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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 #1

The manuscript amt-2020-255 by Conrad and Johnson describes a software heuristic for assisting a remote optical technique (skylight line-of-sight attenuation; skyLOSA) for measuring soot/black carbon emissions in large industrial flames. The technique allows a user to select the most reasonable position to set up the skyLOSA camera for a given set of flare and sky conditions. The computations behind this technique are intensive, so the manuscript spends some time describing a useful pre-computation approach. The pre-computed values are later used as inputs to a Monte Carlo uncertainty calculation.

From the skyLOSA perspective, the manuscript does not present new concepts or results. The main novel concept of this work is to apply the same theory used during detailed analysis to measurement, so that the measurement can be carefully configured to provide optimal results. This is a generally interesting concept, but could be considered as a technical note rather than a manuscript.

I recommend substantially shortening the manuscript's description of skyLOSA in order to reflect the subsequent conclusions. I also have reservations about the assumptions made in the MC analysis, especially the assumption that the flame emits only soot and no volatiles. These and other comments are detailed below.

We thank the Referee for their review of this manuscript and have addressed their comments on a point-by-point basis below. While this manuscript does not present new sky-LOSA *field measurement data*, it does describe novel and significant improvements to the sky-LOSA technique. These noteworthy advancements enable both a thorough general uncertainty analysis and considerably accelerated post-processing of sky-LOSA data, as detailed in our responses below. Moreover, the new open-source software tool developed, tested, and released in conjunction with this manuscript, extends the presented general uncertainty analysis data and, for the first time, allows in-field estimates of uncertainty at the time of measurement, greatly simplifying the use of the sky-LOSA technique and opening it up to others.

Based on the Referee's feedback we have nevertheless made revisions to shorten the manuscript, while further strengthening the discussion and referencing. These include a reduction in the overall length by moving some details of the employed Monte Carlo (MC) method to a new appendix, Appendix A. While the validation of assumptions noted as technical concerns by the reviewer have been largely addressed in previous work, we have made these references clearer in the manuscript and have revised text where necessary to ensure the justification of assumptions in the present work is evident.

General Comments:

Length. *The manuscript often reads like a hybrid between a doctoral thesis and an instrument manual, especially in Sections 2, 3, 4.1.2, and 4.2. The text is well written, but inappropriately long. The audience here is not reading to reproduce skyLOSA calculations, but to understand the general concepts used. Please either cite other work or move this text to a supplement. This text can be replaced by short descriptions focussed on key concepts.*

The presented methodology – specifically the expansion of the scattering phase function, creation of sky categories/groups, and implementation of the variance-reducing MC method – is a novel and significant advancement to the sky-LOSA algorithm that was necessary to enable the presented general uncertainty analysis. Moreover, this new approach is now used in the post-processing phase of sky-LOSA to more-efficiently compute flare BC emissions from image data. Therefore, to explicitly justify necessary assumptions and to support others in the analysis of sky-LOSA data, we chose to describe the procedure in detail. However, we do agree that some of this theory can be placed in an appendix without sacrificing the readability of the manuscript and the understandability of the results and implications.

To reduce the length of the main manuscript, we have made the following revisions:

- Merged original Sections 3.1.1 and 3.1.2 into a single section (Section 3.1.1 in the revised document) that describes the Fourier-Legendre expansion of the scattering phase function (SPF) and provides the updated framework for computing skylight and sunlight inscattering.

- Moved original Section 3.1.3, describing how we model solar irradiance, into Appendix A (Section A.2).

- Moved original Section 3.1.4, describing the truncation of the Fourier-Legendre-expanded SPF, into Appendix A (Section A.1) – see also our response to the Referee's last minor comment.

- Shortened original Section 3.2.1 and merged it with original Section 3.2 – see also our response to the Referee's first minor comment.

