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Abstract

The soot formation process has been investigated at pressures up to 16 bar using a non-premixed laminar coflow flame with nitrogen-diluted ethylene. 2D diffuse line-of-sight attenuation (2D LOSA) and planar laser-induced incandescence (PLII) were used to measure soot volume fraction (SVF). The peak SVF increased exponentially with increasing pressure and the spatial distribution of soot volume fraction changed substantially. At pressures below 6 bar, the two techniques agreed well. At pressures above 6 bar, the techniques began to disagree, with 2D LOSA showing higher peak SVF values at a location lower in the wings of the flame compared to PLII. Errors in the LOSA measurements due to the molecular absorption of PAHs were assessed by performing measurements with bandpass filters centered at 435 nm and at 647 nm. Furthermore, the evolution of polycyclic aromatic hydrocarbons (PAH) in the flame was studied using planar laser-induced fluorescence (PLIF) with the excitation laser set at 282.85 nm and compared to LOSA measurements. Fluorescence signals were captured using bandpass filters (350 nm, 400 nm, 450 nm, and 510 nm) corresponding to increasing PAH size. The peak concentration of PAHs moved closer to the burner nozzle as pressure increased. Absorption by PAH were unable to explain discrepancies between LOSA measurements and PLII measurements. Using the Rayleigh-Debye-Gans approximation for polydisperse fractal aggregates (RDG-PFA), the differences between LOSA and PLII measurements were analyzed, and it was found that LOSA is more sensitive to the soot primary particle diameter due to changes in the scattering to absorption ratio ($\rho_{sa}$). The effect of gate duration on SVF imaging with PLII is also reported.

Keywords:
Soot Formation
High Pressure Non- Premixed Coflow Flame
Laser-Induced Incandescence

2D Diffuse Line-of-Sight Attenuation

Polycyclic Aromatic Hydrocarbons
1 Introduction

Soot formation from incomplete combustion has been shown to negatively impact human health and the environment. Due to the short residence time of soot in the air, there are significant near-term benefits from reducing soot emissions from combustion processes. Thus, studies of soot formation processes are attractive in an effort to mitigate the negative effects of combustion technologies. Substantial effort has been made in studying the soot formation process at atmospheric pressure. Numerous intrusive and non-intrusive techniques have previously been used in these studies as detailed in. Extending our understanding of the soot formation process with pressure is essential as many practical combustion devices operate at higher pressure for an increased thermodynamic efficiency. Due to experimental costs and complexity, there is limited experimental data related to soot formation at these high pressures. A thorough review of previous studies is given in.

One aspect contributing to the complexity of high-pressure diagnostics is the limited access to the combustion environment. Preventing disturbances to the flow, preserving the natural flame chemistry, and limited access to the combustion environment make non-intrusive diagnostics an attractive method to investigate flames, yet these diagnostics typically rely on a knowledge of soot optical properties. These properties have been measured at varying conditions, but concurrent changes in the primary particle size and the aggregate morphology make the optical properties at all locations in the flame difficult to determine. Uncertainty in soot volume fraction (SVF) measurements can be significant due to uncertainty in the scattering to absorption ratio ($\rho_{\text{st}}$) and the refractive index of the soot particles.

Furthermore, as pressure increases, beam steering can affect experimental measurements due to a steepening of density gradients in a flame. Several techniques have been used to limit or eliminate effects from beam steering in an effort to effectively use non-intrusive diagnostics at high pressure. One technique that has become increasingly utilized in high pressure diagnostic is 2D diffuse line of sight attenuation (2D LOSA). The main advantage of this diagnostic is it is relatively immune to beam steering at high pressure. Extinction diagnostic techniques continue to be advanced and applied to challenging combustion environments.
In this work, we implement and compare two commonly used techniques for measuring SVF at pressures between 1 bar and 16 bar in a non-premixed laminar coflow flame with nitrogen-diluted ethylene: 2D diffuse line-of-sight attenuation (2D LOSA), and planar laser-induced incandescence (PLII). Sources of measurement uncertainties are investigated by comparing results from these two methods. Differences between the two measurements are explored with simulations based on the Rayleigh-Debye-Gans approximation for polydisperse fractal aggregates (RDG-PFA). We perform planar laser-induced fluorescence (PLIF) of the polycyclic aromatic hydrocarbons (PAH) to investigate the effects of absorption by PAH on the LOSA measurements and to investigate changes in the soot formation process with pressure. The effects of pressure on both PAH and SVF distributions are reported.

