

8<sup>th</sup> U. S. National Combustion Meeting  
Organized by the Western States Section of the Combustion Institute  
and hosted by the University of Utah  
May 19-22, 2013

## Joint Temperature-Volume Fraction Statistics of Soot in Turbulent Non-Premixed Jet Flames

*Christopher R. Shaddix and Jiayao Zhang*

*Combustion Research Facility  
Sandia National Laboratories  
Livermore, CA 94550 USA*

Soot emissions from internal combustion engines and aviation gas turbine engines face increasingly stringent regulation, but available experimental datasets for sooting turbulent combustion model development and validation are largely lacking, in part due to the difficulty of making quantitative space- and time-resolved measurements in this type of flame. To address this deficiency, we have performed a number of different laser and optical diagnostic measurements in sooting, non-premixed jet flames fueled by ethylene or a prevaporized JP-8 surrogate. Most laser diagnostic techniques inherently lose their quantitative rigor when significant laser beam and signal attenuation occur in sooting flames. However, the '3-line' approach to simultaneous measurement of soot concentration (on the basis of laser extinction) and soot temperature (on the basis of 2-color pyrometry) actually relies on the presence of significant laser attenuation to yield accurate measurements. In addition, the 3-line approach yields complete time-resolved information. In the work reported here, we have implemented the 3-line diagnostic in well-controlled non-premixed ethylene and JP-8 jet flames with a fuel exit Reynolds number of 20,000 using tapered, uncooled alumina refractory probes with a 10 mm probe end separation. Bandpass filters with center wavelengths of 850 nm and 1000 nm were used for the pyrometry measurement, with calibration provided by a high-temperature blackbody source. Extinction of a 635 nm red diode laser beam was used to determine soot volume fraction. Data were collected along the flame centerline at many different heights and radial traverses were performed at selected heights. A data sampling rate of 5 kHz was used to resolve the turbulent motion of the soot. The results for the ethylene flame show a mean soot volume fraction of 0.4 ppm at mid-height of the flame, with a mean temperature of 1450 K. At any given instant, the soot volume fraction typically falls between 0.2 and 0.6 ppm with a temperature between 1300 and 1650 K. At greater heights in the flame, the soot intermittency increases and its mean concentration decreases while its mean temperature increases. In the JP-8 surrogate flame, the soot concentration reaches a mean value of 1.3 ppm at mid-height of the flame, but the mean soot temperature is only 1270 K. Elevated soot concentrations persist for a range of heights in the JP-8 flame, with a rise in mean temperature to 1360 K, before both soot volume fraction and temperature tail off at the top of this smoking flame.

### 1. Introduction

The health effects of fine particulate matter (PM) in ambient air have become increasingly evident over the past decade. These particles are able to deeply penetrate lung tissue and have been shown to have a number of deleterious effects associated with the pulmonary and cardiovascular systems, leading to increased morbidity and mortality [1-6]. Because of this association between fine PM in the atmosphere and deleterious health, soot emissions from internal combustion engines and aviation gas turbine engines have faced increasingly stringent regulation. In addition, national and regional regulatory agencies have been decreasing the maximum fine PM permitted in the atmosphere according to ambient air quality standards and in this regard have shown concern over aircraft emissions associated with airports and military bases. Furthermore, in-flight emission of fine particulates from gas turbine engines has been shown to have effects on contrail/cloud formation and climate forcing [7,8].

In light of these concerns, there is a strong desire among aircraft engine designers to have a truly predictive modeling capability for soot formation and emission, considering the influence of changes in the fuel composition, ambient conditions, and engine design and operation. At this moment, such a capability is still lacking. One reason for the lack of predictive models of soot formation in the complex gas turbine combustion environment is the lack of robust, spatially

and temporally resolved data in turbulent reacting flow fields, specifically for sooting fuels. Such datasets have been in development for many years for soot-free flames [9] and more recently for a slightly sooting methane flame [10], but available experimental datasets for moderate sooting turbulent flames are largely lacking. Such datasets are needed because under moderately sooting flame conditions, believed to be representative of aviation gas turbine combustors, the soot that is formed radiates energy away from the hottest soot-containing regions to the walls and to the cooler soot-containing regions, thus redistributing the reaction enthalpy and influencing the flame chemistry and burning rate in a coupled manner [11]. The ability of computational models to accurately describe these coupled soot formation/radiation/flame chemistry interactions cannot be tested with data from soot-free or lightly sooting flames.

