Application of an Optical Diagnostic (LII/ELS) to Measure
Soot Formation Trends within Turbulent Buoyant Non-
Premixed Flames

by

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Abstract

Flaring in the upstream oil and gas industry generates black carbon, which adversely affects human health and is an important positive climate forcer. Experiments were performed to investigate soot formation and oxidation within these large, flare-type, turbulent, non-premixed, buoyant flames. An optical measurement technique combining simultaneous auto-compensating Laser-Induced Incandescence (LII) and Elastic Light Scattering (ELS) measurements was used to measure the instantaneous soot volume fraction (f_v), primary particle diameter (d_p), and mean aggregate radius of gyration (R_{gml}) of soot black carbon within an optical probe volume. Measurements were completed on two comparable turbulent buoyant flames emanating from a 50.8 mm diameter pipe (lab-scale flare) with and without an internal turbulence generating grid. Centreline profiles of f_v , d_p , and R_{gml} were determined within rigorously quantified uncertainties obtained via Monte Carlo analysis. Results show that, for the conditions investigated, internal pipe turbulence has only a minor effect on the soot formation trends within the flame, suggesting that internal turbulence is likely less important than other factors when modelling and predicting soot formation in flare flames.

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Nomenclature

<i>Symbol</i>	<i>Description</i>	<i>First Equation Usage</i>	<i>First Use Page</i>
α_r	Resistance correction factor	(4.4)	56
$\beta(z)$	Angle between the Lambertian surface and backward collection optics	(4.2)	52
$\Delta\phi_{meas}$	Calibration signal measured at each location along the axis	(4.1)	52
$\Delta(\lambda_c)$	Width of bandpass filter	(4.4)	56
η	Calibration constant for detection channel	(2.3)	29
$\Theta(\lambda)$	Primary detector response	(2.2)	28
λ	Centre wavelength of the emitted radiation	(2.1)	27
ρ	Depolarization ratio of the Lambertian surface	(4.2)	52
σ_g	Width of the log normal distribution	(2.7)	32
$\tau(\lambda)$	Filter transmission	(2.2)	28
$\phi(\lambda, T_p)$	Total radiated power per unit volume of particle	(2.1)	27
Ω_{det}	Solid angle of detection optics	(4.2)	52
$\Omega(\lambda_c)$	Combined response of PMT tubes and filters at the centre wavelengths	(4.4)	56
<i>Symbol</i>	<i>Description</i>	<i>First Equation Usage</i>	<i>First Use Page</i>
A_{AP}	Area of the aperture	(2.2)	28
A_L	Area of the collecting lens	(2.2)	28
c	Speed of light	(2.1)	27
C_s	Structure factor coefficients	(2.9)	32
C_D	Scattering calibration constant	(4.3)	53
d_e	Inner diameter of the burner	(1.1)	18

d_p	Soot primary particle diameter	(2.12)	33
D_f	Fractal dimension	(2.5)	31
D_{filt}	Change in filter arrangement between the calibration and experiment	(4.3)	53
E_D	Energy scattered by the Lambertian surface	(4.1)	52
$E(m_\lambda)$	Soot refractive index absorption function at λ wavelength	(2.1)	27
f_n	Ratio of first two moments of the distribution of aggregate sizes	(2.13)	34
f_S	Stoichiometric mixture fraction	(1.1)	18
f_v	Soot volume fraction	(2.2)	28
Fr_g	Global Froude number	(1.1)	18
$F(m_\lambda)$	Soot refractive index scattering function at λ wavelength	(2.6)	32
g	Acceleration due to gravity	(1.1)	18
G_{exp}	Detector gain	(2.2)	28
h	Planck's constant	(2.1)	27
k	Boltzmann's constant	(2.1)	27
k_f	Fractal prefactor	(2.5)	31
K_{abs}	Absorption coefficient	(2.14)	34
$K_{vv,poly}(q)$	Differential scattering coefficient for a polydisperse distribution	(2.6)	32
M	Magnification	(2.2)	28
N	Number of primary particles per aggregate	(2.5)	31
$\bar{N}$	Arithmetic mean number of primary particles per aggregate	(2.11)	33
N_m	Geometric mean number of primary particles per aggregate	(2.7)	32

$p(N, N_m, \sigma_g)$	Lognormal probability density function for distribution of particle size	(2.6)	32
$\mathbf{q}$	Scattering wave vector	(2.9)	32
r_s	Total reflectivity of the Lambertian surface	(4.2)	52
R_i	Irradiance of incident laser beam	(4.2)	52
R_g	Soot radius of gyration	(2.5)	31
R_{gm1}	Soot mean aggregate radius of gyration	(2.11)	33
R_{vv}	Dissymmetry ratio	(2.8)	32
$S_{poly}(\mathbf{q}R_g)$	Structure factor of a polydisperse distribution	(2.6)	32
$\overline{S(\mathbf{q}R_g)}$	Effective polydisperse structure factor	(2.10)	33
T_p	Particle temperature	(2.1)	27
T_{pe}	Effective particle temperature	(2.3)	29
u_e	Flow exit velocity from the burner	(1.1)	18
V_{exp}	Measured experimental voltage	(2.2)	28
V_{mr}	Volume of measurement volume	(4.3)	53
w_e	Equivalent laser sheet thickness	(2.4)	29
Z	Impedance of the measuring device	(2.2)	28

Acronyms

AC-LII	Auto-Compensating Laser Induced Incandescence
BC	Black Carbon
CI	Confidence Intervals
CLSF	Carleton Lab Scale Flare Facility
DSMC	Direct Simulation Monte Carlo
EF	Emission Factor
ELS	Elastic Light Scattering
FWHM	Full Width at Half Maximum
HHV	Higher Heating Value
LII	Laser Induced Incandescence
MFC	Mass Flow Controller
NIR	Near Infrared
ND	Neutral Density
PAH	Polycyclic Aromatic Hydrocarbons
PM	Particulate Matter
PMT	Photomultiplier Tube
RDG	Rayleigh-Debye-Gans
SLPM	Standard Liters Per Minute (273.15K, 1 atm)
SVF	Soot Volume Fraction
TWG	Test With Turbulence Grid
TWOG	Test Without Turbulence Grid
UOG	Upstream Oil and Gas
VOC	Volatile Organic Compound

Chapter 1 Introduction

1.1 Motivation

Exposure to atmospheric Particulate Matter (PM) has been linked to an increase in cardiovascular and lung diseases (WHO 2018; Cohen et al. 2017; Grahame et al. 2014; U.S. EPA 2010). Black Carbon (BC), a component of particulate matter and soot, is a potent climate forcer due to its light absorbing properties and has been identified as one of the main reasons for accelerated polar glacier melting (Liu et al. 2017; Bond et al. 2013; Anenberg et al. 2012; Ramanathan and Carmichael 2008).

Flaring in the upstream oil and gas (UOG) industry is a well-known source of black carbon soot emissions and other pollutants (Conrad and Johnson 2017; Fawole et al. 2016; Huang et al. 2015). Flaring is a widespread practice in the UOG industry, in which natural gas is burned in a usually open flame at the top of a stack. The most common reason for flaring is to destroy excess gas that is considered uneconomical to use or sell. It is estimated that more than 140 billion cubic meters of gas is flared around the world annually (IEA 2018; Elvidge et al. 2015). In their latest annual flaring and venting report (ST60B), Alberta Energy Regulator (AER) reported 740 million cubic meters of gas were flared in 2017 from UOG sources in Alberta (AER 2018).

Black carbon emitted from flaring in northern regions such as Russia migrates toward the Arctic where it deposits on the surface leading to accelerated melting (Fawole et al. 2016; Stohl et al. 2013; Bourgeois and Bey 2011). Field measurements by Conrad and Johnson (2017) suggest that the mass of black carbon emitted from flaring may be underestimated in current inventories. Furthermore, recent laboratory measurements suggest the light absorption properties (mass absorption cross-section) of flare-generated BC are uniquely high compared to other combustion sources and are consequently underestimated in current climate models (Conrad and Johnson 2019).

Simple, single-value emission factors (EFs, e.g. mass of black carbon per unit volume of flared gas) are generally used to estimate particulate matter and volatile organic

compound (VOC) emissions from flares. The simplicity of single-value EFs ignores myriad factors such as burner geometry, flow regimes, fuel composition, and crosswind which can play an important role in the combustion mechanism (Johnson and Coderre 2012; McEwen and Johnson 2012; Pohl and Soelberg 1986; Dubnowski and Davis 1983). The lack of consideration of flame dynamics while determining EFs can lead to erroneous soot yield estimates (Fawole et al. 2016). In typical flare-type buoyant non-premixed turbulent flames, turbulent flame motion is predominantly generated either by the momentum of the cold flow or by the buoyancy of the rising hot gases (McEwen and Johnson 2012; Canteenwalla 2007; Delichatsios 1993). It is vital to study the influence of turbulence on soot formation and destruction in flare-type flames to determine the validity of current models, which do not explicitly account for turbulence effects, enabling more accurate and precise PM quantification.

1.2 Background

Soot is formed in high temperature regions of the flame where unburnt fuel is present. This unburnt fuel goes through a transformation to become long chain carbon polymers. There are several intermediate steps involved in the formation of soot clusters from unburnt fuel, as can be seen in Figure 1.1 (Michelsen 2017), but the process can be divided into three main sub-processes: inception, growth, and agglomeration (Bockhorn 1994). Wang (2011) outlines the work done on the soot inception particles and different pathways to polycyclic aromatic hydrocarbons (PAHs) formation, but there are still gaps in understanding this mechanism, which is an active area of research. Michelsen (2017) describes the inception and growth mechanism of soot particles and suggests that once formed, these “incipient particles” merge together to grow into bigger spherule-shaped particles 10-50 nm in diameter. These primary soot particles then agglomerate into fractal-type structures. The aggregate size of these agglomerates increases with reaction time in the flame, until they either escape the flame entirely or oxidize and break down again into smaller particles (Stanmore et al. 2001). As the number of available oxygen radicals increase in the flame, more soot is oxidized within the flame resulting in less soot leaving the flame (Nishida and Mukohara 1983; Schug et al. 1980).

Figure 1.1: Schematic representation of soot formation and oxidation process in combustion systems, taken from Michelsen (2017).

Soot formation is a complex phenomenon and is further challenging in the case of turbulent combustion. Turbulence in the flow field of the flame and its impact on chemical reactions affecting soot formation and oxidation are not fully understood but turbulent fluctuations have a direct impact on spatial soot distribution (Decroix and Roberts 2000). Turbulence affects the flame zone by introducing strain (stretching and wrinkling) fluctuations (Tieszen 2002; Köylü and Faeth 1992). These strain fluctuations give rise to toroidal vortices and affect the folding and mixing of flame zone structures inside the flame (Mueller and Schefer 1998). In a recent model presented by Attili et al. (2015) it is shown that the scalar dissipation rate controls soot formation by affecting the turbulent mixing and soot formation precursors. Delichatsios and Orloff (1989) proposed that the soot formation is controlled by the Kolmogorov time scale and that the local soot concentration

is inversely proportional to the strain rate $\sqrt{\frac{\epsilon}{\nu}}$, where ϵ is the dissipation of turbulent energy per unit mass, and ν is the kinematic viscosity, in turbulent flames. Yoo and Im (2007) developed a numerical model to investigate the relationship between soot and inflow turbulence for various turbulence intensities. They found that as turbulence intensity increases, the Kolmogorov eddy scale decreases, and enhanced mixing increases the flame volume. The increased flame volume results in an increase in soot yield, but the enhanced mixing also causes soot pockets to dissipate faster, since the turbulence carries the soot pockets away from high temperature regions (Yoo and Im 2007). This relationship between soot and inflow turbulence is further complicated by the changing mixing characteristics and different flow regimes i.e. laminar, transient, or turbulent, and needs to be understood in greater detail.

1.3 Flow regimes of turbulent non-premixed flames

According to the work done by Delichatsios (1993), for laminar jet flow combustion, after exiting the nozzle of the burner the flow undergoes a transition from laminar to turbulent due to either shear instabilities or buoyancy-induced instabilities. These buoyancy-induced instabilities are commonly seen in buoyant plumes as toroidal vortices made by rising hot gases mixing with the ambient air (Fang et al. 2016). Delichatsios (1993) presented a global Froude number Fr_g , to define different regimes in the flame and to predict if a flame will be buoyancy-dominated or momentum-dominated. It is given as (Delichatsios 1993),

$$Fr_g = \frac{u_e f_s^{\frac{3}{2}}}{(g d_e)^{\frac{1}{2}}} \quad (1.1)$$

where u_e is the burner exit velocity, f_s is the stoichiometric mixture fraction, d_e is the burner inner diameter, and g is acceleration due to gravity. This Fr_g can be seen as the ratio of inertial force on the fluid element to the gravitational force acting on it, and can be expressed as ratio of the momentum length to the buoyant length in flames (Delichatsios 1993). These regimes are shown in Figure 1.2 and are based on the transition mode theory of Delichatsios (1993). The momentum part is negligible for buoyant jet flames, which

means that the transition from momentum-dominated to buoyancy-dominated turbulent combustion will occur very early in the flame.

Figure 1.2: Estimated flare regimes as presented by Delichatsios (1993). In the Froude number used for the abscissa, S is the air to fuel mass stoichiometric ratio.

The shear-type instabilities bring variation in the rate of aerodynamic stress on the exit flow from the burner, and is defined in terms of strain rate (Du et al. 1989). The strain rate at the exit of the nozzle influences soot formation processes and has been studied substantially by many researchers as discussed in the work done by Mahmoud et al. (2018), Qamar et al. (2005, 2009) and the works cited therein. But little emphasis has been given to the effects of in-flow induced turbulence or pipe turbulence on the soot formation and oxidation rates in buoyancy-dominated turbulent diffusion flames. In the UOG industry, flares are burnt at the exit of a long vertical pipe, and so the pipe turbulence can affect the soot formation and oxidation rates in the flame, which prompts further investigation. McEwen and Johnson (2012) performed lab scale ex-situ soot yield measurements on buoyancy and momentum-dominated turbulent non-premixed flames for a range of flame conditions similar to what are found in the UOG industrial flares and found that the soot yield changed with changes in flow conditions. These results show the need for an in-situ

non-intrusive diagnostic technique able to obtain spatially resolved instantaneous measurements with sufficient temporal resolution to correlate the effects of internal flow turbulence only to soot formation rates in buoyant non-premixed turbulent flames.

