Response to Referee #1

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.

Anonymous Referee #1, This paper reported emission factors (EFs) of trace gases and optical properties of aerosol particles during combustion of canopy, litter, duff, dead wood in US and some other fuels in the laboratory. The data obtained in this study are valuable to evaluate the impact of biomass burning on climate and atmospheric environment. However, improvement of the discussion will be necessary before considering the publication in ACP.

General comments

Discussion on the relation between the EF for gaseous species and aerosol optical properties is not enough. Especially, optical properties of BrC should depend on chemical compositions of particles and may be indirectly related to the relative EF of gaseous compounds. I recommend adding discussions on this point.

We agree that it is well-established that certain gases and BC are associated with flaming, while smoldering is associated with BrC and other gases. We now added a description of these indirect associations for completeness on page 10 lines 19-21 as described in detail below.

We are not yet aware of direct mechanistic links between the gaseous species and the aerosol optical properties measured in these fires. We agree aerosol optical properties are related to particle chemistry, but our group did not collect particle chemistry data aligned with the PAX data reported here. Any relevant particle chemistry that was measured by other groups still needs to be analyzed and published. Happily, in the room burn phase of FIREX, our PAXs and extensive other instruments for chemistry and other properties of particles were co-deployed. We think that work will do a better job of addressing the Referee's suggestion than we could do with the data available now. We've provided cross-references to upcoming papers led by other groups already on P3, L8-9 (e.g. Wagner et al, Li et al, both in preparation).

P10, L19-21:

Existing text: "High AAE is an indicator of BrC and relates to smoldering, which is denoted by low MCE and high SSA values. Low AAE, along with low SSA and high MCE values, indicates more flaming combustion."

New text: "High AAE is an indicator of BrC and relates to smoldering, which is denoted by low MCE and high SSA values. Smoldering is also associated with higher EFs for OA, most NMOG, and other gases such as NH3. Low AAE, along with low SSA and high MCE values, indicates more flaming combustion, which is also generally associated with higher EF for BC and "flaming compounds" such as CO2, NOx, and SO2."

Specific comments

1) Page 5, lines 36-39: Scrubber and diffusion dryer were used in this study. Information on the removal efficiency of light absorbing gases and the particle losses should be added.

The manufacturer specification on the scrubber is "minimum removal efficiency of 99.5%" (https://www.purafil.com/wp-content/uploads/2014/12/Purafil-SP-Media-Bulletin.pdf). If the scrubber is not effective this can be detected as absorption even when filtering particles during zeros. However, the scrubber color changes from purple to mostly brown before its effectiveness drops. The scrubber capacity 32 g NO2 per 100 g scrubber combined with the amount we used (~700g) should be sufficient for hundreds of fires, but we exchanged the scrubber pre-emptively partway through the experiment and before any signs of deterioration were observed. We confirm that our drier was a diffusion drier, which we now specify. We did not measure particle losses in the diffusion dryer, but several on-line descriptions claim that "particle loss is minimal in diffusion dryers because the aerosol doesn't contact the desiccant" (e.g. http://dropletmeasurement.com/dmt-diffusion-dryer http://www.tsi.com/diffusion-dryer-3062/ https://www.topas-gmbh.de/en/produkte/ddu-570/). A similar diffusion design is used in the scrubber.

The existing text was: "From the splitter, each separate sample line encountered a scrubber (Purafil-SP Media) to remove absorbing gases and then a drier (Silica Gel 4-10 mesh) to remove water, with this order ensuring that both instruments were sampling at the same relative humidity (varying between 13 and 30%)."

The new text is: "From the splitter, each separate sample line encountered a scrubber to remove absorbing gases (Purafil-SP Media, minimum removal efficiency 99.5%) and then a diffusion drier (Silica Gel 4-10 mesh) to remove water, with this order ensuring that both instruments were sampling at the same relative humidity (varying between 13 and 30%). The scrubber and drier were refreshed before any signs of deterioration were observed (e.g. color change) and the diffusion-based designs should incur minimal particle losses, but losses were not explicitly measured."

