ACCEPTED VERSION

Douglas B Proud, Michael J Evans, Qing N Chan, Paul R Medwell **Dilute spray flames of ethanol and n-heptane in the transition to mild combustion** Combustion and Flame, 2022; 238:111918-1-111918-15

© 2021 The Combustion Institute. Published by Elsevier Inc. All rights reserved.

This manuscript version is made available under the CC-BY-NC-ND 4.0 license <u>http://creativecommons.org/licenses/by-nc-nd/4.0/</u>

Final publication at: http://dx.doi.org/10.1016/j.combustflame.2021.111918

PERMISSIONS

https://www.elsevier.com/about/policies/sharing

Accepted Manuscript

Authors can share their accepted manuscript:

24 Month Embargo

After the embargo period

- via non-commercial hosting platforms such as their institutional repository
- via commercial sites with which Elsevier has an agreement

In all cases accepted manuscripts should:

- link to the formal publication via its DOI
- bear a CC-BY-NC-ND license this is easy to do
- if aggregated with other manuscripts, for example in a repository or other site, be shared in alignment with our <u>hosting policy</u>
- not be added to or enhanced in any way to appear more like, or to substitute for, the published journal article

13 May 2024

http://hdl.handle.net/2440/134698

Dilute spray flames of ethanol and n-heptane in the transition to mild combustion

Douglas B Proud^{a,*}, Michael J Evans^{a,b}, Qing N Chan^a, Paul R Medwell^c

^aSchool of Mechanical Engineering, The University of Adelaide, Adelaide, SA 5005, Australia

^bUniSA STEM, University of South Australia, Mawson Lakes, SA 5095, Australia ^cSchool of Mechanical and Manufacturing Engineering, University of New South Wales,

Sydney, NSW 2052, Australia

Abstract

The structure and stabilisation mechanisms of ethanol and *n*-heptane spray flames are investigated in this study. The burner configuration involves a dilute spray of dispersed droplets which is produced and transported via a carrier gas stream of air to the reaction zone, where the flames are stabilised by a hot coflow of combustion products. A range of coflow conditions were implemented for the different flame cases, allowing the effects of the coflow oxygen (O_2) concentration and temperature to be examined independently. The resulting flames were analysed using three simultaneous laser diagnostic techniques, enabling the combined planar imaging of the hydroxyl (OH) and formaldehyde (CH_2O) radicals, along with the location of droplets. For both fuel types, a noticeable shift in stabilisation behaviour was observed with a variation in the coflow O_2 concentration from 11% to 3%, while the coflow temperature was not seen to have a significant impact. These flames also show an interesting departure from the typical behaviour observed for gaseous and prevaporised flames in a similar configuration, particularly for coflow conditions that are typically associated with the transition to the mild combustion regime.

 $Keywords: \ \ Spray \ combustion, \ Mild \ combustion, \ Laser \ diagnostics, \ Flame stabilisation$

1. Introduction

The use of spray-injected liquid fuels is widespread in practical combustion devices, particularly in applications where a high energy density is a priority, such as the transport and aerospace sectors. Injection of liquids in the form of a spray enhances the combustion process, by increasing the rate of fuel evaporation and improving the mixing between fuel and air. In order to develop lowemissions and fuel-flexible technologies, the ability to accurately predict spray

^{*}Corresponding author. E-mail: douglas.proud@adelaide.edu.au

combustion phenomena under a range of conditions is a necessity [1, 2]. This requires a comprehensive understanding of the physical and chemical processes which take place in spray flames, including turbulence-chemistry interactions, droplet evaporation, and spray break-up.

In practical burners, liquid fuels are typically injected directly into the reaction zone in the form of a "dense" spray, in which droplets are clustered together. This dense region then undergoes spray break-up, with droplets becoming smaller and more dispersed, ultimately evaporating before the fuel burns. This dispersed region of the spray—in which interactions between individual droplets are negligible—is referred to as a "dilute" spray [3]. In order to focus on the fundamental combustion processes in spray flames in an experimental setting, it is useful to form a dilute spray directly, as this facilitates the use of non-intrusive laser diagnostic techniques [4].

A range of detailed measurements have previously been obtained using a dilute spray configuration, providing important insights into the structure and behaviour of spray flames. Experimental findings based on a piloted, dilute spray burner highlighted the complexity of the reaction zones and ignition modes in such flames, with particular focus on the sensitivity to the boundary conditions [5]. Due to this sensitivity, there has been a conscious effort to obtain experimental results for flames with well-characterised initial conditions, forming a major focus of the Workshop on Turbulent Combustion of Sprays (TCS) [6, 7]. These efforts have in turn enabled the development of more accurate and efficient numerical models [8, 9], by providing a detailed database for model validation. However, a lack of understanding regarding the structure and stabilisation of dilute spray flames still persists, particularly in relation to "non-conventional" combustion regimes, such as those encountered in sequential gas turbines and burners featuring recirculation of exhaust products [1].

The dilution of air with exhaust gases—either via recirculation or sequential combustion—can provide increased performance in a range of practical applications. The excess heat from the combustion products enables preheating of the reactants, facilitating an increase in thermal efficiency and improved combustion stability [10]. Additionally, the reduced O_2 concentration resulting from the dilution leads to a less intense combustion process, limiting the increase in temperature and allowing a reduction of CO, NO_x and soot emissions under certain conditions [11, 12]. A particular combustion regime which is of interest in this regard is "mild" combustion, which, in addition to describing the nature of the combustion process, stands for moderate or intense low-oxygen dilution combustion [13]. While initially developed and studied for implementation in furnaces, there is also potential to extend this combustion technique to both land-based and aerospace gas turbines, particularly in applications where greater flexibility is required, both in terms of operating conditions and fuel type [1].

Much of the fundamental experimental work on mild combustion has been carried out using a "jet in hot cross-/co-flow" (JHC) burner configuration [14, 15, 16, 17]. This type of burner allows the temperature and O_2 concentration of the oxidant stream to be controlled and adjusted independently of the fuel

composition and flow rate, allowing the transition to the mild regime to be studied. In the context of gaseous flames, several interesting findings have previously been made regarding the change in stabilisation and lift-off behaviour with variations in the coflow properties [18, 19, 20, 21]. It has been observed that there is a fundamental shift in the stabilisation mechanism of visually lifted flames in the mild regime in comparison with conventional lifted, autoignitive flames, with pre-ignition reactions being detected upstream of the apparent liftoff height [18], and formation of ignition kernels playing a key role in flame stabilisation [19]. This behaviour has been found to be very sensitive to the fuel composition, as well as the coflow temperature and O_2 level [20]. It is important to note that the JHC configuration essentially emulates the conditions required for mild combustion in a simplified configuration, such that the overall appearance of the resulting flames tends to be different to that which is observed under more practical implementations of mild combustion, particularly in downstream locations where the controlled influence of the coflow is diminished [18, 22].

While most fundamental studies of mild combustion have been focussed on relatively simple, gaseous fuels, liquid fuels have also been investigated. Prevaporised flames of ethanol and dimethyl ether (DME) issuing into a hot coflow were found to exhibit similar structures, particularly at a coflow oxygen concentration of 3% by volume, with an increased sensitivity to fuel type at higher O_2 concentrations [23]. In the same experimental configuration, *n*-heptane flames were found to exhibit a significantly different behaviour to prevapourised ethanol and other gaseous flames, with a general deviation away from the mild regime even at very low oxygen concentrations [24]. This was attributed to the more complex chemistry of *n*-heptane and the increased tendency for pyrolysis to occur.

Several studies of mild/flameless combustion of liquid fuels have also been carried out using an enclosed or confined burner configuration. While these configurations typically do not facilitate the analysis of the flame structure and stabilisation mechanisms in as much detail as open flames, the transition to the mild combustion regime can be examined via temperature measurements and analysis of the flue gas composition, in addition to visual observations. Experiments with ethanol in liquid form were performed using a confined burner with recirculation of flue gases and a "blurry injector" configuration to generate a spray, with reduced pollutant emissions and uniform temperatures observed under certain operating conditions corresponding to a range of excess air coefficients [25]. Experiments involving a cyclonic flow configuration emphasised the flexibility of mild combustion, with steady operation spanning across different equivalence ratios and heat inputs for three low-molecular-weight alcohol fuels. while maintaining complete fuel conversion and low pollutant emissions [26]. While these investigations—and indeed others based upon EGR configurations [27, 28]—are very useful for studying the limits of mild combustion in a practical environment under various conditions, it is difficult to draw fundamental conclusions from them with regards to ignition processes and flame structure, largely due to a lack of optical access and challenges with quantifying the level of recirculation in many cases [1]. Thus, while such studies offer important insights which cannot be ascertained from JHC-type experiments, their limitations also emphasise the importance of studying liquid fuels in a JHC configuration, particularly with regards to the development of numerical models.