2 Methods

The setup used for the LOSA, PAH planar laser-induced fluorescence (PLIF), and laser-induced incandescence experiments (described below) is illustrated in Figure 1. The experiments were conducted in a pressure vessel containing a laminar coflow burner. Details about the pressure vessel can be found in 14. A 4 mm ID non-tapered nozzle was used for the fuel flow in the burner. Nozzle and burner geometries are shown in the Supplementary Material with greater detail found in 15. Ethylene diluted with nitrogen was used as the fuel source and air was used in the coflow. The experiments were run at constant fuel and nitrogen mass flow rates, and the coflow was increased with pressure to maintain flame stability. Although previous work done in sub-atmospheric flames has shown increasing coflow rates reduces soot formation of flames 16 the coflow rates were increased to allow experimental results to be comparable to previous work found in 11. Furthermore, high coflow rates can result in blowing off the flames at low pressure. The ethylene, nitrogen, and air coflow rates are presented in Table 1.

The flames were approximately 2 cm tall and stable at all pressures.

<table>
<thead>
<tr>
<th>Pressure (bar)</th>
<th>Ethylene (mg/s)</th>
<th>Nitrogen (mg/s)</th>
<th>Air (g/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>0.85</td>
<td>4.27</td>
<td>0.84</td>
</tr>
<tr>
<td>4</td>
<td>0.85</td>
<td>4.27</td>
<td>1.04</td>
</tr>
<tr>
<td>6</td>
<td>0.85</td>
<td>4.27</td>
<td>1.24</td>
</tr>
<tr>
<td>8</td>
<td>0.85</td>
<td>4.27</td>
<td>1.44</td>
</tr>
</tbody>
</table>
2.1 2D diffuse line-of-sight attenuation (2D LOSA) setup

Details of the 2D LOSA setup and theory are described in \(^\text{10, 15}\). For the light source, a broadband LED lamp with a color temperature of 6500 K was mounted on an integrating sphere for measurements using a 435 ± 20 nm FWHM filter. An LED with a color temperature of 3000 K was used for measurements done with a 647 ± 10 nm FWHM filter. The choice of these two wavelengths is discussed later. The integrating sphere provided a homogenous Lambertian profile that minimized the artifacts from beam steering at high pressure \(^\text{17}\). The light was imaged onto the burner through two achromatic lenses \((f = 100 \text{ mm}, \text{ and } f = 300 \text{ mm})\), magnifying the imaged light by 3x at the burner. Two achromatic lenses \((\text{both } f = 750 \text{ mm})\) with an aperture between them \((d = 10 \text{ mm})\) reimaged the flame to the CCD camera with a resolution of 0.019 mm per pixel. Images were separately acquired using bandpass filters (described above) mounted in front of the CCD camera. By taking a three-point Abel inversion \(^\text{18}\) (we used a corrected version of the deconvolution operator shown in \(^\text{18}\) equation 7, correcting
the final term in equation 7 to be \(-2j\bar{I}_{ij}(0)\) from +2j\bar{I}_{ij}(0) as found in \(^{19,20}\) of the ratio of light extinction through the flame and the incident light, the local extinction coefficient, \(K_{\lambda}^{(e)}\), can be determined at the wavelength of the attenuated light, \(\lambda\), using the Beer-Lambert law \(^{10}\). SVF can then be determined using the local extinction coefficient:

\[
SVF = \frac{K_{\lambda}^{(e)} \lambda}{6\pi(1+\rho_{sa})E(m)},
\]

Equation 1

where \(\rho_{sa}\) is the soot scattering to absorption ratio, \(m\) is the soot refractive index and \(E(m)\) is the refractive index function = \(\text{Im}((m^2-1)/(m^2+2))\). The value of \(\rho_{sa}\) varies within a flame and depends on several parameters \(^{21}\), including, but not limited to: the nanostructure of the soot primary particles, the diameter of the primary particles, the radius of gyration of an aggregate, and the number of primary particles in an aggregate. In \(^{22}\), \(\rho_{sa}\) values were calculated using RDG-PFA, keeping all parameters constant except for the primary particle diameter; \(\rho_{sa}\) was calculated to be 0.05 for a particle with a diameter of 18 nm, and 1.13 for a particle with an effective diameter of 125 nm. These \(\rho_{sa}\) values result in SVF errors of up to ±50%. Uncertainties in \(E(m)\) can also contribute significantly to errors in SVF as reported in \(^{23}\). Values reported for \(E(m)\) vary significantly and are reported to depend on flame conditions, flame height, and in some works the measurement wavelength \(^{23,24}\). In atmospheric pressure premixed flames, relative differences of \(E(m)\) between two flame heights could result in errors in SVF as high as 75% at shorter wavelength. In non-premixed flames, relative differences of \(E(m)\) were much lower and show peak SVF errors of around 15%. Due to the limited data at high pressure, quantifying uncertainties in \(E(m)\) is not possible and future work is needed to investigate \(E(m)\) values in pressurized flames.

This paper uses measured extinction coefficient values from a non-premixed atmospheric coflow flame found in \(^{7}\) to calculate \(\rho_{sa}\) and \(E(m)\). Values of \(\rho_{sa} = 0.26\) and \(E(m) = 0.37\) were used in this study with a maximum uncertainty of 20% at atmospheric conditions \(^{11}\).

