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Instantaneous In-Flame Measurement of Soot Volume Fraction, Primary Particle Diameter and Aggregate Radius of Gyration via Auto-Compensating Laser-Induced Incandescence and Two-angle Elastic Light Scattering

Abbreviated title: Instantaneous soot volume fraction, particle diameter and aggregate size via AC-LII and 2A-ELS

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Abstract

A new combination of soot diagnostics employing two-angle elastic light scattering (ELS) and laser-induced incandescence (LII) capable of producing non-intrusive, instantaneous and simultaneous, in-situ measurements of soot volume fraction, primary particle size, and aggregate radius of gyration within unsteady flames is described. Controlled tests of the new apparatus on a well-characterized laminar flame show good agreement with existing measurements in the literature. From a detailed Monte Carlo uncertainty analysis of the results taking into account all sources of uncertainty, the combined LII/ELS technique was able to make shot-by-shot measurements of soot volume fraction, primary particle diameter and aggregate radius of gyration, with 95% confidence intervals of +37%/-31%, +37%/-31% and -19%/-16% respectively. For comparative measurements in which the properties of soot can be considered invariant among conditions, these uncertainties decrease significantly to +19%/-16%, +15%/-13% and +6.0%/-5.4% respectively. The apparatus is a powerful tool enabling detailed, instantaneous, spatially-resolved, simultaneous measurements of soot volume fraction and morphology within turbulent flames that are not possible with currently available techniques.

1. Introduction

Soot emissions are a known health hazard [1,2] and recently have been shown to be an important forcing factor for global climate change [3–5]. Soot is generally composed of mass-fractal aggregates of spherical carbonaceous particles which are generated as a consequence of incomplete combustion of hydrocarbons. The distribution of the diameter of the primary particles comprising the aggregates are relatively narrow and have been described as monodisperse [6], normal [7], or log-normal [7]. Conversely, the distribution of aggregate sizes, in terms of the number of primary particles per aggregate, N , or the radius of gyration of the aggregates, R_g , is generally quite wide and is commonly described as log-normal [7,8] or sometimes as self-preserving [6,8,9].

Modeling and predicting soot formation in flames is possibly the most challenging problem in the field of combustion given the complex chemistry, aerosol dynamics, short time scales, and large spatial gradients involved. This complexity is further exacerbated considering most practical combustion applications involve turbulent flames. Research on soot formation within turbulent sooting flames is hindered by the difficulty in making quantitative measurements. Large thermal and concentration gradients, optical attenuation, background interference, and beam steering all complicate measurements resulting in a dearth of experimental data for theory development or simulation validation.

A thorough description of soot emissions requires measurement of soot volume fraction, primary particle size, and aggregate size and morphological properties. Laser-induced incandescence (LII) [10–13] and line-of-sight attenuation (LOSA) [14–17] are commonly used to measure soot volume fraction in steady flames. While spatially-resolved measurements of soot volume fraction with LOSA require symmetry to deconvolve the line-of-sight measurements, LII is intrinsically spatially-resolved and thus offers the possibility of time-resolved point measurements in unsteady flames. When LII is collected at

two wavelengths with detectors calibrated to an absolute light intensity scale (known as auto-compensating or AC-LII) [18,19], it becomes possible to determine soot volume fraction without reliance on in situ or ex situ calibration with attenuation measurements [19]. With sufficient temporal resolution of the LII signal and a soot cooling model, it is also possible to infer a soot primary particle size [19–25]. Unfortunately, an important parameter in the soot cooling model is the local gas temperature, making primary particle diameter determination via time-resolved LII extremely difficult in unsteady flames where instantaneous temperature measurements can be prohibitive.

Soot primary particle size can also be measured via elastic light scattering (ELS) [26–28]. Both Sorensen et al. [26] and De Iuliis et al. [27] show that primary particle diameter can be determined via ELS if the soot absorption coefficient and aggregate radius of gyration are known. The soot absorption coefficient, which is also used to determine soot concentration, is commonly determined via extinction measurements with the assumption made that absorption by soot dominates over scattering [26–29]. Teng and Köylü [28] showed that at large scattering angles (i.e. back-scattering), the ELS signal is effectively independent of aggregate size, and is thus dependent only on primary particle diameter and the soot absorption coefficient. The addition of a second scattering detector at a forward scattering angle allows determination of an effective soot aggregate radius of gyration from an ensemble distribution of aggregate sizes [8,27,29]. Various attempts have been made to use scattering measurements at additional angles to discern more information about the aggregate size distribution [29–31]. However, deconvolution of multi-angle scattering measurements to determine the aggregate size distribution is very challenging due to the ill-posed nature of the problem [29,32] and thus a distribution shape (such as lognormal) and width, as well as morphological properties are commonly assumed [33–35].