Unfortunately, many of the laser diagnostic techniques that have been used to provide quantitative space- and time-resolved measurements of flow and scalars in soot-free flames fail or are much more difficult to perform quantitatively in moderately sooty flames. This is largely because these techniques rely on the use of high power, pulsed laser beams, which excite both broadband fluorescence from polycyclic aromatic hydrocarbons (PAH), whose presence is associated with soot formation, and laser-induced incandescence (LII) of the soot itself, which produces a strong background signal throughout visible and near-IR wavelengths. Even if such strong background signals can be managed, for application of a particular laser diagnostic, the presence of significant quantities of soot in the flame results in laser beam attenuation as well as laser diagnostic signal attenuation that varies in intensity in both space and time, making time-resolved quantitative measurements of scalar properties nearly impossible.

In this paper we describe the application of a laser diagnostic technique that actually requires a significant amount of soot loading to generate sufficient signal-to-noise and which simultaneously measures the soot concentration and soot temperature, with high temporal resolution. Thus, joint soot temperature-volume fraction statistics are measured, which is a critical aspect of determining the radiant flux in turbulent, sooty flames [11,12]. This ‘3-line’ diagnostic approach applies laser beam attenuation and 2-color pyrometry, two line-of-sight measurements. To define and to limit the length of the optical probe volume, physical probes are used, which must be designed carefully to avoid unduly perturbing the sampled flame. The measurements reported here are for nonpremixed turbulent jet flames fueled by ethylene and a JP-8 surrogate, and complement other measurements that have been reported elsewhere, such as soot LII and OH planar laser-induced fluorescence (PLIF) [13].

## 2. Methods

Figure 1 shows a schematic of the 3-line diagnostic that was implemented. Laser attenuation was performed with a 632.8 nm HeNe laser, so that soot dimensionless extinction coefficients previously measured at this wavelength could be utilized to interpret the measurement. A reference laser intensity measurement was made using a beamsplitter and a photodiode detector with a 632.8 nm laser line filter (3 nm FWHM). After passing through the flame probe volume, the laser beam was separated from the 2-color soot emission signals with the use of a dichroic beamsplitter. The transmitted beam was collected in a 12-inch diameter integrating sphere, before passing through a laser line filter onto a photodiode detector, in order to remove any influence from turbulent flame beam steering on the attenuation measurement.

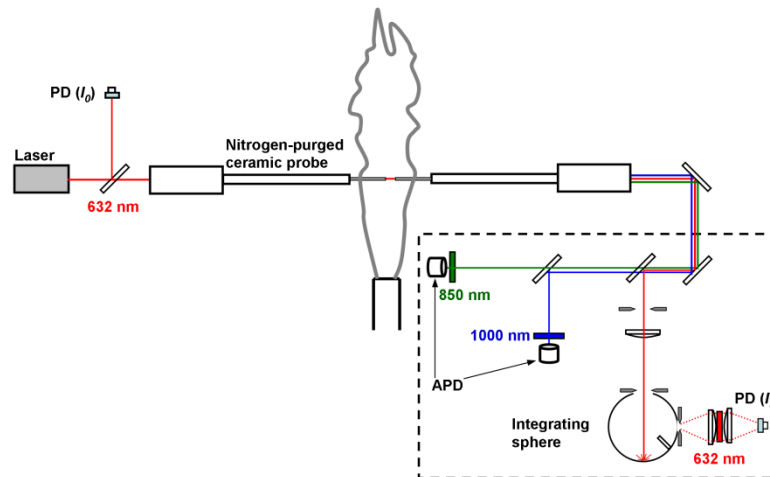


Figure 1. Schematic of diagnostic configuration used to perform 3-line measurements of soot temperature/concentration statistics in the turbulent jet flames.

The soot emission signals were split with a cube beamsplitter and then passed through bandpass filters with center wavelengths of 850 nm and 1000 nm before passing onto thermoelectrically cooled avalanche photodiode (APD) detectors. Calibration of the two-color pyrometry diagnostic was performed using a high-temperature blackbody source and a mirror that redirected the blackbody light towards the avalanche photodiode detectors.

A key aspect of the 3-line diagnostic technique is the need to insert a two-ended probe into the flame to limit the length of the optical interrogation region. In previous studies, these probes have typically been constructed of water-cooled steel or aluminum tubing, in some cases with insulation wrapped around the outside of the probes. With this design approach, the probe tubes are necessarily quite large and also provide a thick thermal quench layer. To minimize probe perturbation of the flow field and flame sheets, we adopted the approach first used by Sivathanu and Faeth [14], with tapered refractory probe ends that are uncooled. Figure 2 shows photographs of the cooled housings containing the outer probe ends and the uncooled probe tips that enter the flame. The alumina probes were found to transmit radiant emission from the broader flame through the walls to the pyrometry detectors at measurable levels. Therefore, the outer surfaces of the probes were painted black with high-temperature paint, which corrected this problem. Furthermore, a small amount of radiation was transmitted to the detectors from the hot tips of the probes when they were located in the hottest regions of the flames, leading to a non-zero radiant background signal. This background signal was subtracted before processing the datasets to determine the soot radiant temperature.

