Experimental Investigation of the Effects of Turbulence and Mixing on Autoignition Chemistry

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Abstract The autoignition of acetylene released into a turbulent coflow of hot air has been the object of a recent experimental study to supplement previous work for hydrogen and *n*-heptane. As with hydrogen and *n*-heptane, autoignition appears in the form of localised spots. Here, we present new information concerning the effects of turbulence intensity, turbulent lengthscale and injector diameter on the location of autoignition. The effects of these parameters on inhomogeneous autoignition have not been investigated experimentally before. The present study establishes that increasing the bulk velocity, while keeping the lengthscale constant, increases autoignition length. For the same turbulence intensity, the autoignition length increases as the injector diameter increases and as the turbulent lengthscale decreases. A simultaneous decrease in turbulence intensity and increase in lengthscale causes a reduction in autoignition length. Further, the frequency of appearance of the autoignition spots was measured. It was found to increase when autoignition occurred closer to the injector, and also at higher velocities. The observed trends are consistent with expectations arising from the dependence of the mixture fraction and scalar dissipation on the geometrical and flow parameters. The data can be used for the validation of turbulent combustion models.

Keywords Autoignition, turbulence-chemistry interactions, turbulence, mixing, flame propagation

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1 Introduction

Autoignition in a turbulent inhomogeneous flow arises as a consequence of an interaction between several complex underlying processes: the turbulent mixing that brings the fuel and oxidizer together; the chemical pathways and associated rates of the slow pre-ignition reactions between the fuel and oxidizer; and the fate of the heat released from these reactions, leading to thermal runaway at localized sites called ‘autoignition kernels’.

In order to predict accurately the emergence (location and timing) of autoignition in a turbulent inhomogeneous field, it is not sufficient to restrict one’s focus to chemical kinetics. The mixing field formed by the reactants must also be considered carefully, as this can have a direct effect on the chemical reaction rates. Unless the chemical and turbulent timescales are completely separated, the chemistry and mixing problems cannot be decoupled, solved separately and superposed to give a correct solution. Models that attempt to split the total ‘autoignition delay time’ into a ‘mixing time’ followed by a ‘chemical delay time’ will fail as soon as turbulence-chemistry interactions are established. It becomes crucial to understand and establish approaches that are capable of predicting accurately the effects of turbulent mixing on the pre-ignition chemistry and consequently on the appearance of autoignition, both spatially and temporally.

Direct Numerical Simulations (DNS) have provided us with some useful insight into the underlying processes that lead to autoignition in a turbulent inhomogeneous field. Simulations have been performed with simple and complex chemistry in two- [1–7], but also three-dimensional turbulence [8, 9]. Specifically, the DNS revealed that autoignition was always observed at a well-defined value of the mixture fraction ξ (normalized gravimetric fuel concentration) termed ‘most reactive’, at locations where the scalar dissipation rate that is defined as proportional to the spatial gradients of ξ , i.e. $\chi \equiv 2D_M (\partial\xi/\partial x_i)(\partial\xi/\partial x_i)$, was lowest. Here, the constant of proportionality D_M is the coefficient of molecular diffusivity. The effect of turbulence was important in influencing χ at locations of most reactive mixture fraction ξ_{MR} , i.e. the conditional $\chi_{MR} \equiv \chi|_{\xi=\xi_{MR}}$. The main postulate of the DNS can be summed up as “enhanced mixing has an

inhibiting/delaying effect on the pre-ignition chemistry, and hence on the appearance of autoignition, depending on the evolution of the scalar dissipation and its fluctuations”. Autoignition of turbulent inhomogeneous flows has been reviewed by Mastorakos [10], where these findings are discussed at length.

Recent experimental studies of autoignition in the ‘Confined Turbulent Hot Coflow’ (CTHC) apparatus have considered the autoignition of hydrogen [11] and *n*-heptane [12] plumes released concentrically into coflows of turbulent preheated air. These studies, together with complementary modelling efforts (as that of Markides et al. [12], amongst others), have established that an increase in the bulk air velocity U_{air} causes an increase in the autoignition length from injection L_{IGN} , as well as in the associated residence time until autoignition $\tau_{IGN} = L_{IGN}/U_{air}$. This important finding is consistent with similar measurements of autoignition made in turbulent counterflowing jets of hydrogen and hot air [13, 14], where it was demonstrated experimentally that increased turbulence in the air stream resulted in a higher critical temperature necessary for autoignition, thus suggesting an inhibiting effect of turbulence on the pre-ignition reactions.

Further consideration of the influence of the air velocity on the flow and mixing fields in the CTHC geometry [15, 16] revealed that at any given spatial location an increase in U_{air} was accompanied by:

- (i) an increase in the turbulent velocity fluctuations, but no significant change to the turbulence integral lengthscale; while in parallel,
- (ii) the mean mixture fraction field remained unaffected, and the mean scalar dissipation rate increased.

The experimental evidence that an increase in mean velocity, while the mean mixture fraction is kept unchanged, will result in an increase in autoignition length and delay time serves as some validation of the link between enhanced mixing and delayed autoignition that was observed in the DNS, since higher velocity fluctuations with an unchanged lengthscale imply a higher scalar dissipation rate.