Liquid sprays issuing into a hot and low-oxygen coflow have also been investigated previously. Measurements of droplet location and gas temperature were obtained for the "Delft spray in hot coflow" (DSHC) flames [29], using a pressure-swirl atomiser to generate droplets. A much more rapid spray breakup and evaporation of droplets was observed for sprays in a hot-diluted coflow compared with a coflow of air. The change in coflow conditions was also found to have a noticeable impact on the underlying flame structure, with a more uniform temperature distribution resulting in lower peak temperatures for the hot-diluted coflow. The existence of distinct inner and outer reaction zones was another important feature of these flames, attributed to the transport of larger droplets away from the spray axis and the flame propagation of a dropletvapour-air mixture towards the centreline [29]. This "double flame" structure was found to be weakened in the presence of a hot and low-oxygen coflow, due to a less pronounced inner flame resulting from a richer gaseous mixture. This structure has also been investigated for pressure-swirl-atomised spray flames in coflows of room-temperature air at a range of coflow velocities (including no coflow) [30, 31], with a transition to a single reaction zone for high coflow velocities. For these flames, partial premixing was found to be crucial to the development of the inner flame structure [31]. The structure of the DSHC flames was also reproduced via large-eddy simulation (LES), in which the formation of multiple flame structures was found to be very sensitive to evaporation rates and chemical time-scales [32], emphasising the importance of understanding these phenomena to further develop modelling capabilities.

A double flame structure has also been identified in a dilute spray configuration [33] which is similar to that used in the current study. For the case of fuel droplets carried by air, it was hypothesised that vaporisation of droplets prior to ignition leads to partial premixing and the formation of locally ignitable mixtures, resulting in an unsteady inner flame front and a broadening of the OH laver [33]. This behaviour was not observed with nitrogen as the carrier gas, with the flame exhibiting a more typical diffusion-like behaviour. The effect of jet boundary conditions and fuel composition has also been investigated using a dilute spray burner, in which simultaneous imaging of fuel droplets and OH/CH₂O-PLIF was performed [34]. While a double flame structure was a consistent feature of all flames studied, a change in behaviour from a bifurcating structure to the occurrence of distinct ignition kernels was identified for ethanol and *n*-heptane flames. While these studies reveal important details of the structure and stabilisation of spray flames in a hot and low-oxygen environment, an open question remains regarding the effect of the coflow conditions, particularly at reduced O_2 concentrations, such that the transition towards the mild regime is realised.

This study builds upon previous experimental work performed with the same burner configuration [34]. While the aforementioned study ([34]) presented results and analysis related to the overall evolution of the flame structures for a range of jet boundary conditions, the current paper is focussed on investigating the stabilisation mechanisms of the flames, and the impact of the coflow composition and temperature on these mechanisms. The burner used is based upon the well-established JHC configuration, allowing the independent effects of coflow temperature and O_2 concentration to be determined while the jet boundary conditions are held constant. Two different liquid fuels are considered in this study; namely ethanol and n-heptane, facilitating additional analysis in terms of the effects of chemistry. These two fuels are also of significant importance in practical combustion applications; ethanol has been identified as a promising low-carbon fuel for the transport sector [35], while *n*-heptane is commonly used as a primary reference fuel for the analysis of internal combustion engines [36]. An ultrasonic nebuliser is implemented to generate a dispersed droplet field, with simultaneous imaging of fuel droplets via Mie scattering, and key intermediate flame species (predominately OH and CH_2O) via planar laserinduced fluorescence (PLIF). This offers a unique insight into the stabilisation and propagation of dilute sprays reacting under conditions relevant to practical, low-emissions and fuel-flexible combustion devices. In particular, an interesting change in the stabilisation behaviour is observed as the O_2 concentration is reduced from 11% to 3%, and fundamental differences in the behaviour of the ethanol and *n*-heptane flames are examined. In addition to developing a deeper understanding of the mechanisms involved in spray combustion, these results also provide a challenging target to test the predictive capabilities of numerical models under a range of boundary conditions.

2. Methodology

2.1. Spray burner description

The burner configuration implemented in this study is identical to that which has been used previously to investigate the effect of jet boundary conditions on spray flames in a hot and low-oxygen coflow [34]. The burner shares many characteristics with the well-studied JHC configuration (e.g. [14] and [37]), while also incorporating the features of dilute spray burners which have previously been used to study piloted flames [6, 38]. An ultrasonic nebuliser is used to generate fuel droplets with a nominal Sauter mean diameter of 30 μ m. These droplets have minimal initial momentum, and are transported to the jet exit via a carrier gas stream of air. The jet has an exit inner diameter (D) of 20 mm, and the Reynolds number based on this diameter (Re_{jet}) for all flame cases was 5000. Both ethanol and *n*-heptane were used as the liquid fuel for different flame cases, with the mass loading of liquid held constant at 0.21 g/s. The hot coflow is produced by the lean premixed combustion of natural gas, H₂ and air in varying quantities, with dilution via N_2 to control the concentration of O_2 in the coflow stream. Further details and a schematic of the burner and nebuliser configuration have been presented in a previous publication [34].

By varying the flow rates of natural gas, H_2 , N_2 and air into the porous bed, four different coflow conditions were produced. These consist of three

	Jet Properties			Coflow Properties				
Case	Fuel	Re_{jet}	Φ_{jet}	X_{O_2}	X_{H_2O}	X_{CO_2}	X_{N_2}	T_{cofl} [K]
HEP-03	<i>n</i> -heptane	5000	2.2	0.030	0.11	0.036	0.83	1400
HEP-08	n-heptane	5000	2.2	0.075	0.11	0.036	0.78	1400
HEP-11	n-heptane	5000	2.2	0.11	0.11	0.036	0.75	1400
HEP-H08	n-heptane	5000	2.2	0.075	0.12	0.061	0.74	1690
ETH-03	Ethanol	5000	1.3	0.030	0.11	0.036	0.83	1400
ETH-11	Ethanol	5000	1.3	0.11	0.11	0.036	0.75	1400

Table 1: Table of flame cases, indicating the jet and coflow properties. Numbers in case names represent the rounded molar percentage of O_2 in the coflow, while the letter "H" in the suffix denotes the higher temperature (1690 K) coflow.

coflows with different O₂ concentrations (all with the same temperature), and two coflows with different temperatures and the same O_2 concentration, enabling the effects of these two parameters to be investigated independently. For the results presented in this paper, the *n*-heptane flames cover all four coflow conditions, while for the ethanol flames only two different coflow conditions are considered. Table 1 displays the various cases for which results are presented, and their corresponding jet and coflow boundary conditions. Also shown in Table 1 is the overall jet equivalence ratio (Φ_{iet}) , which is based on the total flowrates of liquid fuel and carrier air in the main fuel tube. It should be noted that some evaporation of the liquid fuel occurs within this tube, that is, between the nebulising surface and the jet exit. Since air is used as the carrier gas, this prevaporised fuel undergoes premixing within the pipe. In a previous study using a similar dilute spray configuration, the extent of droplet evaporation within the pipe was found to depend on the relative flow of the liquid and carrier gas [6]. Based on these results, it is estimated that approximately 40% of the liquid fuel undergoes evaporation prior to reaching the jet exit plane.

The flame cases described in Table 1 all correspond to a fuel mass flowrate of 0.21 g/s. As a result, the mixture in the jet (i.e. liquid fuel and carrier air) is overall rich for all cases studied, with $\Phi_{jet} = 2.2$ for the *n*-heptane flames and $\Phi_{jet} = 1.3$ for the ethanol flames, noting that this difference is due to the lower stoichiometric air-fuel ratio for ethanol. In a previous study with the same burner [34], flames with different equivalent ratios were analysed, and it was found that the flame structure was very sensitive to these conditions. In particular, leaner mixtures tend to favour the propagation of the inner flame front due to the influence of the air in the jet, with a less prominent double flame structure present. It then follows that the stabilisation and overall evolution of these flames would be less sensitive to the influence of the coflow, hence the focus on rich mixtures in the current study.

2.2. Diagnostic techniques

Three laser diagnostic techniques were implemented simultaneously in this study, to enable combined imaging of the flame boundary and intermediate species, along with the location of fuel droplets. Planar imaging through the central axis of the burner was performed, using vertical sheets of pulsed laser light with a frequency of 10 Hz and a nominal height of 15 mm. Two Nd:YAG lasers and one Nd:YAG-pumped dye laser were used to generate the three laser sheets of different wavelengths. Different axial locations (x) within the flames were captured by traversing the burner in the vertical direction, ranging from the jet exit (x = 0) to 112 mm above the jet (x/D = 5.6). A set of 255 images were collected for each diagnostic at each location, facilitating the analysis of the mean flame structure in addition to capturing instantaneous features.