A limitation of extinction plus scattering measurements is that spatially resolved absorption coefficients must be determined via deconvolution [36] and scattering coefficients are not obtained simultaneously. Thus the methods are typically applied to steady, axisymmetric flames [26,27,31,37]. Yang and Köylü [38], measured localized light extinction in an unsteady flame using both deconvolved LOSA measurements and local LOSA measurements for which a 6.4 mm OD pipe traversed through the flame was used to shield the laser beam from extinction outside of an exposed 2.3 mm sample length. Although this innovative approach permits spatially resolved measurements, the intrusiveness of the probe combined with the inability to determine extinction data concurrently with the ELS measurements means the method remains ill-suited for unsteady flames.

The use of LII in place of extinction measurements offers the possibility of simultaneous measurement of soot absorption and scattering characteristics for the instantaneous determination of soot concentration and aggregate morphological properties. The principle has been demonstrated by several authors [8,39]; however, the LII and ELS measurements were not obtained concurrently and averaged data was typically analyzed.

The objectives of this paper are to present and critically analyze an approach for achieving simultaneous, instantaneous point measurements of soot volume fraction, primary particle diameter, and aggregate radius of gyration via the combination of AC-LII and two-angle ELS. Measurements are performed in a steady laminar diffusion flame for the purpose of apparatus validation and quantitative assessment of measurement uncertainties with the specific goal of enabling future measurements in unsteady flames.

2. Theory

2.1 LII Measurement of Soot Volume Fraction, f_v

The LII method described in [18], including the equivalent filter approximation and numerical determination of the equivalent laser sheet width, was used to calculate soot volume fraction (f_v) via:

$$f_v = \frac{K_{abs,\lambda_{LII}}\lambda_{LII}}{6\pi E(m)_{\lambda_{LII}}} = \frac{V_{meas}}{\eta \frac{12\pi c^2 h}{\lambda_{LII}^6} E(m)_{\lambda_{LII}} w_e \left[\exp\left(\frac{hc}{k\lambda_{LII} T_{pe}}\right) - 1 \right]^{-1}} \quad (1)$$

where $K_{abs,\lambda_{LII}}$ is the soot absorption coefficient at the LII measurement wavelength, λ_{LII} , which is calculated as the equivalent center wavelength of the optical detection system [18]; $E(m)_{\lambda_{LII}}$ is the soot refractive index light absorption function evaluated at λ_{LII} ; V_{meas} is the experimentally-measured voltage; η is the calibration constant obtained using a calibrated tungsten lamp and integrating sphere [18]; c is the speed of light; h is the Planck constant; w_e is the equivalent laser sheet thickness; k is the Boltzmann constant; and T_{pe} is the equivalent heated particle (soot) temperature. The heated soot particle temperature is determined via pyrometry using LII detected at two distinct wavelengths [18] and $E(m)_{\lambda_{LII}}$ is taken to be 0.32 and assumed to be independent of wavelength. However, during the uncertainty analysis, the value of $E(m)_{\lambda_{LII}}$ is allowed to vary with a wavelength-dependence as described in [40].

The equivalent laser sheet thickness, w_{ex} was determined via numerical calculations based on a model of the LII process [41–43] as described in [18]. Calculations were performed for two laser profiles and a range of soot absorption functions, thermal accommodation coefficients, initial gas temperatures and primary particle diameters. The mean value of the resulting distribution of equivalent sheet thicknesses ($w_e = 0.45$ mm) was used for the results presented below, whereas the entire distribution of potential equivalent laser sheet thicknesses was used for the uncertainty analysis.

