Review for Species Correlation Measurements in Turbulent Flare Plumes: Considerations for Field Measurements

General comments

Field measurements of flaring emissions are challenging and unlike those faced by other pollution sources. There are only a few measurements in the literature and most use novel measurement techniques that are difficult to verify. This paper explores an understudied question of the emission variability and its impacts on current measurement techniques and assumptions. A novel measurement method was devised to determine the temporal variability in BC/H_2O ratios measured simultaneously through a laboratory flare plume. The authors clearly present the measurement methods and thoroughly investigate method uncertainties. However, the conclusions and implications are weakly supported. In particular, the variability observed is applied to other sampling conditions without consideration of the effect that sampling technique has on variability.

Specific comments

1) The authors make the assertion that the variably observed in BC/H_2O ratios suggests that fly-by sampling of flares (Gvakharia2017, Weyant2016, and Krause2018) may be prone to large uncertainties and low bias. While these flight passes are only a few seconds and, by most measures are very short samples, they are also much longer than the near-instantaneous optical measures used here. Additionally, the volume of the plume represented by the laser beam is tiny compared to that captured in the flight passes. Considering both the differences in duration and volumes, these flights may capture over 1000 times more of the plume than a single instantaneous optical measure. Aggregating just a few seconds of the optical data would reduce the variability observed, substantially.

While levels of aggregation that better represent flight-passes could be explored in this paper, it isn't clear what amount of aggregation would be appropriate for both the temporal and volume issues. It may be a fundamentally apples-to-oranges comparison to use the variability observed at such small scales to infer the variability of samples made with much greater volumes and duration.

2) The paper seems to be missing arguments about the relationship between the BC/H_2O measured here and the ratios used in the molar/mass balance literature, i.e. BC/combusted carbon or BC/CO_2. Are BC/H_2O ratios equally as variable as BC/CO_2 ratios? It would be natural to assume that the H_2O and CO_2 produced are well correlated from the combustion equation, but the paper currently doesn't mention that this relationship is necessarily assumed.

Another potential related area to explore would be to use the H_2O to infer CO_2 and provide BC emission factor estimates using carbon balance. Presentation in this metric could both better articulate the relevance of the BC/H_2O metric and present variability using metrics readers may be more familiar with. It may also build confidence in the novel measurements conducted here, if they could be grounded around previously measured values.

3) A novel measurement technique is introduced, but not calibrated or verified using other measurement tools. The argument appears to be that because ratios, not absolute magnitudes are presented, this validation is not necessary(?). However, if there are multiplicative errors (instead of additive) then the BC/H_2O ratio variability would be affected.

4) The paper repeatedly suggests that stable BC/H_2O ratios are assumed when using molar/mass balance methods, and suggests that readers should change their understanding of the nature of flare emissions from stability to one that is highly variable. However, most emissions researchers would expect that BC emissions are highly variable; this is observed in all but perhaps the most controlled combustion conditions for any fuel. If stability was the expectation, researchers would be reporting the results of a single measurement, but they do not. (Maybe 1-6 passes of a single flare, but reporting regional averages). If the majority of

readers in the target audience do not have a prior belief that flare emissions are stable, then (in my opinion) it would not be a great choice to lean on this idea so heavily, as it may lose, or worse offend some readers.

Technical corrections

line 9: *"...emission rate (i.e. emission factors)..."* Change to "emission factors".

line 25: Acronym "UOG" not used again, so could be omitted.

line 28-29: 20/100 and 96/34 look like fractions. Perhaps another phrasing to avoid this potential confusion.

line 33: *"...of soot"*. Suggest changing to "of particulate matter", since sometimes soot is considered equivalent to BC.

line 34: Suggest "second most warming atmospheric pollutant", because there are other strongly negative climate forcers, and "important" is subjective.

line 46: Nearly all emission sources are "infrequently" measured; most individual cars, planes, industrial stacks are not measures or are infrequently measured. A stronger case may be made by quantifying the frequency of measurements relative to other sources.

line 61: I suggest changing the sentence *"The veracity of this..."* to "Assessing the veracity of this assumption is necessary to determine representative sampling strategies" (e.g. sampling volumes, number of samples, and perhaps sample timing)"

lines 59 & 63: *"instantaneous measurements"*. The above papers are not reporting instantaneous metrics as is being stated here.

