

Interactive comment on “In-situ measurements of trace gases, PM, and aerosol optical properties during the 2017 NW US wildfire smoke event” by Vanessa Selimovic et al.

Anonymous Referee #2

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This manuscript presents measurements of some aerosol properties and some trace gases in Missoula (US) during approx. one month in August-September 2017. During this period the measurement location was affected by several smoke plumes from wild fires. Some of the fire locations were identified, but several plumes represent aged regional smoke containing emissions from various sources. Altogether this data set contains approx. 500 h of in-plume measurements and can provide valuable information on statistics of flaming vs. smoldering combustion on regional scale. However, the methods need to be described in more detail and different sources of uncertainty have to be assessed before this manuscript can be accepted in ACP.

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Major comments

My main concern is that uncertainties in the analysis are not well quantified. Uncertainties for individual instruments are presented in Section 2, but uncertainty estimates are not presented for any of the data points in the graphs.

Furthermore, it is not clear how “smoke-impacted” periods are distinguished from non-smoke periods. For instance for peak G in Fig. 1: the “smoke-impacted” BC and CO concentrations during afternoon hours are lower than during the following “non-smoke period”. Reliable differentiation between “smoke-impacted” and background periods is essential for accurate definition of excess concentrations and excess mixing ratios especially for more diluted regional smoke (e.g. peaks M, N, R, T in Fig. 1).

Many of the “smoke-impacted” periods last 24h or more. In such cases any diurnal variability in background CO, BC and PM2.5 will be a source of uncertainty, as background is apparently estimated with linear interpolation (see page 6, line 9). Can you estimate how large is the uncertainty in excess mixing ratios due to assumed linear change in background during long smoke-impacted periods?

One more source of uncertainty, which is not very well constrained, is the effect of 3.2km distance between PM2.5 measurements and other measurements. At 1h resolution and for regional scale smoke the distance is probably not an issue, but for the relatively fresh plumes (1-2 h) that distance can make a difference. Is there any difference in the correlation between scattering and PM2.5 for diluted and fresh plumes?

It seems that at the moment only one integrated excess mixing ratio is defined for each smoke-impacted period (page 6, line 9-11). However, many of the smoke-impacted periods represent considerable temporal variability. I recommend calculating excess mixing ratio at e.g. 1h or 5min temporal resolution, which would allow presenting also standard deviation (or other measure of in-plume variability) in addition to mean values in Supplementary Table 1. I think this approach would give also more representative study-average statistics. With the current approach short smoke-impacted periods

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have equal weight to long periods in the study average.

Please include also scattering/CO ratio in the analysis. I believe this would be a valuable reference in the future.

Minor comments

Please indicate the units for excess mixing ratios. Are mass concentrations given in prevailing conditions or e.g. STP?

Page 5, line 4. It seems that no truncation error correction was applied to the scattering coefficient. Please discuss shortly the uncertainty in SSA.

Page 5, line 8. Please define SSA based on scattering and absorption coefficients (Babs, Bscat defined on page 4, line 12).

Page 6, line 20-21: "Other approximate metrics of the relative amount of flaming to smoldering such as BC/CO or CH₄/CO can still be used". Are these ratios calculated as excess mixing ratio or plain concentration ratio? Please make sure that excess concentrations are always indicated with a delta (also in Figures) - now it seems that most excess mixing ratios are written without delta, i.e. as plain concentration ratio.

Page 8, line 3 and Fig. 2. Are there any previous studies to compare CH₄/CO vs. BC/CO dependency?

Page 9, line 9. I agree, but the relationship between MCE and BC/CO is not linear (e.g. Vakkari et al., 2018). Can you estimate the MCE range from BC/CO in your case?

Page 9, line 15. "The Selimovic et al. lab average" Year missing in reference, please check.

Page 9, line 24-25. "Changes in the PM/CO ratio as a plume ages can be used as a metric for the net effect of secondary formation or evaporation of organic and inorganic aerosol (Yokelson et al., 2009; Akagi et al., 2012; Jolley et al., 2012; Vakkari et al., 2014)." This method was recently applied by Vakkari et al. (2018) as well; you may

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consider adding a reference.

Page 9, line 28. "Further our lower BC/CO ratio suggests enhanced smoldering, which should increase the PM/CO." The observations by Vakkari et al. (2014, 2018) seem to indicate the opposite: fresh emission PM/CO decreasing with increasing smoldering. PM emission factor does increase with increasing smoldering, though.

Page 10, line 2-3. "The BC/PM ratio also allows for an estimate of ambient BC from ambient PM data when BC isn't measured, but caution is needed since PM may not be conserved as long as BC." BC fraction may also depend on combustion characteristics (c.f. Vakkari et al., 2014).

Page 10, line 7-8. "A previous study found that smoldering combustion emits anywhere between 4-49 times more PM than flaming combustion (Kim et al., 2018)." It seems that Kim et al. (2018) measured total PM (no size cut in inlet), which could be pointed out here. I would expect PM2.5 or PM1 emission variability be a bit less than TSP.

Page 12, line 12-13. "Figure 5 shows a moderate increasing trend in the SSA at 870 nm, but no significant trend in the SSA at 401 nm." Please state how you checked for statistically significant trend.

Page 12, line 29. "smoke was mostly sourced from a local fire (Rice Ridge)." How far was the fire? Can you estimate the smoke age?

Page 12, line 29. "Our peak-integrated proxy for particle size (4.02, smaller particle size)" Please describe the "peak-integrated proxy for particle size" in Section 2.

Figure 6 (case study). Please add a second panel with high-resolution excess mixing ratios (BC/CO, PM2.5/CO, scattering/CO, trace gases/CO) so that the reader can compare the two peaks.

Page 13, Section 3.6 Diurnal Cycles. I would expect diurnal cycle to be important for near-fire measurements due to diurnal variation in the emissions (e.g. Saide et al., 2015), oxidation and dilution. However, I would not expect much difference in aged

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regional smoke, whether it is observed during morning or evening hours. Here, focusing on extensive properties (PM2.5, BC, CO) is problematic as they depend mostly on dilution. I wonder if the diurnal cycle in Figure 7 has a small increase in morning only because more fresh plumes happened to reach the measurement site during morning hours. I recommend removing this section or concentrating on fresh plumes (e.g. CO > 0.5 or 1 ppm) and intensive properties (excess mixing ratios).

Page 14, line 11-13. "Despite our lower BC/CO ratio our PM/CO ratio was about half that measured in fresh smoke from aircraft. This suggests that OA evaporation, at least near the surface, may typically reduce PM air quality impacts on the time scale of several days." I do not think you can draw such a straightforward conclusion, as PM/CO ratio decreases with decreasing BC/CO. If both fuel and BC/CO are equal, then a lower PM/CO in aged smoke would suggest primary aerosol evaporation. Please check also abstract (page 1, line 18-22).

It seems that all linear fits are calculated with ordinary least squares method, which assumes that there is no uncertainty in x-direction. At least for Figs. 2, 3 and S1 a bivariate method would be more appropriate (see e.g. Cantrell et al., 2008).

Please combine Tables 1 and 5 to avoid repetition. Please also check that you have defined the values in parenthesis in all Table captions. Is the study average a mean of enhancement ratios defined for each plume?

References

Cantrell, C. A.: Technical Note: Review of methods for linear least-squares fitting of data and application to atmospheric chemistry problems, *Atmos. Chem. Phys.*, 8(17), 5477–5487, doi:10.5194/acp-8-5477-2008, 2008.

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