Manuscript ID: amt-2020-255
Title: An uncertainty-based protocol for the setup and measurement of soot/black carbon emissions from gas flares using sky-LOSA
Authors: Conrad, Bradley M.; Johnson, Matthew R.

Point-by-point Response to Comments by Referee

The authors Conrad and Johnson have addressed most of my feedback. I only have two comments remaining.

Length: the authors responded to my request to shorten the manuscript by stating various justifications. These should be added to the text with one or two introductory sentences. The same applies to the authors' response to my comment on the presence of non-BC impurities. Please add a brief summary to the main text.

1) We have revised the introductory paragraph to Section 3 ("General uncertainty analysis methodology") to highlight our justification for the comprehensive methodology section, and specifically that "[t]his new methodology is a significant improvement to the sky-LOSA algorithm that enables accelerated MC-computation of soot column density and, hence, emission rates from sky-LOSA image data".

2) We have included additional text to the paragraph following Equation (3) to highlight that absorption-enhancing non-BC material is not likely to be observed in flare plumes, especially in the near field, according to both laboratory (Kazemimanesh et al., 2019) and field data (Schwarz et al., 2015; Weyant et al., 2016), which justifies the literature-derived probability distributions used in sky-LOSA (Johnson et al., 2013).

Validation of assumptions: the authors' response to my comment ("The manuscript assumes throughout that a perfect skyLOSA measurement gives a perfect result") was "sky-LOSA has been validated [...] from the perspective of first principles." The authors full response included a restatement of the bottom-up work done so far. This work is extensive and thorough, especially in the context of atmospheric measurement techniques, but does not address my comment. The purpose of my comment is to highlight that bottom-up work requires top-down validation (to the extent that such is possible).

For example, Thomson et al. (2004, https://doi.org/10.1016/j.combustflame.2004.11.012) state that "The uncertainty of the LOSA soot volume fraction measurements is estimated to be 20 to 30% (95% confidence interval). The uncertainty is dominated by the uncertainties in the magnitude of and the contribution of scatter to light attenuation measurements." So the authors may add a statement such as "The LOSA technique is estimated to be 20 to 30% accurate. We estimate that the calculation of the inscattering of solar radiation adds an additional X\% to this uncertainty using our MC technique, resulting in an overall uncertainty of Y%." Please add such a statement to the manuscript.

Overall measurement uncertainties vary with field conditions and equipment setup, which is indeed the motivation behind this paper. For most real-world field conditions, overall sky-LOSA uncertainties are in the range of −26/+36% (e.g., Johnson et al., 2013) – we have added text to specifically note this in the penultimate paragraph of Section 2 where the sky-LOSA method is described. Importantly, as detailed in the manuscript, sky-LOSA uncertainties are rigorously calculated on a case-by-case basis via a Monte Carlo method, where fundamental soot properties and field conditions (sky and solar radiation) are treated as random variables. This contrasts with the cited earlier work of Thomson et al., (2005), where uncertainties in their laboratory-based collimated-LOSA technique were estimated by summing selected uncertainties in quadrature (Thomson, 2004): $E(m_\lambda)$, the scatter-to-absorption ratio (related to the single-scatter albedo, $\omega(\boldsymbol{b})$), and diagnostic-specific measurement uncertainties that are unrelated to sky-LOSA.

We agree with the Referee that it is important to validate bottom-up measurements with a top-down alternative to the extent that such is possible. The challenge – as noted by the Referee – is that top-down validation can only be performed if a satisfactory alternative measurement technique exists. Echoing our first response to the Referee, this is unfortunately not the case for sky-LOSA since the current measurement "standard" for flare soot/BC emissions is a visible assessment of plume opacity by a human observer (U.S. EPA, 1974). There are aircraft-based (top-down) techniques that use atmospheric measurements of BC and combustion product concentrations to infer flare BC emissions based on an assumed flare gas composition (Gvakharia et al., 2017; Schwarz et al., 2015; Weyant et al., 2016). However, because BC emission rates are very strongly dependent on fuel composition (e.g., McEwen and Johnson, 2012), and due to the stochastic nature of flare BC emissions (Conrad and Johnson, 2017), it is not obvious how these approaches can resolve robust emission rate statistics sufficient to serve as an additional validation for sky-LOSA.

References

Conrad, B. M. and Johnson, M. R.: Field measurements of black carbon yields from gas flaring, Environ. Sci. Technol., 51(3), 1893–1900, doi:10.1021/acs.est.6b03690, 2017.

Gvakharia, A., Kort, E. A., Brandt, A. R., Peischl, J., Ryerson, T. B., Schwarz, J. P., Smith, M. L. and Sweeney, C.: Methane, black carbon, and ethane emissions from natural gas flares in the Bakken Shale, North Dakota, Environ. Sci. Technol., 51(9), 5317–5325, doi:10.1021/acs.est.6b05183, 2017.

Johnson, M. R., Devillers, R. W. and Thomson, K. A.: A generalized sky-LOSA method to quantify soot/black carbon emission rates in atmospheric plumes of gas flares, Aerosol Sci. Technol., 47(9), 1017–1029, doi:10.1080/02786826.2013.809401, 2013.

Kazemimanesh, M., Dastanpour, R., Baldelli, A., Moallemi, A., Thomson, K. A., Jefferson, M. A., Johnson, M. R., Rogak, S. N. and Olfert, J. S.: Size, effective density, morphology, and nano-structure of soot particles generated from buoyant turbulent diffusion flames, J. Aerosol Sci., 132, 22–31, doi:10.1016/j.jaerosci.2019.03.005, 2019.

McEwen, J. D. N. and Johnson, M. R.: Black Carbon Particulate Matter Emission Factors for Buoyancy Driven Associated Gas Flares, J. Air Waste Manage. Assoc., 62(3), 307–321, doi:10.1080/10473289.2011.650040, 2012.

Schwarz, J. P., Holloway, J. S., Katich, J. M., McKeen, S., Kort, E. A., Smith, M. L., Ryerson, T. B., Sweeney, C. and Peischl, J.: Black carbon emissions from the Bakken oil and gas development region, Environ. Sci. Technol. Lett., 2(10), 281–285, doi:10.1021/acs.estlett.5b00225, 2015.

Thomson, K. A.: Soot Formation in Annular Non-premixed Laminar Flames of Methane-Air at Pressures of 0.1 to 4.0 MPa, University of Waterloo., 2004.

Thomson, K. A., Gülder, Ö. L., Weckman, E., Fraser, R., Smallwood, G. J. and Snelling, D. R.: Soot concentration and temperature measurements in co-annular, nonpremixed CH4/air laminar flames at pressures up to 4 MPa, Combust. Flame, 140(3), 222–232, doi:10.1016/j.combustflame.2004.11.012, 2005.

U.S. EPA: Visual Determination of the Opacity of Emissions from Stationary Sources, Code of Federal Regulations, Title 40, Part 60, Appendix A-4, Method 9, United States of America., 1974.

Weyant, C. L., Shepson, P. B., Subramanian, R., Cambaliza, M. O. L. L., Heimburger, A., Mccabe, D., Baum, E., Stirm, B. H. and Bond, T. C.: Black carbon emissions from associated natural gas flaring, Environ. Sci. Technol., 50(4), 2075–2081, doi:10.1021/acs.est.5b04712, 2016.