One point worth reflecting on briefly here is the DNS finding [1] that, whereas increasing the thickness of the fuel-air interface (and hence decreasing χ_{MR})

reduced the autoignition delay time, the turbulent timescale τ_{turb} did not seem to affect the autoignition time. On the other hand, Mastorakos et al. [2] demonstrated by performing simulations with the same DNS code as in Ref. [1] that enhanced turbulence (that is shorter τ_{turb}) promoted a slight acceleration of autoignition. This is most interesting as it is in subtle contrast to both coflow and counterflow experiments mentioned above. The simulations were performed in two-dimensional shearless mixing layers and did not last long relative to the turbulence turnover time. The method for resolving the fuel-air interface was crucial in determining the initial condition (i.e. the initial value of χ_{MR}). Consequently, the autoignition time was found to also depend strongly on the initial condition, with the turbulence affecting the autoignition time only insofar as it affected the emergence of the lowest value of χ_{MR} . Therefore, autoignition was promoted by fast mixing, due to the earlier emergence of well-mixed ξ_{MR} sites with low χ_{MR} . More recent DNS studies include three-dimensional simulations with detailed chemistry for the investigation of coflows resembling those in the CTHC [9], and also of jet-type flows [17, 18] that examined the interaction between autoignition and flame stabilisation. Phenomenologically, the DNS and experiments [11, 19] are consistent concerning the explosive emergence of localised flamelets that propagate into their unburnt surroundings with complex behaviour. However, a detailed DNS study on the explicit effect of turbulence parameters on autoignition was not performed, most likely due to the enormous cost of these simulations that did not allow a wide range of parameters to be explored.

It becomes essential to clarify experimentally the separate effects of turbulent velocity fluctuations, turbulent lengthscales, and mixing patterns on autoignition. Moreover, the aforementioned experimental observations in the CTHC were restricted to a probing in a fixed geometry of the effect of a single flow parameter on the phenomenon, that of the bulk air velocity U_{air} . In this paper, we manipulate the turbulence characteristics in a parametric study involving a number of geometry modifications. We examine the effects of these changes on the velocity and mixing fields and subsequently associate them with any variations in the spatial appearance of autoignition. Specifically, the effects of the injector diameter size, injection distance downstream of the turbulence-generating grid and of the

hole size of the grid on the autoignition distance from injection are investigated. The velocity and mixing field data in the various configurations, as well as the corresponding autoignition results can serve as a test-bed for future model development.

In summary, the effect of the turbulence character, length and timescales on the magnitude of L_{IGN} (and τ_{IGN}) is in need of elucidation. For the purposes of the current study, the exploration of the sensitivity of autoignition to the turbulence parameters is best attempted in conditions in which the chemical timescales are of the same order, or close to the order, of the turbulent timescales; whence the direct effect of turbulent mixing is most significant.

Furthermore, autoignition in a turbulent inhomogeneous flow will display randomness. Although this has been reported previously, e.g. in the CTHC apparatus [11, 12, 19], no results were made available. In this paper, we also present data concerning the temporal randomness (i.e. the autoignition frequency) of the appearance of autoignition in the CTHC for the first time. Similar observations concerning the temporal evolution of autoignition spots have been presented recently by Oldenhof et al. [20]. However, those experiments focused on the autoignition of high-velocity fuel jets (with bulk injection to coflowing air velocity ratios in the range ~ 3 to 9) in low velocity coflows (with bulk velocities of 4.3 – 4.6 m/s) of vitiated air (with oxygen content in the oxidizer stream of 8.4 – 9.5% by mass). Here we compliment these recent observations, with results obtained in the CTHC apparatus, where the fuel injection velocities are lower (bulk injection to coflowing air velocity ratios around unity), and where electrically preheated turbulent air (oxygen mass fraction 23.2%) is used at considerably higher velocities (bulk velocities 10 – 30 m/s), in place of the vitiated coflow.

2 Experimental Methods

2.1 Apparatus

Figure 1 (left) shows a schematic of the Confined Turbulent Hot Coflow (CTHC) apparatus [11, 12, 15–16, 19]. The radial and downstream axial distances from the central point of injection are denoted by r and z respectively. Nitrogen-diluted acetylene (referred to as ‘fuel’) was issued continuously and concentrically from an injector nozzle with inner diameter d , into a coflow of electrically preheated turbulent air confined in a vacuum-insulated transparent quartz pipe of inner diameter D . The turbulence was generated upstream of injection by a perforated plate, characterized by a hole diameter M and solidity s . The distance of the injection point from the grid is denoted by x .

The fuel and air flowed downstream while mixing and reacting simultaneously, and autoignition was observed at some location in the confining pipe. In the ‘random spots’ regime of operation that all the measurements in this paper were performed, autoignition appeared in the form of statistically steady events that were associated audibly with a popping sound and visually with bright localized spots, streaks, or flashes of blue/yellow light [11, 12]. The ‘autoignition spots’ appeared over a region whose axial distance from the injector nozzle was L_{IGN} , as illustrated in Figure 1 (left).

The phenomenon of autoignition is very sensitive to temperature, which prompted the development of a detailed strategy for radiation and conduction corrections to the raw thermocouple readings. The final reported temperatures are associated with a total error of 0.6% corresponding to an absolute error of approximately 5 K. This can be divided into a 0.3% random and a 0.5% systematic contribution. In addition, the local mean axial temperature $\langle T(r=0, z) \rangle$ along the centreline was measured in the absence of fuel flow, but with the same conditions for T_{air} and U_{air} used in the autoignition experiments. The axial centreline temperature was found to drop linearly by 10 K in the first 100 mm due to heat losses. Finally, the local temperature fluctuations T' were also measured in the coflow at the injection

plane and were found to be small relative to the mean temperature with values ranging from 0.5 K to 2.5 K over the range of T_{air} used.

2.2 Parameter space

In this paper we present two classes of experiments. In the first class, various geometric modifications were made to the basic (or ‘original’) CTHC apparatus in order to investigate the effect of turbulence and mixing on the spatial appearance of autoignition. In the second class, we focus our attention on two CTHC configurations, the ‘original’ and ‘small injector’ geometries, in order to characterise the temporal appearance of the autoignition spots.

Referring to the first investigation concerning the spatial appearance of autoignition, Table 1 provides details of the geometric parameter space explored in the experiments presented in this paper. Three case studies were undertaken specifically to probe our current understanding of how turbulent mixing can affect autoignition in the CTHC configuration. All experiments were done in a pipe with $D = 25.0$ mm. The parametric case studies are also represented graphically in Figure 1 (right). The ‘original’ CTHC geometry used in Refs. [11, 12, 15–16, 19] is highlighted in Table 1 as well as in Figure 1 (right). A detailed description of that apparatus configuration can be found in these references.

