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with Remote Auto-Compensating Laser-Induced Incandescence:
Design & lab-scale demonstration

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Topic Area: Soot and Particulates
Alternate Topic Area: Diagnostics

1. Introduction

1.1 Particulate matter emission characterization from industrial flare plumes

Airborne particulate matter (PM) is a primary atmospheric pollutant that is linked to serious health effects in humans \([1][2]\) and environmental damage \([3]\). PM is further implicated as an important component in climate forcing \([4]\). However, there are critical gaps in our ability to accurately characterize and quantify industrial PM emissions, especially for unconfined sources such as flares, which are used to destroy unwanted flammable gas in the upstream energy industry. Recent satellite imagery data suggest that global gas flaring associated with petroleum production exceeds 135 billion m\(^3\) annually \([5]\). Although flares are known to produce PM in the form of carbonaceous soot, direct emission quantification remains elusive because flares emit directly into the open environment.

Most regulatory standards for PM in plumes are based on a human-observed visual opacity standard, as visually estimated by trained human observers (EPA Method 9 \([6]\)). By nature, this technique is unavoidably subjective. An attempt has been made to modernize the EPA Method 9 standard by using a digital camera \([7][8]\) but this remains qualitative, since broadband opacity is not directly relatable to PM emission rate. Optical measurement methods offer the promise of remote measurement of PM emissions, but until very recently little progress has been made. A diagnostic based on line-of-sight attenuation (LOSA) is under development by Yang et al. \([9][11]\), using sky light as the light source for spectrally resolved attenuation measurement. The technique claims a sensitivity limit of 0.5 mg/s for a 2 m-diameter plume with a wind speed of 4 m/s \([11]\) but has only been demonstrated with lab tests.

The present work investigates the application of laser induced incandescence (LII) for PM concentration measurements in industrial flares. LII is typically used for the measurement of PM in flames, optical engines, or extracted samples. Incandescent emission from soot particles is collected at close range and perpendicular to the axis laser excitation. An alternative optical arrangement will be considered for unconfined industrial flares, where the laser excitation and emission detection axes are near coincident. The set-up is based on the auto-compensating LII (AC-LII) approach \([12]\) and will be labeled ‘remote AC-LII’. In the present work, remote AC-LII is demonstrated on a soot plume from a distance of 1.5 m. Extension of the remote AC-LII to distances relevant to industrial plume measurement is also considered in terms of the system sensitivity and laser beam shaping.

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2. Background

2.1 PM composition of plumes from flares

Flame generated soot consists of nearly spherical, carbonaceous primary particles with 10 and 50 nm diameter, joined together in branch-like, open-structured aggregates. It is well-established[12][15] that the interaction of light with soot aggregates is well described by Rayleigh-Debye-Gans Fractal Aggregate theory (RDG-FA).

2.2 Auto-compensating LII

Laser-induced incandescence (LII) can be used to determine the volume fraction and primary particle size distribution of soot[16][18]. An intense laser pulse is used to heat soot aerosols to, or close to, their vaporization temperature. The incandescence signature from the soot is short-lived as the particles rapidly cool to the ambient temperature principally through conductive cooling and in some cases sublimation. The emission intensity is proportional to the concentration of the soot, but it is also highly dependent on the soot temperature.

Two distinct strategies exist to overcome this temperature dependence. The first, often referred to as high-fluence LII[17], involves imparting sufficient energy to the particles to reach a sublimation temperature of 4000 to 4500 K independent of their initial temperature. A near-proportionality of emission intensity and concentration is then assumed[19] It requires calibration from another calibrated technique in flames of known concentration[20][21]. The second strategy involves measurement of the temperature of the heated soot in real-time by two-color emission pyrometry[12][22]. The diagnostic is less intrusive since the soot temperature evaluation makes it unnecessary to reach sublimation temperatures[12]. The method is referred to as auto-compensating LII (AC-LII). Calibration does not require comparison to other diagnostics or to reference flames. It is also better suited for morphology measurement, since high fluence LII alters soot[23].