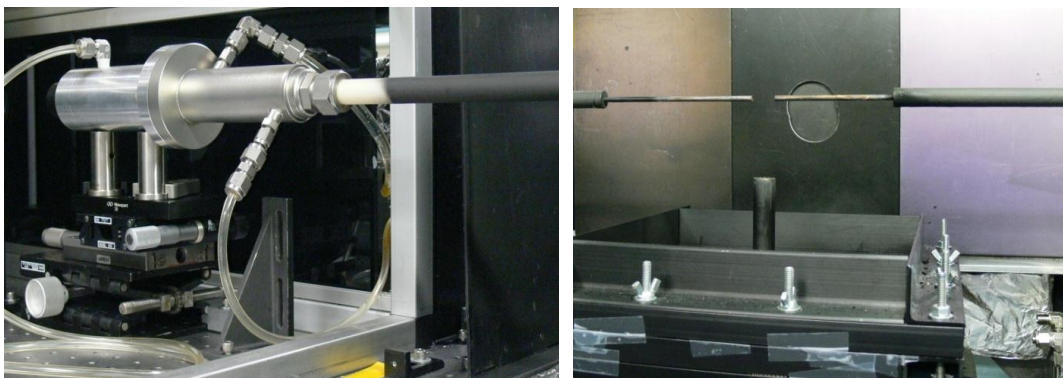


Figure 2. Optical probe for performing 3-line measurements of soot temperature/ concentration statistics in the turbulent jet flame. Aluminum optical housing (left) is water-cooled and provides  $N_2$  purge gas. Refractory probe ends (right) are uncooled.

Data were collected along the flame centerline at many different heights and radial traverses were performed at selected heights. A data sampling rate of 5 kHz was used to resolve the turbulent motion of the soot. Digital time records were collected as 40 sets of 5000 data points (i.e. a 1 sec time record) at a given location in the flame before a computer-controlled X-Y-Z translation stage moved the flame to the next programmed sampling position. A 10 mm probe end separation was used for most of the measurements, but some data were also collected for probe separations of 5 mm and 20 mm. The soot extinction measurement was converted to soot volume fraction according to the Beer-Lambert law with a dimensionless extinction coefficient,  $K_e$ , of 9.3, as determined in laminar flames by Williams et al. [15]. The optical path length of soot extinction was assumed to be equal to 2 mm less than the probe tip separation, to account for the effect of the small purge flow of nitrogen out the tips of the probe.

Measurements were performed in two canonical piloted turbulent nonpremixed jet flames that have been the focus of a number of measurements at Sandia: an ethylene jet flame and a prevaporized JP-8 surrogate jet flame, both with a jet-exit Reynolds number of 20,000 [13,16]. The burner design and experimental operation have been specifically tailored to allow meaningful comparisons with CFD models [16]. The ethylene flame is stabilized on a fuel tube with an ID of 3.2 mm, whereas the prevaporized JP-8 surrogate, composed of m-xylene and n-dodecane (23:77 by liquid volume), is stabilized on a fuel tube with an ID of 2.5 mm. Further details concerning the burner design and operation are available in ref. 16. The ethylene flame does not show any apparent emission of soot in the exhaust (i.e., it is non-smoking), whereas the JP-8 surrogate flame is a lightly smoking flame. Photographs of the investigated flames are shown in Fig. 3.

### 3. Results and Discussion

Figure 4 shows a typical temporal record for laser transmittance and soot emission at mid-height of the ethylene jet flame along the flame centerline, and Fig. 5 shows a time record collected fairly high in the same flame. At the upper height,

there are clearly distinct time intervals when no soot signals are apparent in the probe volume. To ensure that the deduced soot temperature data are not influenced by low signal-to-noise emission data at times when there is little to no soot in the probe volume, cut-off criteria for minimum emission signals have been established at each sampling location, based on the emission signal levels when the laser transmission measurement shows no soot in the probe volume. The emission signals show better sensitivity to low soot concentrations than the laser attenuation measurement, but since 2-color pyrometry relies on the ratio of the emission signals it is very sensitive to noise and a conservative analysis cutoff criteria has been chosen here out of prudence.



Figure 3. Color digital SLR camera photographs of the ethylene jet flame (left) and the JP-8 surrogate jet flame (right).