Case Study 1 (CS1) was performed with a perforated grid with hole size $M = 3.0$ mm and solidity $s = 45\%$. The injector nozzle (inner) diameter was increased from $d = 1.1$ mm (‘small injector’ configuration) to $d = 2.2$ mm (original configuration) by the removal of an injector adapter. This was done in order to ensure that (inner) the air in the coflow around the injector was unaffected by the change to the injector. The doubling of d also resulted in a doubling of the non-dimensional nozzle diameter from $d/M = 0.37$ to $d/M = 0.73$.

In Case Study 2 (CS2), the experiments were all conducted with the $M = 3.0$ mm perforated grid and the $d = 1.1$ mm injector. Hence, the non-dimensional nozzle diameter was kept constant at $d/M = 0.37$. However, an injector extension was used, thus increasing the injection distance from the grid from $x = 66$ mm (small

injector) to 105 mm ('long injector'), or in dimensionless form from $x/M = 22$ to 35 respectively.

Finally, Case Study 3 (CS3) involved the use of a grid with hole size $M = 1.2$ mm and solidity $s = 55\%$ ('small grid'). The axial distance of injection from the grid was adjusted by moving the position of the grid until the turbulence intensity u'/U in the coflow at injection was approximately equal to that in the original configuration, i.e. with the $M = 3$ mm grid placed at distance of $x = 66$ mm upstream of the injection location. Hot wire measurements (presented in Section 3.1) showed that with the $M = 1.2$ mm grid, u'/U at $x = 53$ mm was approximately equal to u'/U at $x = 66$ mm when the $M = 3$ mm grid was used.

2.3 Autoignition measurements

2.3.1 Autoignition length in Case Studies 1, 2 and 3

The autoignition measurement runs were performed over a range of conditions

summarized in Table 2, where $Re_M = \frac{U_{air} M}{\nu}$, $\delta_{fuel} = \frac{\rho_{fuel}}{\rho_{air}}$ and $v_{fuel} = \frac{U_{fuel}}{U_{air}}$.

Each row corresponds to the respective entry in Table 1. After T_{air} reached the desired temperature, final adjustments were made to the fuel and nitrogen flow rates and autoignition was detected in the pipe. Each individual autoignition test run was performed over a period few minutes, with steady conditions of bulk (area-averaged) air temperature T_{air} , bulk air velocity U_{air} , bulk fuel temperature T_{fuel} and bulk fuel velocity U_{fuel} .

The dilution of acetylene (C_2H_2) with excess nitrogen (N_2) in the fuel stream is quantified with the mass fraction of C_2H_2 . All autoignition measurements were done with a single fuel dilution of $Y_{fuel} = 0.65 \pm 0.05$. The resulting density ratio of the injected nitrogen-diluted fuel (C_2H_2/N_2) stream to that of the air stream was $\delta_{fuel} = 1.3 \pm 0.2$. In addition, an equal velocity condition was imposed such that the injection velocity U_{fuel} of C_2H_2/N_2 was approximately equal to U_{air} . A maximum permissible deviation of $\pm 20\%$ was allowed, such that $v_{fuel} = 1.0 \pm 0.2$ was ensured across all runs. Slight changes to the autoignition location due to the

stated variations in Y_{fuel} , δ_{fuel} and v_{fuel} were investigated in a separate set of experiments and found to be at most 10%.

Autoignition was detected by optical measurements of chemiluminescence radiated from the reacting regions, resulting in images similar to that shown in Figure 2 (left). Characteristic emission spectra taken from regions of autoigniting gases for acetylene and spanning the range 250 – 650 nm can be seen in Figure 2 (right). From this we may conclude that the emission is broadband, but also that the hydroxyl radical (OH*) that appears at ~ 310 nm is an excellent marker for the detection of autoignition in the conditions investigated here. Hence, direct OH* images were taken at 10 Hz with a LaVision NanoStar Intensified Charge Coupled Device (ICCD) camera equipped with an Ultra-Violet (UV) lens of focal length 105 mm, and 307 ± 10 nm filter. In the resulting instantaneous images the intensity signal at each pixel can be considered roughly proportional to the OH* emitted along the line-of-sight corresponding to the location of that pixel. The exposure time for each image was set so as to ensure that (on average) the instantaneous images captured as much of the complete history of individual autoignition events, while keeping the number of autoignition spots per image to a minimum. A value of the order of a few ms was chosen based on separate high-speed observations.

An ‘autoigniting region’ was then compiled over a large number of instantaneous images, and hence spots, resulting in time-integrated images such as that illustrated in Figure 1 (left). Referring to that figure, the random autoignition length variable L_{IGN} is defined as the axial distance from the injector to the autoigniting region. This region can be quantified in a number of ways. The measure of L_{IGN} presented here is that of the minimum autoignition length L_{MIN} , based on a 3% rise in the signal from the background, relative to the peak intensity. Due to flame propagation following localised autoignition, the instantaneous images also include contributions from post-ignition flamelets. However, the aforementioned high-speed time-resolved investigation revealed that the true mean location of localized appearance of autoignition $\langle L_{TRUE} \rangle$ lies just downstream of L_{MIN} (within 2 standard deviations, σ_{IGN}) [12]. Figure 3 shows

the main results from the investigation into the discrepancy between L_{MIN} and $\langle L_{TRUE} \rangle$.

2.3.2 Autoignition spot frequency

For a range of selected conditions in the original and small injector configurations, a Hamamatsu Photo-Multiplier Tube (PMT) with a 307 ± 10 nm filter was used to collect OH* chemiluminescence from the whole flow. The PMT voltage was amplified and sampled at 100 kHz. Sampling times varied from 10 to 80 s in order to capture enough events and ensure adequate convergence of the statistics. Simultaneously, a high-speed Phantom v.4.2 Complementary Metal-Oxide Semiconductor (CMOS) camera and a C9546-04MP46 Hamamatsu intensifier were used to provide chemiluminescence images. Typically, 512×152 pixel images were collected at frame rates of up to 12 kHz. A UV lens with focal length 105 mm, and a 307 ± 10 nm OH* optical filter were used. The raw images from the CMOS camera were first processed to remove noise. The fast image sequences were then examined in order to extract the following information:

- (i) the location of the first appearance of light emission from a completely dark background; and,
- (ii) the extent and speed of propagation of the flamelet emerging from the autoignition spot.