In Equation 1, \(\lambda\) is the extinction wavelength. Choice of the extinction wavelength is an important practical consideration when performing 2D LOSA. Longer wavelengths are less attenuated than shorter wavelengths.

Furthermore, \(E(m)\) becomes wavelength independent above the upper visible wavelengths \(^{23}\). At these higher wavelengths, the natural flame luminosity is stronger than at shorter wavelengths and can be even larger than...
the incident light. When combined, these two effects cause a reduction in the signal-to-noise ratio. For this reason, shorter wavelengths have been utilized in several extinction imaging applications \(^{10,13,25}\). However, shorter wavelengths may be subject to molecular absorption by PAHs which can cause systematic errors in the soot measurements. In this study, wavelengths of 435 nm and 647 nm were used and compared. Furthermore, qualitative PAH PLIF was performed and peak signal locations were compared to peak SVF values from 2D LOSA measurements.

### 2.2 PAH PLIF setup

Qualitative PAH PLIF was performed with an ND:YAG pumped rhodium dye laser at 282.85 nm, following the technique described in \(^{26}\). A laser sheet of \(~400\ \mu m\) thickness was formed with a concave cylindrical lens \((f = -50\ mm)\) and focused on the burner with a plano-convex lens \((f = 400\ mm)\). The laser fluence was kept below 20 mJ/cm\(^2\) to avoid generation of an LII signal \(^{27}\). Images were acquired at 90° on a Princeton ICCD camera with a 500 mm UV lens. A set of 50 shots were accumulated using a gate width of 500 ns. The laser prompt began 200 ns after the camera gate opened to ensure the PLIF signal was captured. A delay generator was used to sync the camera and laser. Bandpass filters \((\text{FWHM} = 10\ nm)\) at 350 nm, 410 nm, 450 nm, and 510 nm were separately used in conjunction with 2 WG 305 longpass filters to eliminate elastically scattered light. The bandpass filters allowed the PAH signals to be acquired from two to three ring PAHs, four to five ring PAHs, and larger ring PAHs at each of the respective wavelengths. One source of error with this technique is caused by PAH stacking which can cause signals at higher wavelengths from smaller PAH \(^{26,28}\). Background images of the natural flame luminosity were taken and subtracted from the PAH PLIF shots.

### 2.3 PLII setup

Details of SVF imaging with PLII are described in \(^{27}\). In this experiment, a vertical laser sheet was formed from a pulsed Nd:YAG laser source (Ekspla model NL909-SH #NLL252) at 1064 nm with a 3 ns tophat temporal profile and sent through the flame. The short temporal profile minimized the signal bias towards larger particles at higher pressures \(^{29}\). A variable attenuator control system (Laseroptik AVACS) was used to control the laser power. The laser sheet was formed by first reducing and recollimating the initial beam using an anti-reflective
coated plano-convex lens \((f = 250 \text{ mm})\) and a biconcave lens \((f = -50 \text{ mm})\). The laser sheet was then generated using a concave cylindrical lens \((f = -20 \text{ mm})\) and focused on the burner axis with a convex cylindrical lens \((f = 300 \text{ mm})\). A sheet with a thickness of \(\sim 300 \mu \text{m}\) was measured at the burner nozzle region using burn paper. The vertical laser profile was measured using a power meter with a 1.1 mm slit covering the sensor. The profile was measured in 1 mm increments. The average calculated laser fluence over the vertical profile was \(0.456 \text{ J/cm}^2\) with a standard deviation of \(0.03 \text{ J/cm}^2\). Shot-to-shot noise was calculated as \(\pm 7.7\%\) over 300 shots. With this technique, soot was heated to the sublimation point by the laser pulse causing increased incandescence. The LII signal was imaged with an ICCD camera 20 ns after the laser pulse to avoid \(\text{C}_2\) swan band emissions\(^{30}\), with a 10 ns camera gate duration at an angle of 90° to the laser sheet. The detection wavelength of \(435 \pm 20 \text{ nm}\) was found to be a good compromise between collecting sufficient signal and distinguishing the LII signal from the natural soot luminosity in the imaging line of sight (the temperature of the soot in the flame is as high as 2000 K at atmospheric pressure). Based on Planck’s radiation law, the relative interference of the line-of-sight integrated natural soot luminosity to the LII signal is significantly larger at higher wavelengths and therefore not preferred in this work.

