Response to Referee #1

We thank the Referee for all their comments, which have helped improve the paper as described below. The Referee suggestions are shown in full along with our detailed response/revisions in an "R#, A#" format next.

R1. GENERAL COMMENTS: This study reports emission ratios and optical properties observed in smoke from wildland fires of various ages. The study measured ~500 hours of smoke impacts, making it very thorough in terms of sampling different ages, a variety of origins, and presumably a diversity of fire behavior. The study methods were appropriate, the presentation and discussion is thorough, yet concise. The authors have been careful to properly qualify their conclusions and have clearly pointed out limitations of the data. The discussion and conclusions are supported by the data presented. I found only one general issue that must be addressed. The comparison with prescribed fires somewhat overstates the prescribed fire versus wildfire PM differences. The authors have compared their measurements versus all aircraft measured prescribed fire measurements in May et al. (2014), which were mostly SE coastal plain understory burns or California chaparral. Only two of the May et al. (2014) aircraft prescribed fires, the Shaver fire and Turtle fire in Montane forest seem to be similar ecosystems / fuel types to the source fires that impacted Missoula during their study. From a smoke/air quality impact perspective, the wildfire vs. prescribed fire tradeoff issue is largely a matter of forest fires in the western US. The duration, fuel loading, and total emissions involved with western forest fires significantly exceeds that of chaparral and sagebrush systems (see e.g. French et al., 2011 San Diego County fires). Therefore, in the context of a wildfire vs. prescribed smoke/air quality comparison the authors should compare their wildfire results with the Shaver fire and Turtle fire from May et al. (2014), for which the forest PM1/CO = 0.011+/- 0.01, about 40% higher than the 0.08 value presented in Table 5 and used the discussion. Likewise the Shaver / Turtle fire BC/PM1 is 0.006, much closer to that observed in the current study and similar to Liu et al. (2017).

French, N. H. F., et al. (2011), Model comparisons for estimating carbon emissions from North American wildland fire, J. Geophys. Res., 116, G00K05, doi:10.1029/2010JG001469.

A1. This is a good comment though we assume the Referee meant a PM/CO of 0.11 and a BC/CO of 0.006. The Turtle and Shaver Fires comprise a small data set and it is not clear that brush, chaparral, or grass fires did not impact us in Missoula. E.g. The Rim Fire burned coniferous forests in the Sierras, but also oak and chaparral. In addition, the SE-US prescribed fires in May et al (2014) were also in coniferous ecosystems and help make a larger prescribed fire data set. The Turtle and Shaver prescribed fires may have burned more understory and less overstory than wildfires in the same ecosystem. We also have to be careful because the prescribed fire frequency (time since last burn) could impact emissions and, most of all, we are comparing fresh prescribed fire smoke (May et al, 2014) to more aged wildfire smoke (Missoula 2017) and prescribed fire emissions typically occur at lower air temperatures. In fact, May et al., (2015) observed a significant decrease of OA/CO after five hours of aging for one of the prescribed fire smoke plumes in their 2014 paper. However, we agree that it is worth acknowledging that the geographic location may combine with vegetation type to influence EF so we have added a discussion of the impact of comparing wildfires to a smaller, potentially more relevant, subset of prescribed fires as suggested by the referee.

Changes:
We added a column to Table 5 that shows the data averaged over just the Turtle and Shaver fires.

P 13, L36 Old text: The available PM/CO data for wildfires is consistently higher than for prescribed fires, which has air quality and land management implications.

The available PM/BC ratios are consistently ~20 times higher for wildfires, than prescribed burns, confirming that wildfire smoke is overwhelmingly more organic, which is important partly because many optical properties scale with the BC/OA ratio. In general, our ground-based wildfire study confirms the earlier airborne indications that prescribed fires are less smoky but also less cooling than wildfires.

New text: The $\Delta PM/\Delta CO$ values for fresh wildfire smoke in Liu et al. (2017) and aged wildfire smoke (this study) are about three and 1.5 times higher than $\Delta PM/\Delta CO$ for fresh smoke from prescribed fires in May et al. (2014) when comparing to all their US prescribed fires (Tab. 5). For only prescribed fires in western US mountain coniferous ecosystems (last column Tab. 5), the $\Delta PM/\Delta CO$ for fresh smoke is close to our value for aged wildfire smoke. However, May et al. (2015) noted that $\Delta PM/\Delta CO$ decreased by about a factor of two after several hours of aging on at least one prescribed fire.