AC-LII is based on two-color pyrometry, involving LII acquisition for two wavelengths channels. According to RDG-FA, the emission from soot nanoparticles scales linearly with the volume of soot present in the measurement volume. The theoretical emission per unit volume of soot at temperature $T_p$ and at wavelength $\lambda$ is[12]:

$$
\phi_p(T_p, \lambda_c) = \frac{48\pi^2h}{\lambda^6} \left[ \exp \left( \frac{hc}{k\lambda T_p} \right) - 1 \right]^{-1} E(m_\lambda)
$$

(1)

where $c$ is the speed of light, $h$ is Planck’s constant, and $k$ is the Stefan-Boltzmann constant. $I_i$, the signal measured channel $i$ is then given by Eq. (2) where $f_i$ is the volume fraction of soot, $w_i$ is the laser beam thickness, $D_i$ is the background intensity reading. The term $\eta_i$ accounts for optical characteristics of the demultiplexer box, determined from a calibration with a radiant standard placed at the measurement location. Temperature is calculated from the ratio of LII signals for the two wavelength ranges and $f_i$ can be obtained from either wavelength intensity.

$$
I_i = f_i w_\eta \int_{\lambda} \phi_p(T_p, \lambda_c) \Omega_\lambda d\lambda + D_i
$$

(2)

A key assumption of the equation development is that the laser heats the soot uniformly to a single peak temperature. This is only possible if the laser energy distribution (i.e. the laser fluence) is uniform across the cross-sectional area of the laser beam. The achievement of a ‘top-hat’ laser profile is discussed in Section 3. All of the above equations are functions of time since the LII emission varies as the particles cool down. An effective soot particles diameter, $d_{p_{\text{eff}}}$, can be calculated by substituting the temperature decay into a heat transfer model including the sensible energy of the particle and conduction heat transfer to the combustion gas[24].

2.3 Retro-LII in the literature

Remote LII diagnostics have been developed for soot emission studies in the exhaust of aero engines in order to obtain data on the mass and morphology of emitted soot. Conditions in aero engines are much harsher that in the case of gas turbines or automotive engines and LII collection systems cannot be set up close to the exhaust. This limitation triggered the development of a remote LII set-up capable of collection from a distance of 3 m[25][27].
Published works claimed sensitivity of 5 μg/m³ [26] and 80 μg/m³ [27]. In all three cases, measurements were performed with time-integrated detection at a single wavelength.

Increasing the distance to the measured soot source and applying remote-LII measurements to other sources of PM, such as industrial plume sensing, is an unexplored direction of research. The two-color, time resolved approach used in AC-LII would be a substantial improvement to the existing remote-sensing LII set-up, offering self-calibrated measurement of soot concentration and effective primary particle diameter.

3. Experimental set-up

3.1 Source of soot: inverted burner from a methane/air diffusion flame

Measurements were performed on a soot plume generated from an inverted co-flow non-premixed burner developed by Coderre et al. [28], [29]. Methane is introduced at the top of the burner. The oxidizer, co-flow air is introduced into the annular region between the fuel jet and the outer quartz tube. A secondary dilution air is introduced at the bottom of the quartz tube in order to dilute, cool down and mix the exhaust gas. A converging nozzle is placed at the exhaust outlet to reduce the plume diameter from 50.8 to 12.7 mm in order to provide a well-mixed uniform flow.

3.2 Optical set-up

![Optical set-up diagram](image)

Figure 1. Optical set-up for the retro-LII measurements at 1.5 m

The optical set-up was placed 1.5 m away from the soot source, in order to demonstrate the measurement concept for a close distance range [Figure 1]. A square aperture of 2.5 mm width was placed 700 mm away from the laser head along the beam path, selecting a homogeneous area of the laser and provide homogeneous heating of the soot. The aperture was imaged by a 400 mm-focal length lens, leading to a 10 mm-width beam at the probe volume. The variations of the beam profile [Figure 2] are below 15%, showing a reasonable level of homogeneity.

![Laser-beam profile](image)

Figure 2. Laser-beam profile at the location of the soot source. Intensity scale in arbitrary unit.