2.2 Determination of Primary Particle Diameter

As noted above, since the ratio of elastic light scattering to absorption at large angles (i.e. backward scattering) is effectively insensitive to aggregate size, it is possible to determine the soot primary particle diameter from a measurement of backscattered light without knowledge of the aggregate size distribution as [28]:

$$d_p = \frac{\lambda_s}{\pi} \left\{ 4\pi \frac{E(m)\lambda_s K_{vv}(\theta_2)}{F(m)\lambda_s K_{abs}\lambda_s} \frac{[2 \sin(\theta_2/2)]^{D_f}}{k_f} \right\}^{1/(3-D_f)} \quad (2)$$

where d_p is the primary particle diameter; λ_s is the wavelength of the laser used to produce the scattering signal; $E(m)\lambda_s$ and $F(m)\lambda_s$ are functions of the refractive index of soot and the ratio E/F is estimated to be 0.98 at $\lambda_s=532$ nm based on [44]; K_{vv} is the scattering coefficient for vertically-polarized incident and scattered light which is equal to the measured scattering signal voltage divided by an appropriate calibration constant determined using the method described in [45]; K_{abs,λ_s} is the absorption coefficient of soot at the scattering wavelength (evaluated in the current work based on simultaneous measurements of laser-induced incandescence and adjusted to the scattering wavelength using a correction factor $C_{\lambda_{scat}/\lambda_{LII}} = E(m)\lambda_s \lambda_{LII}/E(m)\lambda_{LII}\lambda_s$ [8]); and θ_2 is the backward scattering angle measured relative to the incident laser direction. The parameters k_f and D_f describe the mass-fractal nature of soot aggregates according to the mass fractal relation [46]:

$$N = k_f \left(\frac{2R_g}{d_p} \right)^{D_f} \quad (3)$$

where N is the number of soot particles in an aggregate, k_f is the fractal prefactor, D_f is the fractal dimension, and R_g is the soot aggregate radius of gyration. The values of k_f and D_f used in the current work are 2.4 and 1.7 [47]. A range of values for fractal prefactor and dimension and the ratio E/F were tested during the uncertainty analysis.

2.3 Determination of Radius of Gyration

Using the mass fractal aggregate theory introduced above combined with an additional forward scattering measurement, it is possible to determine the soot aggregate radius of gyration. In mass fractal aggregate theory for single aggregates [48], the only term dependent on the angle of measurement is the structure function, S , which is typically expressed as a function of the product of the scattering wave vector ($q = \sin(\theta/2) \cdot 4\pi/\lambda$) and the radius of gyration (R_g). De Iuliis et al. [27] presented a structure function for a polydisperse distribution of soot aggregates, S^* , expressed as a function of the product of the scattering wave vector (q) and R_{gm1} , a radius of gyration that is dependent on the mean number of primary particles per aggregate:

$$R_{gm1} = \frac{d_p}{2} \left(\frac{\bar{N}}{k_f} \right)^{1/D_f} \quad (4)$$

R_{gm1} is based on the first moment of N and we have maintained the nomenclature of De Iuliis et al. [27] for clarity. They used a curve-fit of a mean structure function, $\overline{S(qR_{gm1})}$, (determined via numerical calculation assuming N to be log-normally distributed with a distribution width of $\sigma = 2.1$) to represent the “distribution structure function” (S^*). De Iuliis et al. [27] validated the calculation of S^* at measurement angles of 45° and 135° for a range of primary particle sizes and geometric means of the distribution (N_m). The validated curve-fit subsequently allows R_{gm1} to be calculated based on experimental measurements of the ratio of scattering measurements taken at two distinct angles:

$$S^*(qR_{gm1}) = \frac{1}{1 + P_1(qR_{gm1}) + P_2(qR_{gm1})^2 + P_3(qR_{gm1})^3 + P_4(qR_{gm1})^4} \quad (5)$$

$$\frac{I_1}{I_2} = \frac{S^*(q_1 R_{gm1})}{S^*(q_2 R_{gm1})} \quad (6)$$

where the curve fit parameters are $P_1 = 0.03691$, $P_2 = 1.09688$, $P_3 = 0.09523$, $P_4 = 0.01555$, and I_1 and I_2 represent the measured scattering signals from the forward and backward scattering detectors, respectively. In the uncertainty analysis, (5) is not used. Instead, the calculations presented in [27] are

carried out for each iteration in the Monte-Carlo analysis using variates drawn from appropriate distributions of distribution width (σ), fractal prefactor (k_f), fractal dimension (D_f), forward and backward scattering angles (θ_1 and θ_2), and primary particle diameter (d_p). This is the most time-consuming step of the Monte-Carlo analysis by one to two orders of magnitude.