We have also revised text in Section 3 to identify that the presented theory represents the current standard approach to analyzing sky-LOSA data; specifically, "[t]his section describes the MC method used in the present [general uncertainty analysis] including novel updates to the MC approach that are necessary to make this present work tractable and significantly accelerate future sky-LOSA analyses."

Length (continued). *Similarly, too many acronyms are used in these sections and are not used frequently enough to be necessary (including ET, SPF, CM-LHS, ...) and not all symbols are defined next to their equations (e.g. L(b) in Equation 9 and ak next to Equation 18).*

We have deleted unnecessary acronyms in the manuscript and clarified symbols/nomenclature.

Monte Carlo clarification. *A Monte Carlo calculation randomly samples prior distribution(s) and repeats a calculation in order to obtain a posterior distribution of results. The key question here is what priors were assumed, and how accurate are they? The manuscript glosses over this point and takes the MC output as correct without any top-down validation. Please revise Section 3 and Table 2 to emphasize the prior distributions used. The authors have already done this in their earlier work (Johnson et al., 2013, Table 2) by tabulating "Distributions used in MC". I believe the authors did intend to include this information but I do not find it clear enough.*

Prior distributions for the eight fundamental soot properties required by the sky-LOSA method were derived from a thorough literature review of soot generated from flare-like flames. We originally referenced the source for these distributions (Johnson et al., 2013) but have now included them in the updated Table 2 for completeness. Further to the comments of Referee #3, we also now list the properties at the first mention of vector ***b*** in Section 2.

Monte Carlo clarification (continued). *In Table 2 of this work, the last column "MC Implementation" is specified as "MC-randomized" multiple times – this is a meaningless statement. Of course the MC calculation performs random sampling.*

We have removed the term "MC- (Monte Carlo)-randomized" and instead refer to variables perturbed within MC analyses as *random* variables.

Validation of assumptions. *The manuscript assumes throughout that a perfect skyLOSA measurement gives a perfect result. This has not been justified in the manuscript nor in earlier skyLOSA work, to my knowledge. These calculations are not constrained by any direct measurements. The skyLOSA approach is comparable to a satellite retrieval algorithm and requires direct validation. Until directly validated, this limitation must always be repeated.*

In the case of sky-LOSA, there is no comparable quantitative reference standard for time-resolved soot/BC emissions from open flames like gas flares; the current measurement standard is an assessment of plume opacity by a human observer (U.S. EPA, 1974). However, significant validation work of the sky-LOSA approach and underlying assumptions has indeed been performed using a range of alternate approaches, as necessary.

The novel aspect of sky-LOSA is the quantification of soot/BC mass column density along a line-of-sight traversing a plume using radiometric observations coupled with radiative transfer considerations. As the enabling component of sky-LOSA, this theory (presented in Section 2 of the original manuscript) has been the focus of a range of validation efforts. As referenced in the first line of Section 2, the first-generation of the sky-LOSA approach was validated by Johnson et al. (2010) against a commercial laser-induced incandescence instrument and the proven lab-based diffuse-LOSA measurement (Thomson et al., 2008) on which the technique is ultimately based. Proof-of-concept field measurements were subsequently performed in Uzbekistan (Johnson et al., 2011). Johnson et al., (2013) then extended the earliest theory to consider the effect of inscattering of skylight and sunlight that bias the perceived opacity of the plume and, hence, emission rate. In this same work, they derived the prior distributions of soot/BC properties used in the present sky-LOSA method to bound the influence of uncertain soot/BC properties on computed emissions. This theory was used in field measurements in Mexico and Ecuador (Conrad and Johnson, 2017; Johnson et al., 2013).

At this point, sky-LOSA had been validated in its ability to quantify soot/BC mass loading and the theory developed to robustly consider uncertain soot/BC properties and inscattering effects. Validation efforts then turned to two final radiative transfer considerations that could influence sky-LOSA's ability to accurately quantify soot/BC loading. Conrad et al. (2020a) leveraged large-eddy simulations of flares to prove that the effect of refractive index gradient-driven beam steering in flare plumes was negligible in the visible spectrum where sky-LOSA data are acquired. Then, Conrad et al. (2020b) amended the sky-LOSA theory to consider the minor/second-order effect of multiple scattering on the quantification of inscattered light in sky-LOSA analyses. From the perspective of first principles, these analyses completed our efforts to validate the sky-LOSA technique.