The $\Delta BC/\Delta CO$ for prescribed fires is higher than the wildfire average by a factor of ~9 (all prescribed fires) or ~4 (last column), roughly suggesting a higher MCE for prescribed fires. Ignoring smoke age, the $\Delta BC/\Delta PM$ for prescribed fires is higher than the wildfire average by a factor of ~20 (all prescribed fires) or ~6 (last column). The $\Delta BC/\Delta PM$ observations suggest that wildfire smoke is overwhelmingly more organic, which is important partly because many optical properties scale with the BC/OA ratio (Saleh et al., 2014). In general, our ground-based wildfire study confirms the earlier airborne indications that prescribed fires are less smoky but also less cooling than wildfires. Differences in smoke production and chemistry between wild and prescribed fires should be researched more and have air quality and land management implications.

Reference
May, A. A., Lee, T., McMeeking, G. R., Akagi, S., Sullivan, A. P., Urbanski, S., Yokelson, R. J., and Kreidenweis, S. M.: Observations and analysis of organic aerosol evolution in some prescribed fire smoke plumes, Atmos. Chem. Phys., 15, 6323-6335, doi:10.5194/acp-15-6323-2015, 2015.

SPECIFIC COMMENTS

R2. P4, Ln 27-37: Was calibration conducted using the same flow set-up as ambient data, i.e. through scrubber and diffusion drier? Any estimate of particle loss based on other studies?

A2. All the calibrations were done with the same sample line, cyclones, drier, and scrubber in the same location. We only report data where instrument pressures, flow rates, leak checks, etc passed QC checks, and for 401 nm, data that was collected between calibrations where the AAE was within +/- ~2-3 percent of one for fresh propane torch soot. We did not have the specialized equipment to measure any particle losses in the diffusion drier, but using the same drier and scrubber in 2018 with a PM2.5 cyclone gave mass-scattering coefficients for 2.5 micron scattering divided by 2.5 mass that were very close to the middle of the range of numerous other studies indicating that minimal losses are occurring in the drier and scrubber. However, this was an important comment. At the time we set up the PAXs we were aware of websites (at least 3) that suggested drier losses were "minimal." However, upon re-investigating, only two of the three websites still make this claim and a recent paper briefly includes a somewhat relevant measured size-independent particle transmission efficiency (Miyakawa et al., 2017) for their diffusion drier of 84 +/- 5%. We have not applied a correction to our data because we did not measure anything specific to our setup. Referee #2 also brought up one more source of uncertainty; truncation error in the nephelometer. We added new text at P5, L25 to address several poorly characterized sources of error together after defining the relevant parameters.

New text:

P4. L30-32 text changed to: "The scrubber and drier were refreshed before any signs of deterioration were observed (e.g. color change). The diffusion based designs will cause small particle losses, but losses were not explicitly measured."

P5, L24: A few other sources of uncertainty in the measurements and/or calculations are poorly characterized; MAC increases due to coatings, potential particle losses in the drier or scrubber, and truncation error in the nephelometer. Mie calculations provided by the manufacturer suggest the scattering could be underestimated by about 1% at 870 nm and 2.5% at 401 nm due to truncation error (J. Walker, private communication). This would reduce the mass scattering coefficients (Sect. 3.4) and typically, a 1% reduction in scattering would imply approximately a tenth of a percent of value underestimate of SSA. Miyakawa et al. (2017) reported a size-independent particle transmission up to 400 nm of 84±5% in their diffusion drier. Larger particles may be transmitted more efficiently. We did not measure size distribution or transmission efficiency in this study and thus, we did not adjust the data. Size-independent particle losses would reduce scattering, absorption, and derived BC, but should have only a small impact on SSA or AAE. Unlike particle losses, an increased MAC due to "lensing" via coatings would inflate BC values by up to ~30% (Pokhrel et al., 2017).