Item (i) is discussed in detail in this paper, while Item (ii) was presented in Markides and Mastorakos [19].

All autoignition timing (frequency) experiments, the results of which are presented in Section 4.5, were done with nitrogen-diluted acetylene, and again, with the equal velocity condition $v_{fuel} = 1.0 \pm 0.2$ such that the injection velocity U_{fuel} of C_2H_2/N_2 was approximately equal to U_{air} . Two sets of experiments were performed in the original geometry, with $U_{air} = 10.9$ and 17.3 m/s, T_{air} in the range $806 - 825$ K and $850 - 879$ K respectively, and $Y_{fuel} = 0.62 \pm 0.2$. In addition, three sets of experiments were performed in the small injector geometry, with $U_{air} = 15.8, 19.5$ and 29.7 m/s, T_{air} in the range $869 - 884$ K, $870 - 885$ K and $885 - 920$ K respectively, and $Y_{fuel} = 0.77 \pm 0.2$.

3 Flow Field Characterisation

A comprehensive description of the hot wire and Planar Laser-Induced Fluorescence (PLIF) measurements that were used to characterize the velocity and mixing fields in the original CTHC geometry can be found in Refs. [11, 12, 15–16, 19]. In this paper, the procedure is outlined briefly for clarity, with added information on measurements that were performed in the modified geometries, i.e. small injector, long injector and small grid.

3.1 Velocity field

The axial velocity fields in the four configurations were characterized with a hot wire and a Constant Temperature Anemometer (CTA) system, over a range of ‘cold’ (i.e. ambient temperature, non-reacting) conditions that exhibited dynamic similarity with the ‘hot’ autoignition runs described in Table 2. Specifically, coflows of air were established with a range of bulk (area-averaged) velocities U_{air} such that the corresponding range of macroscale Reynolds numbers $Re_M = U_{air}M/\nu_{air}$ was approximately equal to those attained during the autoignition runs (see Table 2). Having set the conditions in the coflow, kinematic similarity with the autoignition runs was ensured by the adjustment of the bulk injection velocity U_{fuel} . Air was passed through the injector with a flow rate such that U_{fuel} was approximately equal to U_{air} , i.e. such that $v_{fuel} = U_{fuel}/U_{air} = 1$ (to within $\pm 20\%$), thus matching the identical constraint in the autoignition experiments (see Table 2). It is emphasised that these ‘equal velocity’ flows were not characterized by the shear layers associated with jets. A selection of important CTA results is summarized in the last two columns of Table 1. Values are reported for turbulence quantities measured in the injection plane, close to the nozzle ($z = 0$, $r/D = 0.1$).

The CTA experiments confirmed that the flow in the regions occupied by the autoigniting spots can be closely modelled as homogeneous decaying turbulence behind the grid. Close inspection of power spectra and Probability Density Functions (PDFs) of velocity revealed that the flow field was as would be expected downstream of a turbulence generating grid, but also confined in the quartz pipe [21]. These experiments provided radial profiles at various axial

locations of time-averaged local axial velocities U , as well turbulence quantities of particular interest including the root mean square (RMS) of the axial velocity fluctuations u' and the corresponding longitudinal integral lengthscale L_{turb} , from which $\tau_{turb} = L_{turb}/u'$ can be inferred. The longitudinal integral lengthscale was obtained by integrating over the normalized autocorrelation function of the axial velocity fluctuations and the use of Taylor's hypothesis.

Specifically, measurements in the small injector configuration confirmed that the presence of the injector adapter did not have a significant effect on the velocity field downstream of the nozzle and that this remained similar to that in the original geometry. Hence, both in the original and small injector geometries, the flow was near-uniform in terms of the mean velocity U and RMS velocity u' , but also in terms of L_{turb} , with the boundary layers confined to regions close to the injector and the pipe walls. The ratio of local mean to bulk velocity U/U_{air} amounted to approximately $1.12 - 1.14 (\pm 0.02)$, staying within 5 % of this value over a region covering $0.15 < r/R < 0.85$, where $R = D/2$. At injection ($x = 66$ mm), $u'/U = 0.11 - 0.13 (\pm 0.01)$ over the region $0.2 < r/R < 0.8$, increasing inside this region with increasing Re_M . At the same location L_{turb} was found to be in the range $3 - 4.5 (\pm 0.5)$ mm, or $L_{turb}/M = 1.0 - 1.5$.

In the long injector configuration the flow was again nearly uniform in terms of U , u' and L_{turb} , with $u'/U = 0.09 - 0.10 (\pm 0.01)$ and $L_{turb} = 3.5 - 5 (\pm 0.5)$ mm over the (radial) injection plane at $x = 105$ mm. The corresponding value of L_{turb}/M was $1.2 - 1.7$. These lower values relative to the original and small injector geometries were expected as the turbulence in the flow decayed downstream of the grid.

Finally, in the small grid configuration u'/U was about 0.11 to $0.13 (\pm 0.01)$ at injection (i.e. at $x = 53$ mm from the $M = 1.2$ mm grid), and hence equal to u'/U at injection in the original geometry (i.e. at $x = 66$ mm from the $M = 3$ mm grid). Moreover, for the same U , the smaller $M = 1.2$ mm grid was associated with increased $L_{turb} = 5 - 7 (\pm 1)$ mm and $L_{turb}/M = 4.2 - 5.8$ at the injection plane. This can be understood in terms of both the decrease in $Re_M = UM/\nu$ and the increase in x/M , consistent with other studies [22, 23]. In Chen and Bilger [23] for example, L_{turb}/M increased from approximately 1.5 to between $3 - 4$ as x/M was increased

from 21 to 43 by decreasing the plug hole diameter from 4 to 2 mm while keeping U unchanged.