To direct the interested reader to these substantial validation efforts, we have revised the first line of Section 2 to reference validation-related publications.

Validation of assumptions (continued). *The concentrations reported by the current approach are a type of "equivalent black carbon" defined by the authors' assumptions.*

As discussed in the seminal works of Andreae and Gelencsér (2006) and Petzold et al. (2013), light absorption measurements of "black carbon" mass are inherently defined by the assumed optical properties of the absorbing particulate. These manuscripts both suggest use of the term "equivalent black carbon" for absorption-based techniques although this term has not been broadly adopted within the literature. Instead, it is generally implied that BC measured using such optical techniques is inherently "equivalent BC".

Thus, we absolutely agree with the Referee and have revised the text in Section 2 to specifically note that sky-LOSA-reported mass emissions are dependent upon the prior soot/BC property distributions:

> *"…, it is worth noting that these prior distributions of soot/BC properties inherently link light absorption measurements and computed mass column density/emissions using sky-LOSA. Thus, in keeping with Andreae and Gelencsér (2006) and Petzold et al., (2013), sky-LOSA-inferred soot/BC mass might be called 'equivalent BC' as is recommended for all light absorption-based diagnostics."*

Validation of assumptions (continued). *My main concern here is with respect to the aerosol optical properties, which have not been discussed at all. Instead, Johnson et al. 2013 is cited. The authors have assumed that the flame emits only soot. What about organics, which may condense when the plume cools? The photograph in Johnson et al. 2013 clearly suggests that the plume may have cooled before measurement. What about inorganics such as sulfates? How pure are the fuels burnt in these flares? Any impurities are likely to influence the aerosol optical properties. Please add calculations where black carbon is assumed to be mixed with organics or other impurities, using reasonable and literature-based assumptions, and show how the conclusions of this work change in response.*

The Referee's main concern is that the presence of non-BC material in flare plumes could bias sky-LOSA-computed soot/BC emissions by their influence on aerosol optical properties. While this is somewhat at odds with the notion of "equivalent BC" (see the Referee's previous comment) – i.e., inferred mass is *operationally defined* by the assumed optical properties – numerous observations in the literature show that non-BC material does not contribute to optical observations of fresh flare plumes in the visible spectrum.

In theory, both internal and external mixtures of soot/BC with other material could bias sky-LOSA-calculated emissions by changing the optical properties of particulate in the flare plume:

1. If emitted soot/BC were internally mixed/coated with a scattering material (via condensation of co-emitted organic species, for example), then emitted BC could have enhanced absorption (e.g., Bond et al., 2006).

2. Alternatively, if emitted soot/BC were externally mixed with significant co-emitted primary (and secondary) aerosols, the effective absorption/scattering by the plume could be different than that of soot/BC alone.

For the specific case of gas flares, it has been observed both in the laboratory and the field that optically active material in the visible spectrum in flare plumes is solely soot/BC. First, in their field measurements in the Bakken region, Schwarz et al., (2015) showed that "flaring BC was not associated with optically significant internally mixed non-BC material or with significant emissions of non-BC-containing primary aerosol". Similarly, Weyant et al., (2016) concluded in their field measurements that the presence of non-BC aerosols in a flare plume is "not statistically different from zero". Importantly, sky-LOSA measurements are performed in the very near-field of the flame – e.g., the noted Fig. 2 in Johnson et al. (2013) shows sky-LOSA measurement of emissions over a control surface that is within meters of the flame tip. While the plume has indeed cooled at this location (likely within 200 K of ambient temperature (Poudenx, 2000)), the soot/BC particulate is very fresh relative to typical atmospheric soot/BC that may be internally mixed with other materials. In fact, the aforementioned field studies were performed well downstream of the flame (at the basin level in the case of Schwarz et al., (2015) and hundreds of meters downstream in the case of Weyant et al., (2016)), where particles would have been subject to much more atmospheric aging and thus much more likely to be mixed with non-BC material.

