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Simultaneous instantaneous measurements of soot volume fraction, primary particle diameter, and aggregate size in turbulent buoyant diffusion flames

Brian M. Crosland [a], Kevin A. Thomson [b], Matthew R. Johnson [a,*]

[a] *Energy & Emissions Research Lab., Department of Mechanical and Aerospace Engineering, Carleton University, Canada*
[b] *Measurement Science and Standards, National Research Council of Canada, Canada*

Abstract

Understanding the mechanisms of soot formation, growth, oxidation, and emission is important for diverse reasons including better combustor design, quantifying soot's contribution to climate change, and mitigating air quality concerns. Instantaneous soot measurement in turbulent flames is difficult, and has mostly been restricted to high-momentum jet flames and pool fires. The current work partially fills this gap by presenting simultaneous measurements of soot volume fraction (f_v), effective aggregate radius of gyration (R_{gm1}), and primary particle diameter (d_p) in combination with separately-acquired measurements of mean gas velocity, within a range of turbulent buoyant non-premixed jet flames burning a fuel mixture representative of associated gas flares in the upstream oil and gas industry. Fifteen cases comprising six possible nozzle exit velocities and four nozzle diameters were studied. Histograms of f_v and d_p support the suggestion that reduced f_v near the flame tip results from increased soot intermittency rather than reduced local f_v. Furthermore, the data indicate that oxidation of mature soot structures occurs rapidly and completely, and is insensitive to the local f_v. For most conditions studied, centerline f_v shows self-similar behavior when scaled by measured flame height in the axial direction. The more buoyancy-dominated flames show a marked increase in peak f_v that occurs lower in flame-length-normalized coordinates, while the more momentum-dominated flames show a decreased peak f_v that occurs higher in flame-length-normalized coordinates. Low in the flame, soot is only present in an annular region where f_v, d_p, and R_{gm1} all grow. Once soot is present on the centerline, d_p growth slows and d_p is relatively constant at all radial locations and all conditions higher in the flame. Aggregate size continues to grow steadily with increased flame height and correlates well with residence time when adjusted to account for differing flow fields close to the burner outlet.

Keywords: Soot morphology; Elastic light scattering; Laser-induced incandescence; Turbulent diffusion flame; Turbulent buoyant flame

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* Corresponding author. Address: Mechanical & Aerospace Engineering, Carleton University, 1125 Colonel By Drive, Ottawa, ON K1S 5B6, Canada. Fax: +1 613 520 5715.

E-mail address: Matthew.Johnson@Carleton.ca (M.R. Johnson).

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1. Introduction

Combustion-generated soot is a known health hazard [1,2], and black carbon, a principal constituent of soot, is a critical climate forcer [3–5]. Recent studies implicate black carbon as the second-most important climate-forcing agent after carbon dioxide [5–7]. Gas flaring in the petroleum industry is a potentially significant source of global soot emissions given that satellite data indicate global flared volumes exceed 135 billion m^3 annually [8]. Accurate estimation of soot emissions to meet reporting regulations and support informed policy decisions is difficult due to the limited literature for buoyancy-dominated turbulent non-premixed flames, the questionable relevance of existing emission factor models (see [9]), and the challenges of in situ field measurements [10,11]. Although flares, like most practical combustion processes, involve turbulent flames, academic research of sooting flames has traditionally focused on laminar flames due to their relative simplicity. Both modeling and measurements of turbulent flames are complicated by their unsteady nature, short time scales, complex chemistry, large spatial gradients, and thermal radiation. Consequently, there are comparatively few works focusing on sooting in turbulent flames. Most previous studies reporting spatially-resolved measurements of soot in non-premixed turbulent flames have focused on momentum-dominated flames [e.g. 12–22] with nozzle exit Reynolds numbers (Re) ranging from 4000 to 83,000. Of these, most use heavier fuels than the methane-dominated compositions found in typical associated gas flares [23]. A notable exception is Qamar et al. [21], who performed 2D laser-induced incandescence (LII) measurements in the Delft Flame III with simulated Dutch natural gas and postulated that the reduction in mean soot volume fraction (f_v) near the flame tip occurs via a reduction in the number of soot sheets, rather than a reduction in f_v within soot sheets. This was supported by instantaneous 2D measurements of f_v [12] in a turbulent non-premixed ethylene jet flame.

To the authors' knowledge, only two studies have reported spatially-resolved measurements of f_v within buoyant non-premixed turbulent flames [24,25]. Xin and Gore [25] performed two-dimensional f_v measurements in turbulent buoyant methane and ethylene flames with Re of ~140. Coppalle and Joyeux [24] reported simultaneous local temperature and f_v measurements in buoyancy- and momentum-dominated ethylene flames with Re of 550, 5700 and 11,800.

Scaling of buoyant non-premixed flames is complicated by the dependence of flame length (L_f) on fuel flow rate. Results in [24] were presented on spatial coordinates normalized by the location of the peak f_v, producing some agreement low in the flame for the two momentum-dominated flames, but generally poor agreement with the buoyancy-dominated flame. Xin and Gore [25] presented results with axial location normalized by nozzle diameter, D_e (as is common practice for momentum-dominated flames where L_f is typically only dependent on fuel properties and D_e), but were unable to directly compare quantitative results between their two fuel types due to optically-thick conditions in the ethylene flame.

The primary objectives of the current research are to (i) use a novel diagnostic technique to explore soot trends in turbulent buoyant flames of practical interest, (ii) investigate the mechanism of soot burnout high in the flame, and (iii) evaluate methods of scaling mean f_v and morphology trends in turbulent buoyant non-premixed flames.

2. Experimental setup and method

2.1. Burner

A schematic of the turbulent non-premixed lab-scale flare (LSF) is provided in [9]. Mass flow controllers feed mixed, gaseous fuel through a diffusing chamber, settling chamber, cubic-contoured converging nozzle, and into an interchangeable burner tip. Burner tip diameters of 25.4, 38.1, 50.8, and 76.2 mm were used in the present experiments. Turbulence grids placed three to five diameters upstream of each burner exit were used to produce cold-flow turbulence intensities (confirmed by hotwire anemometer measurements) of 2–5% at the exit plane. The fuel mixture was selected to be representative of associated gas flare compositions [9,23] and contained 85.3% CH_4, 7.1% C_2H_6, 3.1% C_3H_8, 1.4% C_4H_{10}, 1.9% CO_2 and 1.2% N_2 by volume.

2.2. Measurement strategy

A novel combined laser induced incandescence (LII) and elastic light scattering (ELS) apparatus [26] was used to make simultaneous, instantaneous measurements of f_v, primary particle diameter (d_p) and aggregate radius of gyration (R_{gm1}). Due to vertical space constraints, the burner was fixed in place and a platform supporting the laser head as well as the beam shaping and signal collection optics was positioned using a three-axis traverse. Light from a pulsed Nd:YAG laser operating at the fundamental wavelength of 1064 nm was formed into an 8 × 0.5 mm cross-section sheet and used to induce incandescence and scattering signals from soot present in the flame. The scattering signal was detected at two diametrically opposed locations representing forward scattering (30°) and backward scattering (150°); incandescence signals were also collected from the backward scattering optics. The signals were filtered by wavelength (centered at 447 and