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Title: An uncertainty-based protocol for the setup and measurement of soot/black carbon emissions from gas flares using sky-LOSA
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Point-by-point Response to Comments by Referee #3

This manuscript presented an uncertainty-based guide/instruction for potential users of the sky-LOSA technique to measure soot/black carbon emissions from flares in the oil and gas industry. Although the method itself and the details of various models involved in the data analysis have been published by the authors, the paper is useful to help potential researchers/engineers to better understand and apply this promising technique. The paper is well written as deserves publication. I have few questions/comments for the authors to consider.

We thank the Referee for their positive comments and recognition of the value of the manuscript and software tool to help others to "better understand and apply this promising technique".

1. In line 29 on page 4, please explicitly list the eight soot properties.

We have appended a list of these eight soot properties to the noted paragraph. Further to the comments of Referee #1, we have also included the properties and their distributions in Table 2 of the revised manuscript.

2. In the derivation of Eq. (4), do you need to assume that the mass-normalized extinction cross section of soot is constant everywhere in the flare? Otherwise, it cannot be taken out of the second integral on the right-hand side of Eq. (4).

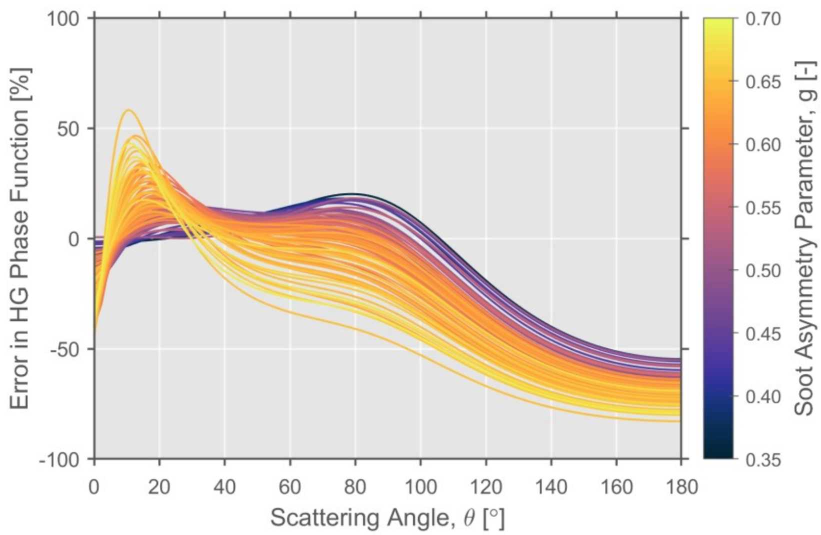
To remove the mass-normalized extinction cross-section from this integral we must assume that it does not vary in space. This assumption is supported by spatially resolved measurements of soot properties within the literature. For example, Köylü and Faeth's (1992) measurements of soot morphology in the overfire region of large-scale buoyancy-driven turbulent diffusion flames have shown that "the structure of soot … is relatively independent of … position in the overfire region" – and stated more conclusively in a later publication (1994): "…soot structure is independent of position in the overfire region…". We have clarified this assumption in Section 2 of the revised manuscript.

Although we inherently assume spatiotemporal uniformity of the soot properties, it is important to note that these soot properties are independently perturbed within sky-LOSA's Monte Carlo analysis to bound the influence of their uncertainty on sky-LOSA-computed emissions.

3. In this work, the authors dealt with the in-scattering term by expanding the SPF in Fourier-Legendre series. As the authors admitted, this expansion requires a large number of terms to have an accurate representation of the SPF, especially when it is highly forward peaked. There are other methods to more efficiently deal with highly-forward peaked SPF, such as the Henyey-Greenstein and transport approximations, have the authors considered such approaches in the calculations of the in-scattering terms?

Although there are a few different options for modelling scattering phase functions (SPFs), the Fourier-Legendre expansion has the advantage of being numerically exact, even if more computationally demanding. Moreover, while not as efficient as the HG phase function, calculation of the Legendre coefficients was quite rapid when following the approach of Schuster (2004) such that their computation was negligible compared to the execution of the Monte Carlo method of the present general uncertainty analysis.