These field observations are supported by laboratory studies of freshly emitted soot/BC from large flare-like flames burning fuels representative of global oil and gas flaring. Specifically, electron micrographs from Kazemimanesh et al. (2019) identify that co-emitted organics are not present as a coating on flare-emitted soot/BC. This is likely because emitted organics are largely unburned fuel (Johnson et al., 2001) which are dominated by highly volatile, low molecular weight hydrocarbons (e.g., Conrad and Johnson, 2019).

Thus, these literature data conclusively show that fresh flare particulate as measured by sky-LOSA is soot/BC, which supports our use of the literature-derived flare soot/BC properties from Johnson et al. (2013).

Validation of assumptions (continued). *A lesser concern is the assumption (Section 3.1.3) of an ideal clean atmosphere. What about background aerosol? Surely the air around an oil field is not perfectly clean.*

The sky-LOSA method does not inherently assume an ideal clean atmosphere. The original Section 3.1.3 (now Appendix Section A.2) discussed how we model ground-level solar irradiance in the context of the sky-LOSA Monte Carlo method. We believe that the Referee is referring to the "ideal extinction for a clean atmosphere at a given relative air mass" ($\sigma^{e^*}(m)$) that is used to compute the ground-level solar irradiance from the extra-terrestrial solar irradiance. Importantly, this ideal extinction is multiplied by the sky-dependent turbidity factor ($T(a)$) to represent extinction through a non-ideal (i.e., polluted) atmosphere. That is, an ideal clean atmosphere is ***not*** assumed, but is scaled to represent a range of realistic atmospheric conditions. To clarify this in the revised manuscript, we have included the new text (in bold):

> *"…σ^{e^*} is the ideal extinction for a clean atmosphere at a given relative air mass, and $T(a)$ is the model-dependent turbidity factor* ***that is used to consider realistic atmospheres and describes*** *the number of clean atmospheres required to represent* ***attenuation through the non-ideal, polluted atmosphere****."*

Minor Comments:

The justification of a quantile-based coefficient of variation in Section 3.2.1 can be shortened.

We have shortened this section as requested and merged it into the preceding section, 3.2.

The word compiled in Section 3.1.5 should probably be changed to grouped. And I am not sure I understand what concept the authors are trying to convey here. Was the grouping done based on skyLOSA results?

We have removed the word "compiled" and amended our original notation to say "sorted into sky 'categories'".

In our original text, we present the concept (emphasis added) that "…there is some additional uncertainty in sky-LOSA-computed soot mass column density through use of a ***single*** CIE sky model in the MC method" due to the inherent error in these simple models. We also present how the sky categories were defined (emphasis added): "…to permit capture of CIE sky model error in the [general uncertainty analysis], like skies were [sorted] into sky '[categories]' ***that have similar properties but differing … directional variability***." Broad descriptions and characteristics of each sky category are located in the final paragraph of Section 3.1.2 and Table 1 of the revised manuscript.

I found the discussion of the total order L(b) in Section 3.1.4 unclear. Is this discussion significant, considering the uncertainties in the assumption of a black-carbon-only aerosol and aerosol-free sky?

As noted in the responses above, we do not assume an aerosol-free sky.

This brief section describes our approach to the truncation of the Fourier-Legendre expansion of the scattering phase function; specifically, the order ($L(\boldsymbol{b})$) at which the expansion was truncated while both ensuring accuracy in sky-LOSA-computed soot/BC mass column density and enabling rapid calculation of sky- and sunlight inscattering. To help reduce the length of the main manuscript (see the Referee's first general comment), we have moved this text to the new Appendix A (Section A.1).

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