line 66: *"..there has not been study attempting..."*. Change to "...no study has attempted to..."

line 67: "the first"

line 94: What is the likelihood that some of the water would have started to condense at this height? Could this impact the measured H_2O?

lines 113-114: Could the "<1 mrad beam steering that is interpreted as additional BC" be quantified in terms of measured BC? Or possibly as a fraction of the average measurement? It is hard for a non-specialist in this technique to interpret the magnitude of this error.

lines 128: Was the BC also averaged over 5 ms?

line 155: BC or soot volume fraction is a niche term. Why not use mass concentration?

line 165: Using a back of the envelope calculation, it seems like this extinction coefficient is large. If the mass extinction cross section (m^2g^{-1}) is around 4 and the particle density is 1.7g cm^{-3}, this value would be around 7x10^6 m^2m^{-3} (m^{-1}). Perhaps a more explicit explanation of the methodology used would clear up the confusion.

lines 167-175: What fraction of the light attenuation at the 1428 wavelength was estimated to be due to BC for a typical measurement?

line 241: What does "similarly averaged" mean? Was there also a wavelength sweep at 1654nm?

Figure 4: "Sample point index" is not very meaningful. Could this be plotted with time on the x-axis instead? My understanding is that this is the time within the 5ms sweep.

line 250: How much of the data was omitted due to the SSE threshold?

lines 260-267: This paragraph feels like it belongs in an "experimental design" section of the methods, rather than in Results.

line 271: A general correlation between the species is not expected "*since the production of BC necessitates the production of* H_2O". Most H_2O is produced through the generation of CO_2. A tiny amount of BC is produced relative to H_2O, thus the variably in this ratio is directly related to the variability in BC formation, which varies due to micro-physical changes in combustion conditions. Whereas, I suspect the variability observed in H_2O is primarily due to dilution. It does not seem surprising, or particularly significant that such variability was observed.

line 276-277: *"..is not explained by a linear model..."* Why should it be? Why isn't a large variance expected? Are we sure these R^2 values should be characterized as "low"?

line 286: Is there a theoretical reason why the ratios and variability were different between the two flare gas mixtures? This result seemed interesting enough to warrant inclusion of a figure, but the implications and interpretation are under-explored.

line 292-293: *"...driven by turbulent dynamics..."* I suspect that this is the main driver of variability, but it isn't clear to me how the comparable skewness of the distributions generated this conclusion.

line 331: At first glance, the skewness observed here appears to be lower than found by Conrad & Johnson (2017), which would suggest that high BC events are somewhat correlated with higher rates of combustion. How do these measurements compare to those observations?

line 349: After *"ratio..."*, add "for that individual flare".

line 366: This description of "path-averaged" and "point-sampling" doesn't correctly describe the differences between the drone samples and the path-averaged optical measurements described here. 1) The drone measurements (as well as those conducted with airplanes) are sampled through the plume, collecting a volume of air for the time it takes to pass through the plume. The assertion that these samples capture less of the plume than a single optical measurement is not convincing; the volume captured in the optical path-averaged measure is tiny. 3) It is also likely that the plume, measured further downstream than here is much better mixed than from 2m from the flare, such that even small samples are likely to have less variability in ratios. 4) The description here also implies that these measures report a single measurement as if it was representative, but they do not. Reporting is made of averages of a number of passes. 5) The description of the path-average sampling seems to suggest that more of the plume is measured than with other sampling techniques, but the volume represented by a laser beam across the plume is small and also fails to capture most of the plume, which varies spatially as well as temporally.

line 375-379: Is methane leaking germane to the discussion about combustion pollutant ratios? The idea that measured variability in BC/H_2O extends to other species should be more grounded in combustion theory. I suspect the BC/H_2O ratios have little bearing on the release of unburnt fuel, but are fundamentally related to the relative emissions of CO_2.

line 415: Is the 150 cm the distance from one side of the optical hoop to the other (from collimator to wedged window in Figure 2)? If so, what are the nodes?

line 434: How can you know the BC was *"correctly estimated to within 0.02 ppb"* without external calibration? Is this actually a measure of measurement uncertainty?

Figure A1b: It isn't clear how features within the beam are determined. Were they measured or modeled?