2) Page 6, lines 15-24: "The emission ratios to CO2 were then used to derive EFs calculated by the carbon mass balance method (CMB), which assumes all of the burned carbon is volatilized and that all of the major carbon-containing species have been measured" "Our estimate of total carbon in this paper includes these three species and all the rest of the C-containing emissions measured by the OP-FTIR and the PAXs." These two sentences are confusing. Did the authors include BC in the estimation of total carbon?

Yes—BC was measured by PAX and then coupled with OP-FTIR data to compile an emission factor (EF BC), which was included in the CMB along with other carbon-containing species that we measured.

We modified this sentence to read "Our estimate of total carbon in this paper includes these three species and all the rest of the C-containing gases measured by the OP-FTIR as well as the C in the particles (i.e. BC and OC) based on the PAX data."

To further clarify on P7, L24-26 we updated the text to read: "We use the qualitative OA to calculate a small term in our CMB that helps account for unmeasured C-species (assuming OA/OC of 1.6), but we do not report OA or OC in the tables as quantitative species."

3) Page 7, lines 15-19: The authors assume the AAE for BC to be 1 to estimate EF abs405 for BrC. However, this assumption is not necessarily correct (e.g., Lack et al. 2010). In addition, the authors assume that the lensing effect was negligible. However, it is strongly depend on the relative amount of OC to BC, as well as combustion conditions. Is it reasonably to estimate that OC (or BrC) did not coat BC even when OC/BC ratio was very high under low MCE conditions? Discussion on the effect of these assumption (and uncertainties) on the uncertainties in EF abs405 for BrC and EF BC should be added.

It is important that the attribution of BrC versus lensing is more uncertain than the absorption data itself. To allude to this we had provided an uncertainty of +/- 20% in the AAE for BC and acknowledged up to 30% absorption from coatings in older room burn smoke (Pokhrel et al 2017). Additional measurements (in preparation) during the FIREX room burns with a thermodenuder indicated that lensing enhancements were typically 5-10% at 870 nm even in smoke 15 minutes to hours old. Thus, again in the 5 second old smoke for stack burns it's likely the lensing effects are smaller as already noted. Without supporting measurements in the stack we can only make a best estimate of the lensing contribution based on closely-related work.

To improve the depiction of this uncertainty in BrC attribution we have added an uncertainty estimate for the BrC attribution (+/- 25%) to line 21 and we now refer to "OA absorption due mainly to BrC at 401 nm" on line 26. We also added an illustrative error bar to Fig. 8 (as also requested by Referee #2) and added the suggested reference to Lack and Cappa, 2010.

4) Page 9, line 20-21: "The lab-measured EFs for these OP-FTIR species and the data for many NMOG species measured by MS and FIREX data in general can thus be used to generate representative EFs or other data for real wildfires." Because the data of MS was not presented, I recommend avoiding the discussion based on MS data.

We think this is a key finding that is important to retain. The representativeness of the FIREX fires is a general issue of great importance to all the participating groups. Our probe/discussion of that issue is not based on MS or other data, but successfully shows that reasonably realistic values can be extracted from the lab fires based on comparing the diverse lab-field overlap species measured by the OP-FTIR. That then has the important implication that the MS and other data are useful to represent real fires.

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.”

5) Page 9, lines 23-32: Because I cannot access to the in preparation papers (Koss et al. and Sekimoto et al.), we (readers) cannot check reasonability of the suggestions.

Koss et al is now available via ACPD and we updated the reference. The point of referring to these papers is not that our approach depends on them, but to make readers aware of other data and approaches that they may find useful. We think it will be useful to retain and update cross-referencing to the other closely related work.

6) Page 9, lines 43-44: “However, for both vegetation types we observed an enhancement in NOx emissions from the litter and canopy components,..” Figure 4 shows EF for NOx from litter and canopy were smaller with “Douglas Fir (Mixed)”.