3. Experimental Setup

3.1 General Arrangement

To support future measurements on an unsteady turbulent flame greater than 1 m tall, the measurement apparatus was designed so that a platform supporting the laser head, beam shaping optics, and signal collection optics could be traversed in three axes about a fixed location burner. The general layout of the experimental excitation and collection system is shown in Figure 1. The 532 nm beam from the Nd:YAG laser is formed into a small sheet and directed into the flame where it induces incandescence and scattering. These signals are picked up by detection optics mounted at a forward angle of -30° and a backward angle of 144° and carried via fiber to a nearby filtering and detector apparatus.

Figure 2 shows a schematic of the signal handling during measurements. Laser excitation (a), induces both elastic light scattering and soot incandescence signals (b), which vary both spectrally (c), and temporally (d). Optical signals were collected at forward and backward angles (e), filtered spectrally (f), and attenuated using neutral density (ND) filters (g), before being focused onto separate detectors (h), which are further detailed below. The signals were then amplified (i), and gated in time (j), before being digitized (k) and stored in a computer (l).

3.2 Laser excitation

A frequency-doubled pulsed Nd:YAG laser (New Wave Research, SOLO 120) was used to produce a scattering signal and to induce incandescence of the soot particles. A schematic of the beam shaping layout is shown in Figure 1. The beam was first passed through a thin film polarizer (CVI Melles-Griot, TFP-532) to ensure only vertically-polarized light remained. The beam was subsequently expanded into a sheet using a pair of cylindrical lenses (focal length, $f = -100$ and 200 mm) and passed through a 7.5 mm x 0.5 mm slit to produce a thin sheet with a mean fluence of 0.8 mJ/mm². The slit was then imaged into the measurement volume using a 100 mm-diameter, $f = 400$ mm spherical lens. Laser beam profiles were taken at three evenly spaced heights within the measurement volume, as shown in Figure 3. In spite of the non-uniformity across the beam profile, the equivalent laser sheet thickness does not vary significantly from one profile to the next.

3.3 Collection optics

The forward- and backward-scattering collection optical assemblies were identical. Scattering and LII emissions first passed through a dichroic sheet polarizer (CVI Melles Griot, 03 FGP 007) to ensure only vertically-polarized light was collected. A mirror was then used to reflect the signals into a vertically-mounted lens tube. The lens tube contained two 50 mm diameter achromatic lenses ($f = 400$ mm and $f = 200$ mm) to collect and focus the emissions onto a 1 mm diameter optical fiber (FG-1.0-UAT, Thorlabs) which carried the signals out of the burner enclosure. Collection optics and tubes were placed at two locations on the measurement platform to allow collection of forward (-30°) and backward (144°) elastic light scattering. LII signals were simultaneously collected through the backward scatter optics as further detailed below. Alignment of the collection optics with the laser sheet was aided by directing a 671 nm diode laser back through the fiber providing a focused spot at the focal point of the collection optics.

A consideration for angular scatter measurements is that the intersection volume of the excitation laser and collection optical axis varies with collection angle and thus soot properties determined from a combination of measurements made at different angles can be biased if the soot properties are not uniform across both measurement volumes [49]. To avoid this problem, the forward and backward scattering collection optics were arranged along a common optical axis (detectors placed on opposite sides of the laser axis at -30° and 144°), such that their measurement volumes were coincident. By collecting the LI signal along this same optical axis, the identical sample volume was observed for both LI and scattering measurements.

3.4 LI Signal Treatment

After exiting the 1 mm diameter optical fiber, the light collected by the lens tube at **Error! Reference source not found**.144° passed through a collimating lens and a high pass dichroic mirror (Semrock model LP12-514RU), reflecting light at wavelengths below 514 nm toward the first LI detection package and transmitting higher-wavelength light through a 532 nm reflective notch filter (532 ± 8 nm, Semrock, NF01-532U) and into the second LI detection package. Each LI detection package contained a bandpass filter (447 ± 30 nm, Semrock model FF02-447/60-25 and 800 ± 32 nm, Coherent 42-7435 T-CXK 811-A, respectively), a calibrated neutral density filter (2.5% and 11.0%, respectively) to avoid detector saturation, and a $f = 50$ mm lens to focus the light onto the surface of a PMT (Hamamatsu H5783-03 or H5783-20 for low and high wavelength signals, respectively). The PMTs were operated at a gain setting of 500 V. The outputs of the PMTs were then amplified by a factor of five with a fast response amplifier (Stanford Research Systems SR240A) and integrated in time using gated integrators (Stanford Research Systems SR250) with a 50Ω termination. The 10 ns duration gates were initiated 6 ns after the onset of the PMT response to maximize the LI signal strength while minimizing interference from scattering. The integrated output was then digitized using two channels of a four-channel simultaneous 16-bit analog-to-digital converter (National Instruments, NI 9205). The detection system was calibrated to an absolute