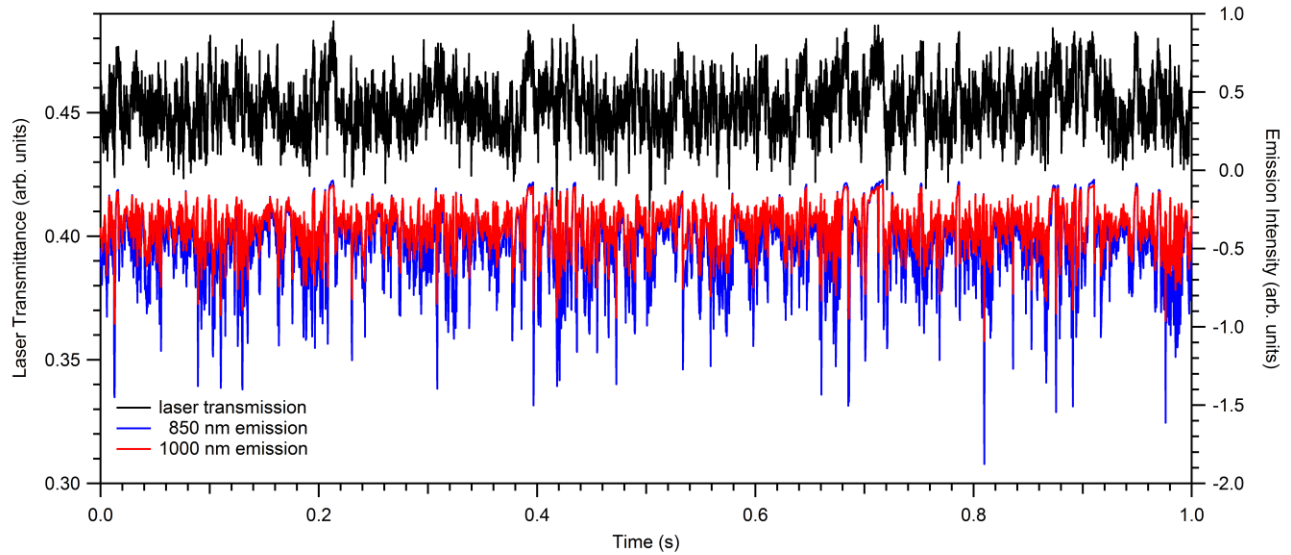


Figure 4. Example time record of laser transmittance and soot emission for a height of 44.8 cm ( $z/d = 140$ ) along the centerline of the ethylene jet flame. Note that the emission signals are recorded as negative values (i.e. increasing signal corresponds to larger negative numbers).

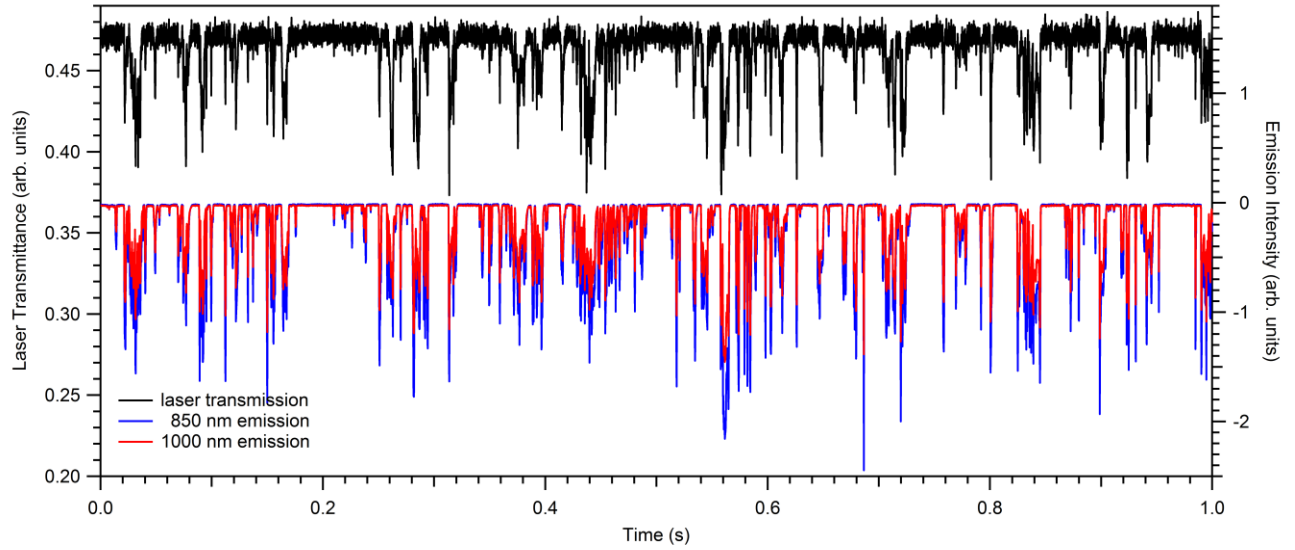


Figure 5. Example time record of laser transmittance and soot emission for a height of 64.0 cm ( $z/d = 200$ ) along the centerline of the ethylene jet flame.