To study the boundary of the reaction zone, the hydroxyl (OH) radical was imaged via the planar laser-induced fluorescence (PLIF) technique. A dye laser, pumped by the 532 nm output of an Nd:YAG laser, was tuned to a wavelength of 282.927 nm to excite the $Q_1(6)$ transition of the OH radicals. This transition was selected as it has a relatively low sensitivity to temperature fluctuations in the range of interest, and provides a stronger signal in comparison to the $Q_1(7)$ transition. The dye laser was operated with an energy of 0.8 mJ/pulse, and the fluorescence from the OH was imaged onto an ICCD camera through an f/3.5 UV lens, with a gate width of 100 ns. The camera lens was fitted with a bandpass filter centred at 310 nm, with a peak transmission > 70% and FWHM of 10 nm. An additional processing step was applied to the OH-PLIF images to facilitate the separate analysis of the inner and outer branches of the flames. This involved the implementation of an algorithm to find the peaks in the radial OH profile for each image, in conjunction with a shape filter for the OH structures, allowing the statistical behaviour of the reaction zone to be explored in more detail.

The PLIF technique was also implemented to detect intermediate species associated with fuel decomposition and pre-ignition reactions, in particular the formaldehyde (CH_2O) species. This was performed using the third harmonic (355 nm) of an Nd:YAG laser, with a measured energy of 125 mJ/pulse. It is important to note that excitation in the UV region is known to lead to the fluorescence of several intermediate species in a flame, leading to difficulty in attributing the signal to a specific chemical species in some instances. A narrowrange bandpass filter was used to filter out interference from unwanted sources. where possible. This bandpass filter was centred at 410 nm with a FWHM of 10 nm (transmission > 45%), allowing fluorescence from the CH₂O species to be targeted. Consequently, the signal associated with the 355 nm laser is expected to be dominated by CH₂O-PLIF, particularly in the near-field region where the flame undergoes stabilisation, as has been observed previously for similar flames stabilised via autoignition [4]. There is, however, the potential for a nonnegligible contribution to the detected signal from other sources which warrants careful discussion. One particular source of interference is from polyaromatic hydrocarbons (PAHs), which are key precursors to the formation of soot, and are therefore expected to contribute to the signal in the furthest-downstream locations where soot begins to form in the n-heptane flames. The presence of droplets can also lead to interferences due to both elastic and inelastic scattering. The elastically scattered 355 nm light is expected to be effectively attenuated by the bandpass filter, which has an optical density of approximately 7.5 at 355 nm, such that the intensity of light transmitted at this wavelength is reduced by a factor of greater than 1×10^7 . In terms of inelastic Raman scattering from the fuel, the wavelength of the scattered light can be estimated from the Raman shift corresponding to the CH bonds, for both ethanol and *n*-heptane [39, 40]. Based on a shift value of 2900 cm⁻¹, the wavelength of the first-order Raman-scattered light is approximately 396 nm. Despite the narrow range of the bandpass filter, the optical density at this wavelength is slightly lower with a value of 3.3, such that the intensity is reduced by a factor of around 2000. While this means that this interference is significantly reduced by the filter, a low-level signal from fuel Raman can be expected. To account for these interferences, the results presented herein refer to the signal resulting from excitation at 355 nm as "UV-PLIF", and the source of the signal is discussed where appropriate. The UV-PLIF signal was detected using a separate ICCD camera with a 100 ns gate width, fitted with an f/1.2 lens.

Imaging of fuel droplets was performed via the Mie scattering technique, using the frequency-doubled 532 nm output of an Nd:YAG laser. The measured energy of the laser was 0.6 mJ/pulse, and the scattered light from droplets was collected on a CCD camera, operated with a gate width of 500 ns. The camera was fitted with an f/5.6 lens and a bandpass filter with a FWHM of 10 nm centred at 532 nm, allowing > 85% transmission at the target wavelength. Additional processing of the Mie scattering images was performed in order to extract the intermittency data relating to the droplet distributions. This involved converting the Mie scattering signal to a binarised image, representing the presence or absence of a droplet at any given location in the detection range. To generate the mean radial droplet distributions, this binarised signal was averaged over a row of 30 pixels centred at each axial location, and a full set of images was then averaged for each case and axial location, yielding the mean probability of droplets being detected at each location. Statistical convergence studies were performed for the radially-integrated droplet probabilities, with results indicating an uncertainty ranging from 1–3% for x/D < 3.0, and less than 5% for any given measurement. The mean data for all other results presented were found to have uncertainties lower than that of the droplet probabilities.

The timing of the laser pulses and camera gates were controlled using a combination of delay/pulse generators, to ensure that each camera detected the signal associated with a single laser only. The OH-PLIF laser pulse occurred first in the sequence; this was followed by the UV-PLIF pulse with a separation of 100 ns, with the Mie scattering pulse delayed by a further 100 ns, such that all laser pulses occurred within 200 ns of each other. The triggering of the camera shutters was synchronised with the corresponding laser pulse for each camera, such that the beginning of each detection in the sequence was also separated by approximately 100 ns from the preceding detection, noting that the Mie scattering camera gate width was 500 ns. It is worth noting that these timescales (that is, of the order of 500 ns) are much shorter than the flow and chemical timescales within the flames, such that the detections can effectively be considered simultaneous [41, 42]. Dark charge, vignetting, and background signal corrections were performed for all imaging techniques, and a beam-profile

correction was also carried out for the OH-PLIF signals. Images were spatially matched to sub-pixel accuracy, with each pixel representing $\sim 130 \,\mu\text{m}$, while the out-of-plane resolution has been estimated to be approximately 400 μm . A 3×3 median filter was applied to the OH- and UV-PLIF images to improve the signal-to-noise ratio, while the Mie scattering images were left unfiltered.

Photographs of the flames were captured using a DSLR camera, with exposure times ranging from 30 s to 250 μ s, although long exposures were not achievable for certain cases due to saturation. The photographs presented in this paper were all captured with an f-number of 16 and an ISO value of 100, with manual focus and white balance.

3. Results and discussion

3.1. Visual observations

Photographs of the six flames in this study are shown in Figure 1. The photographs show the visual appearance of the entire length of these flames, as well as close-up images to highlight the features at the base of the flames. The exposure times used for the photographs are also shown in Figure 1. It is worth noting that the difference in exposures between the *n*-heptane and ethanol flames is a consequence of the increased luminosity of the *n*-heptane flames due to their increased soot loading. Also shown in Figure 1 is the region in which laser diagnostics were performed, from the jet exit plane to x/D = 5.6.

In Figure 1(a), the change in coflow conditions does not appear to have a significant impact on the overall appearance of the flames. For the n-heptane flames, there is little difference in flame luminosity with variations in O_2 concentration or coflow temperature, while there appears to be a slight increase in luminosity as the concentration of O_2 is increased for the ethanol flames. The n-heptane flames all appear to feature a "double flame" structure, with a yellow sooting region which branches away from the blue inner cone in the near-field. This structure has previously been observed for similar flames under a variety of jet boundary conditions [34], and has been attributed to premixing of prevaporised fuel with the carrier air upstream of the jet exit (as described in Section 2.1), along with the radial transport of droplets into the hot coflow. In a previous study involving both air and nitrogen as carriers [33], double reaction zones were only observed for cases with an air carrier, further validating this assumption. The ethanol flames also appear to feature an inner and outer reaction zone; this is most apparent in the ETH-11 case, although it is more difficult to distinguish in comparison to the *n*-heptane cases due to the reduction in soot leading to a less luminous outer flame.

It is worth noting that the influence of the hot coflow extends approximately 100 mm downstream from the jet exit plane $(x/D \approx 5)$, after which the surrounding air begins to mix with the jet and coflow and influences the appearance of the flames. With this in mind, it can be seen that for the *n*-heptane flames, soot forms within the hot coflow-controlled region $(x/D \leq 5)$, which is in contrast to previous studies involving a JHC configuration [18, 22, 24].



(a) Full photographs of flames, with box indicating region in which laser diagnostics were performed.





Figure 1: Photographs of flames captured with a DSLR camera, with exposure times as shown.

This difference is attributed to the presence of carrier air in the jet for these flames, which results in an increase in O_2 concentration as this air mixes with the coflow, leading to an earlier onset of the sooting region than would otherwise be expected for a purely nonpremixed flame in a hot and low-oxygen coflow. It should be mentioned that in one of the aforementioned studies (namely [18]), flame cases with air in the central jet were in fact included, using ethylene as the fuel. Again, soot was not observed for these flames in the coflow-controlled region, although an earlier onset of soot in comparison to the case with nitrogen instead of air was observed. Prevaporised *n*-heptane flames carried by air have also been studied [24], where it was again observed that no soot was present in the coflow-controlled region. A key difference, however, is the fact that the fuel-air mixture in the jet was purely gaseous in these studies [18, 24], whereas the presence of droplets leads to a change in behaviour for the *n*-heptane flames in the present study. This effect was also observed for toluene-doped H₂ flames, where the use of toluene in liquid form was found to lead to increased soot formation in comparison to prevaporised toluene, due to the presence of local fuel-rich regions around individual droplets [43].