radiance scale using a NIST-traceable filament lamp and integrating sphere system (SphereOptics Hoffman LLC, SMS-500) as described in [18].

A drawback to the early gating (6 ns after PMT response) was the possibility of interference from fluorescence of polycyclic aromatic hydrocarbons (PAHs) low in the flame, which would increase the measured signal and possibly influence the measured soot temperature and volume fraction. PAH fluorescence is not expected to be important at the current flame conditions [50]. While the potential influence of PAH fluorescence must be considered on a case-by-case basis in future measurements, the issue could be avoided altogether by using a longer wavelength for laser excitation. This has previously been difficult due to the poor sensitivity of PMTs (typically used for scattering measurements) beyond the visible spectrum. The new method of scattering detector calibration presented in [45] allows the use of other detectors (such as the photodiodes used in the current work) that have suitable sensitivity for scattering measurements in the near infrared.

3.5 Scattering

The backward scattered light was extracted using the reflection from the 532 nm notch filter described above. Each signal was then passed through calibrated neutral density filters (1.17% for forward scattering, 1.14% for backward scattering) and focused onto the surface a photodiode (Thorlabs DET-36A with 50 Ω termination). The detector outputs were integrated in time using gated integrators (Stanford Research Systems SR250) whose outputs were then sampled by the analog-to-digital converter. The scattering calibration was performed via diffuse surface scattering as described in [45], for which a 51 mm diameter by 6.5 mm thick piece of Spectralon Reflectance Material (SphereOptics Hoffman LLC, Contocook, NH) was traversed through the measurement volume in the laser propagation direction in increments of 0.2 mm. The resulting signal was fit using local polynomial regression and integrated numerically. A background value was measured by traversing the diffuse surface away from

the measurement volume a distance four times the full-width half-max of the measured signal, allowing subtraction of secondary light scattering off of other surfaces within the flame enclosure.

As discussed in [45], the diffuse surface scattering calibration method is only applicable to backward scattering measurements and as in [45], the absolute calibration of the backward scatter ELS detector was transferred to the forward scatter ELS detector using the same integrating sphere described previously, but instead illuminated by a small CW Nd:YAG laser (Crystallaser GCL532-075-L). The sphere output port was first arranged facing the backward scattering detector and then repositioned to face the forward scattering detector, with the CW laser beam entering the sphere from the same relative location in each case. The ratio of the detector outputs for the constant integrating sphere input provided a means to transfer the calibration constant from the backward scatter detector to the forward scatter detector.

3.6 Burner

Verification and validation experiments to assess the accuracy and repeatability of the combined LII/ELS approach were performed in an ethylene laminar co-flow diffusion flame previously described in [8] at a height of 42 mm above the fuel tube exit plane on the centerline. Briefly, the burner consists of an inner fuel tube (10.9 mm inner diameter) surrounded by an outer coflow of air (88 mm inner diameter). Ethylene was flowed through the inner tube at a rate of 0.24 grams per minute while air was flowed through the outer tube at a rate of 367 grams per minute. The visible flame height was approximately 65 mm.

4. Results & Discussion

The results of soot volume fraction, primary particle diameter and aggregate radius of gyration measurements in the laminar diffusion flame are shown in Figure 4. All measurements are reported

with measurement uncertainties which are discussed further in the next section. Also plotted for comparison are previously reported data obtained via TEM measurements [51], LI measurements [18], and multi-angle scattering [8], all made at the same location within an equivalent flame at identical conditions.