Fig. 6 shows the corresponding computed soot volume fraction and soot temperature time records for the raw data shown in Fig. 4. From this time record, it appears that the mean volume fraction is approximately 0.5 ppm, with occasional dropouts (i.e. moments with no soot) and instantaneous peak volume fractions of just over 1 ppm. The soot temperature at this location appears to be approximately 1500 K, with excursions down to 1200 K and up to just over 1800 K. Of course, it should be appreciated that because this measurement occurs over a path length of 10 mm in the flame, it is likely that soot at different temperatures is being sampled and the 2-color pyrometry measurement is inherently biased towards the hotter soot that is present in the probe volume [17].

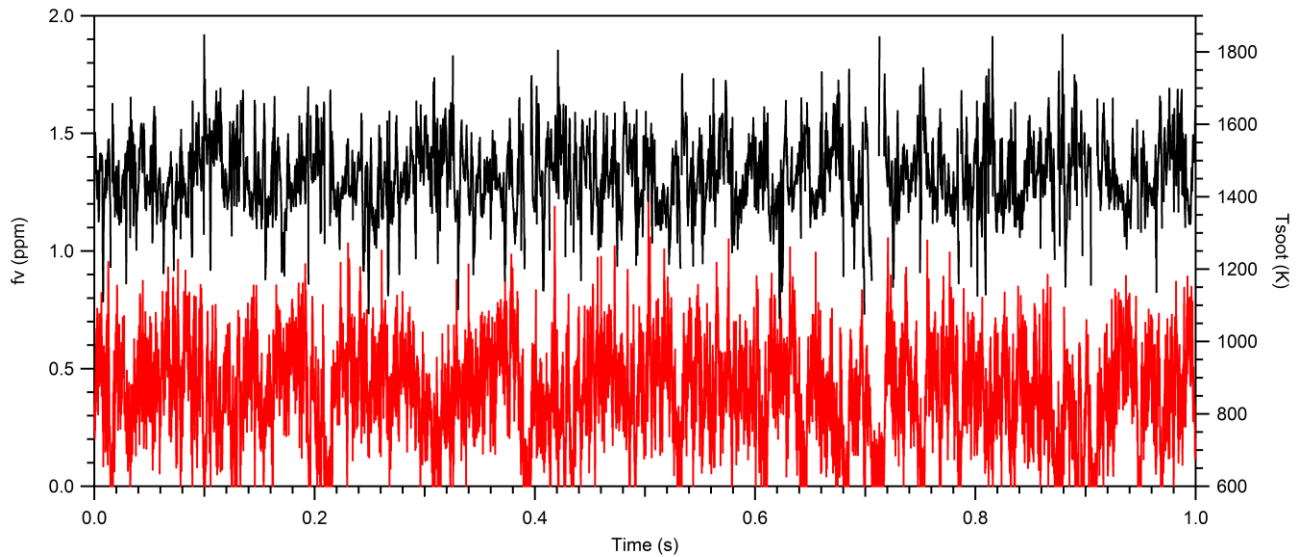


Figure 6. Example time record of deduced soot volume fraction and soot temperature for a height of 44.8 cm ( $z/d = 140$ ) along the centerline of the ethylene jet flame. The volume fraction is shown in red, corresponding to the left axis, and the soot temperature is in black and corresponds to the right axis.

Figures 7 and 8 show the measured probability distribution functions (pdf) for soot concentration and soot temperature at mid-height and at an elevated height of the ethylene flame. Fig. 9 shows the corresponding joint pdfs (i.e. bivariate histograms). The results show a mean soot volume fraction of 0.4 ppm at mid-height of the ethylene flame, with a mean

temperature of 1450 K. At any given instant, the soot volume fraction typically falls between 0.1 and 0.7 ppm with a temperature between 1300 and 1650 K. At greater heights in the flame, the soot intermittency increases and its mean concentration decreases while its mean temperature increases. As is apparent in Figs. 8 and 9, however, occasionally higher local concentrations of soot are seen at greater heights in the flame. Furthermore, lower concentrations of soot are sometimes associated with lower temperatures, which could reflect the occurrence of soot-containing quenched vortices [18].