1.4 Literature review

Table 1.1 summarizes studies that have focused on buoyant turbulent non-premixed flames relevant to industrial applications including flares, open furnaces, and pool fires. From Table 1.1, only a few researchers have performed in-situ soot measurements in buoyancy driven turbulent diffusion flames. Kent and Bastin (1984) performed optical extinction and temperature measurements on acetylene turbulent diffusion flames for various exit Reynolds numbers and found that at low flow-rate flows with $1/\tau < 20$ m/s, where τ is the flame time constant (Kent and Bastin 1984), the soot formation varied linearly with flow mixing rates. As the flow rate increased, the soot formation rate leveled out where the soot formation reaction time scales were slower than the mixing rates in the flame. Coppalle and Joyeux (1994) performed similar measurements on ethylene diffusion flames and found that for different exit velocities and Reynolds numbers, the lower velocity buoyant flame had a higher average soot volume fraction (SVF) both near the exit of the burner and towards the middle of the flame. Xin and Gore (2005) used laser-induced incandescence (LII) to measure SVF in methane and ethylene buoyant flames, and observed that soot only occupies a very small fraction of the space in the flamelets and that the SVF reaches maximum value at about the centre of the flame with high mean SVF fluctuation intensities observed near the annular region.

In a relatively more recent study, Qamar (2009) performed a detailed simulation of the distribution of SVF from a piloted turbulent jet non-premixed flame (Delft Flame 3). He found that a reduction in the number of soot sheets resulted in burnout rather than lesser values of SVF in those sheets, which suggests that the soot is found locally in strained flame sheets which are distorted by the eddies in the flow (Qamar 2009). The work of Mahmoud et al. (2018), Lee et al. (2009), Cardenas et al. (2007), and Qamar et al. (2005) has been concentrated on identifying the effects of varying Reynolds number and strain rate on soot concentration in momentum-dominated turbulent flames. They all generally conclude that the turbulence levels in the flow have an inverse relation to soot formation,

but the impact of exit Reynolds number on soot yield for a given fuel mixture is weaker compared to the impact of higher heating value (HHV) (via a change in fuel mixture) on soot formation. This relationship between the turbulence in the flow field and its impact on soot formation has only been studied for buoyancy-driven non-premixed turbulent flames by Crosland et al. (2015). They varied the burner exit nozzle and fuel flow rates and showed that there are non-linear relationships between residence time and soot concentration in turbulent buoyant non-premixed flames. They concluded that a decrease in mean SVF is linked to a decrease in the probability of the presence of a soot-laden zone, rather than a decrease in the SVF within such zones in the flame.

In continuation of the work of Crosland et al. (2015), Sawires (2017) employed the same combination of diagnostics but scaled up the set-up to allow it to be used with large-scale turbulent flames. This update was successfully tested with a non-premixed laminar flame burning ethylene on a well-characterized co-flow burner previously described in Snelling et al. (2004).

Table 1.1: Summary of research done on buoyant non-premixed turbulent jet flames

Author and year	Measurement	Method	Setup	Results
(Kent and Bastin 1984)	SVF and temperature	Laser light extinction and Kurlbaum temperature measurements	Experimental	<ul style="list-style-type: none"> SVF peaks towards the soot formation region and then declines Rate of soot burnout is dependent on temperature and temperature drop near the tip in slower flames makes them more sooty Delayed onset of turbulence in on-port flames resulted in lower soot formation Soot formation vary linearly with mixing rate for low flow rate flames
(McCaffrey and Harkleroad 1989)	Heat and mass release rate, radiative flux, and soot yield	Gaseous analyzer, soot characterization equipment (ex-situ), and thermocouple and radiometer (in-situ)	Experimental	<ul style="list-style-type: none"> No clear correlation was found between measured temperature and soot and CO yield Soot yield was found to have dependence on the burner diameter Urethanes and crude oil had more tendency to generate soot than propane, heptane or wood
(Sivathanu and Faeth 1990)	SVF and temperature	Laser extinction and two color pyrometry	Experimental	<ul style="list-style-type: none"> In the fuel rich region of a pool like fire, a solid correlation was observed between flame temperature and SVF The trends were found to be similar in acetylene and ethylene laminar flames Optical properties of soot depend on long residence times in fuel rich regions and have direct impact on the SVF state relationships
(Coppalle and Joyeux 1994)	Temperature and SVF	Two color pyrometry and absorption spectroscopy	Experimental	<ul style="list-style-type: none"> Correlation between temperature and SVF was studied It was found that turbulent mixing effects soot formation Buoyancy driven flames found to have more soot near the burner exit Soot concentration fluctuation were high and had low correlation to the measured temperature
(Faeth and Köylü 1995)	Structure of soot aggregates	Transmission electron microscopy (TEM)	Numerical and Experimental	<ul style="list-style-type: none"> Soot aggregate structure properties were found to be relatively independent of fuel types and flame conditions Whereas, soot aggregate size and primary particle size were found to be dependent on fuel type and flame conditions The measured fractal properties were $D_f = 1.82$ and $k_f = 8.5$ for a buoyant turbulent ethylene/air diffusion flame Models unable to predict the decrease in SVF at the outer flame edge
(Krishnan et al. 2000)	Extinction and scattering properties of soot	Light scattering and extinction	Experimental	<ul style="list-style-type: none"> Extinction coefficients were independent of increasing wavelength for 400 – 5200 nm Ratio of the scattering/absorption refractive index function $F(m)/E(m)$ doesn't depend on the fuel types Soot had large aggregate size due to large characteristic residence times

Table 1.1: Summary of research done on buoyant non-premixed turbulent jet flames (*continued*)

Author and year	Measurement	Method	Setup	Results
(Xin and Gore 2005)	SVF	Planar laser-induced incandescence	Experimental	<ul style="list-style-type: none"> Two-dimensional measurement of SVF, horizontal and vertical Soot particles exist in 1 to 2 mm thick streaks SVF ensemble averages increase away from the burner exit, reach a peak, and then decrease towards the flame tip Peak mean SVF is an order lower from the instantaneous measured value Anisotropic spatial correlation of SVF Soot distribution are relatively independent of fuel effects
(Yang and Köylü 2005a)	SVF	Transmission electron microscopy (TEM) and Laser extinction	Experimental	<ul style="list-style-type: none"> SVF measured by TEM overestimated by a factor of 2 to 3 compared to the laser extinction measurements Hypothesized that overestimation is due to deposition of young soot precursor particles, which are translucent to the laser wavelength, onto the probe
(Canteenwalla 2007)	Soot yield	Laser-induced incandescence (ex-situ)	Experimental	<ul style="list-style-type: none"> Soot formation and oxidation from a buoyant turbulent diffusion flame is too complex to be examined on a global level The synergistic effects have lower impact larger diameter burners as compared to the smaller ones
(Bisetti et al. 2011)	SVF, primary particle diameter, number of primary particles per aggregate	DNS with PAH finite rate chemistry by Hybrid Method of Moments soot model	Numerical	<ul style="list-style-type: none"> Scalar dissipation rate and mixture fraction effect the nucleation rate If soot move toward flame sheet it oxidizes faster than soot moving away which grows due to aggregation Regions with rich composition increase average number of primary particles per aggregate
(McEwen and Johnson 2012)	Soot yield per mass of fuel burnt	Gravimetric sampling system and Laser-induced incandescence	Experimental (ex situ)	<ul style="list-style-type: none"> Soot yield changes observed with change in regime from transition buoyant to transition shear Soot yield values approach a constant with fire Froude number 0.003 for different diameter burners

Table 1.1: Summary of research done on buoyant non-premixed turbulent jet flames (*continued*)

Author and year	Measurement	Method	Setup	Results
(Crosland et al. 2015)	SVF, primary particle diameter and mean aggregate radius of gyration	Laser-induced incandescence and two angle elastic light scattering (in situ)	Experimental	<ul style="list-style-type: none"> Studied soot formation in turbulent non-premixed flames for different burner diameters and exit velocities Primary particle diameter initially grows and then becomes stable between 31 to 43 nm In the soot burnout region, the mean SVF decreases due to the increased soot intermittency
(Mahmoud et al. 2018)	SVF	Laser-induced incandescence	Experimental	<ul style="list-style-type: none"> LII measurements were performed for turbulent non-premixed jet flames with different exit strain rates and varying Reynolds number Found that soot concentration is related flame volume and exit strain SVF found to have weak inverse dependence on exit Reynolds number

1.5 Thesis Objective and Outline

The objective of this thesis is to apply the updated set-up of Sawires (2017) to a large-scale buoyant non-premixed turbulent flame to observe the effects of induced flow turbulence on soot properties measured along the burner centreline. These empirical results are important as they are used to gain insight into the complex soot formation and destruction processes and can be used to verify the soot formation and turbulent combustion models that researchers use across the globe to model combustion systems and emission rates from flames.

Chapter 2 describes the theory that is used to measure SVF and soot morphological properties. The experimental set-up is described in Chapter 3, followed by the calibration procedures for the elastic light scattering (ELS) and LII techniques, the flame centreline mapping technique, and calibration validation results from a co-flow diffusion burner with the uncertainty analysis technique in Chapter 4. Chapter 5 presents experimental results for the two cases: with and without a turbulence-inducing grid in the burner exit tube (flare stack). Finally, Chapter 6 presents brief conclusions and recommendations for future work.

Chapter 2 Theory

2.1 General

To understand soot formation and destruction phenomena in conjunction with turbulence, researchers have used various in-situ optical and non-optical techniques to measure soot volume fraction f_v or SVF, primary particle diameter d_p , and mean aggregate radius of gyration R_{gml} , in both laminar and turbulent non-premixed flames. Laser Induced Incandescence (LII) is a widely used optical technique for studies of soot formation. In traditional LII, the soot particles are heated up to near or slightly above their sublimation temperature. In auto-compensating LII (AC-LII), the heated soot particles are deliberately kept slightly below the sublimation temperature using nanosecond-duration laser pulses as excitation. The auto-compensating LII method uses the incandescence signal measured at two different wavelengths to find an equivalent particle temperature through pyrometry and to subsequently determine the SVF. Because the heated soot temperature is measured and accounted for, external calibration is not required, and the soot does not need to be heated to its sublimation temperature.

The LII technique is useful for SVF measurements because it does not strongly depend on soot morphology. However, LII must be paired up with another technique if morphology information is desired. The Elastic Light Scattering (ELS) technique, in which the light scattered by soot particles is measured using photodetectors at one or more angles relative to the excitation laser beam, can be used to infer the aggregate size of fractal soot particles, and when coupled with LII can be used to obtain the primary particle diameter (Sawires 2017; Crosland et al. 2015; Oltmann et al. 2012; De Iuliis et al. 2011; Snelling et al. 2011; Yang and Köylü 2005b; Sorensen 2001). The apparatus and methodology employed in the current work is modeled on those developed by Crosland et al. (2013b, 2015), Sawires (2017), and the works referred to therein.

The general apparatus used in the current research was first described in Crosland et al. (2013b), and a subsequent version was used to make measurements in a small lab-scale flare in Crosland et al. (2015). The apparatus uses a single pulsed laser and a pair of

detection optics focussed on a common point along the laser path to probe a small volume within the flame. The laser light is both absorbed and scattered by soot that is present in the flame along the laser path. The absorbed light heats up the soot, increasing the incandescence of the particles by orders of magnitude. The increased incandescence is measured at two different wavelengths and used to determine the volume fraction of soot present in the measurement volume. The scattered light is picked up by a pair of detectors located at two distinct angular locations relative to the incident laser beam. The ratio of the scattering signals measured at these two angles is used to determine a mean radius of gyration of the soot aggregates in the measurement volume. Finally, both the SVF and the absolute intensity of one of the scattering measurements are used to determine the diameter of the primary soot particles that make up the aggregates.

By careful selection of the wavelengths at which measurements are made, it is possible to see three to five orders of magnitude increase in spectral radiation at typical flame and LII temperatures. Radiated emission from particles along the optical path but outside of the laser-heated zone is negligible compared to the radiated emission from particles in the laser-heated zone.

2.2 Soot volume fraction, f_v

The SVF is calculated using the Rayleigh-Debye-Gans (RDG) theory, which uses the simplifying assumption that the total radiative power emitted by a soot aggregate is equal to the sum of the power radiated by all the constituent particles that make up the aggregate (Snelling et al. 2005; Julien and Botet 1987). Using the notation of Snelling et al. (2005), the total radiated power per unit volume of particle is given by:

$$\phi(\lambda, T_p) = \frac{48\pi^2 c^2 h}{\lambda^6} \left[\exp\left(\frac{hc}{k\lambda T_p}\right) - 1 \right]^{-1} E(m_\lambda) \quad (2.1)$$

where c is the speed of light, h is the Planck constant, k is the Boltzmann constant, T_p is the particle temperature, λ is the centre wavelength of the emitted radiation, and $E(m_\lambda)$ is the soot refractive index light absorption function at λ .

A simple schematic representation (derived from Snelling et al. (2005)) of the light travelling from the probe volume to the PMT is shown in Figure 2.1. The probe volume is imaged on the circular pinhole aperture with a magnification $M = u/v$, where u is the distance between lens and probe volume and v is the distance between lens and the aperture. The measurement volume is approximately cylindrical, and is defined by a cross-sectional area of $M^2 A_{AP}$ and thickness of the laser sheet w_b , where A_{AP} is the area of the pinhole aperture. Assuming that all the soot particles in this volume are being heated to the same temperature with a uniform laser fluence over the whole cylindrical measurement volume, the above total radiated power in equation (2.1) when combined with the detection system calibration can be expressed as (Snelling et al. 2005),

$$V_{exp} = Z G_{exp} f_v M^2 A_{AP} \frac{A_L}{4\pi u^2} w_b \int_{\lambda} \phi_p(\lambda, T_p) \times \Theta(\lambda) \tau(\lambda) d\lambda \quad (2.2)$$

Figure 2.1: Schematic of the optical setup and ray trace for signal from probe volume to detector derived from Snelling et al. (2005).

where V_{exp} is the measured experimental voltage, Z is impedance of the PMT detector, G_{exp} is the detector gain, f_v is the SVF, A_{AP} is the area of the aperture, A_L is the area of the collecting lens (making $\frac{A_L}{u^2}$ the solid angle of detection of the collection optics), $\Theta(\lambda)$ is primary detector response and $\tau(\lambda)$ is filter transmission (Crosland et al. 2013b; Snelling et al. 2005).