Reference: Miyakawa, T., Oshima, N., Taketani, F., Komazaki, Y., Yoshino, A., Takami, A., Kondo, Y., and Kanaya, Y.: Alteration of the size distributions and mixing states of black carbon through transport in the boundary layer in east Asia, Atmos. Chem. Phys., 17, 5851-5864, https://doi.org/10.5194/acp-17-5851-2017, 2017.

R3. P6, L4-21: Please state the criteria used to define smoke impacted periods of sampling?

A3. Any sustained period with $PM_{2.5}$ well above 12.5 µg/m^3, which EPA defines as the upper limit for good air quality, was included in one of the smoke events. Referee #2 commented on the divisions between events. We did not apply a formal algorithm. Instead, for instance, when high PM levels decreased to a local minimum, or more sustained values, near or below the "good" air quality level (12.5 µg/m3) we took this as the end of the "event." In some cases a post-event "cleaner period" was sustained, but sometimes a single point is the end of one event and the start of another. We also elected not to integrate some small or brief peaks that sometimes occurred after adjacent larger peaks. For instance, a small peak after peak G, was not included because of low S:N. The last peak was integrated up to where the CO measurement failed. We verified several times that the integrals for events are dominated by the large values and insensitive to small shifts in the endpoints at lower levels.

P7, L20: Added: Sustained periods when $PM_{2.5}$ was elevated well above the 12.5 $\mu g/m^3$ EPA standard for “good” air quality were designed as events and assigned a letter in Fig. 1 and Tab. S1.

R4. P6, L4-21: Was a diurnal variation observed in CH4 for background conditions (e.g. due to constant source + varying mixed layer depth)?

A4. From 2017 and 2018 there is some variability in CH_4 during smoke-free periods, but it is not well-defined enough to confidently calculate a new baseline under peaks and we have no evidence that it contributes overall bias to the integrals. Most likely the variability contributes to a higher standard deviation for our measured ΔCH4/ΔCO ratios than we might have seen otherwise. This topic is also addressed in some detail in the response to Referee #2. Basically, the concept of a measureable background was usually not applicable due to the widespread (often synoptic scale) impacts.

R5. P7, L33-38 & Table 2: The authors should include CH4/CO ratios form Urbanski (2013) as the wildfires reported on in that study are most similar to the MT/ID/BC fires that impacted Missoula.

A5. We added the Urbanski CH4/CO (0.0946 +/- 0.0108) to Tab.1. Note: we were also impacted by fires in WA, OR, CA.

R6. P7, L 37: “Our higher study average ER of CH4 is indicative of smoldering, or specifically glowing combustion (Yokelson et al., 1997).” This statement implies CH4/CO for glowing combustion is different from smoldering pyrolysis, which is at odds with ground-based field study of Reisen et al. (2018). Please comment on this apparent discrepancy. (Reisen, F., Meyer, C. P., Weston, C. J., & Volkova, L. (2018). JGR - Atmospheres, 123, 8301–8314. https://doi.org/10.1029/2018JD028488)

A6. Reisen et al “visually” “broadly” sorted samples into glowing or pyrolysis, but pure pyrolysis cannot actually occur alone since pyrolysis requires heat. Glowing can occur “alone” briefly in a lab if no fresh fuels are left to pyrolyze. Yokelson et al., (1997) reported that CH_4 was enhanced from glowing compared to other organic gases, but CO could have also been enhanced from glowing so the Referee is correct that additional analysis would be needed to scope out effects on the □CH_4/□CO ratio due to glowing/pyrolysis. That is beyond scope of this study, especially since fuel type may have an impact and authentic field conditions make it harder to isolate processes than in lab. We decided to remove any speculation about smoldering sub-types since the sub-type of smoldering is secondary here to our main point that □CH_4/ □CO tends to increase with smoldering. We do note that □CH_4/ □CO increased with smoldering (lower MCE) in Reisen et al (as in many other studies), which supports our interpretation of our Figure 2.

Change in text: We deleted “, or specifically glowing combustion” and added the Reisen et al reference on line 38.

R7. P9, L1: Reference needed.

A7. The references on line 2 supported both of the first two sentences. We moved Bond 2004 reference to the previous line to clarify that literature support exists for both sentences.