An important observation from the photographs in Figure 1(b) is the apparent change in lift-off behaviour under the different coflow conditions. The

HEP-03 and ETH-03 flames—which correspond to the 3% O₂ coflow—appear to be lifted, while the flames with 7.5% and 11% O₂ in the coflow can be seen to be attached. It is worth noting that previous findings related to prevaporised ethanol flames show a non-monotonic trend in lift-off height as the O₂ in the coflow is varied, with an initial increase in lift-off as the level of O₂ is increased [24]. This behaviour is thought to be related to a shift in the location of the most reactive mixture fraction in relation to the shear layer, which plays an important role in flame stabilisation in the mild regime [20]. While it is difficult to draw conclusions from the two ethanol flames included here, it appears that an increase in O₂ causes the flame to stabilise nearer to the jet, suggesting a different stabilisation process due to the presence of droplets.

In addition to the change in lift-off behaviour, the close-up images indicate that the onset of the luminous, sooting region occurs closer to the jet exit for the cases with increased O_2 in the coflow. This is particularly evident for the ethanol flames, where a transition to an orange flame can be seen in the ETH-11 close-up image, while this transition occurs further downstream for the ETH-03 case. It is also worth noting the differences in the intensity of the blue region near the flame base in the different cases. For the 3% O₂ cases, this region is relatively faint compared with the higher O_2 cases, with a less clearly defined structure. This result is interesting, since the blue region of the flame is thought to correspond to combustion of the prevaporised fuel [33, 34, 44], which mixes with the carrier air upstream of the jet exit to form a premixed stream and appears to be stabilised by autoignition at the flame base. However, if this were the case, then the temperature of the coflow would be expected to be the dominant factor in the appearance and lift-off behaviour of this blue region in the near-field, rather than O₂ concentration. Comparing the HEP-08 and HEP-H08 flames, although there is a slight increase in intensity for the case with higher coflow temperature (HEP-H08), it does not seem to have a significant impact on the flame structure in the near-field. This behaviour is further explored in the context of the laser diagnostics results in the following sections.

3.2. Instantaneous OH-PLIF, UV-PLIF and Mie scattering signals

To allow the structure and stabilisation of the flames to be examined, simultaneous OH-PLIF, UV-PLIF and Mie scattering signals were obtained. This facilitates the instantaneous visualisation of the reaction zone boundaries, along with the formation of flame precursor species and the location of fuel droplets. Figure 2 displays typical single-shot images of these signals for the HEP-08 case at an axial location of x/D = 1.5, in addition to a superimposed image showing the spatially matched signals together.

Focussing on the OH signal in Figure 2, the formation of a double flame structure can be seen, with inner and outer reaction zones evident on either side of the centreline. The inner flame front appears to be less stable, with discontinuities in the OH signal which suggests the presence of local extinction and/or ignition events—this is particularly evident on the left-hand side of Figure 2(a). It should be noted that it is unlikely that these discontinuities are a result of out-of-plane wrinkling of the flame, due to the continuous nature of the UV-PLIF signal which lies inside of the inner OH layer, and the fact that



Figure 2: Typical instantaneous signals of (a) OH-PLIF, (b) UV-PLIF and (c) Mie scattering, and (d) the three signals superimposed. Images correspond to flame case HEP-08 and are centred at x/D = 1.5.

these flames are dominated by streaming flow in the axial direction. It is also worth noting that there is background OH signal resulting from the equilibrium OH in the coflow, although this cannot be observed in Figure 2 due to the much higher signal from the flame fronts. This "coflow OH" concentration is higher in regions closer to the jet exit, with an estimated mole fraction of 3×10^{-5} for the 1400 K coflows and 3×10^{-4} for the 1690 K coflow, based on equilibrium calculations.

Another interesting feature, apparent in Figure 2, is the overlap between the OH and UV signals, particularly in locations where there are discontinuities in the inner OH layer. This supports the notion that the isolated regions of OH represent ignition kernels, with the UV signal corresponding to the build-up of pre-ignition species. The broader regions of UV-PLIF which can be seen between the inner and outer flame fronts are also worth noting; these are associated with the formation of soot precursors in the outer diffusion flame, hence the very strong signal on the right-hand side of Figure 2(b). There is also evidence of some interference in the UV-PLIF signal from the presence of fuel droplets in Figure 2(b). This is represented by the distinct "spots" in the UV-PLIF image, which appear to share some spatial overlap with the Mie scattering signal in the superimposed image. This is hypothesised to be a result of Raman scattering from fuel droplets, which, as discussed in Section 2.2, is expected to produce some low-level interference in the UV-PLIF signal.

To examine the behaviour of the double flame structure in greater detail, it is useful to analyse separately the OH signal corresponding to the inner and outer reaction zones. This in turn enables quantification of the statistical features relating to the flame structures, and facilitates comparisons between different cases. This analysis involves the classification of the instantaneous OH signals into four separate categories, as shown in Figure 3. Table 2 presents the statistical data relating to the occurrence of these structures for the different flame cases at various axial locations, in terms of the frequency of detection over a series of images.



Figure 3: OH-PLIF images displaying four different types of behaviour; (A) continuous and distinct inner and outer structures, (B) merged/bifurcating inner and outer structures, (C) unsteady inner structure/ignition kernels, and (D) outer OH structure only. Images correspond to flame case HEP-08 and are centred at x/D = 1.5.

Table 2: Frequency of detection of different OH structures (as shown in Figure 3) for the different flame cases at x/D = 1.5.

Case	Type A $[\%]$	Type B $[\%]$	Type C $[\%]$	Type D $[\%]$
HEP-03	11	18	8	63
HEP-08	19	8	29	45
HEP-11	37	2	28	32
HEP-H08	45	1	31	23
ETH-03	17	81	2	0
ETH-11	37	63	0	0

From Table 2, it is evident that the coflow conditions have an impact on the formation of the different OH structures shown in Figure 3. For the 3% O₂ *n*-heptane case (HEP-03), the majority of frames displayed an outer OH layer only, with a relatively large number of instances in which there was a "merged" structure (i.e. Type B). The prevalence of a distinct and stabilised inner OH layer (Type A) can be seen to increase with O_2 concentration and coflow temperature, accompanied by a decreased likelihood of merged flame structures. The presence of ignition kernels—characterised by the Type C structures—can be seen to increase significantly from the 3% to 7.5% O₂ *n*-heptane case, while a further increase in O_2 does not have a noticeable effect. The ethanol flames can be seen to display a significantly different behaviour, which is characterised by an increased tendency of a merged/bifurcating flame structure (i.e. Type B), and a negligible percentage of frames which display either ignition kernels or an outer OH structure only. Similar to the *n*-heptane flames, the increase in O_2 leads to an increased likelihood of a distinct inner and outer reaction zone. The frequency of the occurrence of these different structures is further discussed in the following sections in the context of the stabilisation mechanisms and mean flame structures.

3.3. Flame stabilisation

To illustrate the stabilisation mechanisms of the flames in this study, Figure 4 displays the OH, UV and Mie scattering signals at the flame base for the various cases. The superimposed images are centred at x/D = 0.35, with the laser sheet extending from the jet exit to x/D = 0.7. Images were selected based on being representative of typical images for each case.



Figure 4: Instantaneous, superimposed images of OH-PLIF, CH₂O-PLIF and Mie scattering, for the six different flames at x/D = 0.35.

In Figure 4, the HEP-03 case can be seen to have much lower OH-PLIF signal in comparison to the other cases. This is consistent with the flame photographs shown in Figure 1, where the HEP-03 flame appears lifted from the jet exit. In saying this, the presence of UV-PLIF signal in Figure 4 suggests that there are in fact reactions taking place in the visually lifted region for the HEP-03 flame. It should be noted that there is also OH-PLIF signal in the HEP-03 image in Figure 4, although it is difficult to distinguish from the coflow equilibrium OH. Interestingly, the equivalent ethanol flame (ETH-03)—which is also visually lifted—has quite a strong OH signal at the jet exit. It has previously been observed that *n*-heptane has a greater tendency for lift-off in comparison to ethanol [24], which would explain the difference in the OH signals between the two fuels in Figure 4.

With the exception of the HEP-03 flame, Figure 4 indicates that the flames are stabilised at the jet exit. It is interesting to note that, for the n-heptane

flames and the 11% O_2 ethanol flame, the OH structure which is stabilised at the jet exit appears to correspond to the "outer" flame; this is substantiated by the fact that the continuous OH layer can be seen to spread radially outward in the images. Referring back to the flame photographs (Figure 1), the inner blue cone is always angled towards the centreline, and the outer structure is not visible until further downstream. The OH-PLIF imaging therefore reveals that the flames are actually stabilised by the outer reaction zone, which is barely visible in the photographs even in the cases with 11% O_2 , at least with the exposure times shown in Figure 1. The fuel within the inner region then undergoes ignition, supported by the heat release and radical build-up from the outer flame.

In the ETH-11 image in Figure 4, the bifurcation of the flame into an inner and outer reaction zone can be seen. The CH_2O layer lies within the inner region, which is consistent with this inner flame front being related to mixing of the prevaporised fuel and the carrier air, as observed in previous studies of similar spray flames [33, 34]. In contrast, the *n*-heptane flames only feature a single OH structure on either side of the centreline. However, the CH_2O layer can be seen to branch away from the OH layer at the flame base (for all *n*heptane cases except for HEP-03, as previously discussed), which indicates that the prevaporised fuel within the inner region has begun to thermally decompose. This fuel in the inner region then undergoes ignition further downstream; this can be seen for the HEP-08 case in Figure 2.