To avoid reliance on a soot source for calibration, the auto-compensating LII technique is used (Snelling et al. 2005). In AC-LII two detectors, typically photomultiplier tubes (PMTs), detect laser-induced incandescence at two distinct wavelengths, λ_1 and λ_2 . The incandescence signals at these two wavelengths are then used to calculate an effective particle temperature (T_{pe}) using the two-color pyrometry (Wien's law) technique:

$$T_{pe} = -hc \frac{\lambda_2 - \lambda_1}{\ln \left(\frac{V_1 \eta_2 \lambda_1^6 E(m_{\lambda_1})}{V_2 \eta_1 \lambda_2^6 E(m_{\lambda_2})} \right) k \lambda_1 \lambda_2} \quad (2.3)$$

By using the above expressions and collecting the properties of the detection system into a calibration constant, the SVF can be expressed as:

$$f_v = \frac{V_1}{\eta_1 \frac{12\pi c^2 h}{\lambda_1^6} E(m_{\lambda_1}) w_e \left[\exp \left(\frac{hc}{k \lambda_1 T_{pe}} \right) - 1 \right]^{-1}} \quad (2.4)$$

where V is the measured voltage on a particular LII measurement channel (here channel 1 is used to refer to the lower wavelength channel), η_1 is the calibration constant for this channel (discussed in Chapter 3), and w_e is an equivalent laser sheet thickness, discussed below. Based on a literature survey described in Crosland et al. (2013b), a value of 0.348 is used at $\lambda_1 = 447$ nm, and a value of 0.303 is used at $\lambda_2 = 775$ nm (such that the ratio between them is $\frac{E(m_{\lambda_1})}{E(m_{\lambda_2})} = 1.15$). The above wavelengths are close to the ones selected by Liu et al. (2009a) as optimal for LII based on a compromise between LII signal strength, sensitivity, and shot noise.

To account for the non-uniform heating of soot particles, the laser sheet thickness w_b in equation (2.2) is replaced in equation (2.4) with an equivalent laser sheet thickness w_e . The equivalent laser sheet thickness is calculated using a numerical simulation for a given fluence distribution in the laser sheet (Crosland et al. 2013b; Snelling et al. 2000, 2005; Smallwood et al. 2002). The simulation source code was provided by Dr. Fengshan Liu (Snelling et al. 2000, 2005) to Dr. Brian Crosland, who then modified it for his work to run in multiple loops with variable model parameters to allow for the creation of probability distribution functions needed for Monte-Carlo uncertainty propagation (Crosland et al. 2011). This modified version is used here with permission from Dr. Brian

Crosland to calculate equivalent laser sheet thickness. The simulation algorithm (Liu and Smallwood 2012) used a modified version of a Direct Simulation Monte Carlo (DSMC) model for fractal aggregates to model heat conduction between these aggregates and the surrounding gas. The algorithm splits molecular motion and collisions between molecules into separate time steps. This type of conduction is the main heat loss mechanism for the soot particles. This simulation uses the harmonic mean of the continuum and free-molecular regime expressions to determine the heat transferred by conduction. For the free molecular and transition regimes, Liu et al. (2006) found that the Fuchs boundary-sphere model (Fuchs 1963) calculates the conduction heat loss rate more accurately than other models. This simulation requires the laser fluence profile of the laser sheet to be input along with a range of following parameter values including:

- soot index of refraction absorption function, $0.21 < E(m_{\lambda_2}) < 0.41$ and $0.8 < \frac{E(m_{\lambda_2})}{E(m_{\lambda_1})} < 1.2$;
- thermal accommodation coefficient, $0.2 < \alpha < 0.5$;
- initial gas temperature, $1500 \text{ K} < T_g < 2100 \text{ K}$; and
- primary particle diameter, $15 \text{ nm} < d_p < 60 \text{ nm}$.

The range for the soot index of refraction absorption function $E(m_{\lambda})$ is derived from the data presented by Coderre et al. (2011), Crosland et al. (2011), Snelling et al. (2004), Krishnan et al. (2001), and Köylü and Faeth (1994), while the thermal accommodation coefficient range is derived from Bladh et al. (2011), Crosland et al. (2011), and Snelling et al. (2004). The initial gas temperature range and primary particle diameter range are used from the work presented previously by Crosland et al. (2011). The laser fluence profile is measured using a beam profiler and a beam energy meter described further in Chapter 3.

The first part of the simulation generates a database for the temperature history of the soot aggregate of varying size d_p for various fluence values ranging from 0 to 2 mJ/mm^2 . The second part of the simulation derives an effective laser sheet width from

the data set by applying the user-supplied laser fluence profile. Snelling et al. (2005) have shown that for a non-uniform Gaussian laser profile, the maximum increase in the equivalent laser sheet width within 100 ns after onset of a low fluence (non-sublimating temperature, i.e. < 3400 K) laser beam is 5 %.

The largest source of uncertainty when performing the equivalent sheet width calculation for turbulent flames is the initial gas temperature. Though the primary effects of varying gas temperature are captured by the T_{pe} term in equation (2.4), there is also a higher-order effect seen in the equivalent sheet width calculation (Bejaoui et al. 2015). The equivalent sheet width code was run at initial gas temperatures of $1800 \text{ K} \pm 300 \text{ K}$, which caused the equivalent sheet width to increase by an average of $\pm 20 \%$ (Appendix D). The uncertainty in the equivalent sheet width, which includes the uncertainty due to unknown initial gas temperature, is carried forward to the determination of SVF and primary particle diameter as discussed later in Chapter 4 Section 4.5.

2.3 Mean Aggregate Radius of Gyration, R_{gm}

The mean aggregate radius of gyration is determined using the mass fractal aggregate theory of a polydisperse distribution of aggregates to interpret a measurement of the ratio of scattered signals at two different angles (Köylü et al. 1995a; Julien and Botet 1987). The number of primary particles in an aggregate, N , is given by (Julien and Botet 1987):

$$N = k_f \left(\frac{2R_g}{d_p} \right)^{D_f} \quad (2.5)$$

where k_f is the fractal prefactor, D_f is the fractal dimension, R_g is the radius of gyration, and d_p is the primary particle diameter. The fractal dimension D_f describes how compact the soot aggregate mass fractal is and when combined with the prefactor k_f , or the structure factor and aggregate size, can give us the scattering properties of the aggregate (Sorensen 2001; Köylü et al. 1995a; Sorensen et al. 1995).

Snelling et al. (2011) used RDG theory for light scattering from fractal aggregates to obtain the differential scattering cross-section of aggregates via a correlation between the soot aggregate's morphological and optical properties. The differential scattering coefficient for a polydisperse N -distribution is given as (Snelling et al. 2011):

$$K_{vv,poly}(q) = \frac{f_v}{\frac{\pi}{6}d_p^3} k^4 F(m_\lambda) \left(\frac{d_p}{2}\right)^6 \frac{\int_0^\infty N^2 p(N, N_m, \sigma_g) dN}{\int_0^\infty N p(N, N_m, \sigma_g) dN} \times S_{poly}(\mathbf{q}R_g) \quad (2.6)$$

where d_p is the primary particle diameter, k is the light wave vector ($2\pi/\lambda$), $F(m_\lambda)$ is the refractive index function for scattering, $S_{poly}(\mathbf{q}R_g)$ is the RDG structure factor for a polydisperse population of N aggregates, and $p(N, N_m, \sigma_g)$ is the lognormal probability density function for the distribution of the number of particles in an aggregate and is defined as:

$$p(N, N_m, \sigma_g) = \frac{\exp\left[-\frac{1}{2}\left(\frac{\ln N/N_m}{\ln \sigma_g}\right)^2\right]}{\sqrt{2\pi}N \ln(\sigma_g)} \quad (2.7)$$

where N_m and σ_g are the geometric mean and width of the log normal distribution, respectively. N_m is the unknown variable that will be determined based on the measurement.

Measuring the scattering at two different angles (30° and 150° , chosen based on the work of Teng and Köylü (2006)) enables the calculation of a ratio of two scattering coefficients wherein many terms in (2.6) cancel out, leaving a dissymmetry ratio R_{vv} given by:

$$R_{vv} = \frac{K_{vv,poly}(\theta_1)}{K_{vv,poly}(\theta_2)} = \frac{\int_0^\infty N^2 S(\mathbf{q}(\theta_1)R_g(N)) p(N, N_m, \sigma_g) dN}{\int_0^\infty N^2 S(\mathbf{q}(\theta_2)R_g(N)) p(N, N_m, \sigma_g) dN} \quad (2.8)$$

The structure factor for a single aggregate, $S(\mathbf{q}R_g)$, depends on θ and thus exhibits variation in scattering from the aggregate due to change in the scattering angle. A handful of simplified expressions for the structure factor of aggregates have been proposed by Sorensen (2001). As detailed in Crosland et al. (2013b), the choice of structure factor is treated as a modelling uncertainty. For ease of calculation the structure factor proposed by Lin et al. (1990) for a single aggregate is first used:

$$S(\mathbf{q}R_g) = \left(1 + \sum_{s=1}^4 C_s(\mathbf{q}R_g)^{2s}\right)^{\frac{-D_f}{8}} \quad (2.9)$$

where $\mathbf{q}$ is the scattering wave vector, $\mathbf{q} = \sin\left(\frac{\theta}{2}\right) 4\pi/\lambda$, and the values for the constant C_s are: $C_1 = 8/(3D_f)$, $C_2 = 2.5$, $C_3 = -1.52$, and $C_4 = 1.02$. An effective structure factor for a polydisperse population of aggregates can then be derived as (Snelling et al. 2011):

$$\overline{S(\mathbf{q}R_g)} = \frac{\int_0^\infty N^2 S(\mathbf{q}(\theta)R_g(N)) p(N, N_m, \sigma_g) dN}{\int_0^\infty N^2 p(N, N_m, \sigma_g) dN} \quad (2.10)$$

By using the expressions from in (2.7) and (2.9) in (2.10), and substituting the expression in (2.10) into (2.8), the only unknown (after measuring R_{vv} experimentally) is N_m . However, to facilitate comparison with existing literature, it is convenient to express the aggregate size not in terms of N_m , but instead in terms of an average radius of gyration. Considering again equation (2.5), if we take the first moment of the distribution $p(N)$ described in (2.7) and use the mean number of primary particles per aggregate as described in De Iuliis et al. (1998), a mean aggregate radius of gyration can be derived as:

$$R_{gm1} = \frac{d_p}{2} \left(\frac{\bar{N}}{k_f} \right)^{1/D_f} \quad (2.11)$$

where $\bar{N}$ is the arithmetic mean number of primary particles N , and is given by $\bar{N} = N_m e^{\ln\frac{(\sigma_g)^2}{2}}$. While equation (2.11) includes a primary particle diameter term, Crosland et al. (2013b) found that the R_{gm1} determined in this way is effectively independent of the assumed value of d_p . For calculation purposes, a value of 30 nm is assumed for d_p in the current work.

2.4 Primary Particle Diameter, d_p

Based on the theory presented above, the primary particle diameter can now be calculated by substituting equation (2.4) into equation (2.6) and solving for d_p , as described in De Iuliis et al. (1998),

$$d_p = \left(\frac{\lambda_s^3 4\pi E(m_{\lambda_s}) K_{vv}(\theta_2)}{\pi^3 F(m_{\lambda_s}) f_n \overline{S(q \cdot R_g)} K_{abs, \lambda_s} k_f (2R_{gm1})^{D_f}} \right)^{\frac{1}{3-D_f}} \quad (2.12)$$

where λ_s is the scattering wavelength (1064 nm in the current work), K_{vv} is the scattering coefficient for vertically polarized light, $F(m_{\lambda_s})$ is the scattering function of the soot refractive index at the scattering wavelength (0.31 (Crosland et al. 2013b; Yon et al. 2011)), f_n is the ratio of first two moments of the distribution of aggregate sizes, given by:

$$f_n = \frac{\int_0^\infty N^2 p(N) dN}{\left(\int_0^\infty N p(N) dN\right)^2} \quad (2.13)$$

and K_{abs} is the absorption coefficient, given by,

$$K_{abs,\lambda_s} = \frac{6\pi E(m_{\lambda_s}) f_v}{\lambda_s} \quad (2.14)$$

Chapter 3 Experimental Setup

The LII/ELS optical diagnostic setup layout is shown in Figure 3.1. Each component of this setup is discussed separately below. An Nd:YAG laser operating at its fundamental wavelength of 1064 nm was used to create a vertically fanned beam that passed through the flame where it was both absorbed and scattered by the soot particles. These scattered light (ELS) and incandescence (LII) signals were then collected by optical systems at a forward (30°) and a backward (150°) angle and measured by detectors. The detector signals were processed by gated integrators, digitized, and stored on a computer running LabVIEW software.

Figure 3.1: Overall layout of the LII/ELS experimental setup with optical and non-optical components

3.1 Optical Layout

The optical setup was based on the work presented in Crosland et al. (2015) and modified by Sawires (2017) to be used with the Carleton Lab Scale Flare (CLSF) facility's large turbulent flames. Key components of the system include a laser excitation system and forward and backward collection optics, which were mounted on a 1.5 m diameter Invar

hoop, shown schematically in Figure 3.2. Invar was chosen as the material for the hoop due to its low thermal expansion coefficient, which ensured stable alignment of the optical components attached to the hoop even under high thermal radiation from the 1 m to 1.6 m tall flames.

Figure 3.2: 3D schematic of the Invar hoop and mounted optical arrangement

3.2 Laser excitation system

Soot particles were excited with a New Wave Research SOLO 120 PIV Pulsed Nd:YAG laser. The second harmonic generator was removed so that light at 1064 nm with a pulse width of approximately 5 ns was generated, and only one of the two laser heads was used. It was triggered externally at 15 Hz with a TTL signal generated by a Berkeley Nucleonics Model 577 delay/pulse generator. As depicted in Figure 3.3, the laser intensity was regulated by passing it through a thin film polarizer and then a beam attenuator (Thorlabs VBA05), which consists of an adjustable $\lambda/2$ waveplate. The reduced-power beam was then passed through two cylindrical lenses of focal lengths -50 mm and 150 mm (Thorlabs LK1336RM-C and LK1419RM-C), to expand it into a thin vertical sheet. The edges of the sheet were then clipped by passing it through a 3 mm $\times$ 0.2 mm rectangular slit aperture

(S200R). The slit aperture was then relay imaged onto the measurement plane at the centre of the invar hoop using a spherical focusing lens of focal length 750 mm; two mirrors (NK1-K14) were employed to provide a more compact beam path. After passing the centre, the laser sheet was then captured in a beam dump placed at the far side of the hoop.

Figure 3.3: Photograph of the laser head mounted on the invar hoop with components as indicated. The laser beam path is indicated with thick red arrows.

The beam profile was measured using a Gentec Beamage-4M beam profiler. Figure 3.4 shows the spatial variation in laser energy for the whole beam at the location of the measurement volume (i.e. at the centre of the hoop). The precise measurement volume is thus the intersection of the profile in Figure 3.4 and an ellipse approximately $0.9 \text{ mm} \times 1.8 \text{ mm}$ in size. Shot-to-shot variation in the laser profile was negligible. The total energy of the beam was then measured using a Gentec QE25 energy meter and used to calculate the mean fluence of 0.89 mJ/mm^2 , which was sufficiently low to eliminate the risk of non-linearity in the emitted LII signals due to soot sublimation (Bladh and Bengtsson 2004). These data were used to calculate the equivalent laser sheet width (w_e) as discussed earlier in Chapter 2.

