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**Title:** Species Correlation Measurements in Turbulent Flare Plumes: Considerations for Field Measurements
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## Point-by-point Response to Comments by Referee #2

### *General comments*

*This paper describes the application of a novel device to investigate the short-term variability of the BC/H2O ratio in a lab-scale flare using two different fuel compositions representative of the Bakken and Ecuador regions. The authors performed a Monte Carlo simulation to estimate variation and skewness in the BC/H2O ratio. They found that high ratios can be related to high BC production and further discuss the impact it could have on uncertainty and field measurements that assume a constant ratio.*

*The topic of this study is in the scope of the Atmospheric Measurement Techniques Chemistry journal and addresses an important subject relevant to field measurements of flaring emissions. Nevertheless, the implications of the study would benefit with more clarification and the Conclusions need to be improved since they read very similar to the Abstract.*

We thank for your thoughtful input on our manuscript and your highlighting of the importance of this subject for emissions quantification. We have responded to your comments in detail below and have revised the conclusions to reduce overlap with the abstract as requested. Thank you for your help in improving the clarity of our manuscript.

### *Specific comments*

1) *In Section 4.1 the authors mention the impact that crosswind might have on the short-term variability of the ratio. As part of the experimental method they also use data from CFD simulations of flares under crosswind conditions. However, the authors use a vertical flame to conduct the measurements of the BC/H2O ratio and in the manuscript it is not mentioned if they conducted experiments under crosswind conditions and why. My question is if the results obtained with a vertical flame can be representative also for crosswind conditions. What are the implications for the experimental device / setup to explore that? Are the histograms of both the sample mean and ratio distribution subject to change, and if so, to what extent?*

Although crosswind experiments are not possible in the current facility, as discussed in Section 4.1, we expect the presented experimental results for vertical flares without crosswind represent the most conservative case for minimizing species variability effects. Under crossflow conditions the radial symmetry of the plume is lost and experiments (Poudenx, 2000) have shown that crosswinds can cause separation of gas phase species into different regions of the plume, with different local carbon conversion efficiencies. Further experiments (Johnson et al., 2001) and flow visualization (Johnson and Kostiuk, 2002) have also demonstrated how crosswinds can cause intermittent bursts of unburnt fuel (methane) to be emitted from the underside of the wind-deflected flame. These bursts of unburnt fuel are spatially separate from the $CO_2$-rich plume and are thermally distinct, suggesting that $CO_2$ and methane would be uncorrelated in the plume. Although these results speak to variations in gaseous species ratios in crossflow, it is reasonable to assume that they would only serve to further complicate measurements reliant on well-correlated species assumptions. However, looking toward future experiments, the presented device and methodology could be directly applied in future measurements in a wind tunnel or similar facility. This is presently being explored by close collaborators.

*2) Regarding the implications of the study, the authors suggest that based on the results of the BC/H2O ratio, aircraft-based estimates may be under-sampled and the estimated black carbon emission rate would be biased low. However, the techniques might not be comparable between each other. Factors like a large variability in the operating conditions which can impact the estimates substantially even from day to day [Conrad and Johnson, 2017], or the difference in the volume sampled between these approaches are not discussed in detail in the manuscript. In addition, the measurements in the paper were obtained with freshly emitted BC which might not be the case for those obtained with an aircraft, especially those sampled kilometers* *downwind. A brief discussion considering differences between non-intrusive and extractive sampling techniques will add more clarity to the paper.*

We agree that different techniques will be more or less susceptible to species ratio variations, and have added further discussion to the implications section (4.1) to make this more clear. As noted in the text, our optical system was not intended to reproduce any specific aircraft or other measurement approach, but "to determine whether variations in species ratios in a turbulent plume could be detected, and if so, to examine how these might affect field measurements that rely on the assumption of a fixed ratio as part of a molar- or mass-balance to quantify pollutant emission rates or combustion/destruction efficiency." Ultimately, the results showing both strong variation and skewed distributions of path-averaged $BC/H_2O$ ratios do have important implications for a range of approaches. As noted in the revised text, "in general, shorter path measurements (e.g., extractive samples from fixed probes or stationary drones) are expected to observe greater variability in species correlations without the benefit of long path averages to smooth out relative species variations." Spatial coverage of the inhomogeneous plume (see Poudenx, 2000) is also important as elaborated in Sect. 4.1. The potential relevance of sample volume would depend on the relative portion of the plume being captured, where we note,

> *This may be less of an issue in full-sized aircraft sampling approaches (e.g., Gvakharia et al., 2017; Weyant et al., 2016) which would be expected to measure a much larger volume than the presented technique; however, in general, the required data averaging to obtain representative results will depend on the relative plume volumes sampled at that downstream measurement position, sampling duration, and how the technique interrogates the plume (e.g. stationary point or line-of-sight, transect downwind or crosswind, etc.).*

It is also important to distinguish between variable BC emission rates and variable $BC/H_2O$ ratios which can be quite different. The cited time-resolved measurements in Figures 1 and 2 of Conrad and Johnson (2017) show that mean and instantaneous BC emission rates [g/s] are highly variable. However, prior to the present work it was not clear from the literature whether BC would be well-correlated with other combustion products, or specifically if species ratios, e.g. $BC/H_2O$ or $BC/CO_2$, might still be stable. The results suggest that variability in species ratios (e.g. $BC/H_2O$) is an added factor to be considered (i.e. in addition to variability in raw BC emission rates), where both are important in designing measurement campaigns to maximize accuracy.