To further explore the formation and stabilisation of the double flame structure for the various cases, Figure 5 displays the averaged OH-PLIF signal, from the jet exit to x/D = 1.85 for the *n*-heptane cases with varying O₂ concentrations (averaged images for the ethanol flames are included in the Supplementary Material). These images highlight the change in flame structure for the $3\% O_2$ coflow case (HEP-03) in comparison to the other two n-heptane flames at the same coflow temperature. The HEP-08 and HEP-11 cases appear very similar, with a consistent outer flame front stabilised at the jet exit, and a less prominent inner flame which begins to branch away at approximately x/D = 1. The HEP-03 case, in contrast, does not show a stabilised outer flame until approximately x/D = 1.15, which is consistent with the apparent liftoff height (Figure 1(b)). Interestingly, there is in fact OH signal upstream of this location, albeit at a much lower magnitude relative to the HEP-08 and HEP-11 cases. This low-magnitude signal, which can be seen from approximately x/D = 0.4in Figure 5, does not appear to spread radially outward in the same way as the outer flame front for the cases with more O_2 in the coflow. This difference can be explained by changes in droplet evaporation as the concentration of O_2 is varied, which is explored in Section 3.4. For the HEP-08 and HEP-11 cases, droplets which are transported radially into the hot coflow begin to evaporate and burn rapidly, forming radicals and releasing heat, such that the increased temperature causes more evaporation and leads to a stabilised diffusion flame at the jet exit. For the 3% O₂ case, the droplets still evaporate at a similar rate initially; however, the low level of O_2 leads to slower chemical timescales and a reduced tendency for droplets to react near the jet. As a result, the flame cor-



Figure 5: Averaged OH images for the HEP-03, HEP-08 and HEP-11 cases, from the jet exit to x/D = 1.85.

responding to the prevaporised fuel in the inner region is stabilised prior to the formation of an outer diffusion flame for the HEP-03 case. For the equivalent ethanol case (ETH-03), a weakened outer flame front is again apparent, with a relatively strong signal from the inner flame near the jet exit. This emphasises the importance of the carrier air on the stabilisation of these flames, particularly under highly vitiated conditions.

Referring again to the prevaporised flames of Ye et al. [24], some interesting comparisons can be made regarding the flame structure. One of the key findings of the aforementioned study [24] was the observation that the *n*-heptane flames maintained a "transitional" flame structure as the O₂ concentration was varied from 9% to 3%, characterised by the presence of a weak OH signal below the apparent lift-off height. This was in contrast to the ethanol flames (along with ethylene and natural gas), for which this structure was only observed for the $9\% O_2$ cases. This transitional structure is considered to be indicative of a shift away from the mild regime [24, 45], suggesting a greater difficulty in achieving mild combustion for *n*-heptane. In the present study, the 3% O₂ cases show similarities with the corresponding prevaporised cases [24], with the HEP-03 flame displaying a very weak OH signal upstream of the apparent lift-off height, while the ETH-03 case has a relatively strong signal (Figure 4). As the concentration of O_2 in the coflow is increased, however, a change in behaviour is observed due to the presence of droplets, resulting in the flames being stabilised by the outer reaction zone at the jet exit.

3.4. Droplet evaporation and distribution

It has previously been observed that the behaviour of droplets in the nearfield can have a significant impact on the overall structure of a spray flame [34]. It is therefore crucial to predict these features in order to enable accurate modelling of these flames. To analyse the distribution and evaporation of droplets under the different coflow conditions, Figure 6 displays the radial profiles of droplet intermittency for the various cases, with results corresponding to



Figure 6: Radial distribution of droplets at a range of axial locations, for all flame cases.

different axial locations grouped together. These plots are generated based on the probability of droplet detection from the Mie scattering signal, as outlined further in Section 2.2.

In Figure 6, the droplet distributions at the four axial locations can be seen to follow a similar overall behaviour with variation in the coflow conditions. This is to be expected, since all cases had the same initial fuel loading and jet Reynolds number. A noticeable feature for all cases is the tendency for droplets to cluster near the pipe walls at the jet exit, as shown by the peaks at |r/D| = 0.5. This has been observed previously for similar conditions, and is related to the low-Stokes flow phenomena of Saffman lift and turbophoresis [46]. These droplets are immediately exposed to the hot coflow after exiting the jet, and proceed to evaporate relatively quickly, as evidenced by the rapid decrease in droplets at this radial location. An interesting difference in the HEP-03 plot in comparison to the other n-heptane cases is the fact that the peak at approximately |r/D| = 0.5 remains evident at x/D = 1.5 (red dotted line), while the profiles have flattened out at this location for the other cases. This supports the explanation from Section 3.3 regarding the change in the flame stabilisation mechanism for the HEP-03 case, in which it was theorised that the slower chemistry and reduced temperatures in the case of lower oxygen leads to a reduction in droplet evaporation and combustion in the near-field, which leads to the premixed portion of the fuel undergoing ignition prior to the formation of the outer flame structure. This difference is also evident for the ethanol cases, where an increase in droplets in general can be observed for the ETH-03 case.

An interesting difference between the *n*-heptane and the ethanol droplet distributions can be observed for the downstream plots, in particular at x/D = 4.5. In all of the *n*-heptane cases, there is a sharp reduction in the presence of droplets from x/D = 3.0 to x/D = 4.5, whereas droplets near the central axis are shown to persist further downstream in the case of ethanol. In fact, for the ethanol cases, the likelihood of droplets being detected along the centreline does not appear to decrease at all with increasing axial location; this is also the case for the *n*-heptane profiles at x/D = 0.35 and x/D = 1.5, where there even appears to be a slight increase in probability. It should be mentioned that this does not mean that there is no evaporation occurring along the central axis; rather, it suggests that the majority of the droplets have not yet undergone complete evaporation by x/D = 4.5 for the ethanol cases, whereas the droplets evaporate more rapidly from x/D = 1.5 to x/D = 4.5 in the case of *n*-heptane. This is further discussed in the context of the flame structure in the following section.

3.5. Mean flame structure: radial profiles

To investigate the effect of the coflow O_2 concentration on the near-field flame structure, Figure 7 displays the mean radial profiles of OH- and UV-PLIF up to x/D = 1.5, for the 3% and 11% O_2 ethanol and *n*-heptane cases. The radial signals are generated via the averaging of 15 rows of pixels, centred at the heights stated in the figures. The OH and UV signals are respectively normalised against the maximum average OH and UV signal intensity from each "pair" of cases, at all axial locations up to x/D = 1.5. A single normalising value is therefore used for each of the *n*-heptane cases shown in Figure 7, while a separate value is used for the ethanol cases. In addition to time averaging, the signals on each side of the centreline were also averaged.

A noticeable change in the near-field flame structure with variation in the coflow O_2 concentration is apparent in Figure 7. For both the *n*-heptane and ethanol cases, distinct peaks in the OH signal can be seen at x/D = 1.5 for the 11% O₂ cases, while only a single peak is present for the 3% cases. This confirms that there are distinct, stabilised inner and outer reaction zones for the HEP-11 and ETH-11 cases at this location, while this double reaction zone is less prominent for the 3% O₂ cases. Previously, temperature measurements of ethanol spray flames in air and hot-diluted coflows have been attained [29], with distinct local maxima in the radial profiles observed for the case of an air coflow, suggesting the presence of separate inner and outer reaction zones. For the hot and diluted coflow cases in the same study, the results suggested that a double reaction zone still exists, but with a reduced-intensity inner flame front due to the reduced stoichiometric mixture fraction and hence richer local mixture [29]. In the plots shown in Figure 7, it is interesting to note the location of the peaks in the OH profiles for the ETH-03 case. At each of the three axial locations shown, the peak occurs at approximately |r/D| = 0.5. This peak corresponds



Figure 7: Mean radial profiles of OH- and UV-PLIF for cases HEP-11, HEP-03, ETH-11 and ETH-03, from x/D = 0.35 to x/D = 1.5. Signals are normalised against the maximum OH- or UV-PLIF intensity amongst cases of the same fuel type.

to the inner, partially premixed flame front, since the radial location coincides with the inner peaks in the ETH-11 case, and a slight irregularity in the OH profile can be seen at greater radial locations (i.e. at $r/D \approx 0.9$ for x/D = 0.75and x/D = 1.5), representing the outer flame front. This indicates that the inner flame is dominant in the near-field for the ETH-03 case, contrasting with the results of Rodrigues et al. [29]. It should, however, be noted that the results being compared against were obtained with a pressure-swirl atomiser to generate the spray without an air carrier, such that there is not expected to be any partial premixing between fuel and oxidant. This again emphasises the role of partial premixing on the formation of the inner flame front—and in turn the double flame structure—with the burner configuration implemented in the current investigation.

