosol and identification of brown carbon chromophores, J. Geophys. Res., 122, doi:10.1002/2016JD026230, 2017.

Chakrabarty, R. K., H. Moosmuller, L.-W. A. Chen, K. Lewis, W. P. Arnott, C. Mazzoleni, M. Dubey, C. E. Wold, W. M. Hao, and S. M. Kreidenweis.: Brown carbon in tar balls from smoldering biomass combustion, Atmos. Chem. Phys., 10(13), 6363-6370, 2010.

Peat and rice straw (and dung) are very minor components of this study used only briefly to check fuel chemistry effects or compare to field data to further investigate the possibility of reasonably realistic simulations in the laboratory. In addition, an exhaustive discussion of these fuels could potentially include some previous lab studies that may have had less realistic results. For instance some previous lab studies of peat fire emissions reported unrealistic EC emissions by the thermal method or C2H2/CH4 ratios >1 where the latter shows that the emissions sampled were actually dominated by the propane torch used for ignition. We prefer not to engage in a lengthy discussion of these issues in this paper about wildfires. Finally, there are recently

published, more extensive, lab and field comparisons for peat and rice straw combustion, which are noted in our new text revised as follows:

P12, L13: Cited Pokhrel et al., 2016 peat AAE paper after first "AAE"

P12, L26: We added "briefly" before "summarized"

P12, L28: We appended to the end of the paragraph: "More comprehensive, recent discussions of these fuels can be found elsewhere (Stockwell et al., 2016a, b; Jayarathne et al., 2017a, b)."

References:

Jayarathne, T., Stockwell, C. E., Bhave, P. V., Praveen, P. S., Rathnayake, C. M., Islam, Md. R., Panday, A. K., Adhikari, S., Maharjan, R., Goetz, J. D., DeCarlo, P. F., Saikawa, E., Yokelson, R. J., and Stone, E. A.: Nepal Ambient Monitoring and Source Testing Experiment (NAMaSTE): Emissions of particulate matter from wood and dung cooking fires, garbage and crop residue burning, brick kilns, and other sources, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-510, in review, 2017a.

Jayarathne, T., Stockwell, C. E., Gilbert, A. A., Daugherty, K., Cochrane, M. A., Ryan, K. C., Putra, E. I., Saharjo, B. H., Nurhayati, A. D., Albar, I., Yokelson, R. J., and Stone, E. A.: Chemical characterization of fine particulate matter emitted by peat fires in Central Kalimantan, Indonesia, during the 2015 El Niño, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-608, in review, 2017b.

3. P5,L24-42: References for the PAX instrument including reciprocal nephelometer are mostly missing.

P5, L25: We added a reference to an earlier prototype instrument with some similarities to our PAXs in that they combined a reciprocal neph with a PAS (Lewis et al., 2008). We had already cited a recent detailed "PAX description and evaluation" paper by Nakayama et al., 2015

Reference:

Lewis, K., Arnott, W. P., Moosmuller, H., and Wold, C. E.: Strong spectral variation of biomass smoke light absorption and single scattering albedo observed with a novel dual-wavelength photoacoustic instrument, J. Geophys. Res., 113, D16203, doi:10.1029/2007JD009699, 2008.

4. P7, L29: Replace "The EFs for scattering and absorption: : :" with "The EFs for scattering and absorption cross-sections: : :" to better define what you are actually reporting.

We updated the text here to read: "The EFs for scattering and absorption optical cross-sections…".

5. P8,L30-31: ” It is important to compare our FIREX lab fire emissions data to field measurements of real wildfires to assess how representative and useful the lab-based data are, especially for the many species measured in the lab, but not the field.” This seems pretty nonsensical, how do you compare laboratory data with field data for species that weren’t measured in the field. Please explain!

As noted in our response to comment 4 of Referee #1: To clarify on P8, L31 we added: “We assess representativeness by comparing the EF results for species measured in both the field and our laboratory fires.”

6. P8, L41-44: “: : :because the lab fires had higher average MCE (i.e. a higher fire-integrated flaming/smoldering ratio) than the real wildfires sampled to date, most likely due to some unavoidable drying of the fuels during storage.” The second reason may be that in the laboratory, one burns fairly small pieces of fuel, while in the field larger pieces (e.g., tree trunks) may smolder for days.

The largest diameter dead/down woody debris fuels are referred to as 1000 hr fuels and are over 7.6 cm in diameter (Table S1). We burned some of these fuels, but upon re-checking we do find that they were under-represented by our team of forest fire “experts” compared to the FOFEM-recommended amounts. We thank the Referee for bringing this to our attention and have appended the following to the end of the sentence: “and some under-representation of the largest diameter fuels (Tab S1).”