Early in this research we did consider the Henyey-Greenstein (HG) phase function as suggested by the reviewer, but ultimately decided that it was not suitable for the sky-LOSA technique. To elaborate, the HG phase function is a simple one-parameter model that approximates the soot/Legendre coefficients via $\Phi_l = g_{HG}^l(2l+1)$ (e.g., Boucher, 1998), where g_{HG} is the "anisotropy factor" of the phase function that is usually estimated via least-squares fitting to the RDG-computed SPF (e.g., Daun et al., 2008). While the HG phase function renders calculation of the soot coefficients trivial, our experience has shown that the single-parameter HG phase function may not always capture the highly asymmetric SPFs that are typical of soot, especially larger aggregate populations. The below figure plots the angle-resolved error of the HG phase function for a range of soot properties obtained using the prior distributions in the manuscript – here, the colour of each plot represents the asymmetry parameter of the soot population. The figure shows that errors can exceed 50% but, more importantly and perhaps unsurprisingly, these errors tend to be highest for large aggregates where the SPF is more asymmetric. This implies that the HG phase function is a poorer alternative in the context of sky-LOSA, where measurement uncertainty is largely controlled by the forward scattering of sky- and sunlight into the camera (as discussed in the text surrounding Fig. 4 in the original manuscript).



4. In this paper, the authors did not provide the detailed soot properties, but reference to a previous study. It appears that the authors assume that the soot properties are uniform over the entire flare under consideration and also remain the same from one flare to another. Is this correct? If yes, the authors need to justify this assumption. The soot properties in a flare with soot emission reduction measure (such as partial premixing) may not be the same as those without any such measures.

While we defensibly assume spatiotemporal uniformity of soot properties *within a given draw* of the Monte Carlo (MC) analysis (see the Referee's second comment), it is important to note that these soot properties are perturbed within the MC method over notably broad prior distributions. These prior distributions were derived from an exhaustive literature review of soot property data (Johnson et al., 2013), including studies of soot generated by flames of varying scales and configurations (premixed/non-premixed) burning fuels that extend well beyond typical flare gas compositions. Despite the likely variability of soot properties from flare to flare, the breadth of these priors suggests that sky-LOSA confidence intervals are likely to bound the ground truth emission rate; for this reason, the same prior distributions of soot properties are used in all sky-LOSA analyses.

5. The choice of sky model group seems not straightforward and has a strong influence on the measurement. I wonder if the authors can provide more useful 'tips' to make such a choice for new users of sky-LOSA.

Sky categories/groups are selected using criteria laid out in the rightmost column of Table 1. These criteria are based on simple observations by the sky-LOSA user such as whether the sun is obstructed or unobstructed and whether the sky is overcast, partly cloudy, or clear. Sky-LOSA-calculated soot emissions are indeed sensitive to the selected CIE sky model; however, *within* each of the defined sky categories/groups (e.g., highly turbid overcast skies (category/group "A")), different sky models generally have similar effects on computed emissions and uncertainties. In fact, it is this weak variation in the effect of the models within a sky category/group that allows us to define the sky categories/groups and ultimately treat the sky model as a random parameter within the MC analysis with minor impact on measurement uncertainty.

To clarify how a user might select the most appropriate sky category/group, we have added text to Section 3.1.2 of the revised manuscript noting the simple selection criteria.

References

Boucher, O.: On Aerosol Direct Shortwave Forcing and the Henyey–Greenstein Phase Function, J. Atmos. Sci., 55(1), 128–134, doi:10.1175/1520-0469(1998)055<0128:OADSFA>2.0.CO;2, 1998.

Daun, K. J., Thomson, K. A. and Liu, F.: Simulation of Laser-Induced Incandescence Measurements in an Anisotropically

Scattering Aerosol Through Backward Monte Carlo, J. Heat Transfer, 130(11), 1–10, doi:10.1115/1.2955468, 2008.

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.

Köylü, Ü. Ö. and Faeth, G. M.: Structure of overfire soot in buoyant turbulent diffusion flames at long residence times, Combust. Flame, 89(2), 140–156, doi:10.1016/0010-2180(92)90024-J, 1992.

Köylü, Ü. Ö. and Faeth, G. M.: Optical properties of overfire soot in buoyant turbulent diffusion flames at long residence times, J. Heat Transfer, 116(1), 152–159, doi:10.1115/1.2910849, 1994.

Schuster, G. L.: Inferring the Specific Absorption and Concentration of Black Carbon From Aeronet Aerosol Retrievals, Pennsylvania State University., 2004.