Incandescence emissions are directly proportional to SVF in the flame with caveats described in \(^{31}\). In this work, the acquired LII signal was quantitatively related to SVF by calibration with the 647 nm 2D LOSA data. Calibration values were determined for each experimental pressure to account for changes in LII decay times \(^{32}\). The values were taken at a location 0.2 mm off center and at 15.5 mm above the burner (HAB) using the average SVF value in a 0.1 x 0.1 mm area. This location was chosen for two reasons; first, there are lower gradients of SVF found in this area; second, to avoid noise found at the centerline due to the inversion process. The calibration value for the 4 bar condition was used for the 2 bar condition due to the large amount of noise found in the 2 bar 2D LOSA measurements. Within the same camera settings, calibration factors between the different pressures had a maximum difference of 34%. Two settings were used on the camera. From 2 to 6 bar, the intensifier was set to a maximum value of 255. From 8 to 16 bar the intensifier was set to 0. The calibration values were found to increase with pressure with both camera settings, although the rate of change of calibration value was different.
between the two camera settings. The differences between calibration values at each pressure can be attributed to changes in $E(m)$ as well as changes to $\rho_{sa}$ with pressure. Currently there is a gap in knowledge of how these values change with pressure, and thus they were kept constant at all pressures.

3 Results and Discussion

3.1 SVF light extinction imaging

The 2D LOSA images acquired with the 435 nm and 647 nm filter are shown in Figure 2 with the 435 nm filter images on the top and the 647 nm filtered images on the bottom. Fifty images were captured and ensemble-averaged to account for shot-to-shot variation. Tomographic inversion (see section 2.1) was performed separately on each half of the flame to find the local extinction coefficient. SVF was determined using Equation 1, assuming a constant $\rho_{sa}$ throughout the flame. The flame halves are presented together where some dissymmetry is visible. Performing the abel-inversion on the images introduced significant noise in the image centerline. Furthermore some images are shown to have lower SVF values on the centerline with increasing SVF values at a position slightly radial to this. This is not a physical result, but rather a result from the inversion process. Shot-to-shot noise was negligible, with the largest sources of uncertainty coming from uncertainty in soot optical properties and from averaging the two flame halves. At 1 bar, orange soot luminosity is visible in the

![Figure 2: 2D diffuse LOSA measurements (435 nm on top and 647 nm on bottom) of SVF from 2 to 16 bar. The false color scheme represents SVF in units of ppm. At 1 bar, soot was not detected. As pressure increases, the peak soot volume fraction shifts into the wings of the flame at both detection wavelengths.](image)
flame tip but soot loading is not detectable with extinction imaging, hence it is not shown in this work. Both imaging wavelengths showed similar trend. That is as pressure increased, soot began to form near the flame tip. As pressure further increased, the peak SVF location moved into the wings of the flame. These trends are consistent with flames under similar conditions where 2D SVF profiles were found by using point extinction measurements with a HeNe laser at 632.8 nm. 

There are two main differences between the 435 nm filter and the 647 nm filter. The first is the higher noise found at 2 bar in the 647 nm image as compared to the 435 nm image. Second, the 435 nm SVF values are consistently larger than the 647 nm filtered images at all pressures above 2 bar. This can be attributed to a number of factors. First, while previous studies such as show a decrease in $E(m)$ with increased wavelength, $E(m)$ was held constant for both wavelengths due to high uncertainty in the value itself. This would result in larger SVF values with the 435 nm filter. Second, SVF may be larger with the 435 nm images due to absorption by PAH, thus affecting peak SVF values. The average difference between the peak values at the centerline is 19% and the average difference in the wings is 31% with maximum differences being found at 2 bar for the centerline and 4 bar for the wings. Both sets of images are included since numerous studies using 2D diffuse LOSA have been done in both wavelength ranges.

SVF is generally reported to scale with pressure using an exponential $P^n$ function. In , LII images were calibrated using laser extinction and peak SVF was found to scale as $n = 1.7$ between 1 and 16 bar. In , they used 2D diffuse LOSA with a bandpass filter centered at 450 nm and measured $n$ to be 1.98 for the local SVF between 1 and 8 bar in an undiluted ethylene/air non-premixed coflow flame. In , LOSA was used with a laser wavelength of 632.8 nm and SVF was found to scale as $n = 2.0$ in the flame centerline and 2.3 in the flame wings between 4 and 16 bar in a non-premixed ethylene diluted with nitrogen coflow flame. In the current 2D LOSA measurements peak SVF values were found at the centerline between 4 and 16 bar between 0.1 to 0.28 mm off center on each flame half to avoid noise from the inversion process, and in the wings at all areas below 14 mm. 

At 435 nm, SVF scaled as $n = 1.82 \pm 0.06$ and $2.37 \pm 0.20$ in the centerline and wings, respectively. For measurements taken with the 647 nm filter, SVF scaled as $n = 1.70 \pm 0.30$ and $2.37 \pm 0.48$ in the centerline and
wings. Standard deviations were calculated from the differences between the left and right side of the images and were highest in the flame wings in both cases. The above reported 95% confidence intervals were much larger with the 647 nm filter due to two reasons. First, differences between the flame halves in the wings, were all less than 10% except at 4 bar and 8 bar where there were differences of 29% and 33% respectively. Second, the centerline standard deviations were large due to slight dissymmetry in the two halves caused by a reflection from the light source imaging lens. A comparison of the scaling values at 435 nm show scaling values found in the flame wings are comparable to the work found in 11. Scaling values in the centerline are between the values found in 33 and the work found in 11,25 at 435 nm. Measurements at 647 nm in the flame wings are closer to values found in 11, and similar to values found in 33 at the flame centerline.