## *Technical corrections*

*Ln 7. Briefly state why these measurements are important.*

We have added this sentence to the abstract,

> *Incomplete combustion from these processes results in emissions of black carbon, unburnt fuels (methane), $CO_2$, and other pollutants.*

*Ln 21. Replace 'should be easily avoidable'. Consider rephrasing.*

We have rephrased this to, "can be avoided."

*Ln 27. Consider rephrasing 'up 3% from 2018' to something like (just an example) 'up to 3% higher than in 2018'.*

This sentence now reads,

> *…estimated to be 150 billion $m^3$, an increase of 3% from 2018, …*

*Ln 28. Maybe mention that methane is also considered a Short Lived Climate Pollutant (SLCP). It is indirectly mentioned for Black Carbon on Ln 38.*

We have revised the sentence to read:

> *Of the pollutants produced by gas flaring, black carbon (BC; the carbonaceous, strongly light-absorbing component of particulate matter) and methane are both Short-Lived Climate Pollutants (SLCP) that, along with $CO_2$, constitute the three most climate warming pollutants in the atmosphere (e.g. Bond et al., 2013; IPCC, 2013; Jacobson, 2010; Ramanathan and Carmichael, 2008).*

*Ln 66. Change 'been study' to 'been a study'.*

Thank you for catching this typo. The line now reads,

> *…no study has attempted to directly inspect this potential issue*

*Ln 77. This sentence is not clear. Are these gas mixtures representative for both off-shore and on-shore facilities worldwide, or just for specific regions? A brief sentence would be good to clarify and help introduce the oil regions on Ln 86.*

We have removed the reference to flare gas mixtures in this opening sentence and to improve clarity, the following sentences now include the composition-relevant text originally at line 86 followed by a new sentence explaining the selection of these mixtures considering the relationship between heating value on sooting propensity.

*Ln 78. remove 'in schematic'.*

Removed as part of previous edit.

*Ln 84-85. Consider moving this sentence to the previous paragraph and merge with Ln 81.*

Thank you for the suggestion. We have merged the sentence into the previous paragraph as part of the previous two edits.

*Figure 1. Please increase the font size. It can be hard to read.*

We have increased the font size in Figure 1 as suggested.

*Ln 96. Consider changing 'well away' to 'far enough' or similar wording.*

Changed to, "…far away from the flame."

*Ln 190. It is mentioned that the profile fitting technique was implemented to laminar flames. Is it specific for laminar flames? What are the basis to use it for turbulent flames like those in the paper?*

No, the technique is not specific to laminar flames. Although Liu et al. (2007) experimentally demonstrated the profile fitting technique on laminar flames measuring $H_2O$, the fitting approach was introduced more generally for nonuniform flow fields. More recently, Grauer et al. (2018) successfully used the same profile fitting approach to measure heated, turbulent gas escaping a vertical nozzle. Nevertheless, as detailed in Appendix A.1, as part of this paper we conducted extensive simulations of the technique using large eddy simulation data, where we directly tested the ability of the algorithm to correctly reproduce known path-averaged temperature and $H_2O$ concentrations. Results of this effort further proved the applicability of the technique and informed the subsequent Monte Carlo uncertainty analysis.

*Ln 274. Why is the error in H2O volume fraction much higher than the error in BC volume fraction?*

The $H_2O$ measurement is derived from the profile fitting technique, in which absorption measurements across two $H_2O$ absorption lines are used to simultaneously quantify the path-averaged $H_2O$ volume fraction and temperature distributions (see previous response and Sect. 3.3). BC volume fraction, however, is directly measurable from the attenuation signal on the 1654 nm laser. The difference in uncertainties is a direct result of the added steps required for the $H_2O$ measurement.

*Ln 286. "an apparent dependence on flare gas composition." It is an interesting result but it was only mentioned and not discussed with more detail. Did you consider the effect of the composition on soot chemistry ? According to Table 1, Ecuador composition has more branched-chain isomers. It could produce different intermediate radicals, which affect the formation of molecular soot precursors [Lei et al., 2020].*

Thank you for this suggestion. Studies by McEwen and Johnson (2012) and Conrad and Johnson (2017) have demonstrated a relationship between fuel higher heating values and black carbon yield. In our experiment, we would expect the higher BC production observed from the Ecuador mixture because of its larger heating value (see Table 1 in the manuscript). Although the Ecuador mixture produces a higher BC yield, its production of $H_2O$ is roughly the same as the Bakken mixture, resulting in an increase BC/$H_2O$ ratio. We have updated the manuscript to include this discussion.

*Ln 705. Figure 2. Change 'shown' to 'show'.*

Typo corrected, thank you.

*Figure 3. include the meaning of qamb/Tamb, etc also in the Figures' description.*

The figure caption now includes relevant definitions. Thank you for pointing this out.

*Figure 5. Spell out 'NM' in the Figure's description.*

Spelled out "Nelder-Mead" as suggested.

*Ln 291. Remove the hyphen, it might be read as a negative value.*

Removed as suggested.

*Ln 340. remove 'etc.'*

Removed as suggested.

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