The strong correlation between soot volume fraction and primary particle diameter in Figure 4 underscores the importance of accurate soot volume fraction measurements when calculating primary particle diameter by this methodology. The mean soot volume fraction measurement of 3.4 ppm is 9% higher than the previous LI measurement [18] and the primary particle diameter of 25 nm is 12% lower than TEM measurements [51]. The present R_{gmi} data was compared to previous aggregate size measurements by first converting those measurements to a common form using consistent soot morphological and optical properties. Snelling et al. [8] reported the geometric mean number of particles in the aggregate size distribution determined via TEM. This was converted to R_{gmi} via (4) using $d_p = 28.3$ nm [51] and fractal parameters of $K_f = 2.4$ and $D_f = 1.7$ [47]. Snelling et al. [8] and Link et al. [29] report scattering coefficients at multiple angles. Using measurements at 35° and 145°, these were converted to R_{gmi} via (6) using $K_f = 2.4$ and $D_f = 1.7$ [47] and $\sigma = 2.1$ [27]. The present mean result of 85.4 nm is above the scattering and TEM measurements of Snelling et al. [8] and slightly below the scattering measurements of Link et al. [29].

4.1 Measurement Uncertainty

The strongly non-linear nature of the soot volume fraction equation, (1), suggests that a first-order linear error propagation method such as in [52] will produce erroneous results. Instead, Monte Carlo simulation similar to [40] was used for the propagation of elemental errors through to soot volume as well as to primary particle diameter and radius of gyration.

Most sources of uncertainty were characterized based on published literature values or manufacturer's specifications as detailed in Table 1. The soot absorption coefficient at the lower LII wavelength was taken to be a normal distribution truncated between 0.21 and 0.41 and centered at the nominal value of 0.32 with a standard deviation of 0.023 [40]. The equivalent laser sheet width distribution was calculated as described in [40] (based on measured laser profiles) and used to create a custom distribution of sheet widths from which random variates were drawn. The resulting distribution of sheet widths was approximately lognormal in shape with 95% of the values falling between 0.42 mm and 0.52 mm. Table 1 provides a summary of the Monte-Carlo simulation input parameters, their source, and the resulting 95% confidence intervals on f_{iv} , d_p and R_{gmm} , when all other terms are fixed.

$S^*(qR_{gmm1})$ is a curve fit through data points which assumes a distribution width $\sigma = 2.1$ and fractal parameters $K_f = 2.4$ and $D_f = 1.7$. However, there is uncertainty in these parameters as outline in Table 1. To account for these uncertainties on the scattering model, the Monte Carlo calculation numerically solved the scattering equations presented in [27] for each draw rather than via the curve fit provided in [27]. The Monte Carlo results indicated that the resultant R_{gmm1} is most sensitive to variation in σ .

Uncertainty analysis results of the Monte Carlo simulation are presented in Table 2. The analysis assumed the most challenging case of instantaneous measurements (i.e. assuming only single measurements are used and there is no time-averaging of the measured signals). The first of the three results columns represents the total uncertainty in each measured quantity. The uncertainty in the soot volume fraction and, as a result, in the primary particle diameter, is dominated by uncertainty in the soot absorption function $E(m_i)$. While the precision uncertainty was constant in the steady flame studied, it will be a function of the measured LII signal in the case of an unsteady flame making proper detector characterization a necessity in evaluating the uncertainty of such measurements. The major

contributor to the aggregate radius of gyration uncertainty is in the width of the log-normal distribution of aggregate sizes, σ .

The next column of Table 2 fixes values related to the characterization of soot ($E(m)$, $F(m)$, D_p , k_p , and σ) while allowing all remaining parameters to vary as outlined above. These results represent the relevant uncertainty when comparing one single measurement to another, since for any given flame location it is reasonable to assume that these soot properties are not changing significantly from one measurement to the next. The final column presents the uncertainty attributable only to measurement noise, and is instructive when considering possible improvements to the apparatus. For this latter case, the soot volume fraction uncertainty remains large due to the high photon shot noise associated with the use of PMTs used in detection of the LII signals [45]. The soot primary particle diameter uncertainty is dominated by the uncertainty in the soot volume fraction, while the uncertainty in the radius of gyration is quite low since the process of ratioing two measurement signals cancels out any shot-to-shot variations in the laser energy or flame movement.