Scaling was also done for the peak carbon conversion efficiency ($\eta_s$) as described in 6,34. The soot density ($\rho_s$) was assumed to be 1.9 g/cm$^3$ and the flame acceleration constant ($a$) was assumed to be 32 m/s$^2$ 11. Peak $\eta_s$ also follows a $P^n$ function. Work in a non-premixed laminar ethylene/nitrogen flame scaled peak $\eta_s$ as $n = 1.36$ between 10 and 35 bar using spectral soot emission 35 and $n = 1.8$ between 4 and 16 bar 11 using LOSA with a HeNe laser at 632.8 nm. In 25, $n$ was found to be 1.62 between 1 and 8 bar using 2D LOSA at 450 nm. In this work, peak $\eta_s$ scaled as $n = 1.31 \pm 0.17$ for 435 nm and $n = 1.47 \pm 0.21$ at 647 nm between 4 and 16 bar. The standard deviation of both measurements falls within the range found in 35. Scaling values may be slightly lower than values reported in 11 due to differences in diagnostics or differences in measurement resolution.
The 435 nm and 647 nm LOSA show slight differences (within the standard deviation) of peak \( \eta \) scaling. Figure 3 also shows differences in peak carbon conversion location with 435 nm having a peak conversion value at a location nearer to the burner, especially at pressures above 8 bar. Both measurement filters show peak conversion approaching the burner as pressure increases.

![435 nm LOSA and 647 nm LOSA](image)

**Figure 3:** Carbon conversion versus height above burner (HAB) with the 435 nm LOSA on the left and the 647 nm LOSA on the right. As pressure increases, peak carbon conversion moves toward the burner. Peak carbon conversion is located higher in the flame for the 647 nm LOSA compared to the 435 nm LOSA.

### PAH PLIF

While both peak SVF scaling and carbon conversion for the 435 nm and 647 nm filters were found to fall within their respective confidence intervals, there were minor differences in peak SVF location between the 435 nm filter and 647 nm filter, as shown in Figure 4. One reason for this shift is likely due to absorption by PAH. In they use spectral resolved line of sight attenuation (Spec LOSA) at several heights in the flame and found that attenuation measurements are not affected by PAH in the upper visible and infrared region but may be affecting absorption measurements at lower wavelengths.

Fluorescence from PAH PLIF was captured at several wavelengths to view the overlap between regions of PAH and soot, and also to investigate how soot formation changes with pressure. Figure 4 (top) shows the peak PAH signal location and the upper and lower locations of 75% of the maximum PAH signal (from any radial location) where there was sufficient signal. The location of the peak SVF from the 435 nm and 647 nm LOSA with one sigma standard deviation are also shown. Similar to the soot measurements, sufficient PAH PLIF signal could not be acquired for any of the filters with the 75% threshold at 1 bar. At 2 and 4 bar, signals from 350 nm and 510 nm filters were nearly homogenous in the PAH region. Thresholding eliminated nearly all of the signal and thus
the upper and lower locations were excluded at this pressure. As the pressure increased, the peak PAH location migrated toward the burner. At pressures below 8 bar, PAHs were located throughout the flame, and there was a clear overlap of peak soot locations and PAH. Above 8 bar, the location of the peak PAH shifted towards the burner tip. Figure 4 (bottom) shows this shift as pressure increases. The shifting rates of the peak PAH signal relative to the burner nozzle were calculated. The 350 nm peak signal location versus pressure fit a nearly linear trend from 2 to 16 bar (disregarding 6 bar) shifting to the burner at a rate of 0.13 mm/bar. The 400 nm filter peak signal location showed a sharp shift in peak signal location between 4 bar and 6 bar. Following this, the peak signal location shifted at a linear rate of 0.28 mm/bar. The 450 nm filter peak signal location remained relatively constant from 2 to 4 bar then between 4 and 6 bar the peak signal showed a large shift toward the

![Figure 4: (Top) Peak PAH signal with the upper and lower location of 75% of the maximum PAH signal at each pressure (offset for clarity) and the peak SVF from LOSA. (Bottom) Binarized overlay of PAH and SVF at 8 bar and 16 bar with thresholds (TH) at 10% and 75%.

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burner with a relatively linear shift toward the burner after this of 0.33 mm/bar. The 510 nm peak signal location showed a rapid shift toward the burner between 4 and 8 bar and then shifted at 0.24 mm/bar toward the burner after 8 bar. Images of PAHs at each wavelength are available in the supplementary material.