The LII signal is collected with a 300 mm focal length, 75 mm diameter lens that focuses the LII emission signal onto a 2 mm-diameter circular aperture, which defines the collection volume. The emission is then collimated inside the demultiplexer box, and split into a blue (445 nm) and a red (753 nm) channel using dichroic mirrors and narrow band interference filters. The LII emission signal is collected via photomultipliers connected to fast A/D boards.
Overlap of the collection volume and the laser beam over the full depth of the measurement is needed for the AC-LII approach. The laser axis has a 2.9 degree-angle with the collection axis and collection volume itself varies with the distance, but they overlap completely from 1480 to 1520 mm away from the collection lens, as shown in Figure 3. This allows soot concentration measurements in flows with diameters up to 40 mm.


4. Results

4.1 Sensitivity of the set-up at 1.5 m

The sensitivity of the current remote AC-LII set-up was compared to the sensitivity of the High Sensitivity Auto-Compensating LII (HS-LII) setup which was developed at NRC to measure ambient level PM concentration. The HS-LII set-up is able to detect soot concentration down to 0.01 ppt or 20 ng/m$^3$, but the value of 100 ng/m$^3$ was used here as a reference. An expected sensitivity of the remote AC-LII set-up can be extrapolated by comparing the properties of the optical components involved in the two set-ups.

The collection efficiency limits the sensitivity of the retro-LII set-up. It is controlled by the f-number of the focusing lens and the aperture diameter, which defines the probe volume diameter. The f-numbers for HS-LII and remote AC-LII are 3.75 and 21.75, and the collection volume cross-sections have diameters of 16 and 8 mm, respectively. Hence, the remote AC-LII set-up will collect approximately 135 less light than the HSLII set-up for a comparable measurement depth. The equivalent width of the remote AC-LII is defined by the flow diameter, 11.86 mm when adjusted for the cylindrical shape. The HS-LII equivalent depth was 8.339 mm. Hence, the remote AC-LII set-up should be 95 times less sensitive than the HS-LII set-up. The target detection limit is thus 95 µg/m$^3$ or 5 ppt.

4.2 Soot concentration measurements

Soot concentration was measured for two laser fluencies (86 and 112 mJ/mm$^2$). The values for the peak temperature, soot volume fraction and primary particle diameter are listed in Table I. The table also includes reference values for the inverted burner: peak temperature measured in 2009 using another NRC AC-LII (Mobile-II), soot volume fraction from gravimetric analysis of physical samples, and primary particle diameter from TEM analysis.

The peak temperatures are slightly lower than the values obtained with Mobile II, which could relate to small differences in the absolute intensity calibrations. The measured soot volume fraction varies with laser fluence, which is a known anomaly of the AC-LII method applied to cold soot sources. It has been found that best agreement of AC-LII from cold sources to other measurement methods (such as gravimetric analysis) is achieved when the soot is heated closer to its sublimation temperature. In the present case, the measurement obtained at a laser fluence of 112 mJ/mm$^2$ is more representative, hence concentration is over-estimated by about 70%. Future work must include
validation of the alignment and calibration of the various systems. Nonetheless, this first proof-of-concept test is highly encouraging with initial measurements in the correct range.

Table 1 Retro-LII measurement in the inverted burner used with methane and air

<table>
<thead>
<tr>
<th>Laser fluence [mJ/mm²]</th>
<th>Remote AC-LII</th>
<th>Mobile II</th>
<th>86</th>
<th>112</th>
</tr>
</thead>
<tbody>
<tr>
<td>Peak temperature [K]</td>
<td>Remote AC-LII</td>
<td>2556</td>
<td>2819</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Mobile II</td>
<td>2875</td>
<td>3250</td>
<td></td>
</tr>
<tr>
<td>Soot volume fraction [ppb]</td>
<td>Remote AC-LII</td>
<td>67.5</td>
<td>111.8</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Gravimetric[29]</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>TEM[29]</td>
<td>37.4</td>
<td>39.3</td>
<td></td>
</tr>
</tbody>
</table>

5. Perspectives

For soot concentration in the field, the remote AC-LII set-ups will have to be placed 20 to 30 m away from the measured plume. Various aspects need to be investigated before obtaining a field-ready remote AC-LII set-up.