Another important observation from Figure 7 is the difference between the HEP-03 and ETH-03 cases. Recalling from Figure 1 that both of these flames are visually lifted to a height of approximately x/D = 1, the radial OH- and UV-PLIF profiles centred at x/D = 0.35 show that there is in fact a flame below this apparent lift-off height for both cases. This is much more noticeable for the ETH-03 case, which displays an appreciable signal above that of the coflow OH, and of comparable magnitude to the ETH-11 case. The HEP-03 case can also be seen to show a peak in the OH, although this signal is only marginally above that which corresponds to the coflow equilibrium OH, and much lower than the 11% O₂ case. The HEP-03 OH signal is also relatively low at x/D = 0.75, after which it can be seen to increase significantly, which is consistent with the apparent lift-off height. It is interesting to note that at x/D = 0.75, the location of the OH peak is closer to the centreline for the HEP-03 case compared with the HEP-11 case, indicating that the outer flame is stabilised further downstream for the 3% case; this is also evident in Figure 5. At x/D = 1.5, the HEP-03 peak can be seen to shift radially outwards to $r/D \approx 0.8$, suggesting that the outer flame is stabilised at this point. At the same axial location, the HEP-11 case shows two distinct peaks in the OH profile, with the outer peak occurring at $r/D \approx 0.9$ and the inner peak at $r/D \approx 0.4$. Similar to the ETH-03 case, the HEP-03 profile also displays an asymmetric shape in the OH profile at x/D = 1.5. A key difference, however, is that the ETH-03 case displays a dominant "inner peak" with a broadening of the curve at greater radial distances, whereas the opposite is true for the HEP-03 case. This is consistent with the results displayed in Table 2, which shows that the merged or bifurcating OH structure is the most prevalent for ETH-03 flame (occurring in 81% of frames), while the HEP-03 flame is most likely to feature an outer reaction zone only, with a smaller percentage of frames in which an inner flame is detected. Importantly, the shift in the location of the peak at x/D = 1.5 for the HEP-03 case, and the broadening of the profile in the ETH-03 case at the same location, indicates that the visually lifted nature of the $3\% O_2$ flames can be attributed to the lack of a stabilised outer flame front upstream of this location.

Similarly to Figure 7, radial profiles of the mean OH and UV signals for cases HEP-H08 and HEP-08 are shown in Figure 8, highlighting the effect of the

coflow temperature on the near-field flame structure. The coflow temperature can be seen to have a relatively minor effect on the flame structure compared with the O_2 concentration, with the HEP-08 and HEP-H08 profiles showing a similar behaviour. For both cases, the UV signal can be seen to peak radially inward with respect to the OH layer at x/D = 0.35, suggesting pre-ignition reactions of the prevaporised fuel, as discussed in Section 3.3. Once the inner flame is established further downstream (i.e. at x/D = 1.5), the radial location of the peak in the UV can be seen to coincide with the inner peak in the OH signal, again highlighting the strong overlap between OH and CH₂O during the autoignition stage. It is worth noting that this behaviour is also evident for the HEP-11 flame (Figure 7), while the UV signal peak at x/D = 1.5 for the ETH-11 case occurs closer to the centreline in relation to the inner OH peak. This difference can be attributed to the change in the stabilisation of the inner flame front between the *n*-heptane and ethanol flames. Rather than undergoing spontaneous ignition, the inner flame tends to branch away from the outer flame front near the jet exit for the ETH-11 case, forming a bifurcating structure (see Figure 4). Consequently, the inner OH structure is more stable for the ethanol flame, and there is less overlap between the OH and CH_2O in this region.

Although the radial profiles are very similar for the HEP-H08 and HEP-08 cases, a difference in the OH structure can be seen at x/D = 0.75 in Figure 8. For both cases, the outer OH structure is dominant, with the peaks occurring at a similar radial location (approximately |r/D| = 0.7). For the higher temperature case (HEP-H08), a slight peak in the OH profile can be observed at |r/D| = 0.5, indicating that the inner flame is at least partially stabilised at this axial location. The HEP-08 case, on the other hand, only features a single peak in the OH profile, indicating that the inner flame is stabilised further downstream. This behaviour is to be expected, since the lower coflow temperature leads to a delayed ignition of the prevaporised fuel mixture. It should be noted that, due to the averaging process and the turbulent nature of the flames, the occurrence of a single peak in the OH profile does not necessarily dictate that there is no inner flame front at x/D = 0.75 for the HEP-08 case; rather, it shows that there is a less prominent double flame structure, with the outer flame dominating the OH profile.

To show the structure of the flames further downstream, Figure 9 displays the OH profiles for all cases, from x/D = 3.0 to x/D = 5.3. The plots are grouped in pairs, to show separately the effects of coflow oxygen concentration (for both fuel types) and temperature (for *n*-heptane). It is interesting to note that, despite the clear difference in near-field flame structure between the HEP-03 and HEP-11 cases as seen in Figure 7, the two appear to exhibit similar overall structures for $x/D \ge 3.0$, with distinct inner and outer reaction zones present for both cases. In saying this, there are some important differences to note between the two cases. While it is not possible to directly compare the signal magnitudes between cases due to varying quenching effects, it is possible to compare the relative magnitudes between the inner and outer peaks at different axial locations. With this in mind, it can be seen at x/D = 3.0 that the inner and outer OH peaks are approximately equal in magnitude for the 11% O₂ case,



Figure 8: Mean radial profiles of OH- and UV-PLIF for cases HEP-H08 and HEP-08, from x/D = 0.35 to x/D = 1.5. Signals are normalised against the maximum OH- or UV-PLIF intensity from both cases.

whereas the inner OH peak is greater for the 3% O₂ case. This indicates that the outer reaction zone is significantly less intense for the HEP-03 flame, consistent with previous findings for gaseous flames under similar conditions [14]. For both cases, the centreline OH signal can be seen to peak at x/D = 4.5, representing the tip of the inner flame. This is consistent with the droplet distribution plots shown in Figure 6, which showed a sharp reduction in the presence of droplets at x/D = 4.5, indicating that there is rapid evaporation of droplets in this region due to the influence of the inner flame. Although both cases reach their peak centreline OH magnitude at x/D = 4.5, there is a much more rapid reduction in OH from x/D = 4.5 to x/D = 5.3 for the HEP-11 case, highlighted by the reversal in the relative magnitudes of the OH peaks between these two axial locations which is not the case for the HEP-03 flame. This suggests that the tip of the inner flame occurs slightly further downstream for the HEP-03 flame, which is likely a result of the reduced thermal back-support from the weakened outer flame front.

Comparing the HEP-H08 and HEP-08 profiles in Figure 9, the ≈ 300 K difference in coflow temperature does not appear to have a major impact on the downstream flame structure. Although the OH signal magnitude is significantly higher for the higher temperature coflow (HEP-H08), as expected, the relative magnitudes between the inner and outer OH structures can be seen to be consistent between the two cases at each of the axial locations. Similarly to the 3% and 11% O₂ cases, the peak centreline OH occurs at approximately x/D = 4.5, with the outer flame dominating further downstream. In general, the flame structure at $x/D \geq 3.0$ is shown to be very consistent across all of the *n*-heptane flames in Figure 9. In a previous study of similar flames with varying jet boundary conditions and constant coflow conditions [34], the downstream behaviour—in particular the presence of a double flame structure was shown to be very sensitive to the initial conditions. This highlights the relative dominance of the jet boundary conditions and near-field structure.

For the ethanol cases shown in Figure 9, the OH profiles indicate a clear change in behaviour in comparison with the n-heptane flames. For both the ETH-11 and ETH-03 cases, the inner and outer flame fronts are noticeably less distinct, with the inner OH peak occurring at an increased radial distance and the outer peak closer to the centreline, relative to the n-heptane flames at the equivalent axial location. It is interesting to note that in the near fieldspecifically at x/D = 0.35 in Figure 7—the location of the OH peaks for the ETH-11 and HEP-11 cases is seen to coincide. This suggests that the difference observed in Figure 9 is not a result of a shift in the stoichiometric mixture fraction (as has been hypothesised previously [34]), since this difference would manifest itself at the point of stabilisation. It can instead be concluded that the change in shape of the OH profiles is a result of increased interaction between the two flame fronts in the case of ethanol, with thermal back-support and the transport of radicals leading to the two structures stabilising closer to one another. The absence of an OH peak at the centreline for the ethanol cases also suggests that the inner reaction zone persists further downstream in comparison



Figure 9: Mean radial profiles of OH-PLIF from x/D = 3.0 to x/D = 5.3. Plots are grouped to show the effect of coflow oxygen concentration and temperature separately.

to the *n*-heptane cases. Referring again to the droplet distributions shown in Figure 6, the lack of centreline decay of droplets in the case of ethanol can be linked with the inner flame front occurring at a greater radial location.