R8. P9, L14-15: Should note that Hobbs et al. (1996) were mostly prescribed fires of logging slash.

A8. We compared specifically to the subset of fires described a wildfires as now noted:

Old text: “The Hobbs et al. (1996) is notably”

New text: “The Hobbs et al. (1996) average value for their two fires specifically identified as wildfires is notably”

R9. How robust is BC = f(MCE) from Selimovic ?

A9. BC/CO correlates with MCE, but with considerable noise and in non-linear fashion. To acknowledge this, on page 9, line 16 we appended “, which tends to enhance BC emissions.” We added a plot of BC/CO versus MCE from the Selimovic study to Fig. 2 in response to this comment and Referee #2 and some brief new text described in that response

R10. Fig 5. Adding date labels to a few ticks on the x-axis would be helpful.

A10. Done.

R11. P9, L16-23: Does “annual” refer to 2011 or average over some period of time?

A11. We changed “Liu et al. (2017) calculated an annual CO production from western US wildfires of 5240 ± 2240 Gg, which they reported was in good agreement with an EPA estimate from the 2011 National Emissions Inventory (4894).”

to “Liu et al. (2017) calculated an average annual CO production from western US wildfires for 2011-2015 of 5240 ± 2240 Gg, which they reported was in good agreement with an EPA estimate based on a similar burned area in the 2011 National Emissions Inventory (4894).”

On line 23 we added the year (2006) for the Mao et al (2015) study.

R12. Section 3.5. Please note the value of PM2.5/CO over these periods.

A12. Unfortunately, the PM monitor had its few missing hourly values during peak “W”.

R13. P13, L29-30: This should be restated, prescribed fires do not allow control over dispersion conditions, but allow one to ignite fires when dispersion conditions are favorable and/or manipulate ignition in a manner that enhances dispersion, e.g. mass ignition that puts smoke above mixed layer.

A13. This sentence now ends “… and can be ignited when conditions are favorable for minimizing air quality impacts (Liu et al., 2017)”

TECHNICAL

R14. Mixing of units notation, superscripts and “/”, e.g. L minˆ-1 and L/min, throughout paper

A14. Fixed

R15. P4, L36: missing “nm” after 401

A15. Fixed

R16. P5, L13: “BrC” should not be subscript

A16. This could be OK or could be parenthetical format, but be consistent.

R17. P10, L35: missing “nm” after 401

A17. Fixed

Voluntary updates.

We made three additional voluntary minor changes (described next). We did some very light editing (updating references, added a missing word, etc) that is indicated in “track changes.”

P2, L29: Added Tomaz et al., 2018 reference.

P6, L37: Changed “British Columbia experienced a record fire season…” to “Over 1.2 million ha burned in British Columbia in 2017.” The previous record was broken in 2018.

P12, L24, after Ansmann citation: We re-inserted a link that had been accidentally removed to a NASA website that described how Labor Day weekend smoke from the NW US reached Europe.

Table 4. The MSC and MAC values between 870 and 401 nm were adjusted slightly using a more accurate method of extrapolation. We note that both calculation methods produce MSCs in excellent agreement with the literature when used with our 2018 data that was collected with PM2.5 cyclones on the PAXs. With both procedures, the MSC values are lower with PM1 cyclones on the PAXs. We think the PM1 cyclones likely do a good job of isolating the combustion generated aerosol, but that super-micron dust and vegetative debris gets entrained in smoke plumes, transported, and affects the optical properties, which has prompted us to switch to PM2.5 cyclones for continued monitoring. Change “linear” in text to indicate based on power law fit.

P11, L35: Old text: “…were calculated using a linear regression using the calculated averages.”

New text: “…were calculated with a power law fit using the calculated average.”

Figure 3. Added caption: “b) Lab averaged BC/CO ratio versus modified combustion efficiency (MCE) separated into bins by 0.1 of MCE.

Figure 4. We had the caption for parts a and b reversed and that has been corrected.

Figure 7. Added “shown for the entirety of the monitoring period” to caption.

Figure 8. Added “BC and PM shown for the entirety of the monitoring period, but %401-Absorption by BrC only shown for when the PAX 401 was operational.”