3.2 Mixing field

The mixing fields in the original, small diameter and small grid configurations were investigated with PLIF of acetone in non-reacting (i.e. lower temperature) conditions [15, 16]. Measurements were done over a range of coflow conditions that were dynamically similar to those used in the CTA velocity measurements, and hence also those used for the actual autoignition runs. This was ensured by setting the bulk air velocities U_{air} such that the macroscale Reynolds number Re_M matched the necessary range of values. Also, as with the CTA and autoignition experiments, the bulk injection velocity U_{fuel} was set to be within $\pm 20\%$ of the coflow air velocity U_{air} , such that the fuel-to-air velocity ratio $v_{fuel} = 1.0 \pm 0.2$. Moreover, actual nitrogen-diluted acetylene plumes seeded with acetone were issued from the injector to ensure that the dimensionless injection density $\delta_{fuel} = \rho_{fuel}/\rho_{air} = 1.5 \pm 0.1$ was kept close to that used in the autoignition runs (see Table 2). Even so, buoyancy is not expected to play a role here, due to the high momentum of the coflow. Measurements were made inside a two-dimensional plane passing through the centreline of the CTHC apparatus.

The PLIF measurements of the fluorescent conserved scalar (i.e. acetone) field provided information on the mixture fraction ξ , and from this on the scalar dissipation rate χ . Measurements of the local volumetric/molar concentration of the injected acetone-laden fuel were suitably normalized and corrected for local density variations to obtain instantaneous maps of ξ . The two-dimensional scalar dissipation rate χ_{2D} was calculated from the sum of the axial and radial gradients of the measured two-dimensional planar ξ , and from local estimated values of the molecular diffusion coefficient D of acetone into the multi-component air/fuel mixtures [15]. The Region of Interest (RoI) in the original and small diameter flows occupied the region from the injector nozzle to a downstream distance of about $z = 60$ mm. In the small grid flows the largest downstream distance in the RoI was increased to $z = 110$ mm, to allow observations of the longer plumes.

Figure 4 compares the axial decays of the mean mixture fraction along the centreline $\langle \xi(r=0, z) \rangle$ between the small injector ($d = 1.1$ mm) and original ($d = 2.2$ mm) geometries (left) and between the original ($M = 3.0$ mm) and small grid ($M = 1.2$ mm) geometries (right). The resulting data reveal that the choice of equal velocity between injection and the coflow ($v_{fuel} = 1$) in all flows set up in the CTHC for this study, resulted in mixing patterns traditionally associated to diffusion downstream of a low momentum release of a scalar from a point source into a uniform background flow of homogeneous turbulence. Consequently, even though in the vicinity of the source the effect of the finite sized source can be seen, farther away from the source the plumes decayed axially as z^{-2} .

It is evident from Figure 4 that in all geometric configurations the axial plume extent was not affected significantly by the velocities and hence Re_M .

Furthermore, Figure 4 (left) shows that $\langle \xi(r=0, z) \rangle$ was only slightly affected by the change in d . In the CS1 experiments the removal of the small injector adaptor resulted in a slight increase in the axial plume extent. By contrast, Figure 4 (right) shows that in the CS3 experiments the small grid plumes mixed slower than in the original configuration. This may be explained by the fact that the large lengthscale of the turbulence (recall that the small grid plume resulted in large lengthscale at the nozzle location) relative to the initial plume thickness causes the plume to meander without appreciable mixing.

Figure 5 (left) provides further confirmation of the conclusion that the low momentum fuel release from the injector into the (almost) uniform background homogeneous turbulent flow results in ‘Gaussian’ mixing plumes, i.e. that extend radially as $\exp[-r^2/2\sigma^2]$. We note that the small injector plumes are somewhat narrower than those established in the original geometry, and that the small grid plumes were the narrowest, as would be expected from the corresponding axial profiles in Figure 4.

In more detail, Figure 5 (right) shows that from a distance of about $z = 5$ mm, before which the plume in this near-source region is still developing, the spreading of the plumes is determined mainly by the background turbulence in the

coflowing air, in line with predictions from the Gaussian plume model at ‘short lengths’,

$$\langle \xi(r, z) \rangle = \langle \xi(r=0, z) \rangle \exp \left[-\frac{1}{2} \left(\frac{r}{\sigma} \right)^2 \right]; \frac{\sigma}{z} = \frac{u'}{U} \quad (1)$$

where $r_o = \sqrt{2\sigma^2}$ is the plume width at which $\langle \xi \rangle$ reaches 1/e of its centreline value [24]. This is reflected by the flat σ/z that decreases very slowly as the turbulence levels downstream of the grid decay. At long z , the near source spreading is replaced by a ‘long length’ behaviour described by,

$$\langle \xi(r, z) \rangle = \langle \xi(r=0, z) \rangle \exp \left[-\frac{1}{2} \left(\frac{r}{\sigma} \right)^2 \right]; \frac{\sigma}{z} = \sqrt{2C_\mu \frac{u'}{U} \frac{L_{urb}}{z}} \quad (2)$$

Finally, in Figure 6 we consider the centreline evolutions of the RMS of the mixture fraction fluctuations $\langle \xi'^2 \rangle^{1/2}$ (left) and the mean two-dimensional scalar dissipation rate $\langle \chi_{2D} \rangle$ (right). Both increase from zero to a maximum that occurs between $z = 5 - 10$ mm or $z/d = 2$ to 5, and then decay downstream. This is not surprising as it was already shown in Markides and Mastorakos [15] that the two quantities were closely connected via the simple model $\langle \chi \rangle = \langle \xi'^2 \rangle / \tau_{mix}$, except in close proximity to the injector ($z/d < 2 - 3$). Hence, we may conclude that at shorter z , though not in the immediate vicinity of the nozzle, and certainly from the region $z > 10$ mm (or $z/d > 5$) that is relevant to the appearance of autoignition, the fluctuations in ξ decrease as z increases. It was also found (not shown here) that, similarly to ξ , the measured fluctuations in χ_{2D} also increased at shorter z . With increasing Re_M the two quantities increased and the maximum shifted to shorter lengths, although it is pointed out that this effect was not as pronounced in $\langle \chi_{2D} \rangle$ and was not evident in $\langle \xi'^2 \rangle^{1/2}$ with the small injector.