Although the total uncertainties of 31-37% for instantaneous measurement of primary particle size and soot volume fraction shown in Table 2 are non-negligible, they are dominated by uncertainties in soot properties which are a common concern among all optical diagnostics for soot. For relative measurements in which soot properties can be assumed invariant among a given set of conditions, the achievable uncertainties of $\leq 18\%$ are quite encouraging. Moreover, the combined LII/ELS technique for the first time enables instantaneous, simultaneous measurement of soot volume fraction, primary particle diameter and radius of gyration within a turbulent sooting flame, and to the authors' knowledge there is currently no well-characterized alternative measurement apparatus that can produce equally detailed, non-intrusive, spatially-resolved, instantaneous data.

5. Conclusions

Combined laser-induced incandescence and two-angle elastic light scattering was shown to be a viable method of calculating instantaneous soot volume fraction, primary particle diameter and radius of gyration. A new experimental arrangement that places the two scattering angles on opposite sides of the incident laser beam was introduced in an effort to maximize measurement volume overlap. Measurements in a previously-studied flame show good agreement with published soot volume fraction, primary particle diameter and aggregate radius of gyration results. While the total uncertainty of the soot volume fraction (+37%/-31%) primary particle diameter (+37%/-31%) and the aggregate radius of gyration (+19/-16%) measurements are considerable, they are dominated by imperfect knowledge of soot properties which plagues all optically-based techniques for measuring soot aggregates and are thus not specific to the current apparatus. Likewise, the total uncertainty of the aggregate radius of gyration (+19/-16%) is dominated by uncertainty in the width of the polydisperse distribution of aggregate sizes, which is similarly difficult to determine in any experiment. Uncertainties for relative measurements, in which soot properties can be considered fixed among conditions, are much more modest at +19/-16%, +15/-13%, and +6.0/-5.4% for soot volume fraction, primary particle diameter and aggregate radius of gyration, respectively. Because the LI signals were decreased by 10 to 20 times using a neutral density filter to avoid detector saturation, with the present technique it should be possible to make measurements in more weakly-sooting flames by adjusting the filtering without increasing the levels of uncertainty reported here. By offering the possibility of simultaneous, instantaneous, and spatially resolved measurements of soot volume fraction and morphology within turbulent flames, the new combined LII/ELS technique has potential for acquiring critical data not available to other techniques.

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Figure and Table Captions

Figure 1 - Laser beam shaping and detector arrangement.

Figure 2 - Signal treatment flow chart.

Figure 3 - Profiles of laser fluence taken at four heights within the measurement volume.

Figure 4 – (a) Scatter plot comparing instantaneous measurements of soot volume fraction and primary particle size obtained via combined LII/ELS in the present work with 95% confidence intervals of total uncertainty for a single instantaneous measurement, uncertainty with fixed soot properties, and uncertainty considering instrument noise only. Also shown are soot volume fraction results from [18] corrected for the current choice of $E(m)$ plotted versus primary particle diameter results presented in [51] and calculated from the results in [8]. b) Histogram showing instantaneous measurements of soot aggregate radius of gyration with 95% confidence intervals of total uncertainty on a single instantaneous measurement, uncertainty with fixed soot properties, and uncertainty considering instrument noise only. Radius of gyration calculated from TEM measurements in [8] and calculated from scattering data from [8] and [29] are shown for comparison. All measurements taken on the burner centerline at a height above the burner exit of 42 mm (~65% along the visible flame length)

Table 1 - Input parameters used in the Monte-Carlo uncertainty propagation analysis.

Table 2 - Properties of the distributions resulting from Monte-Carlo uncertainty propagation. Mean values are followed by upper and lower bounds of the 2σ (95%) confidence intervals expressed both in absolute terms and as a percentage of the mean.