Figure 3.4: Laser beam profile with normalized fluence vs the height and width of the rectangular beam profile measured at the centre of the hoop

3.2.1 Backward collection optics

The backward collection optics consisted of two 3-inch achromatic doublet lenses (Newport PAC 09) with focal lengths of 750 mm and 250 mm. The longer lens collected and collimated light originating from its focal point; the shorter lens took collimated light from the longer lens and focused it to a point with a magnification of 1/3. Located at this point was a pinhole aperture, whose diameter when combined with the magnification of the lens system determined the size of the measurement volume. The intersection of the field of view of these optics and the excitation laser sheet formed an elliptical cylinder optical probe volume where soot was measured.

Light collected by the achromatic lenses was turned perpendicularly via a silver coated mirror (Thorlabs PF10-P01) with high reflectance over the 450 nm – 20 μ m range and focused onto a 300-micron pinhole aperture that restricted the elliptical measurement volume to $1.8 \text{ mm} \times 0.9 \text{ mm} \times 0.2 \text{ mm} \approx 0.25 \text{ mm}^3$. This choice was used to provide a

strong enough signal within the measurement volume relative to the total signal along the line of sight, both within and outside of the measurement volume, which resulted in low measurement noise and high spatial resolution (Sawires 2017).

As depicted in Figure 3.5, after passing through the pinhole aperture, the light was collimated using a 50 mm spherical collimating lens (Edmund Optics 32-323). This collimated light passed through three short pass filters:

- Shortpass filter # 1 – 400 nm (Edmund Optics 69-216) – For blue PMT
- Shortpass filter # 2 – 500 nm (Edmund Optics 69-214) – For blue PMT and alignment laser
- Shortpass filter # 3 – 850 nm (Edmund Optics 69-221) – For red PMT and scatter detector at 1064 nm

which reflected the unwanted radiation from higher wavelengths prior to reaching the PMT detectors. The radiation from the higher wavelength is reflected at 90° by these shortpass filters, which are mounted at 45° inside dichroic filter cage cube mounts (Thorlabs CM1-DCH).

A Hamamatsu H10721-110 PMT was used to measure LII signals for the lower wavelength following spectral filtering with a Semrock FF01-447/60-25 bandpass filter (centre wavelength 447 nm, full width at half maximum, FWHM, 60 nm), while an H10721-20 PMT spectrally filtered with a Semrock FF01-785/62-25 bandpass filter (centre wavelength 785 nm, 62 nm FWHM bandpass) was used for the higher wavelength (775 nm). At the scatter position, Shortpass filter # 3 reflected the filtered signal towards the Thorlabs detector (DET-10C) that measured the scatter signal at 1064 nm.

Figure 3.5: Schematic of the backward side detection system showing the filtration of the incoming broadband incandescence and scatter signal by various shortpass filters

A laser diode (Thorlabs CPS532) of wavelength 532 nm was placed at short pass filter # 2 to direct an alignment laser beam backward through the optical system to ensure that the probe location projected from the 300-micron pinhole aperture was concentric with the probe location projected by the forward side optical system. Alignment lasers at the forward and backward side helped to ensure the forward- and backward-side collection optics overlap exactly.

Figure 3.6: Backward collection optics assembly

All individual photosensors and detector locations were also equipped with motorized neutral density (ND) filter wheels (Thorlabs CFW6), which were remotely controlled through a LabView VI to adjust for the changing flame luminosity at different heights above the burner.

3.2.2 Forward collection optics

The forward collection optics, shown in Figure 3.7, were functionally identical to those used for the backward collection optics. Light was collected and focused on a pinhole aperture by a pair of 3-inch achromatic doublet lenses with focal lengths of 750 mm and 250 mm. A silver coated mirror (Thorlabs PF10-P01) turned the captured light 90° toward the (Thorlabs P300D) 300 micron pinhole aperture. A 50 mm focal length spherical collimating lens (Edmund Optics 32-323) then collimated the signal and sent it towards the 850 nm short pass filter (Edmund Optics 69-221), which then reflected the scatter signal through an ND filter in a motorized mount and onto the photodetector (Thorlabs DET-10C). An alignment laser diode (Thorlabs CPS633) of wavelength 633 nm was used

to align the probe volume from the forward collection optics with the probe volume from backward collection optics.

Figure 3.7: Forward collection optics assembly

3.2.3 Signal Processing

The LII and ELS signals from both the backward and forward collection optics were gated using temporally gated integrators (Stanford research system SR250), as shown in Figure 3.8. The output of the PMTs and photodiode detectors were terminated with $50\ \Omega$ terminations at the gated integrators. A 10 ns gate was used for the LII signals, which were delayed by 30 ns from the initial response to reduce uncertainty in equivalent laser sheet thickness, w_e , while still maintaining good signal strength for LII (Crosland et al. 2013b; Snelling et al. 2005).

To fully capture the ELS signal, which has a duration of about 15 ns, and to neglect any erratic scatter signal a gate width of 30 ns was used on the signal from the photodiode detectors. The timing between the incandescence signal onset and the gate placements were kept the same on both the LII channels using the inbuilt math/averaging function of

the WaveSurfer 3000z Oscilloscope. The gate placements were adjusted on the SR250 module until the deviation over a sample of 1000 pulses was < 0.3 ns.

Figure 3.8: Gates and signal time graph for LII and ELS signals from a co-flow diffusion burner Ethylene laminar flame. (a) Shows the LII signals from the upper and lower wavelength PMTs with 10 ns gates placed at 30 ns after onset of the signal. (b) Shows the ELS signals from the backward and forward scatter photodetectors with 30 ns gates placed

3.3 Non-optical setup

3.3.1 Carleton Lab Scale Flare Facility

The Carleton lab scale flare (CLSF) facility is designed to produce pipe flares representative of the upstream oil and gas industry. Flames can be up to ~ 3 m high at typical fuel flow rates of up to ~ 600 standard liters per minute (SLPM, at 0°C and 101.325 kPa). Figure 3.9 shows a schematic of the facility. To replicate different flare gas compositions, compressed gas bottles of pure species (C1-C4 straight-chain alkanes, CO_2 , N_2) are regulated and connected through various Bronkhorst® and Brooks® thermal Mass Flow Controllers (MFCs) and combined in a mixing manifold prior to being directed to the flare. Higher carbon fuels stored in liquid state (C5 to C7 hydrocarbons) are metered by Bronkhorst® Coriolis mass flow controllers (MINI CORI-FLOW™ M13 and M14) and

then vaporized by a Bronkhorst® CEM evaporator (W-303B) before being sent to the mixing manifold. The burner is located in a sunken hexagonal pit inside the lab, surrounded by two concentric fiberglass wire mesh screens to calm eddies or turbulence present in the room air so that the flame is free of impact from such external factors. A supplementary air supply system is also used to supply make up air to the pit via a perforated pipe to mitigate the effects of the irregular pit structure and air blockage due to the pit walls. This setup is discussed in detail in Jefferson (2017), Corbin (2014), and McEwen (2010).

A single fuel mixture denoted Alberta Heavy 9 was used in this study. The mixture properties are summarized in Table 3.1. This mixture had a slightly higher than average sooting tendency than the nominal average flare gas mixture for UOG sites in Alberta (Conrad and Johnson 2019). The fuel flow rate was set at 156 SLPM.

Figure 3.9: Schematic of the Carleton Lab Scale Flare (CLSF) facility with the traverse system and optical hoop

Table 3.1: Alberta Heavy 9 fuel composition, molecular weight (MW), volumetric higher heating value (HHV_v), molar air to fuel ratio (AF_m), stoichiometric mixture fraction (Z_{st}), kinematic viscosity (ν), and adiabatic flame temperature (T_{ad}), derived from Conrad and Johnson (2019)

Species	Mole fraction	MW [g/mol]	HHV _v [MJ/m ³]	AF _m	Z _{st}	ν [m ² /s]	T _{ad} [K]
Methane	0.7568	21.77	57.77	11.88	0.059	1.21×10 ⁻⁵	2235.16
Ethane	0.1152						
Propane	0.0606						
Butane	0.0260						
Pentane	0.0078						
Hexane	0.0023						
Heptane	0.0021						
Nitrogen	0.0170						
Carbon Dioxide	0.0122						

3.3.2 Traverse

A three-axis Parker Hannifin traverse was used to position the optical hoop relative to the flare. Figure 3.10 shows a 3D model of the traverse system. The traverse was controlled using a LabView VI via an ACR9000 controller system, which controls the servo drives for each of the three traverse axes. The controller can move each axis of the traverse in 1 mm steps, which provides high spatial resolution for optical measurements. All three axes were equipped with Heidenhain EnDat® 8D absolute encoders with resolution of 19 bits (524,288 pulse per unit), 4096 turns.

Figure 3.10: 3D model of the three-axis Parker Hannifin traverse system

3.3.3 Burners

3.3.3.1 Turbulent Diffusion Flame Burner

A turbulent diffusion flame (pipe) burner designed and used by Canteenwalla (2007), shown in Figure 3.11, was used in the CLSF facility to replicate the flare-type non-premixed turbulent flames on a lab scale. Fuel from the mixing manifold (Figure 3.9) entered the burner (Figure 3.11) and passed through a diffuser filled with 5 mm glass beads, followed by a settling chamber with fine mesh screens to uniformly distribute the flow. This distributed flow then passed through a converging nozzle which produced a uniform top-hat velocity profile at its exit as verified by hot-wire anemometry in Canteenwalla (2007). The burner exit inner diameter used in the current work was 50.8 mm.

Figure 3.11: Pipe flow burner schematic (Canteenwalla 2007)

An optional turbulence generating grid, shown in Figure 3.12, is installed near the burner exit to induce pipe turbulence similar to fully developed turbulent pipe flows. The grid was designed by Canteenwalla (2007) based on the work done by Deschamps (1990) and its design is discussed in detail in Appendix A.1 of Canteenwalla (2007). The turbulence intensity results for different Reynolds number measured by hotwire experiments for this grid are shown in Table 3.2.

Table 3.2 Turbulence intensity results from the turbulence generating grid showing, diameter (d), location of the grid in the burner, blockage ratio (σ), Reynolds number of the flow (Re), turbulence intensity ($u'/\bar{u}$), and turbulent length scale (l_t), taken from (Canteenwalla 2007)

Grid Number	d (mm)	σ (blockage ratio)	Re	$u'/\bar{u}$ (%)	l_t (mm)
10	50.8	0.49	208	2.0	0.1
			1162	5.3	0.8
			2145	3.1	0.3

To investigate the effects of internal flare stack turbulence on soot formation and oxidation, ELS and LII measurements were taken at the centreline of the flame with the turbulence grid installed in the burner and without the turbulence grid installed in the burner.

Figure 3.12: The turbulence generating grid that was placed five diameters upstream of the burner exit nozzle to induce flow turbulence

3.3.3.2 *Laminar Co-flow Burner*

A standard laminar co-annular, non-premixed burner was used to verify the calibrations as shown in schematic in Figure 3.13 (Snelling et al. 1999). The burner consisted of a central 10.9-mm inner diameter fuel tube surrounded by a 100-mm inner diameter concentric co-flow annular air nozzle. The standard operating condition used 99.9% pure ethylene at a flow rate 0.24 g/min (mean velocity of 0.034 m/s) and filtered air from the lab pressurized air line at a flow rate of 367 g/min (mean velocity of 0.695 m/s) producing a laminar diffusion flame with a visible flame height of ~65 mm. The fuel and air flow were controlled by a pair of SLA 5800 series Brooks® MFCs, calibrated using a Sensidyne Gilibrator 2 air flow calibration system for the ethylene MFC and a Bronkhorst® Fluical Portable Calibrator for the air MFC. This properties of soot present in this flame have been well-studied previously, with multiple measurements of f_v , d_p and R_{gml} available on the centreline at a height of 42 mm above the burner (Sawires 2017; Crosland et al. 2013b; Link et al. 2011; Snelling et al. 2011; Tian et al. 2006; Thomson et al. 2005).

Figure 3.13: Schematic of the co-flow annulus diffusion flame burner (Snelling et al. 1999)

Chapter 4 Calibrations and Methodology

4.1 Scatter (ELS) Calibration

The scattering calibration method was based on the work done by Crosland et al. (2013a). First, the detection system used to measure scattering at the backward angle was calibrated to an absolute radiance. Then a relative calibration between the backward and forward scattering detection systems was determined to allow for the measurement of the dissymmetry ratio described in Chapter 2 Eq (2.8).

To obtain a known scattering signal for the backward scattering detection system, a Lambertian surface was placed in the measurement volume to scatter the excitation laser light. The Lambertian surface produces a known calibration signal that is many orders of magnitude larger than Rayleigh scattering with common gases, enabling the use of photodiode-based detectors. Prior to the publication of the Lambertian-surface scattering calibration method researchers were obliged to calibrate via Rayleigh scattering. Though photodiode detectors are generally sufficient to measure flame scattering, the requirement to calibrate using very weak Rayleigh scattering signals necessitated the use of very sensitive detectors such as photomultiplier tubes. Photomultiplier tubes then needed to be heavily filtered during in-flame measurement to avoid detector saturation, thus reducing the number of photons being measured which in turn increases the photon shot noise component of measurement noise. The primary drawback of the Lambertian surface method is that the surface needs to be traversed through the measurement volume to calculate the total calibration coefficient, a step that is not required for Rayleigh scattering due to the optically thin nature of the gases that are typically used.

The use of photodiode detectors in place of photomultiplier tubes also enabled measurement at higher wavelengths (1064 nm) than commonly employed previously (Liu et al. 2013; Olofsson et al. 2013; Oltmann et al. 2012; Snelling et al. 2011; Reimann et al. 2009; Witze et al. 2001). Measurement at 1064 nm was important for a combined LII/ELS measurement system since LII that is excited at 532 nm is thought to be susceptible to interference from nearby C₂ swan bands (Bengtsson and Aldén 1995).

A Spectralon® diffuse reflectance surface standard puck of diameter 15 mm was used for calibration of the backwards-facing scattering optics, as shown in Figure 4.1.

Figure 4.1: Schematic depicting the backward scatter calibration arrangement with the Lambertian surface puck and the z-axis along which the measurements are taken

The Lambertian diffuse surface had a very high reflectance ($\sim 99\%$). Because the intensity of scattered light decreased following the Lambert's cosine law of reflectance as the detection angle moved away from the surface normal, it was only used to calibrate the backward side photodetector (for which the incidence angle to the surface normal was 30°). Average signals were collected as the diffuse surface was traversed across the measurement volume in 1 mm increments along the z-axis, as shown in Figure 4.2.