Comparing the two configurations, it is noted that $\langle \xi'^2 \rangle^{1/2}$ in the small injector plumes was lower than what was observed in the case of the original geometry. Furthermore, at low Re_M (i.e. U_{air}), $\langle \chi_{2D} \rangle$ was higher at short z (< 10 mm), but

decreased quickly at longer z . At high Re_M the profiles of $\langle \chi_{2D} \rangle$ for the original and small injectors converged.

The main conclusions from this section concerning the effects of the various geometric parameters such as d , x and M , as well as of Re_M on the turbulence character of the air stream and consequently on the mixing field are summarized below:

- Increasing U_{air} , while keeping the geometry (M, x, d) the same, resulted in similar mean ξ distributions, but higher χ . The latter conclusion concerning χ was almost absent in the small injector configuration.
- Decreasing d resulted in a slightly shallower decay of the mean ξ . Also, at low U_{air} , χ was initially higher (closer to the source), but decreased quickly downstream becoming lower compared to the larger injector. At high U_{air} , χ was not affected significantly by the choice of d .
- Decreasing M , while keeping u'/U unchanged, resulted in significantly thinner and longer plumes.
- These outcomes are expected to affect the appearance of autoignition in the CTHC. In Section 4, we will use these results to explain the trends observed in the autoignition case studies.
- The fluctuations in both ξ and χ increased at shorter z , in the region relevant to the appearance of autoignition. Higher U_{air} resulted in an increase in both ξ and χ .

4 Autoignition Results and Discussion

4.1 Comparison with Previous Results with Other Fuels: Effect of chemistry

The investigated parameter space for the autoignition of nitrogen-diluted acetylene in the original geometry presented in Tables 1 and 2 resulted in L_{MIN} in the range 26 – 88 mm, over the range $T_{air} = 806 – 904$ K and $U_{air} = 10.5 – 28.4$ m/s. Before we proceed, it is interesting to compare this with results from our previous studies in the same geometrical configuration but with other fuels, and specifically hydrogen [11] and *n*-heptane [12]. Table 3 summarises the results.

It can be seen that the autoignition of *n*-heptane in the ‘random spots’ regime of operation required the use of the highest $T_{air} = 1104 – 1138$ K, followed by the autoignition of hydrogen that required $T_{air} = 941 – 966$ K, with acetylene autoignition possible at lower T_{air} . Within a range of comparable U_{air} , hydrogen autoignition spots appeared at the shortest $L_{MIN} = 18 – 63$ mm, while acetylene spots appeared at intermediate $L_{MIN} = 26 – 88$ mm. Finally, *n*-heptane spots were observed at even longer $L_{MIN} = 34 – 103$ mm, even though T_{air} was higher and Y_{fuel} was larger. The trends reflected in these findings are consistent with expectations from the homogeneous autoignition of these fuels [25–27].

From this point onwards we restrict ourselves to our chosen fuel (i.e. nitrogen-diluted acetylene) and investigate the effects of turbulence and mixing on the appearance of autoignition in a parametric study involving a number of geometrical modifications.

4.2 Case Study 1 (CS1): Effect of injector nozzle diameter

Referring to Figure 7, we consider first the open symbol data points generated in the original geometry with the larger injector. Three datasets were generated at constant $U_{air} = 10.9, 17.3$ and 24.7 m/s. An increase in T_{air} for a given U_{air} results in a decrease in L_{MIN} , and an increase in U_{air} for the same T_{air} gives rise to an increase in L_{MIN} . This conclusion is consistent with Refs. [11, 12] that considered

hydrogen and *n*-heptane respectively in the same geometry. The two trends can be understood as follows:

- (i) for a given U_{air} the chemical reaction rates are enhanced by the increase of T_{air} , and hence autoignition occurs closer to the injector; whereas,
- (ii) for a given T_{air} an increase in U_{air} causes u' to increase (maintaining an approximately constant u'/U ; see Section 3.1). Since L_{turb} is also approximately constant, this leads to faster $\tau_{turb} = L_{turb}/u'$ and therefore higher χ (see Figure 6), that inhibits autoignition.

When using the smaller injector geometry similar effects are noted, although L_{MIN} appears to be less sensitive to variations in T_{air} and U_{air} . The insensitivity in L_{MIN} with the small injector can be explained by the fact that χ is not affected significantly by Re_M (or U_{air}), as revealed by the mixing measurements (see Figure 5). Moreover, the tendency of the autoignition spots to propagate upstream and cause ‘flashback’ at high T_{air} (and hence short L_{MIN}) was found to lessen when the smaller injector was used.

The two datasets shown in Figure 7 for the small injector were taken with $U_{air} = 20.5$ and 29.5 m/s, which bracket the original geometry dataset taken at $U_{air} = 24.7$ m/s. We observe that the original geometry is associated with longer L_{MIN} and steeper slopes. At the highest T_{air} and U_{air} , the measured L_{MIN} from the two different injectors converge. We recall that:

- (i) the plume extent, in the mean, is not affected substantially by the changes in d ;
- (ii) χ is higher initially, but also decays faster with the small injector, and is affected only slightly by changes to Re_M (and thus U_{air}) when compared to the original configuration. Hence, χ is lower with the small injector for $z > 10$ mm at low Re_M , whereas at higher Re_M the dissipation profiles converge.