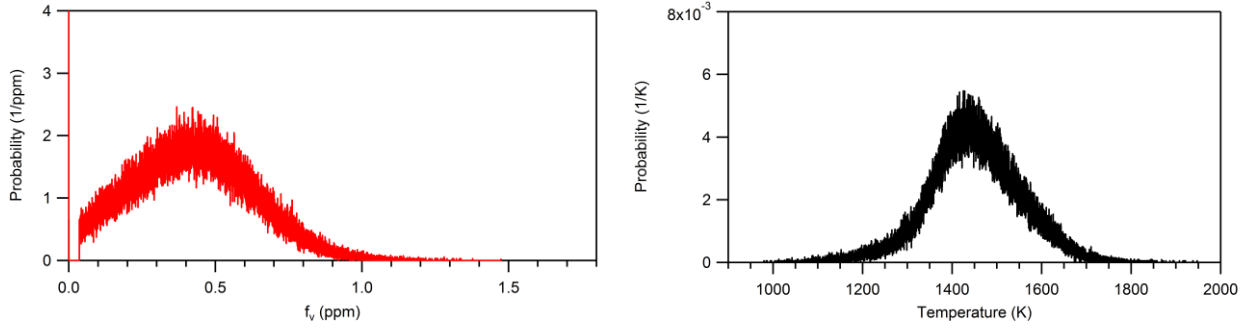


Figure 7. Soot volume fraction pdf (left) and soot temperature pdf (right) at a height of 44.8 cm ( $z/d = 140$ ) along the centerline of the ethylene jet flame.

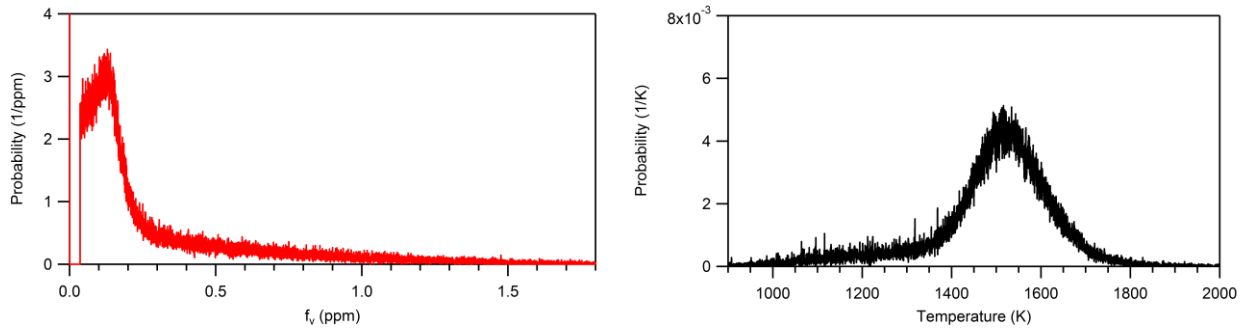


Figure 8. Soot volume fraction pdf (left) and soot temperature pdf (right) at a height of 64.0 cm ( $z/d = 200$ ) along the centerline of the ethylene jet flame.

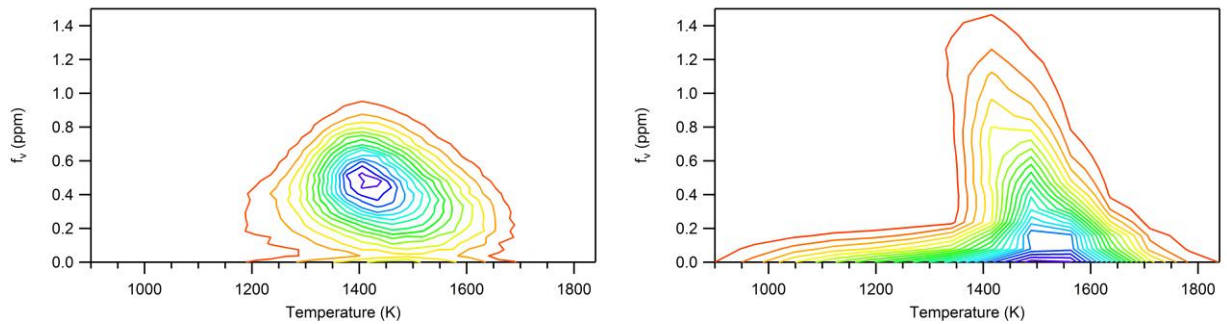


Figure 9. Joint pdfs of soot volume fraction and soot temperature at a height of 44.8 cm ( $z/d = 140$ ) (left) and at a height of 64.0 cm ( $z/d = 200$ ) along the centerline of the ethylene jet flame.

Corresponding results from the prevaporized JP-8 surrogate flame are shown in Figures 10-12. In the JP-8 surrogate flame, the soot concentration reaches a mean value of 1.3 ppm at mid-height of the flame, but the mean soot temperature is only 1270 K. Elevated soot concentrations persist for a range of heights in the JP-8 flame, with a rise in mean temperature to 1360 K, before both soot volume fraction and temperature tail off at the top of this smoking flame. The probability distribution function shapes for soot volume fraction and temperature are similar for the JP-8 surrogate flame in comparison to the ethylene flame, except that the temperature distribution for the JP-8 flame at intermediate heights



shows evidence of a small peak towards higher temperatures riding on the shoulder of the main peak. The joint pdfs also are similar to those observed for the ethylene flame, except for the evidence of the small higher temperature ‘arm’ that is present at intermediate heights, as well as the lack of increase in temperature of the most probable soot measurement with increasing measurement height.