Figure 4.2: Backward side scatter calibration signal with 1 mm step locations across the measurement volume with error bars showing 95 % Confidence Interval (CI) on the mean of 1000 measured voltages at each position step

These collected signals from the diffused surface were then integrated over the entire length of the measurement volume along the z-axis:

$$\int_z \Delta\phi_{\text{meas}}(z)dz = \eta_D \int_z E_D dz \quad (4.1)$$

where $\Delta\phi_{\text{meas}}$ is the calibration signal measured at each location along the z-axis, η_D is the optical calibration constant and E_D is the scattered energy based on the Lambertian cosine law of reflectance:

$$E_D = R_i r_s \frac{(1 - \rho) \cos \beta(z)}{\pi} \Omega_{\text{det}}(z) A(z) \quad (4.2)$$

where R_i is the irradiance of the incident laser beam, r_s is the total reflectivity (99 %) of the surface, $\beta(z)$ is the angle between the diffuse surface and the backward collection optics (0°), $A(z)$ is the irradiated area of the diffuse surface visible to the backward signal collection optics, ρ is the depolarization ratio of the diffused reflectance surface (0.48 from

Crosland et al. (2013a)), and $\Omega_{det}(z)$ is the solid angle of detection. The origin of the z axis was approximately in the centre of the measurement volume, though the exact placement was unimportant.

Since the measurement volume length was small as compared to the length between the diffuse surface and the detection optics, $\Omega(z)$ and $\beta(z)$ were considered independent of the variable z . The calibration constant C_D was defined by combining equations (4.1) and (4.2) and isolating the invariant terms as described in Crosland et al. (2013a):

$$\eta_D R_i \Omega_{det} V_{mr} = C_D = \frac{\int \Delta\phi_{meas}(z) dz}{r_s \frac{(1-\rho) \cos \beta(z)}{\pi}} \quad (4.3)$$

The raw scatter signals coming from the photodetectors were amplified by a factor of approximately five by passing them through a SR 240A Quad Fast Preamplifier followed by a SR250 gated integrator module, which was operated such that it amplified its output signal by a gain factor of 1 V/5 mV (Volts out/Volts in). Since the calibrations had to be done on the same instrument gain conditions that were used during the in-flame measurements, the strong signal produced by the Lambertian surface puck needed to be filtered during calibration to avoid detector saturation. Two Thorlabs NIR ND filters were installed in tandem to avoid saturation of the photodetector during calibrations. The typical transmission and uncertainties for the Thorlabs ND filters were: 54.6 % $\pm$ 0.2 % for NENIR02B ND filter and 0.75 % $\pm$ 0.2 % for NENIR20B ND filter at 1064 nm. For six repeated tests, the absolute calibration constant with repeatability error was calculated to be 99.9 V m $\pm$ 0.9 V m.

To transfer this calibration to the optics on the forward-scattering side, a diffuse surface integrating sphere was used. The integrating sphere was also a Lambertian coated surface and diffusely reflected the incident radiation following the Lambert's cosine law. As depicted in Figure 4.3 (a), the output reflectance port of the integrating sphere was first oriented to capture excitation laser light while remaining visible to the backward collection optics. After recording the signals captured by the backward-scattering optics, the process was repeated with the forward-scattering optics, as shown in part (b) of Figure 4.3. The ratio of the two measured values was used to transfer the calibration constant from the

backward collection optics to the forward collection optics as shown earlier in work done by Crosland et al. (2013b).

Figure 4.3: Transferring scatter calibration from backward side optics (left) to forward side optics (right) using an integrating sphere

For six repeated tests, the absolute calibration constant of $56.9 \text{ V m} \pm 0.49 \text{ V m}$ was measured at the forward side scatter detector location with a forward to backward signal ratio of 0.57. The spread of the measured voltages is shown in Figure 4.4. From the repeated tests, the error on the calibration procedure was found to be 2% at a 95% confidence level.

Figure 4.4: (a) Box and whisker plot for six calibrations with mean and 95% CI calculated by a t-test are plotted with the blue dashed lines, (b) Measured signal voltages vs. position in the measurement volume gathered during these six different calibrations

4.2 Incandescence (LII) Calibration

An integrating sphere illuminated by a calibrated light source was used for the absolute intensity calibration of the LII detection system. The light source, a 100 W Rhodium lamp, was attached to the integrating sphere along with a radiometer which provided the absolute radiance of the Rhodium lamp measured directly from the integrating sphere. Using this radiometer, the absolute radiance of the Rhodium lamp at the exit port of the integrating sphere was found to be $0.89 \text{ mW/cm}^2\text{-sr}$. The spectral radiance at each LII measurement wavelength was then determined by scaling the spectral radiance reported on the integrating sphere calibration certificate by the ratio of the radiometer readings during LII calibration and from the certificate. The spectral radiances for each LII measurement channel were thus $0.12 \text{ mW/cm}^2\text{-sr-}\mu\text{m}$ at 445 nm and $0.75 \text{ mW/cm}^2\text{-sr-}\mu\text{m}$ at 784 nm. The LII calibration theory was based on the work presented by Crosland et al. (2013b), and Snelling et al. (2005).

Figure 4.5: Labsphere® Integrating Sphere used for both LII calibrations and to transfer the ELS calibrations from backward side optics to forward side optics.

As the spectral radiance at the sphere output port was known for a given wavelength, the voltage measured at the PMT was described by (Snelling et al. 2005):

$$V_{cal} = G_{cal} Z M^2 A_{AP} \frac{A_L}{u^2} \alpha_r R_s(\lambda_c) \Omega(\lambda_c) \Delta(\lambda_c) \quad (4.4)$$

where G_{cal} is the PMT gain setting, Z is the impedance of the measuring device, M is the magnification of the optics, A_{AP} is the area of the aperture, A_L is area of the lens, u is the distance between the measurement volume and the collection optic lens, α_r is the resistance correction factor, $R_s(\lambda_c)$ is the spectral radiance of the light source, $\Omega(\lambda_c)$ is the combined response of the PMT tubes along with the filter transmission and $\Delta(\lambda_c)$ is the width of the bandpass filter.

A calibration constant, η , was defined by isolating the invariant terms as (Snelling et al. 2005):

$$Z M^2 A_{AP} \frac{A_L}{u^2} \Omega(\lambda_c) \Delta(\lambda_c) = \eta = \frac{V_{cal}}{R_s(\lambda_c) \alpha_r G_{cal}} \quad (4.5)$$

Rather than relying on manufacturer's specifications relating the PMT gain control voltage to the actual PMT gain, the calibration process was repeated for a range of gain settings expected to be used during in-flame measurements. This allowed the G_{cal} term in Equation (2.2) to be removed, leaving only the voltage measured during calibration, V_{cal} , and the spectral radiance at the wavelength of interest, $R_s(\lambda_c)$, in the calibration coefficient expression. Because PMTs respond non-linearly at much lower light levels when exposed to continuous light rather than pulsed light, the lamp in the integrating sphere was partially blocked with an aperture and the PMT output current was monitored with an ammeter to ensure the PMTs were not being saturated during calibration. The output from the radiometer in the integrating sphere was used to determine the spectral radiance inside the lamp, after the aperture. Once it was verified that the PMTs were in their linear regime, the PMT output was connected to the gated integrator system where the output was terminated with a resistor to convert the PMT current output into a voltage output. During calibration a large resistance of 50 k Ω was used to amplify the very weak PMT output to a voltage suitable for measurement by the gated integrator. Since the integrating sphere provided a continuous signal, the slow time response imposed on the signal by the 50 k Ω resistor was acceptable. During in-flame measurements the 50 k Ω resistor was replaced with a 50 Ω resistor to ensure a fast temporal response and a resistance correction factor α_r was added to the calibration calculations to account for this change.

The calibration constants measured were $2.34 \times 10^{-11} \pm 0.045 \times 10^{-11} \text{ m}^3/\text{A}$ for the blue PMT and $7.67 \times 10^{-12} \pm 0.086 \times 10^{-12} \text{ m}^3/\text{A}$ for the red PMT. Due to the very stable output radiance of the Rhodium lamp light source, the calculated calibration constants remained unchanged for a given single sphere radiance over six repeated calibrations.

4.3 Flame Imaging and Centreline Mapping

The buoyancy driven flames with low fuel exit velocity tended to drift away from the centre axis of the burner, presumably due to room air currents in the vicinity of the flare. Therefore, it was vital to map the true centreline of the flame to ensure soot measurements were made on the centreline. A pair of PCO Edge 5.5 sCMOS cameras was used to acquire images of the flame from two different view points perpendicular to each other, as shown in Figure 4.6. The frame rate of cameras was chosen to ensure flame images were

independent considering the typical residence time of the flame and so both the cameras were simultaneously triggered at 1 Hz for 20 minutes, acquiring a total of 1200 images from each camera to be averaged. Two pairs of sample images from the set of 1200 images are shown in Figure 4.7. Each individual image was binarized at a threshold of approximately 50 % of the change in brightness between the background and the flame. The 1200 binarized images were then averaged to produce an image representing probability of flame presence, which in turn was binarized at a value of 0.5 yielding an image (see Figure 4.8) depicting a zone where the flame is expected to be present > 50 % of the time.

Figure 4.6: CLSF schematic depicting camera positions, the fuel bottle arrangement and the exhaust system of the pit. The cameras were installed 1.7 m above the pit floor on a height adjustable railing system.

Figure 4.7: Still images of the flame taken from camera 1 and 2 at same instances from two different angles

For each row of pixels, the flame centreline was then defined as the midpoint between the two edges of the $> 50\%$ zone. The computed centrelines are shown overlaid on the $> 50\%$ flame presence zones in Figure 4.8

These centrelines were then located in the co-ordinate system of the traverse at ten discrete heights above the burner. This was achieved by mounting an LED at the centre of the hoop and adjusting the co-ordinates of the traverse in three dimensions until the LED appeared in live images from both the cameras on the centreline found in the previous paragraph. The co-ordinates on the traverse were then recorded for use later in the day during flare testing. This process was tested four times, with the flame centreline showing a similar inclination angle of $9^\circ \pm 2^\circ$ from the centreline of the burner. The entire process was performed before each of the two experimental tests to ensure the measurements were made as close to the flame centreline as possible.

Figure 4.8: Averaged contour (50 %) of the flame with overlaid centreline from each camera

4.4 Verification of Calibrations with a Laminar Diffusion Flame

Immediately before each measurement on the turbulent buoyant non-premixed flare flame, the calibration of the optical setup was verified using the laminar co-flow burner described earlier in Chapter 3. These reference laminar flame results are shown in Figure 4.9, and are referred to as “pre-TWG” and “pre-TWOG” to distinguish between the laminar flame verification done in advance of the flare test with a turbulence-generating grid (TWG) and the laminar flame verification done in advance of the flare test without a turbulence-generating grid (TWOG). The 95 % prediction intervals for single instantaneous measurements of each parameter are indicated by the dashed error bars, calculated using the Monte-Carlo error propagation method further detailed in Section 4.5. The total uncertainties were dominated by the uncertainty in the soot optical properties used to calculate f_v , R_{gml} , and d_p . Considering the overall uncertainties, the mean values for the pre-TWG and pre-TWOG cases agreed well with each other and with results from the literature (pre-TWG / pre-TWOG, $f_v = 4.9 \text{ ppm}/4.9 \text{ ppm}$; $d_p = 30 \text{ nm}/29 \text{ nm}$, and $R_{gml} = 63 \text{ nm}/62 \text{ nm}$).

Figure 4.9 also shows results from literature that were made in the same laminar flame configuration, at the same location: TEM measurements used to calculate d_p (Tian

et al. 2006), LII measurements used to calculate f_v , (Snelling et al. 2005), multi-angle scattering measurements used to determine R_g (Link et al. 2011; Snelling et al. 2011), and combined two-angle ELS and AC-LII measurements to calculate f_v , R_{gml} , and d_p (Sawires 2017; Crosland et al. 2013b). The TEM data from Snelling et al. (2011) were converted to comparable values by Crosland et al. (2013b) using common soot optical and morphological properties. These identical soot optical and morphological properties were then used by Sawires (2017) for different soot measurement volumes to calculate the f_v , R_{gml} , and d_p , which are compared with the present results. A summary of these previously reported measurements and a comparison with our measured mean values for f_v , d_p , and R_{gml} are summarized in Table 4.1.

Figure 4.9: Scatter plot for the measurements from co-annular burner laminar flame taken immediately before the TWG and TWOG cases on the flare flame. All measurements were taken at the centre of the laminar flame at a height of 42 mm above the burner exit. Scatter plots show instantaneous measurements of, (a) SVF (f_v) and primary particle diameter (d_p), and (b) mean aggregate radius of gyration (R_{gml}) and primary particle diameter (d_p), with 95% prediction interval of total uncertainty for a single instantaneous measurement represented by dotted lines. The solid lines represent measurement uncertainty assuming fixed soot properties (i.e. attributable to instrument noise and calibration). Included for comparison are results of previous measurements by others on the same type of burner and flame as presented earlier in Crosland et al. (2013b).

These comparisons show general agreement between the mean measurements presented in the current work and the previously presented measurements as summarized in Table 4.1. The mean f_v values in the current work were 16 % to 19 % higher than all the reported values of Crosland et al. (2013b), and Snelling et al. (2005), but matched those of Sawires (2017) using the same measurement volume, optical setup, and burner setup. This difference might also be due to small discrepancies in alignment of the system to the measurement point on the centreline of the laminar flame, 42 mm above the burner exit. Some flicker was also visibly apparent in the laminar flame tip, which may have affected the repeatability of the burner in comparison to previous measurements in the literature. The measured d_p values also matched those of Sawires (2017). The mean R_{gm1} values in the current work were 3.1 % and 3.6 % higher than the values of Snelling et al. (2011) and Link et al. (2011) as adjusted and reported in Crosland et al. (2013b) and 27% lower than those of Sawires (2017). The alignment, calibration and testing on laminar flame were repeated several times to match the mean R_{gm1} values reported previously by Sawires (2017) but the values couldn't be repeated. Overall this comparison demonstrates the ability of the system to reproduce standard measurements well within uncertainties, which tend to be dominated by large uncertainty in the soot optical properties and laser sheet thickness as further discussed in the following section.