4.3 Case Study 2 (CS2): Effect of injection distance from grid

Figure 8 shows three constant U_{air} datasets related to CS2, two taken in the small injector ($U_{air} = 20.5$ and 29.5 m/s) and one in the long injector ($U_{air} = 32.1$ m/s)

configuration. In all datasets L_{MIN} decreases with increasing T_{air} as with previous measurements [11, 12] (see also Section 4.1; Figure 7). It was established that when x was increased and hence L_{MIN} shortened, a higher U_{air} was necessary to avoid flashback and to establish the statistically-steady random spots autoignition behaviour. For this reason random spots experiments in the long injector configuration were performed at higher U_{air} .

In more detail, with the small injector nozzle at $x = 66$ mm from the grid, L_{MIN} was between 20 to 30 mm over the range of $T_{air} = 885 - 910$ K. However, when x is increased in the long injector geometry to 105 mm, L_{MIN} is notably shorter (at about 15 mm), even at higher $U_{air} = 32.1$ m/s. This can be explained by the fact that at longer x from the grid the velocity measurements showed lower u'/U and higher L_{turb} for the same U_{air} (and hence Re_M). Therefore, $\tau_{turb} = L_{turb}/u'$ is larger, leading to lower χ , which promotes earlier autoignition.

4.4 Case Study 3: Effect of grid hole size

In CS3 results were generated in the small grid geometry over a range of T_{air} for three datasets corresponding to constant $U_{air} = 10.5, 16.5$ and 22.6 m/s, as shown in Figure 9 (filled symbols). These are compared with original geometry data (empty symbols). Within each small grid dataset, an increase in T_{air} brings autoignition closer to the injector and an increase in U_{air} moves autoignition to longer L_{MIN} , as with the previous experimental configurations. Also, the propensity for ‘flashback’ at high T_{air} (and hence short L_{MIN}) was found to increase in the small grid configuration.

Referring to the small grid data in Figure 9, the occurrence of random spots autoignition shifts to lower T_{air} and is possible at longer L_{MIN} than in the original geometry. At low T_{air} there is a significant increase in L_{MIN} with respect to the original geometry at otherwise similar conditions, whereas at high T_{air} the measured L_{MIN} is actually shorter than that in the original geometry.

From our flow field characterization measurements we are aware that in the small grid flows the plumes in the mean are elongated axially (from Figure 4). Indeed,

we observe that at low T_{air} the small grid L_{MIN} is longer for comparable U_{air} than in the original configuration. However, the higher L_{turb} in the small grid flows correspond to increased $\tau_{turb} = L_{turb}/u'$ for a given U_{air} (since u'/U is kept unchanged), which in turn should be associated with reduced χ , favouring earlier autoignition. This goes some way towards explaining the shorter L_{MIN} in the small grid relative to the original configuration, observed at higher T_{air} and hence when autoignition is occurring close to the injector nozzle. Also, it might perhaps explain the increased sensitivity of L_{MIN} to T_{air} (steeper gradients) and the greater propensity for flashback exhibited in the small grid flows.

4.5 Temporal randomness

Figure 10 shows the OH* chemiluminescence registered with the PMT simultaneously with a high-speed image sequence. Also, Figure 11 shows a magnification of a single autoignition event. Immediately before the explosive event, the camera records a dark background and the PMT records zero signal. Following autoignition, flamelets propagate radially outwards in all directions and the size of the reacting region grows, but the chemiluminescence intensity decreases. The explosive emergence of chemiluminescence occurs around 0.5 ms after the initiation of the event. The post-ignition decay of chemiluminescence takes about 3 ms to reach 10% and 6 ms to reach 3% of the peak value.

Figure 12 shows typical timeseries of OH* for a situation with long (left) and short (right) L_{IGN} . The experiment with short L_{IGN} produces more frequent spotting, hence we refer to this as a ‘fast spotting’ condition. It is evident that the slower spotting flow produces well-defined autoignition events, which are detected by a sharp rise in the global OH* emission above the dark background. Quite often a spot will consist of ensuing, secondary peaks coming very close after the primary rise. We believe that some of these secondary peaks in the OH* signal are due to localized autoignition (‘secondary events’) triggered by the narrow proximity of an earlier (primary) autoignition event, i.e. autoignition assisted by the flamelet propagation following from autoignition elsewhere. This is shown more clearly in the image sequence in Figure 11, but also in Figure 13. In Figure 13, an independent small spot is evident in Frame 15. In Frame 16 we

can see the flamelet growth around it, and also a second explosive event just downstream of the initial flamelet. Autoignition spots separated by a large distance are also evident, e.g. Frame 4 and in Figure 13, which may also produce OH* peaks quite closely spaced in time.

In the fast spotting regime it is difficult to distinguish between individual events, although a close examination of the time-series shows the same features of an explosive rise of OH* and a slower decay, which is interrupted by another sharp rise. The reacting region now resembles the lifted flames in vitiated air of Cabra et al. [28], although a continuous flame sheet is still not clearly evident.

Figure 14 shows that the time-averaged OH* signal decreases quickly with increasing L_{MIN} in both the original (left) and small injector geometries (right). The RMS of the signal also decreases with increasing L_{MIN} . This can be explained by the fact that spots appear more frequently at short L_{MIN} conditions, which is reported in more detail in the next paragraphs and in Figure 17. So, we would expect the generated OH* to follow f_{IGN} .

But further, note the higher instantaneous OH* intensity of each spot in the fast spotting flow in Figure 12. In Markides and Mastorakos [19] it was reported that the average physical size of the spots formed by the reaction of the fuel in the immediate vicinity of each explosion event before extinction of the propagating post-ignition flamelets increases when L_{MIN} increases. However, this was by about 10% over the entire range $L_{MIN} = 40 - 85$ mm. On the other hand, the average propagation velocity of the flamelets was found to increase by about 50% over the same range of L_{MIN} . We assume that the flamelets can only be sustained in a bounded range of fuel-air mixture compositions surrounding the original location of autoignition. We also know that the fuel plume is narrower nearer the injector. Hence, since faster spotting occurs at shorter L_{MIN} , it is possible that flamelets originating from autoignition spots at shorter L_{MIN} , where the fuel is less well-mixed with air, are advected quickly to consume more of the available surrounding mixture, at least in their immediate vicinity. A higher U_{air} (and hence fuel flow rate since in these experiments $U_{fuel} \approx U_{air}$), with autoignition at the

same L_{MIN} , results in larger average spot sizes and greater propagation flamelet velocities, resulting in greater fuel consumption and increased mean emitted OH*.