P11, L11: deleted “all the” so the sentence doesn’t imply “perfection.”

In addition, in the conclusions P13, L13: We changed: “Using a simple procedure to account for the flaming to smoldering ratio, we generated EF values from the lab data that were in agreement with the field data for ….” to “Despite some underrepresentation of the largest diameter fuel class we were able to use a simple procedure to account for the flaming to smoldering ratio and generate EF values from the laboratory data that were in agreement with the field data for …”

7. P8, L43 & P9, L30: Please define the “flaming/smoldering ratio”!

This is explained in different words on page 6 associated with the description of MCE. To clarify, on P6, L36, we appended to the end of the sentence “and an MCE of 0.9 would indicate roughly equal amounts of flaming and smoldering (i.e. a flaming/smoldering ratio of ~1)”

8. Error bars must be added to figs. 2, 6, 7, and 8.

We added representative error bars to each of these figures.

REFERENCES Chakrabarty, R. K., H. Moosmuller, L.-W. A. Chen, K. Lewis, W. P. Arnott, C. Mazzoleni, M. Dubey, C. E. Wold, W. M. Hao, and S. M. Kreidenweis (2010). Brown Carbon in Tar Balls from Smoldering Biomass Combustion. Atmos. Chem. Phys., 10(13), 6363-6370.

This was added as noted above.

We've also made some minor voluntary corrections and updates as described next:

P1, L17: "Subalpine Fire" changed to "Subalpine Fir".

P1, L28: After a last-minute addition of a comparison to the one previous wildfire airborne NH3 measurement, we forgot to update the abstract and conclusion.

The existing text was: "This is especially valuable for species not yet measured in the field. For instance, the OP-FTIR data alone show that ammonia (1.65 g kg-1), acetic acid (2.44 g kg-1), nitrous acid (HONO, 0.61 g kg-1) and other trace gases such as glycolaldehyde and formic acid are significant emissions not previously measured for US wildfires."

This now reads: "This is especially valuable for species rarely or not yet measured in the field. For instance, the OP-FTIR data alone show that ammonia (1.65 g kg-1), acetic acid (2.44 g kg-1), nitrous acid (HONO, 0.61 g kg-1) and other trace gases such as glycolaldehyde and formic acid are significant emissions previously poorly, or uncharacterized, for US wildfires."

P1, L35: removed unmatched ")" after "kg-1".

P4, L12: After "poplar shavings" added "(aka "excelsior")" to connect to name in supplemental tables.

P6, L14 and also on P12, L37: We've added two new gases (C2H2 and C2H4) to the list that we analyzed for in the room burns due to a recent (post-submission) request.

P6, L31: After smoldering we added, "where "smoldering" is an approximate term for all non-flaming processes (e.g. glowing and pyrolysis) as explored in more detail elsewhere (Yokelson et al., 1996, Koss et al., 2017; Sekimoto et al., in preparation)"

P8, L33: "Compositions" corrected to "Composition" (not plural) in SEAC4RS.

P9, L43: Appended "because of a transition to flaming combustion during the second half of the fire."

P10, L25: Corrected quoted lab average AAE from "2.19 ± 0.24" to "2.80 ± 1.57" consistent with conclusions and Table 4.

P10, L36: Liu et al reference, we added year

P11, L14: We removed an unnecessary sentence about Tables 2 and 3.

P11, L15: Added citation to reflect that the Rim Fire AAE was from Forrister et al., 2015

P12, L9 sect 3.7 header: changed to "Trace gas …" (no plural)

P12, L13: added “emissions” after “BC”.

P12, L14: corrected EFCH4 from “10.83” to “10.39”.

P12, L17: We expanded “(BC extremely small and gases within 31%)” to “(EF BC extremely small compared to most biomass burning (Akagi et al., 2011) and gases within 31%)” to clarify “small”

P12, L22: Added “EF” before “BC” to clarify as above.

P13, L7: changed the “with BrC accounting for nearly 100% and 78% of the absorption at 401, respectively, for these fuel components.” to “with BrC accounting for nearly 100% and 94% of the absorption at 401, respectively, for these fuel components (using data only from fires with measurements at two wavelengths).”

P13, L17: The NH3 uniqueness retracted by adding “rarely, or” before “not previously” for reasons explained above.

Table 4: We removed un-needed “EF” from the “Lab AVG EF” column header.

Table 5: The MCE variability for duff was missing a zero and has been fixed.