Table 4.1 Summary of the reported values measured on the co-flow diffusion flow burner laminar flame with comparison to our measured mean values

Sources	f_v [ppm]	d_p [nm]	R_{gm1} [nm]	Deviation from present value		
				f_v compared	d_p compared	R_{gm1} compared
Present Work	4.9	29	63	-	-	-
Sawires 2017	5	29	80	2.6%	-0.3%	27%
Crosland 2013	3.8	36	70	-23%	23%	12%
Snelling 2011 TEM	4	25/28	69	-19%	-3.4 to -13%	9.7%
Snelling 2011 ELS	4	25	61	-19%	-3.4%	-3%
Link 2011 ELS	4	25	60	-19%	-3.4%	-3.5%

4.5 Uncertainty Analysis

A Monte Carlo (MC) simulation derived from the work of Crosland et al. (2011) was used to calculate total uncertainties in f_v , d_p , and R_{gm1} . MC analysis was ideal for this type of

complex measurement system in which several input parameters with different probability distributions were non-linearly related to the outputs. To quantify the uncertainty distributions in f_v , d_p , and R_{gm1} , each parameter was calculated $N = 20,000$ times using input parameters that were drawn at random from representative distributions. Upper and lower 95% CI were then calculated from the resulting distributions of the N values of f_v , d_p , and R_{gm1} .

The mean, standard deviation, and distribution type for each of the input variables and its contribution to the overall uncertainty on each of the three output parameters are shown in Table 4.2. The soot morphological properties were derived from Crosland et al. (2013b). Other measured uncertainty parameters such as uncertainty in calibration factors and measured voltages from co-flow burner measurements taken before TWG are discussed below. The contribution of each individual variable to the overall uncertainty (95% CI) on f_v , d_p , and R_{gm1} was estimated by performing a standalone MC analysis and letting only a single parameter vary while fixing all other parameters.

Table 4.2 Input parameter used in the MC simulation to calculate measurement uncertainties and their relative contribution to the 95% CI on f_v , d_p and R_{gm1} measurements in the reference laminar flame when other terms are fixed. The variable distributions and their sources are taken from (Crosland et al. 2013b)

Input	Parameters of Distribution	Source	Contribution to 2σ of		
			f_v	d_p	R_{gm1}
Variables					
Soot refractive index light absorption function at lower LII wavelength $[E(m_{\lambda_1})]$	Normal: $\mu = 0.348$ $\sigma = 0.045$	(Coderre et al. 2011; Yon et al. 2011; Bond and Bergstrom 2006; Snelling et al. 2004; Schnaiter et al. 2003; Krishnan et al. 2000; Köylü and Faeth 1996; Dobbins et al. 1994)	+31/ -22%	+19/- 20%	0%
Ratio of soot refractive index light absorption function at lower and upper LII wavelengths $[E(m_{\lambda_1})/E(m_{\lambda_2})]$	Normal: $\mu = 1.15$ $\sigma = 0.11$	(Coderre et al. 2011; Yon et al. 2011; Snelling et al. 2004; Dobbins et al. 1994)	+46/ -39%	+41/- 28%	0%
Soot refractive index light scattering function at scattering wavelength $[E(m_{\lambda_s})]$	Normal $\mu = 0.31$ $\sigma = 0.04$	(Yon et al. 2011)	0%	+24/- 17%	0%
LII calibration constant at lower wavelength, η_1	Normal: $\mu =$	Calculated	± 4.8 %	+3.9/ -3.6%	0%

	$2.34 \times 10^{-11} \frac{m^3}{A}$, $\sigma =$ $0.045 \times 10^{-11} \frac{m^3}{A}$				
LII calibration constant at upper wavelength, η_2	Normal: $\mu =$ $7.67 \times 10^{-12} \frac{m^3}{A}$, $\sigma =$ $0.086 \times 10^{-12} \frac{m^3}{A}$	Calculated	+5.3/ -5.1%	+4.1/ -3.9%	0%
Equivalent laser sheet thickness, w_b	Normal: $\mu = 1.499$ mm $\sigma = 0.005$ mm	Numerical simulations (Crosland et al. 2011)	+6.9/ -6.2%	±5%	0%
Fractal dimension, D_f	Normal: $\mu = 1.7$ $\sigma = 0.15$	(Köylü et al. 1995b)	0%	+37/ -38%	+4.1/ -3.4%
Fractal prefactor, k_f	Normal: $\mu = 1.7$, $\sigma = 0.4$	(Sorensen 2001)	0%	+53/ -28%	0%
Width of log-normal distribution of aggregates, σ_g	Uniform: Min = 1.7 Max = 2.5	(Link et al. 2011; Sorensen et al. 1995)	0%	+13/- 12%	+27/ -23%
Experimental measurement					
Forward detection angle, θ_1	Uniform: Min = -29° Max = -31°	Measured	0%	±0.3 %	±0.4 %
Backward detection angle, θ_2	Uniform: Min = 149° Max = 151°	Measured	±2.9 %	±2.5 %	±0.3 %
Forward scat. calibration constant, η_3	Normal: $\mu = 58.10$ V m $\sigma = 0.49$ V m	Measured	0%	±1.7 %	±2.4 %
Backward scat. calibration constant, η_4	Normal: $\mu = 99.87$ V m $\sigma = 0.86$ V m	Measured	0%	+3.2/ -3%	+2.4/ -2.5%
Input voltage for LII at λ_1	Normal: $\mu = 3.41$ V $\sigma = 0.081$ V	Measured	+6.5/ -5.8%	+4.6/ -4.8%	0%
Input Voltage for LII at λ_2	Normal: $\mu = 0.645$ V $\sigma = 0.017$ V	Measured	+12/ -12%	+9.8/ -8.8%	0%
Forward scatter	Normal: $\mu = 2.33$ V $\sigma = 0.018$ V	Measured	0%	±1.5 %	±2.2 %
Backward scatter	Normal: $\mu = 2.64$ V $\sigma = 0.014$ V	Measured	0%	±1.9 %	±1.5 %
Various calibrated neutral density filters	Normal: $\sigma = 2\%$ of nominal value	Based on spec. of calibration device	±5%	±2%	±0%
Modeling errors					
Equivalent filter approx.	Normal: $\mu = 1$ $\sigma = 0.005$, on T_{pe}	Calculated for $T_{pe} = 3500$ K	+10/ -9%	±3%	0%
LII signal trapping	Normal: $\mu = 1$; $\sigma = 0.025$, on f_v	(Liu et al. 2009b)	±5%	±2%	0%

Assumed monodisperse d_p	Normal: $\mu = 1$; $\sigma = 0.1$, on d_p	Calculated based on Tian et al. (2004)	0%	$\pm 20\%$	0%
ELS signal trapping	Normal: $\mu = 1$; $\sigma = 0.033$, on K_{vv}	Calculated from f_v (Snelling et al. 2005)	0%	$\pm 1.3\%$	$\pm 3\%$
ELS intra-aggregate scattering	Normal: $\mu = 1$; $\sigma = 0.05$, on K_{vv}	Based on Sorensen (2001)	0%	$\pm 1\%$	0%
Choice of structure factor	Normal: $\mu = 1$; $\sigma = 0.03$, on R_{gml}	Calculated	0%	$+4/-3\%$	$\pm 6\%$

The convergence criterion for the minimum number of iterations required to obtain a stable output distribution was taken from Thompson et al. (1992) in which the MC simulation was considered converged once the 95th percentile values of all three variables (f_v , d_p , and R_{gml}) were estimated to be within 1 % of their stable value. The stable value was approximated by performing a single MC simulation with a very large number of iterations, in this case 100,000. This convergence test was performed for the measurements taken from the flare test TWG. For each MC simulation, 20,000 draws were deemed sufficient as a good compromise between computation time and sufficient draws to get total uncertainty values with low variance. Figure 4.10 shows both the coefficient of variance of the mean or the relative error percentage and the upper CI (95%) of R_{gml} converging at ~20,000 iterations for the MC convergence analysis done on the minimum and maximum voltage measured by the detectors during the flare test TWG. Similar trends were observed for f_v , and d_p , hence, 20,000 iterations were used for MC simulation for the uncertainty analysis of lab scale flare flame measurements during both TWG and TWOG.

Figure 4.10: Convergence criteria for number of Monte Carlo simulation draws. (a) Plot of the relative error on the upper CI (95%) of R_{gm1} vs number of iterations on Monte Carlo draws. (b) Coefficient of variance of the mean R_{gm1} vs number of Monte Carlo simulation draws for both minimum measured voltage and maximum measured voltage by the detectors during flare test TWG

As can be seen from Table 4.2, the soot optical properties were the biggest contributing factors to the overall uncertainty on f_v , d_p , and R_{gm1} , while the uncertainties due to instrument noise and calibrations were comparatively low. For the LII measurement of SVF f_v , the two biggest contributing factors to the overall uncertainty were the soot refractive index light absorption function at lower LII wavelength, $E(m_{\lambda_1})$, with a contribution of +31/-22%, and the ratio of soot refractive index light absorption function at lower and upper LII wavelengths, $E(m_{\lambda_1})/E(m_{\lambda_2})$, with +46/-39% contribution. Figure 4.11 shows the results of two MC simulations to isolate the effects of $E(m)_\lambda$ and its ratio on the uncertainty of f_v . The strong dependence illustrates why these two variables were responsible for up to 69% of total uncertainty in f_v .

Figure 4.11: Plots showing varying SVF (f_v) with varying (a) soot refractive index light absorption function at lower LII wavelength $E(m_{\lambda_1})$, and (b) the ratio of soot refractive index light absorption function at lower and upper LII wavelengths $E(m_{\lambda_1})/E(m_{\lambda_2})$, with other measured and assumed values held constant

The largest contributing factor to the overall uncertainty on mean aggregate radius of gyration R_{gml} was the width of probability distribution used to describe the population of aggregates, σ_g , with a contribution of +27/-23 %. The largest contributor to the uncertainty of primary particle diameter d_p , was the fractal prefactor k_f (+53/-28 %). The next two largest contributors were the fractal dimension D_f (+37/-38 %), and the uncertainties propagated from the SVF (+55/-37 %). Figure 4.12 shows the relationship between primary particle diameter and (a) fractal dimension, D_f and (b) the fractal prefactor, k_f , each obtained from MC analyses where other measured and assumed values were held constant. Figure 4.12 (b) shows that for the given range of fractal prefactor k_f , the primary particle diameter could vary from 10 nm to 90 nm.

Figure 4.12: Distribution of the primary particle diameter with (a) the fractal dimension, D_f , and (b) the fractal prefactor k_f , with other measured and assumed values held constant

If it is assumed that the soot optical properties remain unchanged at a particular measurement location in the flame, then relevant uncertainties when making comparisons are due solely to measurement noise. For this, a MC simulation was performed on the first test dataset pre-TWG, while keeping the soot optical properties and other soot property variables constant. The results for these two cases are summarized in Table 4.3. Though the uncertainties are higher than those reported by Crosland et al. (2013b), this is expected since the smaller solid angle of detection used in the current apparatus resulted in fewer photons being collected, which in turn increased the photon shot noise (Crosland et al. 2013b).

Table 4.3 Comparison of total uncertainty and measurement noise uncertainty only shown by mean values with upper and lower bound of the 95% CI

Measured Property	Total Uncertainty	Fixed soot properties
Soot Volume Fraction f_v	5.0 {8.2 ppm 63%} 2.8 ppm 45%	4.9 {5.6 ppm 16%} 4.1 ppm 15%
Primary Particle Diameter d_p	29.9 {57.2 nm 91%} 12.7 nm 58%	30.0 {34.9 nm 17%} 25.7 nm 14%
Mean Aggregate Radius of Gyration R_{gm1}	62.4 {78.8 nm 24%} 46.4 nm 28%	63.1 {65.9 nm 4%} 60.1 nm 5%

4.6 Filters Used for Soot Trends

Soot is highly intermittent within these buoyant non-premixed flames, being absent at most locations more often than it is present. When negligible amounts of soot are measured, the non-linear nature of the equations combined with the very weak LII and scattering signals can produce extreme, non-physical results for f_v , d_p , and R_{gml} . These non-physical results can sometimes cause algorithms to converge very slowly, or not to converge at all. It is thus crucial to remove non-sooting data points before performing any calculations. The criteria discussed in the next two subsections were used to filter out points where soot was reasonably believed not to be present:

4.6.1 2σ of the background

At each new location in the flame, background signals were acquired without any laser excitation. A filtering threshold was then set at two times the standard deviation of the flame background to remove points with only negligible laser-excited signals. This filter was implemented in the LabView VI used to gather instantaneous measurements during the tests.

4.6.2 R_{vv} between 1 and 6

As discussed in Chapter 2, R_{vv} is the dissymmetry ratio between the forward- and backward-scattering signals. The uncertainty in R_{gml} starts to increase asymptotically at values of R_{vv} above about 6 because R_{gml} (as propagated from R_{vv}) begins to increase asymptotically (Crosland et al. 2013b). Thus, this optical measurement setup is only useful for R_{vv} values between 1 and ~ 6 to avoid non-physical R_{gml} measurements due to the associated high uncertainty in modelling. This filter was implemented for every soot point measured at any given height as a pre-processing step before running the MC uncertainty analysis. The MATLAB code that is used for MC analysis is attached in the Appendix A.

While performing the $N = 20,000$ MC iterations associated with each measured data point, the MC MATLAB code skipped any iterations where the randomly generated N_m was less than 1 (physically impossible) or greater than 200, which approximately corresponds to a R_{vv} of 5-6 depending on the random values of the other parameters.

Figure 4.13 depicts the number of successful (i.e. not skipped) solves for each of the 650 and 872 soot data points measured at 50 cm above the burner for the two tests. As R_{vv} approached 1, the number of successful MC solves fell rapidly from $N = 20,000$ (all solves successful) to below 10,000. Even though a particular set of $N = 20,000$ MC iterations was based on an input $R_{vv} > 1$, some individual MC iterations randomly drew values that produced a value of R_{vv} less than 1 and thus were non-physical and unsolvable resulting in lesser number of successful MC solves. There were still some unsolved cases at mid-range R_{vv} points mostly due to the occasional random draws that fell many standard deviations away from nominal values. A similar decrease in the number of successful solves occurred as R_{vv} approaches 5. Only the data gathered from these valid points were retained and used in the following Chapter 5.