Similarly to the *n*-heptane flames, the reduction in the coflow O_2 concentration can again be seen to cause a weakening of the outer reaction zone in terms of the relative peak signal magnitudes for the ethanol flames. This is particularly evident at x/D = 3.0 in Figure 9, where the inner flame front dominates the OH profile and only a single peak is observed for the ETH-03 case. This indicates that the change in O_2 concentration has an increased effect for the ethanol flames compared with *n*-heptane, in terms of the formation of the double flame structure. Once again, it should be stressed that the lack of a separate outer peak at x/D = 3.0 for the ETH-03 case does not disprove the presence of separate inner and outer reaction zones for this case, rather that these two structures are less distinct, such that the two structures become "blurred" in the mean profile.

It is evident from the profiles shown in Figures 7 to 9 that the averaging process leads to a "blurring" effect, such that the variation in the position of the reaction zone due to turbulent fluctuations leads to the inner and outer structures being difficult to distinguish in some instances. To examine the flame structure for the different cases in a more quantitative sense, it is therefore useful to perform calculations on the instantaneous images prior to averaging. As mentioned in the Methodology, an algorithm was developed to separate the inner and outer OH structures in the instantaneous images. Using this method, it is then possible to calculate the width of the outer reaction zone for the different cases. For this analysis, the full-width at half maximum (FWHM) value of the radial OH signal was used to provide a measure of the reaction zone thickness, similar to previous studies [37, 47]. For further details regarding this process the reader is directed to the Supplementary Material, where an example of the separation between the inner and outer structures and the subsequent analysis are provided. The FWHM results are plotted against the axial location in Figure 10, for the *n*-heptane flames from the jet exit to x/D = 5.3. For the HEP-03 case, the results are only included for x/D > 1.5, since the lack of a stabilised outer flame upstream of this point prevents a meaningful analysis of the flame width.

The FWHM values displayed in Figure 10 highlight some key differences between the flame cases, which are not evident from the mean profiles alone. From the mean FWHM plots, it can be seen that a reduction in O_2 concentration leads to a narrowing of the outer reaction zone, with the HEP-03 case in particular having a noticeably narrower reaction zone in comparison to the higher O_2 cases for $x/D \ge 3$. This is an interesting result, since previous studies of mild combustion have found that the reaction zone tends to become wider as the O_2 concentration is reduced, which is attributed to a shift in the stoichiometric mixture fraction towards the oxidant side resulting in lower strain rates [37]. The change in behaviour observed in the current study is likely due to the presence of the inner reaction zone, which influences the outer flame front differently in the various cases. Recalling from Figure 7 and the accompanying



Figure 10: Mean and RMS values of the FWHM reaction zone thickness for the *n*-heptane cases, from x/D = 0.35 to x/D = 5.3.

discussion that the HEP-03 case only displays a single peak in the mean OH profile at x/D = 1.5 while the HEP-11 case features distinct peaks, it follows that the double flame structure is less prominent for the 3% case, with increased interaction between the two reaction zones in the HEP-03 case; this is substantiated by the increased likelihood of the merged flame structures being detected for this case (Table 2). Somewhat paradoxically, this merging of the inner and outer structures leads to a broadening of the reaction zone when it occurs, hence the much higher RMS values for the HEP-03 case at x/D = 1.5. However, it should be noted that this behaviour only accounts for approximately 18% of the images analysed, while the majority show only a stabilised outer flame. It is therefore hypothesised that the presence of a stabilised and distinct inner reaction zone corresponds with a broadening of the outer flame front, whereas the lack of a stabilised inner flame for the HEP-03 case results in a build-up of radicals within the inner region which supports the outer reaction zone, ultimately leading to a narrowing of the OH structures. This behaviour is also consistent with the shift in the OH peak towards the centreline for the HEP-03 radial profiles at x/D = 1.5 in Figure 7 and at x/D = 3.0 in Figure 9. This also explains the increased width for the HEP-H08 case, for which the inner flame was shown to have greater stability and separation from the outer structure in Table 2 and Figure 8.

The change in the reaction zone width with increasing distance from the jet exit is also interesting to note. For the cases with > 3% O₂, there is an initial increase in the FWHM values from the jet exit to x/D = 1.5, followed by a narrowing of the reaction zone up to x/D = 4.5, after which the width appears to increase again. The broadening observed in the near-field region is likely a result of the radially outward shift of the outer reaction zone (shown in Figures 7 and 8), which is accompanied by reduced strain rates. Interestingly, the reaction zone continues to move radially outwards up until $x/D \approx 3.0$, yet there is a sharp reduction in the reaction zone width from x/D = 1.5 to x/D = 3.0 for all cases. This is consistent with the explanation that the presence of the inner reaction further downstream leading to a narrower outer structure. As mentioned, the tip of the inner flame is reached at approximately x/D = 4.5 for the *n*-heptane cases, which is consistent with the increase in the outer reaction zone width downstream of this location.

4. Conclusions

The stabilisation features and flame structures of dilute sprays of ethanol and *n*-heptane have been experimentally analysed under a range of coflow conditions. Three laser diagnostic techniques were performed simultaneously, enabling the combined visualisation and analyses of the reaction zones and droplet distributions. This work highlights the complex nature of the reaction zones and stabilisation processes for spray combustion, specifically under highly vitiated coflow conditions which are typical of the mild combustion regime. A transition from a stabilised flame base at the jet exit to a visually lifted flame was observed with reduction in the coflow O_2 concentration to 3%, for both *n*-heptane and ethanol flames. The instantaneous imaging, however, revealed the occurrence of flame radicals and intermediate species in this apparently lifted region, indicating the presence of a relatively weak reaction zone extending to the jet exit for the 3% O_2 cases. Analysis of the radial droplet distributions revealed that there is a tendency for droplets to cluster near the pipe walls at the jet exit, after which they undergo rapid evaporation in the presence of the hot coflow. This leads to a stabilised diffusion flame at the jet exit for coflows with >3% O_2 concentrations, with the heat release in this region leading to increased evaporation rates in comparison to the reduced O_2 cases. Droplets were found to persist further downstream in the ethanol cases, which is attributed to the increased interaction between the inner and outer flame fronts which results in the inner flame propagating further radially from the central axis in comparison to the *n*-heptane flames.

The mean OH- and UV-PLIF results revealed interesting changes in the structures of the flames studied. The 3% O₂ cases were found to display a less prominent double flame structure, particularly in the near field, due to the weakened outer flame front which is stabilised further downstream. A broadening in the mean radial OH profile was observed near the apparent lift-off height, for both the HEP-03 and ETH-03 cases, suggesting that the lack of a stabilised "outer flame" at the jet exit is responsible for the visually lifted nature of the flames. It was also observed that there is an increased probability of merging between the inner and outer flame fronts for the HEP-03 case in comparison to the other n-heptane flames, leading to greater variability in the reaction zone width for this case in the near-field. Interestingly, and in apparent contrast to gaseous flames in similar configurations, the reduction in O_2 was found to lead to a narrowing of the mean outer reaction zone, and the cases with higher O_2 concentrations displayed a non-monotonic behaviour with respect to the axial location. This indicates that the presence of a stabilised inner reaction zone leads to a broadening of the outer reaction zone, which is in competition with the reduced strain rate at greater axial locations.

Acknowledgements

The authors thank Dr Jingjing Ye for her contribution in the design of the burner used for these experiments, and Dr Thomas Kirch for his assistance with data collection. The support from the University of Adelaide is acknowledged, along with the financial support provided by the Australian Research Council (ARC) and the Future Fuels Cooperative Research Centre (CRC): Project RP1.10–04.

References

 A. A. V. Perpignan, A. Gangoli Rao, D. J. E. M. Roekaerts, Flameless combustion and its potential towards gas turbines, Prog. Energy Combust. Sci. 69 (2018) 28–62.