5.1 Sensitivity of the set-up with telescopes

The sensitivity of the system was predicted as a function of the collecting lens characteristics for systems placed 5 to 30 m away from the measured plume, with a model similar to the optical model used in section 4.1. The laser beam illumination was assumed homogeneous at 2 mJ/mm², with a 11 mm diameter collection volume (reasonable value for 330 mJ/pulse laser energy). A 2 m-diameter plume was considered (2 m equivalent width).

Figure 4. Sensitivity of the retro-LII set-up as a function of the distance to the plume. Soot measurement for a plume diameter of 2 m. Analysis for commercial telescopes.

Commercial telescopes seem interesting collection optics since they include large diameter collection lens. Various models have been considered, with lens diameter ranging from 4 to 8” (Celestron NexStar 4SE” to 8SE”) and 16” (Meade 16” LX200-ACF). All f-numbers were 10 except for the NexStar 4SE” which had an f-number of 13. The calculated sensitivities are plotted vs. the plume distance in Figure 4 which also includes the 3”-diameter lens used for measurements from a distance of 1.5 m. The sensitivity of the remote AC-LII set-up with a 3”-diameter lens would be 5.1 µg/m³ at 20 m. With a 6”-diameter telescope, the detection limit would be 1.1 and 2.5 µg/m³ at 20 and 30 m, respectively. A 16”-diameter telescope should allow soot concentration measurements down to 0.15 and 0.35 µg/m³ at 20 and 30 m.

There are very few literature references to quantitative emission measurements from industrial flares. The existing literature does not directly report data on mass emission rates of soot and anyways considers very heavily sooting flares, which are not representative of low-emission solution gas flares. Equally, there are few examples of other
diagnostics suitable for remote PM emissions from industrial flares to compare against. A single exception is the sky-LOSA diagnostic developed by Johnson and coworkers[11], which reports a sensitivity limit of 0.5 mg/s for an industrial flare with a diameter of 2 m and wind speed of 4 m/s. This target corresponds to a concentration of \(25 \mu g/m^3\). All of the telescopes shown here should be able to measure soot concentration for such emission rates.

5.2 Laser beam shaping and absolute light calibration

Providing a fluence of \(2 \text{ mJ/mm}^2\) in a homogeneous 11 mm diameter beam could be difficult to achieve at a distance of 20 to 30 m from the laser head, all the more since the fluence profile should remain constant across the plume width. The outdoor use of high power laser beams also raises safety issues. Specific authorizations must be granted by Transport Canada because of possible airplane accidents induced if a laser beam reaches the cockpit.

Applying the typical AC-LII calibration approach seems unlikely if the soot source is found 30 m away from the collection box. In typical AC-LII measurements, the LII collection system is calibrated with an integrating sphere whose light intensity is accurately known, placed at the measurement location. Solution to this problem likely involves extrapolation of the calibration values from calibration measurements made from a closer distance.

Finally, the remote AC-LII method would provide a measure of the amount of soot present along one line of sight. A full characterization of the plume emission rate would require rapid scanning of LII system across the plume width. Information about the velocity of the plume perpendicular to the measurement plane would also be necessary.

6. Summary

The present work shows a preliminary investigation of a novel technique for quantifying soot (PM) emissions in flare plumes, centered on the auto-compensating laser induced incandescence optical diagnostics (AC-LII), but configured for remote measurement. A plume of known concentration produced by an inverted flame was used to test the method at close range. The preliminary results were correct to within a factor of 1.7. Further work is needed in order to improve the agreement of the measurement. An extrapolation of the set-up sensitivity to a field setting indicated that the remote AC-LII system should be able to measure soot concentration in typical gas flare plumes. However, delivering a homogeneous small diameter laser beam at a distance of 30 meters will be a challenge, as well as dealing with safety issues for outdoor laser use.

7. Acknowledgements

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