Figure 4.13: The number of data points that were solved within the $N_m = 1$ to 200 range for any given soot point measured at the height of 50 cm above the burner (a) for TWG, and (b) for TWOG in the pipe burner

Chapter 5 Results

5.1 Turbulent Buoyant Non-Premixed Flame Results

This chapter presents soot measurement results for the flare type flame described in Chapter 3 with and without its turbulence-inducing grid installed. The chosen flow rate of 156 SLPM (standard liters per minute at 0°C and 101.325 kPa) was intended to represent a nominal flaring condition in the UOG industry with a flame in the turbulent buoyant (turbulent shear regime) regime, shown earlier in Figure 1.2 (McEwen 2010; Delichatsios 1993). From the global flare gas mixtures discussed in Conrad and Johnson (2019), the “Alberta Heavy 9” fuel mixture was chosen as representative of a more strongly sooting mixture for upstream flares in Alberta. The flow parameters for this flame are shown in Table 5.1. The internal stack flow was selected to be near the boundary between the turbulent buoyant (transition shear) and turbulent buoyant (transition buoyant) flame regimes as defined by Delichatsios (1993) and discussed in Chapter 1. At standard conditions (0°C), the pipe flow parameters produce a Reynolds number of > 6000 , but heating of the flare stack by the flame preheats the fuel as it approaches the burner exit. Although accurate measurement of this exit fuel gas temperature is difficult without disturbing the flow or incurring measurement errors due to thermal radiation from the flame, thermocouple estimates suggest the actual gas temperature at the flare exit could be 60°C or greater. At this temperature, the Reynolds number decreases to 5430 and the flow appears to relaminarize at the burner exit in the absence of a turbulence generating grid. An attempt was made to quantify turbulence intensities and scale with the flare operating using a Dantec Dynamics Flow Explorer LDA system, but the system was determined to be giving spurious results and needed to be sent back to the manufacturer for repair. Another attempt will be made to make these measurements in the flame after the system is repaired.

Table 5.1 Fuel flow conditions, with estimated theoretical flame length based on theory presented in Delichatsios (1993)

Diameter of the burner tip [mm]	Fuel flow rate [LPM]	Burner exit velocity [m/s]	Reynolds number (Re)	Theoretical flame length [m]	Global Froude Number (Fr_g)
50.8	156 (at 0°C) 190 (at 60°C)	1.28 (at 0°C) 1.45 (at 60°C)	6520 (at 0°C) 5430 (at 60°C)	1.58 (at 0°C) 1.68 (at 60°C)	0.025 (at 0°C) 0.028 (at 60°C)

Figure 5.1 depicts the variation of the time-averaged f_v along the burner centreline as a function of height above the burner. This average includes zero-value f_v readings and thus represents the time-averaged volume fraction at the measurement location. As discussed in Section 4.5, soot was assumed to be absent ($f_v = 0$) in all instances where the measured PMT voltages were within 2σ of the measured background without laser excitation. Eighteen thousand measurements were made at each height.

Figure 5.1 shows the mean SVF increasing axially towards the middle of the flame and then decreasing slightly towards the flame tip for both the test with the turbulence grid (TWG) and the test without the turbulence grid installed in the burner (TWOG). The time-averaged f_v is consistently lower in the TWG case than the TWOG case along the centreline of the flame. The overall trend with higher time-averaged mean f_v for the TWOG case in the axial profile (from 35 cm to 105 cm) suggests that, as discussed in Yoo and Im (2007), the effect of dissipation of soot pockets due to enhanced mixing in the TWG case dominates over the increase in SVF due to the increase in flame volume. In both cases the SVF reaches a maximum at around 90 cm above the burner and starts decreasing near the tip of the flame. This could be caused by one or more mechanisms, including i) partial oxidation of the soot within sooty regions of the flame, ii) complete oxidation of sooty regions of the flame, causing a reduction in the number of sooty regions, iii) mixing with surrounding air decreasing the frequency with which sooty regions are encountered. Lee et al. (2009) proposed that a similar trend was due to soot oxidation by air entrainment and dissipation of soot pockets. It should be noted that the time-averaged f_v does not go to zero towards the tip of the flame, indicating that there were emissions from the flame, i.e. the soot was not fully destroyed or oxidized before leaving the flame. One notable outlier is the time-averaged f_v measurement at 124 cm in the TWOG case. This outlier is a non-physical

measurement and is thought to have occurred due to instrument error as further discussed in Appendix B.

Figure 5.1: Axial profile of the mean SVF of all 18,000 datapoints along the centreline of the flame centreline with (red) and without (green) a turbulence grid installed in the flare stack. Error bars indicate the standard error of the mean. A visible flame length of 1.85 m was observed for both the TWG and TWOG cases.

The influence of turbulence on SVF can be further explored by considering only those instances where some soot is detected. This can be described in terms of intermittency. Intermittency is expressed as $1-\Omega$, where Ω is the intermittency index, defined as the ratio of the number of measurements where soot is present N_s , i.e. instances when scatter and laser-induced incandescence signals were observed, to the total number of measurements N ($\Omega = N_s/N$) (Crosland et al. 2015). Figure 5.2 plots the axial profile of the mean intermittency of the flames with (TWG) and without the turbulence grid (TWOG) installed. A mean value for the intermittency was calculated by subdividing the $N = 18,000$ temporally ordered data points into 10 temporally ordered sets of 1,800 points, determining the intermittency of each subset, and then calculating the 95 % CI of the mean of the 10 subsets. Figure 5.2 also shows the intermittency calculated by subdividing the $N = 18,000$ temporally ordered data points into 8 and 12 temporally ordered sets of 2,250 and 1,500 points respectively, which demonstrates that the intermittency trends are independent of chosen size of the subgroups.

Axial profiles of the mean intermittency (shown in Figure 5.2) exhibit a similar trend but are mirrored relative to the time-averaged f_v in Figure 5.1. The TWOG case has soot present more often (lower intermittency) than the TWG case. The soot is most intermittent near the exit of the stack and the least intermittent at around 90 cm above the burner. Similar to the trends observed above, after 90 cm the air entrainment results in soot oxidation in the flame and soot is observed less often (Lee et al. 2009). Crosland et al. (2015) also observed that the reduction in f_v towards the tip of the flame is due to an increase in intermittency.

Figure 5.2: Plot showing mean intermittency at the centreline of the flame with increasing height above the burner for both test conditions. Mean intermittency values and 95% CI bars shown were calculated by dividing the dataset into 10 subsets and analyzing the subsets. Mean intermittency is also shown for different subset sizes. A visible flame length of 1.85 m was observed for both the TWG and TWOG cases.

Since the trends for the time-averaged f_v and the intermittency are similar, it is important to plot the SVF without including the effects of intermittency to determine if the differences in the time-averaged f_v curves for the TWG and TWOOG cases are due to increased intermittency. Figure 5.3 shows the average axial profile of f_v^s , where superscript s in the f_v^s indicates the calculation only includes those measurement points where some soot was detected. Note that the axial profile of f_v^s differs from the trends shown above in Figure 5.1 by a factor equal to the mean intermittency index (Ω) at each point. This axial profile of f_v^s is produced by taking the mean of the medians produced by the MC analysis for each non-zero measurement (soot) point at each height above the burner. The smaller error bars (solid lines) represent the 95% CI of the measured values, i.e. uncertainties due only to instrument noise and shot-to-shot variation in SVF. The larger dashed error bars in Figure 5.3 represent the 95% CI of the total uncertainty calculated for each mean value, i.e. including all elemental sources of error as well as dominant contributions from uncertainty in soot properties. In the context of the total uncertainties, the trends in Figure 5.3 are not significantly different between the TWG and TWOOG cases. Between 37 cm and 92 cm above the burner, the f_v^s is ~21 % to 33 % lower with the grid than without and beyond the relative uncertainties; however, the difference is still well within the total uncertainties of the measurement. These trends suggest that, at least for the present test conditions, induced inflow turbulence has little effect on f_v^s for these buoyant non-premixed turbulent flames. This observation is consistent with the findings of Mahmoud et al. (2018) for turbulent non-premixed jet flame soot trends with different exit Reynolds number.

Figure 5.3: Axial profile of the mean f_v^s (i.e. mean f_v of measurement points where soot was detected) along the centreline of the flame with (red) and without (green) the turbulence grid installed in the flare stack. Error bars indicate relative uncertainties from measurement noise (solid) and total uncertainties (dashed) in mean f_v^s . A visible flame length of 1.85 m was observed for both the TWG and TWOG cases.

5.2 Influence of Internal Stack Turbulence on Soot Morphology

Figure 5.4 shows axial profiles of average (a) R_{gml} and (b) d_p . The averages and CI are calculated in the same way as for f_v^s , i.e. using the population of medians produced by the MC analysis for each non-zero-soot measurement to calculate a mean and associated CIs. The profiles show no obvious effect of the induced in-flow turbulence on either d_p or R_{gml} . Figure 5.4 thus suggests that the structure of the soot, when present, is not affected by the presently induced change in in-flow turbulence. Figure 5.3 further suggests that when soot is detected, the volume fraction is similarly unchanged. Instead, the increased intermittency in Figure 5.2 is likely the result of either i) enhanced dilution of the flame region due to mixing with ambient air, decreasing the probability of sooting regions passing through the measurement volume, or ii) rapid and complete oxidation of sooting regions of the flame at a time-scale that is fast relative to the acquisition rate of the measurement system.

The R_{gml} trends are almost identical for the TWG and the TWOG cases. The R_{gml} starts from 75 nm at the height of 25 cm where soot is first detected low in the flame. The

average aggregate size increases with height above the burner (which is correlated with residence time), as nascent soot particles collide and form aggregates throughout the flame, similar to that observed by Crosland et al. (2015) and Hu et al. (2003).

The mean axial d_p (Figure 5.4(b)) starts high near the exit of the burner and decreases with height in the flame for both the TWG and TWOG cases. Decreasing d_p near the tip of the flame is in line with the idea that soot is produced in the annular region low in the flame before significant radial mixing has occurred, is carried upward with buoyancy-driven flame streaks or eddies, and then oxidized higher in the flame (Yang and Köylü 2005a). These axial profiles show trends similar to those made in reference to momentum-dominated turbulent non-premixed flames by Mahmoud et al. (2018), Henriksen et al. (2009), Qamar et al. (2009), and Hu et al. (2003). Histograms of all f_v , d_p , and R_{gml} at various heights are attached in Appendix C.

Figure 5.4: Axial profile of the mean (a) R_{gm1} and (b) d_p of detected soot along the centreline of the flame with (red) and without (green) the turbulence grid installed in the flare stack. Error bars indicate relative uncertainties from measurement noise (solid) and total uncertainties (dashed). A visible flame length of 1.85 m was observed for both the TWG and TWOG cases.

While the present results show that in-flow turbulence has the potential to alter the soot formation patterns, they also suggest that the effects are likely to be small relative to other factors – most notably fuel chemistry (Conrad and Johnson 2017; Corbin 2014). Further study of the influence of in-flow turbulence is recommended to better understand and model in-flame processes; however, the influence of this turbulence on the overall emission trends of lab- and full-scale flares seems likely to be of secondary importance.

Chapter 6 Conclusion

A simultaneous auto-compensating LII and two-angle ELS diagnostic technique was used to measure f_v , d_p and R_{gml} in a turbulent buoyant diffusion flame. The system was first characterized on a well-established laminar flame, prior to being applied to a large turbulent buoyant diffusion flame (flare) in the Carleton University flare facility. Measurement uncertainties were rigorously quantified using a Monte Carlo method. Relative uncertainties (considering only measurement noise) of +16/–15 % in f_v , +17/–14 % in d_p , and +4/–5 % in R_{gml} were achieved, which are very low considering the instantaneous nature of the measurement and the high spatial resolution ($\sim 0.25 \text{ mm}^3$) of the system. Total uncertainties including dominant contributions from uncertainty in soot optical and morphological properties were +63/–45 % in f_v , +90/–58 % in d_p , and +22/–19 % in R_{gml} . The soot characteristics that dominate the uncertainties are sufficiently difficult to measure that these high uncertainties are not considered to be a drawback specific to this measurement technique.

The f_v , d_p , and R_{gml} measurements were taken at multiple heights above the burner on a turbulent buoyant diffusion flame for two conditions: with and without a turbulence grid installed in the burner exit stack. This allowed investigation of the effect of induced turbulence on soot production and oxidation in these flare-type flames. To the author's knowledge, no previous study has been done linking effect of in-flow/pipe turbulence to soot formation in a flare-type flame. The time-averaged f_v along the flame centreline was lower for the case with pipe turbulence (TWG) than the case without pipe turbulence (TWOOG) by anywhere from 1.5 % to 43 % along the height of the flame. The soot intermittency showed similar but mirrored trends for these same conditions, with higher intermittency in the TWOOG case than in the TWG case, depicting higher soot dissipation due to enhanced mixing in the flame. When only non-zero soot measurement points were considered to calculate a mean SVF (f_v^S) without the intermittency affecting it, the trends were not significantly different between TWG and TWOOG cases, consistent with recent experiments by Mahmoud et al. (2018) at different exit Reynolds numbers. When soot was

present, measured data for d_p , and R_{gml} showed that there was very little effect of induced in-flow turbulence especially in the context of the very strong effects of fuel chemistry in influencing soot formation and optical properties in these types of flames (Conrad and Johnson 2017, 2019; McEwen and Johnson 2012). Nevertheless, this simultaneous AC-LII and two-angle ELS diagnostic setup has the capability of measuring soot properties and concentration with high spatial and temporal resolution and should be further used for different test cases with changing fuel composition, turbulence levels, and flow regimes (which could more strongly influence soot formation, (Delichatsios 1993)).

6.1 Future Improvements

Due to the intermittent nature of the flame it is vital that the trends be observed for different burner configurations and fuel compositions to get a quantitative relationship between induced flow turbulence and soot characteristics in the flame. This is limited in the current results by the limited fuel storage capacity at CLSF compared to the high rates of fuel flow required to run the experiment. Six methane gas bottles were used for a single measurement run, which included finding and plotting the centreline and taking 18,000 measurements at a rate of 15 Hz for each height location. This was both time-consuming and expensive. This could be overcome by increasing the methane storage capacity or connecting a pressurized natural gas supply.

The optical setup was designed for precise measurements in a small measurement volume, but the alignment and calibration process required utmost precision and was prone to misalignment and erroneous results due to any misalignment. The small sensor size of the photo detectors and multiple reflecting surfaces the incoming beam must pass through – be it the mirrors or the short pass filters in the setup – made it very difficult to achieve a usable alignment. Updating the setup to use optical fibers to gather the signals collected by the achromatic doublet lenses and then transmitting them straight to the short pass filters and detectors may reduce chances of partially hitting the lenses, mirrors, and sensor area of the detector.