Thank you. We fixed the sentence to say, “Additionally, we observed an enhancement in NOx emissions from the litter and canopy components in Ponderosa Pine.” We also updated the sentence in the conclusions P12, L41-43 to read: For instance, emissions of some NMOG were enhanced from a Douglas Fir rotten log and emissions of NOx were enhanced from Ponderosa Pine litter and canopy components.

7) Page 10, lines 16-18: “As mentioned previously, we measured absorption and scattering coefficients directly at 401 and 870 nm. For the first 31 stack fires, which includes most of the studied fuel types, we have both 401 and 870 nm data. For the remaining 44 stack fires, we only report data at 870 nm as we used our 401 nm PAX for intercomparison studies that will be reported elsewhere.” Same information was given many times (introduction section and experimental section). I recommend avoiding duplicate contents.

We chose to duplicate these comments because several of our coauthors suggested “refreshers” for the reader. Some readers may skip directly to the optical property section of the paper, so we opted to leave this as is.

8) Page 10, lines 23-24: “Table 3 does not reveal a strong ecosystem dependence among coniferous ecosystems tested for optical properties, but does indicate that chaparral fire aerosol is relatively more absorbing and that there are significant contributions of absorption by BrC at 401 nm among all ecosystems." The description may be incorrect. Table 3 indicates that EF Babs 870 and EF Babs 401 for chaparral fire aerosols were not necessarily greater than those for Lodgepole Pine and Ponderosa Pine aerosols.

The Referee is right as written. We changed “is relatively more absorbing” to “has consistently lower SSA than coniferous fire aerosol” – the new text is correct and should clarify the point we actually intended to make.

9) Table 4: Because lab. average EF for BC would be calculated from average EF Babs by multiplying a constant factor, I think that the relative uncertainties for them should be same. Why are the ratios 0.53/0.58 and 3.20/5.16 different?

The initial EF for BC was incorrect. Rather than being the average for all 75 stack fires where 870 nm data was available, we incorrectly only reported the average of the first 31. We fixed this to now correctly account for all 75 stack fires. It has been corrected to: 0.67 (1.09). The EF Babs 870 number was correct and was not changed.

Why did the authors choose different types of function (linear and power law) for EF for Babs401 and EF for Babs401(BrC) for the equation to estimate EF from MCE?

This was empirical, we just chose whatever fit best based on the r^2 value, but confirmed the fit looked reasonable especially near the field-average MCE for wildfires.

Fitting uncertainties (or reasonability) for each equation should also be added. For example, no clear relations between EF Babs 401 and MCE and between EF Babs401(BrC) and MCE are observed in Fig. 8.

We've added the r-squared value for each equation in a new column. Due to a few outliers the EFBabs at 401 is not highly correlated with MCE, but the equation nevertheless estimates reasonable looking values averaged over the scatter for MCE values near the wildfire field average MCE. This is now clarified in a footnote at the head of the r-squared column "The low r2 equations return reasonable values near the field average MCE." In general, the application of these equations is too extract a reasonable value from the data at an important field-measured characteristic value rather than demonstrate high correlation, which may not in fact exist. Stated alternatively, EFBabs401 is likely impacted by many factors and doesn't correlate highly with MCE, nonetheless a reasonable guess at the appropriate EFBabs401 at the field average flaming/smoldering characteristics of wildfire is important and can be obtained from the equations presented.

10) Figure 8: I recommend adding error bars.

We have added representative error bars to Fig. 8 and other figures as requested by Referee #2.

11) Same terms should be used throughout the text. For example, EFabs 401, EF_{abs}401, and EF Babs 401 are used for EF for Babs at 401 nm.

We fixed all terms to now either say EF B_{abs}401 or EF B_{scat}401. The same goes for the 870 nm values.

References

Lack, D. A. and Cappa, C. D.: Impact of brown and clear carbon on light absorption enhancement, single scatter albedo and absorption wavelength dependence of black carbon, Atmos. Chem. Phys., 10, 4207–4220, doi:10.5194/acp-10-4207-2010, 2010.

We've added this reference.