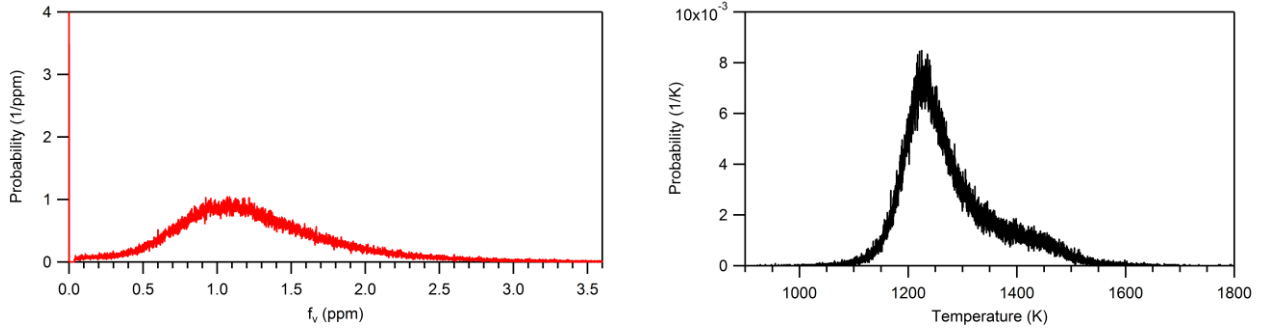


Figure 10. Soot volume fraction pdf (left) and soot temperature pdf (right) at a height of 37.5 cm ( $z/d = 150$ ) along the centerline of the JP-8 surrogate jet flame.

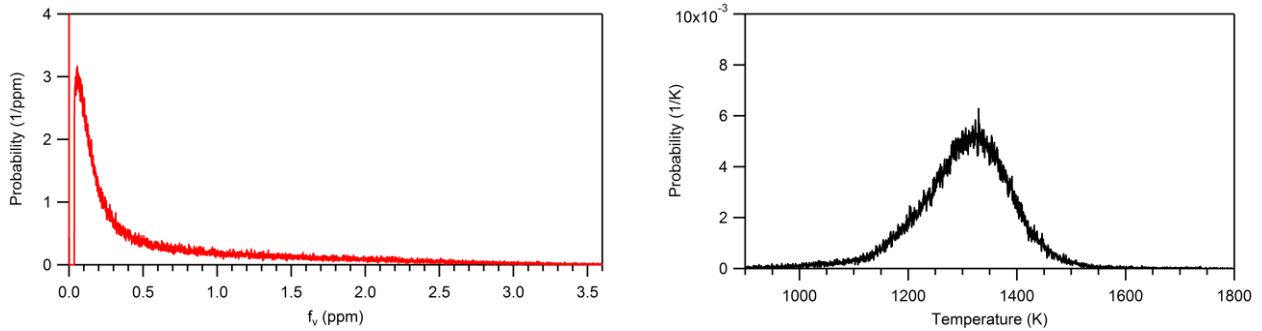


Figure 11. Soot volume fraction pdf (left) and soot temperature pdf (right) at a height of 68.75cm ( $z/d = 275$ ) along the centerline of the JP-8 surrogate jet flame.

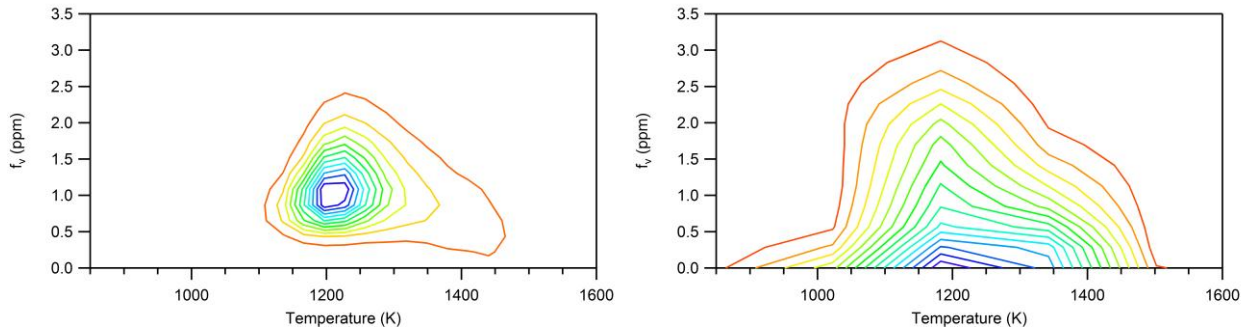


Figure 12. Joint pdfs of soot volume fraction and soot temperature at a height of 37.5 cm ( $z/d = 150$ ) (left) and at a height of 68.75 cm ( $z/d = 275$ ) along the centerline of the JP-8 surrogate jet flame.