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Appendix A MATLAB Code for MC simulation of measured data

This section presents the MATLAB code that was used for the uncertainty analysis using MC simulation of the raw recorded voltages for soot points from the flare tests for TWG and TWOOG cases. The filters applied to find these soot points are displayed here in the code with comments.

```
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
This section has all the constant values assigned here at the beginning
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
clc;
clear all;
clearvars;
rng('default');
dp = 30*10^-9; %30 nm primary particle diameter assumed
lam1 = 445.784*10^-9; %nm shorter wavelength in the blue region for PMT
lam2 = 783.5*10^-9; %nm larger wavelength in the red region for PMT
lam_scatt = 1064*10^-9; %nm scatter wavelength from 1064 nm laser beam
h = 6.6207004*10^-34; %m2kg/s
kb = physconst('Boltzman'); %m2kg/s2K
c = physconst('lightspeed'); %m/s
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
Here we read in the raw data files from the two folders. The files are named in the
order they are acquired e.g. the first run at 10 cm height above the burner will say
Run1HAB10cm.csv and so on. The flame radiation background at each height is also stored
in seeparate cells to be used later on.
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
HAB = [10 20 30 40 50 60 70 80 90 100 110 120];
x = length(HAB);
[y, z] = size(csvread('29th Oct\Run1HAB120cm.csv',23,0));
[y1, z1] = size(csvread('30th Oct\Run1HAB120cm.csv',23,0));
M = zeros(y,z,x);
M1 = zeros(y1,z1,x);
for j=1:length(HAB)
    filename1 = [cd,'\29th
Oct','\Run',num2str(j,'%g'),'HAB',num2str(HAB(j),'%g'),'cm','\'.csv'];
    M1(:, :, j) = csvread(filename1,23,0);
    BackgroundSD1(:, :, j) = csvread(filename1,20,0,[20,0,20,3]);
    filename = [cd,'\30th
Oct','\Run',num2str(j,'%g'),'HAB',num2str(HAB(j),'%g'),'cm','\'.csv'];
    M(:, :, j) = csvread(filename,23,0);
    BackgroundSD(:, :, j) = csvread(filename,20,0,[20,0,20,3]);
end
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
In this section we save the raw signal data for all the non-zero soot detection points
and distribute them into separate voltage files (cells) for each detector for all heights
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
Raw = []; Raw1 = []; soot = []; soot1 = [];
```

```

for j=1:x
    for k = [1 2 3 4]
        Raw = [Raw,M(:,k,j)];
        Raw1 = [Raw1,M1(:,k,j)];
    end
    for k = [5 6 8]
        soot = [soot,M(:,k,j)];
        soot1 = [soot1,M1(:,k,j)];
    end
    nonzind = find(soot(:,3*j-2));
    nonzind1 = find(soot1(:,3*j-2));
    nonzdata{:,j} = Raw(nonzind,4*j-3:4*j);
    nonzdata1{:,j} = Raw1(nonzind1,4*j-3:4*j);
end
for j = 1:x
    temp = cell2mat(nonzdata(:,j));
    temp1 = cell2mat(nonzdata1(:,j));
    vbs{:,j} = temp(:,1); vbs1{:,j} = temp1(:,1); vbpmt{:,j} = temp(:,2); vbpmt1{:,j} = temp1(:,2);
    vrpmt{:,j} = temp(:,3); vrpmt1{:,j} = temp1(:,3); vfs{:,j} = temp(:,4); vfs1{:,j} = temp1(:,4);
end
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
num are the number of iterations used for the MC analysis for each soot point and the eta
and cal are the calibration constants for both the PMTs and scatter detectors
respectively.
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
num = 20000;
EtaBPMT = normrnd(2.34*10^-11,0.0446217*10^-11, num, 1); %m3/A
EtaRPMT = normrnd(7.67*10^-12,0.086457*10^-12, num, 1); %m3/A
CalBS = normrnd(99.87,0.86, num, 1); %V m
CalFS = normrnd(58.10,0.49, num, 1); %V m
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
These are all the parameters (soot properties, and laser sheet width and angle) and their
respective distributions that are used for the MC simulation. Their names, sources, and
impact on the calculated soot properties are discussed in Table 4.2
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
theta1_TU = unifrnd(29*(pi/180),31*(pi/180),num,1);
theta2_TU = unifrnd(149*(pi/180),151*(pi/180),num,1);
sig_g_TU = unifrnd(1.7,2.5,num,1);
kf_TU = normrnd(1.7,0.4,num,1);
df_TU = normrnd(1.7,0.15,num,1);
ratio_TU = normrnd(1.15,0.11,num,1);
Em_TU = normrnd(0.348,0.045,num,1);
Em_scat_TU = normrnd(0.32,0.04,num,1);
Fm_scat_TU = normrnd(0.31,0.04,num,1);
wb_TU = normrnd(0.1499,0.005,num,1);
we_TU = (wb_TU./sin(theta2_TU)).*0.001; %mm
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
This is the first level for loop to loop through the measured values at each height for
both the test caases. For the case TWOG, the raw measured data is being stored with
suffix 1.
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%

```

```

for k = 1:12
    VBS = cell2mat(vbs(k)); VBSV1 = cell2mat(vbs1(k)); VBPM TV = cell2mat(vbpmt(k));
    VBPM TV1 = cell2mat(vbpmt1(k));
    VRPM TV = cell2mat(vrpmt(k)); VRPM TV1 = cell2mat(vrpmt1(k)); VFSV = cell2mat(vfs(k));
    VFSV1 = cell2mat(vfs1(k));
    VBS_SD = BackgroundSD(1,1,k); VBPM TV_SD = BackgroundSD(1,2,k);VRPM TV_SD =
    BackgroundSD(1,3,k);VFSV_SD = BackgroundSD(1,4,k);
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
This nested second for loop is to loop through the soot points at each of height above
the burner measurement location and introduces the noise of the flame background in the
signal by randomly distributing each detected voltage with background signal with laser
off i.e. the electronic noise of the detector and fluctuations due to flame radiation
only, as standard deviation.
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
    CI_TU = [];
    for m = 1:length(VBSV)
        VBS = normrnd(VBSV(m),VBS_SD,num,1); % Measured voltages at the backward side
scatter detector with the SD from the background
        VBPM TV = normrnd(VBPM TV(m),VBPM TV_SD,num,1); % Blue PMT signals
        VRPM TV = normrnd(VRPM TV(m),VRPM TV_SD,num,1); % Red PMT signals
        VFS = normrnd(VFSV(m),VFSV_SD,num,1); % Forward side scatter signals
        BS_Absolute = (VBS./CalBS.*1);
        FS_Absolute = (VFS./CalFS.*1);
        Rvv = (FS_Absolute./BS_Absolute.*1); % Ratio of the absolute forward side scatter
signal to the backward side
        Tp_TU = (-h*c*(lam2-
lam1))./(log((VBPM TV.*EtaBPMT.*lam1^6)./(VRPM TV.*EtaBPMT.*ratio_TU.*lam2^6)))*k*lam1*lam2);
% Temperature of the soot particelles
        SVF_TU = (VBPM TV.*(exp(h*c./(k*Tp_TU.*lam1))-
1))./(EtaBPMT.*Em_TU.*we_TU.*12*pi*(c^2)*h./(lam1^6));%Soot Volume Fraction
        soot_TU = SVF_TU.*10^6;%Adjusting from ppm to one digit numbers (easier to read)
        Soot_TU_CI = [mean(soot_TU), std(soot_TU), median(soot_TU), prctile(soot_TU,[2.5
97.5])];%finding the mean, standard deviation, median, lower CI value, and upper CI value
of SVF out of the 20,000 MC simulations while neglecting all the zero, Inf and NaN terms
        Nm = zeros(num, 1) + NaN;
        fn = zeros(num, 1) + NaN;
        SqRgBar = zeros(num, 1) + NaN;
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
This nested third parfor loop is to solve the integrals discussed in section 2.3 to get
the dissymmetry ratio  $R_{vv}$ .in order to get value for  $R_{gm1}$  by solving  $N_m$  analytically
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
        parfor index = 1:num
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
This if statement is the first filter in which we filter out all the soot points that are
out of detection range of our optical setup and would produce erroneous results. This is
discussed in Section 4.6
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
            if ((Rvv(index)>1) && (Rvv(index)<6))
                q1 = sin(theta1_TU(index)./2).*4.*pi./lam_scatt; % These are for radians
                q2 = sin(theta2_TU(index)./2).*4.*pi./lam_scatt;
                Rg = @(N) dp./2.*(N./kf_TU(index)).^(1./df_TU(index));
                S_theta1 = @(N) (1 + (8./(3.*df_TU(index))).*(q1.*Rg(N)).^2 +
2.5.*(q1.*Rg(N)).^4 - 1.52.*(q1.*Rg(N)).^6 + 1.02.*(q1.*Rg(N)).^8).^(-df_TU(index)./8);
                S_theta2 = @(N) (1 + (8./(3.*df_TU(index))).*(q2.*Rg(N)).^2 +

```

```

2.5.*(q2.*Rg(N)).^4 - 1.52.*(q2.*Rg(N)).^6 + 1.02.*(q2.*Rg(N)).^8).^(-df_TU(index)./8);
    p = @(N, Nm) exp(-
1/2.*(log(N./Nm)./log(sig_g_TU(index))).^2)./(sqrt(2.*pi).*N.*log(sig_g_TU(index)));
    % Create objective function and solve it
    obj = @(Nm) Rvv(index) - integral(@(N) N.^2.*S_theta1(N).*p(N, Nm), 0,
inf,'abstol', 0, 'reltol', 1e-3, 'ArrayValued', true) ...
    ./ integral(@(N) N.^2.*S_theta2(N).*p(N, Nm), 0, inf, 'abstol', 0,
'reltol', 1e-3,'ArrayValued', true);
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
This second if statement is filter out results and include only those that have a value
for  $N_m$  between  $0.1 < N_m < 200$ .
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
    if( obj(200)>0 || obj(0.1)<0 )
        continue
    end
    Nm_temp = fzero(obj,[0.1 200]); % checking big intervals
    % Get fn
    p1 = @(N) exp(-
1/2.*(log(N./Nm_temp)./log(sig_g_TU(index))).^2)./(sqrt(2.*pi).*N.*log(sig_g_TU(index)));
    fn(index) = integral(@(N)N.^2.*p1(N),0,inf, 'ArrayValued',
true)./(integral(@(N)N.*p1(N),0,inf, 'ArrayValued', true).^2);
    SqRgBar(index) = (integral(@(N) N.^2.*S_theta2(N).*p1(N), 0, inf,
'ArrayValued', true))./(integral(@(N) N.^2.*p1(N),0,inf, 'ArrayValued', true));
    Nm(index) = Nm_temp;
    end
end
good_points = sum(~isnan(Nm)); % getting rid of NAN values from the  $N_m$  output
Nbar = (Nm.*exp((log(sig_g_TU).^2)./2));
gyr_TU = (dp./2).*(Nbar./kf_TU).^(1./df_TU); % solving and getting mean aggregate
radius of gyration
Rgm1_TU = (gyr_TU.*10^9);
Rgm1_TU_CI = [nanmean(Rgm1_TU), nanstd(Rgm1_TU), nanmedian(Rgm1_TU),
prctile(Rgm1_TU,[2.5 97.5])]; %finding the mean, standard deviation, median, lower CI
value, and upper CI value of  $R_{gm1}$  out of the 20,000 MC simulations while neglecting all
the zero, Inf and NaN terms
Q_abs_TU = (4*pi*sin(theta2_TU./2))./lam_scatt;
K_absolute_TU = real((Em_scatt_TU.*SVF_TU.*6*pi)./lam_scatt);
Dp_TU =
((2.*BS_Absolute.*(lam_scatt^4))./((3*(pi.^3)*Fm_scatt_TU.*SqRgBar.*SVF_TU.*kf_TU.*((gyr_TU
.*2).^df_TU).*fn)).^(1./(3-(df_TU.*1)))); % solving and getting primary particle diameter
Particle_dia_TU = real(Dp_TU.*10^9);
Particle_dia_TU_CI = [nanmean(Particle_dia_TU), nanstd(Particle_dia_TU),
nanmedian(Particle_dia_TU), prctile(Particle_dia_TU,[2.5 97.5])]; %finding the mean,
standard deviation, median, lower CI value, and upper CI value of  $d_p$  out of the 20,000 MC
simulations while neglecting all the zero, Inf and NaN terms
CI_TU(m,:) = [Soot_TU_CI, Particle_dia_TU_CI, Rgm1_TU_CI good_points mean(Rvv)];
fprintf('Current completion percentage for HAB %d', k)
fprintf(' is %.3f%% \n',m/length(VBSV)*100)
end
filename = sprintf('%s_%d','CIatHeight',k);
col_header={'SVF mean','SVF std','SVF median', 'SVF lower CI','SVF upper CI','dp
mean','dp std','dp median', 'dp lower CI','dp upper CI','Rgm1 mean','Rgm1 std','Rgm1
median', 'Rgm1 lower CI','Rgm1 upper CI','Good points','Mean Rvv'}; %Row cell array
(for column labels)

```



```
xlswrite(filename,col_header,1,'A1'); writing the mean, standard deviation, median, lower  
CI value, and upper CI value of SVF,  $R_{gmL}$ , and  $d_p$  for each soot point  
xlswrite(filename,CI_TU,1,'A2');  
end
```

Appendix B Discussion of Anomaly at 124 cm HAB

One notable outlier from the trend shown in Figure 5.1 is the high f_v at a height of 124 cm above the burner, seen in both f_v^s and time-averaged f_v in Figure 5.3 and Figure 5.1. After close examination this appears to be due to an instrument error as the incandescence signal from the lower wavelength PMT suddenly increased and remained high until the end of the test, as can be seen in Figure B.1. This is believed to be due to an instrument issue with the gain controller on the SR250 Gated Integrator. The data at all other heights from both test cases were closely examined and no other such error was found.

Figure B.1: Signal / time graph of the blue/lower wavelength PMT for height (a) 113 cm above the burner, and (b) 124 cm above the burner, for the TWOG case

Appendix C Histograms depicting trends of soot measurements for the two tested cases

C.1 Histogram for SVF with TWG and TWOG cases, for different HAB

Figure C.1: Histogram of SVF for the two cases with relative frequency on the ordinate axis

C.2 Histogram of R_{gml} for the two cases

Figure C.2: Histogram of the R_{gml} measured for the two cases with relative frequency on the ordinate axis

C.3 Histogram of d_p measured for the two cases

Figure C.3: Histogram of d_p measured for the two cases with relative frequency on the ordinate axis

Appendix D Effect of initial gas temperature on laser sheet thickness and SVF

The effect of initial gas temperature assumption on the equivalent laser sheet width w_e is shown in Figure D.1. Since T_{pe} is calculated using the calculated w_e , the effect of assumed initial gas temperature only affects the equivalent laser sheet width. The actual laser sheet thickness (w_b) was measured at 0.15 mm and was found to vary by about $2\sigma = 0.025$ mm. Figure D.1 shows the results of running the LII simulation code (Crosland et al. 2013b; Liu et al. 2009a; Snelling et al. 2000, 2005) with a variety of initial gas temperatures to determine the effect on T_{pe} . The resultant error on T_{pe} is of 17 %.

Figure D.1: Plot showing effect of variation of assumed initial gas temperature on (a) Equivalent laser sheet width w_e , along with linear fit results and 95 % CI bars depicting lower and upper CI from spread of data due to other varying variables in the simulation (b) SVF which is just the inverse of the trend shown in (a). A linear fit has been used to calculate the R-squared value of the variation introduced.