PAH location shifting towards the burner was expected, as the rate of pyrolysis increases with increasing pressure \(^{11,37}\). Previous non-premixed coflow experiments with an ethylene/nitrogen mixture running at similar dilutions reported that particle growth with pressure is highest between 4 and 8 bar and mostly ceases between 8 and 12 bar \(^{11}\). PLIF measurements show that this is likely due to limited PAHs at higher flame locations.

Furthermore, numerical studies have demonstrated decreased HACA surface growth as pressure increases due to decreased H and OH radical concentrations and increased PAH condensation rates as pressure increases \(^{38}\).

Transmission electron microscopy (TEM) images of soot sampled thermophoretically at 5, 10, 15, and 20 bar show significant increases in primary particle diameter between 5 and 10 bar that levels off after these pressures \(^{39}\). This is likely due to high availability of PAHs throughout the flame at pressures below 8 bar and limited availability of PAHs above 8 bar thus inhibiting surface growth.

Peak SVF locations of the 435 nm LOSA and the 647 nm LOSA show the 435 nm LOSA having a peak SVF location lower than the 647 nm LOSA at all pressures except for 6 bar. The greatest difference between peak SVF locations is at 16 bar, but is less than 14% different. Peak SVF location differences between the 435 nm and 647 nm LOSA are highest at low pressures.

For peak SVF values, there is no apparent trend of differences between the two filters in the center line, but differences in the wings generally decrease with increasing pressure. At 16 bar, the difference between the 435 nm LOSA and 647 nm LOSA are 22% in the centerline and 11% in the wings. The differences between the two wavelengths could be due to absorption by PAH, changes in the emissivity of soot with wavelength, or differences in \(\rho_{sa}\) due to \(\lambda\) (and other parameters discussed later) at the two wavelengths.

### 3.2 SVF imaging with LII

Figure 5 shows the LII signal (captured using a 10 ns camera gate width) starting 20 ns following the initial laser prompt. The laser enters the flame from the left side of the image and shows some attenuation at pressures of 8
bar and above. Peak SVF values and locations agree well between LII and LOSA images between 1 and 6 bar. At 8
bar, the peak SVF locations for the LII and 2D LOSA measurements begin to diverge and at 12 and 16 bar, the
LOSA images show the peak SVF location moving down into the wings (Figure 2) while in the LII images, peak SVF
remains at a higher location (Figure 5). At 16 bar, the LOSA image shows a peak of 64 ppm at 435 nm and 58
ppm at 647 nm in the wings, while LII shows a peak value of 27 ppm in the wings. It should be noted that while
the LII calibrations come from the 2D LOSA measurements, the differences of overall trends between LOSA and
LII are significant rather than the calibrations themselves. For example, a calibration value for the LII
measurements from the flame wings would result in a higher average SVF, but these values would be found
throughout the flame rather than only the wings as is found in the 2D LOSA measurements.

Scaling factors were found for LII measurements between 4 and 16 bar. Errors were determined from shot to
shot noise differences as well as differences between the two sides of the flame. Peak SVF scaled as $n = 1.71 \pm
0.18$ and $n = 1.72 \pm 0.24$ in the centerline and wings, respectively. The centerline scaling factor was nearly the
same as LOSA as would be expected from scaling the images at this location. Scaling values in the wings of the LII
measurements fell within the 95% confidence interval of LOSA measurements done at 647 nm but not within
the standard deviation of LOSA done at 435 nm. Peak carbon conversion scaled as $1.37 \pm 0.43$ also falling within
the error of both 2D LOSA measurements.
The camera gate time was increased to 500 ns and the LII signal was biased towards particles with a larger diameter or larger apparent diameter. This is because smaller particles produce a shorter LII signal lifetime due to their larger surface-to-volume ratio, hence faster cooling. The peak SVF scaling values for LII shifted to $n = 1.70 \pm 0.23$ and $n = 1.85 \pm 0.42$ in the centerline and in the wings respectively. The carbon conversion as a function of HAB is shown in Figure 6. Peak carbon conversion scaled as $n = 1.48 \pm 0.56$. By increasing the gate time from 10 ns (left side) to 500 ns (right side) the peak carbon conversion shifted nearer to the burner at pressures of 8 bar and above. Peak carbon conversion values also begin to deviate at 8 bar and above between the 10 ns gated LII and the 500 ns gated LII.

In Figure 7, a comparison of the 16 bar case shows how better agreement of peak SVF (left) and peak location (right) can be found between LII and LOSA when the LII is weighted toward larger particles. The peak signal location shifts toward the burner by increasing the camera gate duration. Increasing the gate duration also results in higher SVF measurement in the wings of the flame. This is because the particles in the wings have a larger diameter than the particles near the tip, thus the integrated signal is higher in these locations. One should keep in mind that the LII signal lifetime at 16 bar is much shorter than the 500 ns gate (with our LII model, we calculated that the LII signal from a 30 nm particle decays within less than 150 ns after the laser. We ignored the effects of bath-gas heating. Thus, a 500 ns camera gate convolutes the entire LII signal.
The above comparison shows significantly higher peak SVF values in the wings for the extinction measurements.