The soot volume fraction-temperature joint pdfs measured in these turbulent jet flames are qualitatively similar to those reported by Murphy and Shaddix [18] along the centerline of a 2-m diameter JP-8 pool fire, with an increasing spread of both volume fraction and temperature as one proceeds up the flame. In fact, the joint pdf at mid-height of the JP-8 surrogate jet flame ( $z/d = 150$ ) is nearly identical (even in quantitative values) to the joint pdf measured at mid-height of the JP-8 pool fire ( $z/d = 0.5$ ).

#### 4. Conclusions

Soot concentration (volume fraction) and temperature have been measured in turbulent non-premixed jet flames fueled by undiluted ethylene and prevaporized JP-8 surrogate, respectively, using the combined laser extinction/2-color emission pyrometry approach over an optical pathlength of 10 mm. Both the mean soot concentration values and the statistical distributions show approximately twice the soot concentrations in the JP-8 surrogate flame as in the ethylene flame. Further, the soot in the JP-8 surrogate flame is approximately 200 K cooler than the soot in the ethylene flame. For both flames, the distribution of soot concentrations and temperatures widens with increasing flame height, even as the fraction of time in which no soot is present in the probe volume increases. This data should aid in the development and validation of non-premixed turbulent flame simulations that include soot formation and radiation.

#### Acknowledgements

This work was supported by the U.S. DoD's Strategic Environmental Research and Development Program (SERDP). The authors thank Robert Harmon of Sandia for his technical assistance with experiments. Sandia is a multi-program laboratory operated by Sandia Corporation, a Lockheed Martin Company, for DOE's National Nuclear Security Administration under Contract DE-AC04-94AL85000.

#### References

- [1] F. Laden, L.M. Neas, D.W. Dockery, J. Schwartz, *Envir. Health Persp.* 108 (2000) 941-947.
- [2] C.A. Pope, R.T. Burnett, M.J. Thun, E.E. Calle, D. Krewski, K. Ito, G.D. Thurston, *JAMA* 287 (2002) 1132-1141.
- [3] C. de Haar, I. Hassing, M. Bol, R. Bleumink, R. Pieters, *Toxic. Sciences* 87 (2005) 409-418.
- [4] R.B. Schlesinger, N. Kunzli, G.M. Hidy, T. Gotschi, M. Jerrett, *Inhalation Toxic.* 18 (2006) 95-125.
- [5] M.R. Heal, P. Kumar, R.M. Harrison, *Chem. Soc. Rev.* 41 (2012) 6606-6630.
- [6] A.C. Rohr, R.E. Wyzga, *Atmos. Envir.* 62 (2012) 130-152.
- [7] R. Watson, J. Houghton, D. Yihui, B. Metz, O. Davidson, N. Sundararaman, D. Griggs, D. Dokken, *Aviation and the Global Atmosphere*, a special report of the Intergovernmental Panel on Climate Change (IPCC), 1999, available from <http://www.ipcc.ch/ipccreports/sres/aviation/index.htm>.
- [8] T.C. Bond, S.J. Doherty, D.W. Fahey, et al. *J. Geophys. Res. Atm.* doi: 10.1002/jgrd.50171.
- [9] R.S. Barlow, *Proc. Combust. Instit.* 31 (2007) 49-75.
- [10] N.H. Qamar, Z.T. Alwahabi, Q.N. Chan, G.J. Nathan, D. Roekaerts, K.D. King, *Combust. Flame* 156 (2009) 1339-1347.
- [11] A. Gupta, D.C. Haworth, M.F. Modest, *Proc. Combust. Instit.* 34 (2013) 1281-1288.
- [12] P.J. Coelho, *Prog. Energy Combust. Sci.* 33 (2007) 311-383.
- [13] J. Zhang, C.R. Shaddix, R.W. Schefer, Proceedings of the Western States Section of the Combustion Institute, Paper 09F-57, Oct. 26-27, 2009, UC Irvine, Irvine, CA.
- [14] Y.R. Sivathanu, G.M. Faeth, *Combust. Flame* 81 (1990) 150-165.
- [15] T.C. Williams, C.R. Shaddix, K.A. Jensen, and J.M. Suo-Anttila, *Int. J. Heat and Mass Transfer* 50 (2007) 1616-1630.
- [16] J. Zhang, C.R. Shaddix, R.W. Schefer, *Rev. Sci. Instr.* 82 (2011) 074101.
- [17] J.J. Murphy, C.R. Shaddix, *Combust. Flame* 143 (2005) 1-10.
- [18] J.J. Murphy, C.R. Shaddix, *Combust. Sci. Technol.* 178 (2006) 865-894.