- [2] A. E. E. Khalil, A. K. Gupta, Fuel flexible distributed combustion for efficient and clean gas turbine engines, Appl. Energy 109 (2013) 267–274.
- [3] G. Faeth, Mixing, transport and combustion in sprays, Prog. Energy Combust. Sci. 13 (4) (1987) 293–345.
- [4] A. R. Masri, Turbulent Combustion of Sprays: From Dilute to Dense, Combust. Sci. Technol. 188 (10) (2016) 1619–1639.
- [5] A. R. Masri, A. Kourmatzis, W. O'Loughlin, J. D. Gounder, From Dilute to Dense Turbulent Sprays: Combustion, Auto-Ignition and Atomization, in: B. Merci, E. Gutheil (Eds.), Experiments and Numerical Simulations of Turbulent Combustion of Diluted Sprays, ERCOFTAC Series, Springer International Publishing, Cham, 2014, pp. 1–29.
- [6] J. D. Gounder, A. Kourmatzis, A. R. Masri, Turbulent piloted dilute spray flames: Flow fields and droplet dynamics, Combust. Flame 159 (11) (2012) 3372–3397.
- [7] B. Merci, E. Gutheil (Eds.), Experiments and Numerical Simulations of Turbulent Combustion of Diluted Sprays, Vol. 19 of ERCOFTAC Series, Springer International Publishing, Cham, 2014.
- [8] P. Jenny, D. Roekaerts, N. Beishuizen, Modeling of turbulent dilute spray combustion, Prog. Energy Combust. Sci. 38 (6) (2012) 846–887.
- [9] S. Hochgreb, Mind the gap: Turbulent combustion model validation and future needs, Proc. Combust. Inst. 37 (2) (2019) 2091–2107.
- [10] M. Katsuki, T. Hasegawa, The science and technology of combustion in highly preheated air, Symp. (Int.) Combust. 27 (2) (1998) 3135–3146.
- [11] J. A. Wünning, J. G. Wünning, Flameless oxidation to reduce thermal no-formation, Prog. Energy Combust. Sci. 23 (1) (1997) 81–94.
- [12] M. de Joannon, G. Langella, F. Beretta, A. Cavaliere, C. Noviello, Mild Combustion: Process Features and Technological Constrains, Combust. Sci. Technol. 153 (1) (2000) 33–50.
- [13] A. Cavaliere, M. de Joannon, Mild Combustion, Prog. Energy Combust. Sci. 30 (4) (2004) 329–366.
- [14] B. B. Dally, A. N. Karpetis, R. S. Barlow, Structure of turbulent nonpremixed jet flames in a diluted hot coflow, Proc. Combust. Inst. 29 (1) (2002) 1147–1154.
- [15] R. Cabra, T. Myhrvold, J. Chen, R. Dibble, A. Karpetis, R. Barlow, Simultaneous laser raman-rayleigh-lif measurements and numerical modeling results of a lifted turbulent H2/N2 jet flame in a vitiated coflow, Proc. Combust. Inst. 29 (2) (2002) 1881–1888.

- [16] J. Sidey, E. Mastorakos, Visualization of MILD combustion from jets in cross-flow, Proc. Combust. Inst. 35 (3) (2015) 3537–3545.
- [17] C. M. Arndt, M. J. Papageorge, F. Fuest, J. A. Sutton, W. Meier, M. Aigner, The role of temperature, mixture fraction, and scalar dissipation rate on transient methane injection and auto-ignition in a jet in hot coflow burner, Combust. Flame 167 (2016) 60–71.
- [18] P. R. Medwell, P. A. M. Kalt, B. B. Dally, Imaging of diluted turbulent ethylene flames stabilized on a Jet in Hot Coflow (JHC) burner, Combust. Flame 152 (1) (2008) 100–113.
- [19] E. Oldenhof, M. Tummers, E. van Veen, D. Roekaerts, Ignition kernel formation and lift-off behaviour of jet-in-hot-coflow flames, Combust. Flame 157 (6) (2010) 1167–1178.
- [20] M. J. Evans, A. Chinnici, P. R. Medwell, J. Ye, Ignition features of methane and ethylene fuel-blends in hot and diluted coflows, Fuel 203 (2017) 279– 289.
- [21] M. J. Evans, P. R. Medwell, H. Wu, A. Stagni, M. Ihme, Classification and lift-off height prediction of non-premixed MILD and autoignitive flames, Proc. Combust. Inst. 36 (3) (2017) 4297–4304.
- [22] M. J. Evans, P. R. Medwell, Z. Sun, A. Chinnici, J. Ye, Q. N. Chan, B. B. Dally, Downstream evolution of n-heptane/toluene flames in hot and vitiated coflows, Combust. Flame 202 (2019) 78–89.
- [23] J. Ye, P. R. Medwell, K. Kleinheinz, M. J. Evans, B. B. Dally, H. G. Pitsch, Structural differences of ethanol and DME jet flames in a hot diluted coflow, Combust. Flame 192 (2018) 473–494.
- [24] J. Ye, P. R. Medwell, M. J. Evans, B. B. Dally, Characteristics of turbulent n-heptane jet flames in a hot and diluted coflow, Combust. Flame 183 (2017) 330–342.
- [25] C. G. de Azevedo, J. C. de Andrade, F. de Souza Costa, Flameless compact combustion system for burning hydrous ethanol, Energy 89 (2015) 158–167.
- [26] G. B. Ariemma, P. Bozza, M. de Joannon, P. Sabia, G. Sorrentino, R. Ragucci, Alcohols as Energy Carriers in MILD Combustion, Energy Fuels 35 (9) (2021) 7253–7264.
- [27] S. Kruse, B. Kerschgens, L. Berger, E. Varea, H. Pitsch, Experimental and numerical study of MILD combustion for gas turbine applications, Appl. Energy 148 (2015) 456–465.
- [28] J. Ye, P. R. Medwell, E. Varea, S. Kruse, B. B. Dally, H. G. Pitsch, An experimental study on mild combustion of prevaporised liquid fuels, Appl. Energy 151 (2015) 93–101.

- [29] H. C. Rodrigues, M. J. Tummers, E. H. van Veen, D. J. E. M. Roekaerts, Spray Flame Structure in Conventional and Hot-Diluted Combustion Regime, Combust. Flame 162 (3) (2015) 759–773.
- [30] J. A. Friedman, M. Renksizbulut, Investigating a methanol spray flame interacting with an annular air jet using phase-Doppler interferometry and planar laser-induced fluorescence, Combust. Flame 117 (4) (1999) 661–684.
- [31] S. K. Marley, E. J. Welle, K. M. Lyons, W. L. Roberts, Effects of leading edge entrainment on the double flame structure in lifted ethanol spray flames, Exp. Therm Fluid Sci. 29 (1) (2004) 23–31.
- [32] L. Ma, D. Roekaerts, Numerical study of the multi-flame structure in spray combustion, Proc. Combust. Inst. 36 (2) (2017) 2603–2613.
- [33] W. O'Loughlin, A. R. Masri, The Structure of the Auto-Ignition Region of Turbulent Dilute Methanol Sprays Issuing in a Vitiated Co-Flow, Flow, Turbulence and Combustion 89 (1) (2012) 13–35.
- [34] D. B. Proud, M. J. Evans, P. R. Medwell, Q. N. Chan, Experimental investigation of the flame structure of dilute sprays issuing into a hot and low-oxygen coflow, Combust. Flame 230 (2021) 111439.
- [35] N. Leplat, P. Dagaut, C. Togbé, J. Vandooren, Numerical and experimental study of ethanol combustion and oxidation in laminar premixed flames and in jet-stirred reactor, Combust. Flame 158 (4) (2011) 705–725.
- [36] H. J. Curran, P. Gaffuri, W. J. Pitz, C. K. Westbrook, A Comprehensive Modeling Study of n-Heptane Oxidation, Combust. Flame 114 (1) (1998) 149–177.
- [37] P. R. Medwell, P. A. Kalt, B. B. Dally, Simultaneous Imaging of OH, Formaldehyde, and Temperature of Turbulent Nonpremixed Jet Flames in a Heated and Diluted Coflow, Combust. Flame 148 (1-2) (2007) 48–61.
- [38] A. R. Masri, J. D. Gounder, Turbulent Spray Flames of Acetone and Ethanol Approaching Extinction, Combust. Sci. Technol. 182 (4-6) (2010) 702–715.
- [39] H. Zheng, E. Qiao, Y. Yang, T. Duan, Determination of inner pressure for fluid inclusions by Raman spectroscopy and its application, Geosci. Front. 2 (3) (2011) 403–407.
- [40] T. Müller, G. Grünefeld, V. Beushausen, High-precision measurement of the temperature of methanol and ethanol droplets using spontaneous Raman scattering, Appl. Phys. B 70 (1) (2000) 155–158.
- [41] M. J. Evans, P. R. Medwell, Understanding and Interpreting Laser Diagnostics in Flames: A Review of Experimental Measurement Techniques, Frontiers in Mechanical Engineering 5 (2019) 65.

- [42] S. M. Mahmoud, G. J. Nathan, P. R. Medwell, B. B. Dally, Z. T. Alwahabi, Simultaneous planar measurements of temperature and soot volume fraction in a turbulent non-premixed jet flame, Proc. Combust. Inst. 35 (2) (2015) 1931–1938.
- [43] M. J. Evans, D. B. Proud, P. R. Medwell, H. Pitsch, B. B. Dally, Highly radiating hydrogen flames: Effect of toluene concentration and phase, Proc. Combust. Inst. 38 (1) (2021) 1099–1106.
- [44] Y. K. Jeong, C. H. Jeon, Y. J. Chang, Evaluation of the equivalence ratio of the reacting mixture using intensity ratio of chemiluminescence in laminar partially premixed CH4-air flames, Exp. Therm Fluid Sci. 30 (7) (2006) 663–673.
- [45] P. R. Medwell, B. B. Dally, Experimental Observation of Lifted Flames in a Heated and Diluted Coflow, Energy Fuels 26 (9) (2012) 5519–5527.
- [46] T. C. W. Lau, G. J. Nathan, Influence of Stokes number on the velocity and concentration distributions in particle-laden jets, J. Fluid Mech. 757 (2014) 432–457.
- [47] E. J. Welle, W. L. Roberts, C. D. Carter, J. M. Donbar, The response of a propane-air counter-flow diffusion flame subjected to a transient flow field, Combust. Flame 135 (3) (2003) 285–297.