Extinction measurements are one method typically used to calibrate PLII measurements. These calibrations assume an extinction coefficient or a value for $\rho_{sa}$ and the emissivity of soot. Values for these numbers are typically found from atmospheric flames due to limited data on high pressure flames. Due to the increased particle diameter found in the flame wings, typical assumptions of the extinction coefficient or $\rho_{sa}$ would likely result in higher errors if the extinction measurements are performed at locations that are lower in the flame.

One way to reduce these uncertainties would be choosing a calibration location for PLII at a higher flame locations. In $^{39}$, thermophoretic sampling of soot was performed at 3 locations in a flame between 5 and 20 bar. The lowest location showed a 40.9% difference between the mean value of primary particle sizes at 5 and 20 bar. The central location showed a difference of 27%, and the top location showed a difference of 5.7% between the 5 bar and 20 bar case.
In Figure 8, a comparison between diagnostics of peak SVF location and peak carbon conversion location is shown for pressures between 4 and 16 bar. Measurement uncertainties for the LOSA measurements are based on differences between peak SVF and carbon conversion location between the flame halves as shot to shot noise was negligible. LII uncertainties are based on shot to shot variation within the flame. The peak SVF location for each measurement are within the 95% confidence interval at 4 bar and 6 bar. Above this pressure, there is a significant deviation between the LII with the 10 ns gate and the 2D LOSA at both wavelengths. The peak SVF location for the LII with the 500 ns gate is found at a location between the 2D LOSA peak values and the peak value of the 10 ns gated LII. The location of peak carbon conversion is consistently lower for the LOSA measurements at all pressures compared to the LII. Increasing the gate duration from 10 ns to 500 ns only slightly lowered the location of the peak carbon conversion. The lower peak SVF location of the 435 nm LOSA measurement as compared to the 647 nm LOSA measurement is likely due to higher absorption by PAH for the 435 nm LOSA. Although the peak SVF location is lower for the 435 nm LOSA measurements, SVF scaling is within measurement uncertainties for the two wavelengths.

![Figure 8: A comparison of peak SVF location and peak carbon conversion location between 2D LOSA at 435 nm and 647 nm and LII with a 10 ns gate and LII with a 500 ns gate. Peak SVF locations are within the LII 95% confidence intervals at 4 and 6 bar. At 8 bar and above, there is significant deviation between the measurements. For peak carbon conversion, peak locations maintain a similar relative distance at all pressure.](image)

4 Discussion

The comparison in Figure 7 suggests that the SVF distributions measured by the LOSA extinction images may be biased towards larger particles. Therefore, the scattering-to-absorption ratio was calculated from RDG-PFA theory, following the procedure detailed in $^9,42$. In Figure 9 (left image) $\rho_{sc}$ was calculated as a function of the
particle diameter with fractal dimension $D_f = 1.74$, fractal prefactor $k_f = 8.0$, geometric standard deviation of aggregate size distribution $\sigma_g = 2.3$, and geometric mean of number of primary particles in an aggregate $N_g = 31.4$. Since numerous values for $E(m)$ and $F(m)$ have been reported in literature, lower and upper values reported in literature were used in Figure 9: $E(m) = 0.37$, and $F(m) = 0.65$, and $E(m) = 0.2294$ and $F(m) = 0.1804 \; ^7$.

Values for $D_f$, $k_f$, and $\sigma_g$, were measured from thermophoretically sampled soot using TEM imaging in an atmospheric pressure non-premixed ethylene/air Gülder burner in \(^6\) $E(m)$ and $F(m)$ were derived from extinction coefficients in an ethylene coflow flame from \(^7\). These flames are most similar to flames we used. A maximum particle diameter of 125 nm was chosen based on values measured from light scattering in a similar flame \(^1\). This value is much higher than the peak mean value reported in \(^3\) of 48 nm at 15 and 20 bar found using thermophoretic sampling, but in their work, sampling was done by inserting the grids through the entire cross-section of the flame at different flame heights. The same work shows measured peak particle sizes of between 95 nm and 100 nm at 15 and 20 bar. These values are well outside of the Rayleigh approximation of $\pi d_p / \lambda < 0.3 \; ^{44}$.