Figures and Tables

Figure 1

Figure 2

Figure 3

Figure 4

Table 1

Input	Parameters of Distribution	Source	Contribution to 2σ of $f_w d_w R_{sml}$
Normalized laser irradiance	Normal: $\mu = 1$; $\sigma = 0.02$	Manufacturer Spec.	0%, $\pm 4\%$, 0%
Soot refractive index light absorption function at lower LI wavelength [-]	Normal: $\mu = 0.32$; $\sigma = 0.023$ Min = 0.21; Max = 0.41	[40] and references therein	+20/-16%, $\pm 2.7\%$, 0%
Ratio of soot refractive index light absorption function at lower and upper LI wavelengths [-]	Normal: $\mu = 0.99$; $\sigma = 0.056$; Min = 0.8; Max = 1.2	[40] and references therein	+26/-24%, +20/-16%, 0%
Soot refractive index light absorption function at scattering wavelength [-]	Linear interpola ⁿ from $E(m_{\lambda_1})$ and $E(m_{\lambda_2})$. Contribution calculated using $\mu = 0.32$; $\sigma = 0.029$	[40], assumed linear change with wavelength	0%, +18/-14%, 0%
Ratio of soot refractive index light absorption to scattering functions at scattering wavelength [-]	Normal: $\mu = 0.98$; $\sigma = 0.098$	[44] and references therein	0%, $\pm 15\%$, 0%
LI calibration constant at lower wavelength, $\eta_{\lambda LI-1}$	Normal : $\mu = 3.90 \frac{m^3}{A}$, $\sigma = 0.016 \frac{m^3}{A}$	Calculated via constituent elemental errors	$\pm 4\%$, $\pm 4\%$, 0%
LI calibration constant at upper wavelength, $\eta_{\lambda LI-2}$	Normal : $\mu = 1.39 \frac{m^3}{A}$, $\sigma = 0.0095 \frac{m^3}{A}$	Calculated via constituent elemental errors	$\pm 4\%$, $\pm 4\%$, 0%
Equivalent laser sheet thickness	Custom, similar to Log-normal: $\mu = \ln(0.46 \text{ mm})$; $\sigma = \ln(1.06 \text{ mm})$	Numerical simulations [40]	+9/-12%, +10/-7%, 0%
Fractal dimension	Normal: $\mu = 1.7$; $\sigma = 0.15$	[47]	0%, $\pm 16\%$, +10/-8%
Fractal prefactor	Normal: $\mu = 2.4$; $\sigma = 0.2$	[47]	0%, +16/-12%, <0.5%
Width of lognormal distribution of aggregates	Uniform: Min = 1.7; Max = 2.5	[27]	0%, $\pm 2\%$, +14/-13%
Backward detection angle	Uniform: Min = 142° ; Max = 146°	Measured	0%, $\pm 2\%$, $\pm 3\%$
Forward detection angle	Uniform: Min = -28° ; Max = -32°	Measured	0%, $\pm 3\%$, $\pm 1\%$
Backward scattering calibration constant	Normal: $\mu = 24 \text{ m}^{-1}$; $\sigma = 0.65$	[45]	0%, $\pm 2\%$, <0.5%
Forward scattering calibration constant	Normal: $\mu = 7.8 \text{ m}^{-1}$; $\sigma = 0.39$	[45]	0%, $\pm 2\%$, $\pm 1\%$
Experimental measurement	LI at λ_1	Measured	+7/-6%, +5/-6%, 0%
	LI at λ_2	Measured	$\pm 12\%$, +10/-9%, 0%
Backward scatter	Normal: $\mu = 4.8 \text{ V}$; $\sigma = 0.024 \text{ V}$	Measured	0%, $\pm 2\%$, $\pm 1\%$
	Forward scatter	Measured	0%, $\pm 3\%$, $\pm 3\%$
Background during experimental measurement	LI at λ_1	Measured	<0.5%, <0.5%, <0.5%
	LI at λ_2	Measured	<0.5%, <0.5%, <0.5%
Backward scatter	Normal: $\mu = -0.00086 \text{ V}$; $\sigma = 0.015 \text{ V}$	Measured	<0.5%, <0.5%, <0.5%
	Forward scatter	Measured	<0.5%, <0.5%, <0.5%
Various calibrated neutral density filters	Normal: $\sigma = 1\%$ of nominal value	Based on specs of calibration device	0%, $\pm 4\%$, $\pm 4\%$

Table 2

	Total Uncertainty	Fixed Soot Properties	Measurement Noise Only
Soot volume fraction, f_v	3.4 { 4.8 ppm 37% } 2.4	3.4 { 4.0 ppm 19% } 2.8	3.4 { 3.7 ppm 14% } 2.9
Soot primary particle diameter, d_p	25 { 34 nm 37% } 17	25 { 29 nm 15% } 22	25 { 28 nm 12% } 23
Soot aggregate radius of gyration, R_{gIII}	85.4 { 103 nm 19% } 72	85.4 { 91.1 nm 6.0% } 81.3	85.4 { 87.8 nm 2.3% } 83.9