As particle size increases $\rho_{s\alpha}$ also increases. Uncertainty in $\rho_{s\alpha}$ also increases based on $E(m)$ and

A sensitivity analysis (Figure 9, right image) was performed on $\rho_{s\alpha}$ again using RDG-PFA theory, with initial parameters of $D_f = 1.5$, $k_f = 6$, $\lambda = 435$ nm, $N_g = 30$, $\sigma_g = 2.3$, $E(m) = 0.37$, $F(m) = 0.65$, and $d_p = 20$ nm. For the sensitivity analysis, we have chosen the $D_f$ and $K_f$ values which lie within the range of commonly reported values

Figure 9: (Left) The scattering-to-absorption ratio as a function of the primary particle diameter at 435 nm and 647 nm using RDG-PFA theory. (Right) A sensitivity analysis of $\rho_{s\alpha}$ using RDG-PFA theory. $\rho_{s\alpha}$ is shown to be most sensitive to the primary particle diameter. $F(m)$.
for soot aggregates. Each parameter was doubled while all other parameters remained constant, and then the percent difference was found between the new and the original $\rho_{sa}$. Fractal dimension for a linearly chain aggregates is 1 whereas for a compact sphere it is 3. Doubling the Df = 1.5 still results in a physically possible value. Doubling the Df value, used for scattering to absorption ratio, for sensitivity analysis will be physically unreasonable. As shown on the left image of Figure 9, the values chosen for $E(m)$ and $F(m)$ contribute to uncertainty in $\rho_{sa}$ but changing $E(m)$ to 0.2294 and $F(m)$ to 0.1804 had a limited effect on results from the sensitivity analysis. The sensitivity analysis showed that $\rho_{sa}$ is most sensitive to the primary particle diameter. Previous work at atmospheric pressure suggests changes in $\rho_{sa}$ in the radial direction have limited effects on the results and add corrections for $\rho_{sa}$ only in the vertical direction of the flame \cite{36,42}. Measurements done in \cite{11} and in this work show significant variation in primary particle size between the centerline and wings of the flame. In this work, a comparison between measurements obtained using LOSA as compared to LII show that as pressure increases, assuming $\rho_{sa}$ changes only in the axial direction is no longer a valid assumption.

6 Conclusions

In this work 2D LOSA was performed at a shorter wavelength of 435 nm, similar to several previous studies \cite{10,25,36}, as well as at a higher wavelength (647 nm) to determine the significance of absorption by PAH. These measurements were compared with PLII performed at 2 different gate widths. By comparing the results from these two methods, sources of measurement uncertainties were determined and the following conclusions can be drawn.

- Quantitative SVF data at conditions relevant to practical combustion devices are reported. Peak SVF for diffuse LOSA at 435 nm scaled as $P^n$ with $n = 1.82 \pm 0.06$ and $n = 2.37 \pm 0.2$ in the centerline and wings respectively. Peak SVF for diffuse LOSA at 647 nm scaled as $n = 1.70 \pm 0.3$ and $n = 2.37 \pm 0.48$ in the centerline and wings respectively. LII was found to scale as $n = 1.71 \pm 0.18$ and $n = 1.72 \pm 0.24$ in the centerline and wings respectively. Centerline scaling is within the uncertainty between the two LOSA measurements. Discrepancies in the wings between LII measurements and LOSA measurements are a result of assuming constant optical properties throughout the flame.
• Carbon conversion efficiency scales as \( n = 1.31 \pm 0.17 \) and \( n = 1.47 \pm 0.21 \) for the 435 nm and 647 nm LOSA measurements. It scales as \( 1.37 \pm 0.43 \) for the 10 ns LII. Carbon conversion efficiency scaling shows better agreement between the measurements than peak SVF scaling.

The following conclusions on the effects of pressure on PAH and soot formation are drawn based on 2D LOSA, PLII, and PAH PLIF measurements:

• Qualitative PAH measurements are reported; additional data is available in the Supplementary Material. PAH formed nearer to the burner as pressure increased. This results in limited soot surface growth caused by PAH addition as the pressure increases.

• PAHs imaged with 350 nm, 400 nm, 450 nm, and 510 nm bandpass filters moved toward the burner at different rates with the highest changes occurring between 4 and 6 bar for the 400 nm and 450 nm filters, and 4 and 8 bar for the 510 nm filter.

• PAH absorption had a limited effect on the LOSA measurements. Absorption by PAH resulted in peak SVF locations for LOSA at 435 nm being located in a position lower than LOSA at 647 nm while peak SVF scaling at the two LOSA measurement wavelengths fell within measurement uncertainties.

• Quantitative soot profiles measured with 2D diffuse LOSA at high pressures have high uncertainty due to significant changes in soot morphology and soot optical properties throughout the flame. Knowledge of these properties is necessary to provide adequate measurement corrections. Modelers should use caution in making comparisons between simulations and experimental data obtained using extinction techniques.

• Soot volume fraction imaging with PLII is sensitive to the camera gate width. Longer gating times bias the results towards larger particle-size classes. SVF measurements obtained with PLII with longer gating times show better agreement with measurements taken using diffuse LOSA indicating LOSA measurements are biased toward larger particles.
• Uncertainty in $\rho_{sa}$ is highest in the flame wings where differences between the PLII measurements and LOSA measurements are highest. Calibration values for LII at high pressures should be determined at locations near the flame tip to decrease uncertainty in $\rho_{sa}$.

A greater understanding of how $\rho_{sa}$ changes in the radial and axial direction as well as with pressure is necessary to increase the reliability of extinction diagnostics.

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References