The time between spots can be characterized by measuring the time delay Δt_{IGN} between peaks in the OH* signal (e.g. Figure 12). These have been located by a peak-detection algorithm. Figures 15 and 16 show that the PDFs of the time delay between peaks have a similar shape for all experiments, i.e. for all L_{MIN} and in both geometric configurations. The PDFs peak at about 0.3 – 0.5 ms and have long tails at longer delays, which increase substantially as we move from fast spotting to slow spotting conditions. It is also evident that the probability of peaks closer than about 0.2 ms is negligible and that this limit seems to be independent of the flow conditions. Note that although for clarity the insert in Figure 15 only shows six of the lines that are drawn in the main figure, all PDFs adhere to the trend suggested in the insert; that is for the function to shift the right and to peak at higher values as the mean delay $\langle \Delta t_{IGN} \rangle$ decreases.

The minimum time delay between such primary and secondary events is about 0.2 ms in the present experiments. With a speed of sound of about 650 m/s for the conditions tested, this corresponds to a distance of 130 mm. This is much longer than the physical distance between the spots, which does not exceed 25 mm. Hence, the induced ignition from the primary to a secondary spot is probably not due to a localised pressure wave, e.g. as in engine knock or a developing detonation, but due to diffusion or convection of heat associated with a propagating flamelet. The fact that autoignition events can be triggered by an earlier nearby autoignition is relevant to HCCI engines, as this phenomenon has been considered responsible for the quick growth of the combustion zone across the cylinder visualised in such engines [29].

Figure 17 shows that, in both geometries, the inverse of the mean delay between autoignition spots $1/\langle \Delta t_{IGN} \rangle$, which is not exactly the same as the mean autoignition frequency $\langle f_{IGN} \rangle$, increases very quickly as L_{MIN} decreases (i.e. T_{air} increases for the same U_{air} , or $U_{fuel} \approx U_{air}$ decrease for the same T_{air}). Specifically, it increases by 3 orders of magnitude from about 1 Hz to about 1 kHz for the

conditions tested. In the small injector geometry, when autoignition in the random spots regime is possible at shorter L_{MIN} without flashback, the rise in $1/\langle\Delta t_{IGN}\rangle$ is particularly steep at very short L_{MIN} (i.e. high T_{air}).

We postulate that, locally, the intermittent availability of ζ_{MR} , but also the fluctuations in χ that controls thermal runaway, would also determine the frequency of appearance of the autoignition events, through f_{IGN} . It is possible that the higher f_{IGN} at short z are driven by the larger fluctuations in ζ and χ that were measured here (refer to Section 3.2 and Figure 6). If our postulate concerning the role of the fluctuations of ζ and χ is correct, then we would also expect that increasing U_{air} while keeping L_{MIN} the same would result in an increase in $1/\langle\Delta t_{IGN}\rangle$, and also $\langle f_{IGN}\rangle$, since we know from the investigation of the mixing field that higher U_{air} (i.e. Re_M) increase the fluctuations of ζ and χ (see Figure 6). In fact, we would expect this effect to be more pronounced in the original geometry, as we know that in the small injector flows the fluctuations of ζ and χ are not as strongly affected by changes to U_{air} (i.e. Re_M). Indeed, this is precisely what is observed in Figure 17.

It is instructive to compare these findings with those reported in Ref. [20] regarding the autoignition of ‘jet-in-hot-coflow flames’, where results are presented for two coflow conditions: (i) Case DJHC-I: maximum coflow temperature 1540 K, maximum coflow velocity 4.6 m/s, coflow oxygen mass fraction 8.4%; and, (ii) Case DJHC-V: maximum coflow temperature 1460 K, maximum coflow velocity 4.3 m/s, coflow oxygen mass fraction 9.5%. The effect of the coflow conditions was significant, with mean autoignition frequencies measured for DJHC-I at least twice higher than those measured for DJHC-V. Given that DJHC-I was associated with a shorter ‘lift-off height’ (akin to our L_{MIN}) than DJHC-V, this is consistent with our observations. Further, for Case DJHC-I, the mean autoignition frequency changed by no more than a factor of 2 across a range of jet Reynolds numbers from 3,000 to 9,500. For Case DJHC-V, the mean autoignition frequency was more sensitive to changes in jet Reynolds number, especially for jet Reynolds number larger than 4,000. For one fuel the frequency decreased by a factor of almost 5 over a range of Reynolds numbers from 4,000 to 6,000. For a second fuel the frequency decreased by a factor of

about 3 over the range of Reynolds numbers 4,000 – 9,500. A direct comparison between these DJHC flows and our CTHC flows is difficult. Nevertheless, the authors' attempt to explain this result concerning the decrease in autoignition frequency with increasing jet Reynolds number in the light of the expected higher strain rates at the higher Reynolds numbers, is in line with our postulate concerning the effect of χ on $\langle f_{IGN} \rangle$. Interestingly however, in both coflow cases and for all fuels, a slight increase of approximately 20% in the autoignition frequency was reported as the jet Reynolds number was increased from 3,000 to 4,000.

5 Conclusions

The autoignition of ‘equal velocity’ (between the air and fuel streams) acetylene plumes in turbulent coflows of hot air has been studied to supplement a previous database of hydrogen and *n*-heptane autoignition, and to provide new information on the effects of turbulence intensity, turbulent lengthscale and injector diameter on the autoignition location (i.e. length). These parameters have not been studied experimentally before in the context of inhomogeneous autoignition problems. Autoignition appears in the form of localised ‘random spots’, associated with an explosive emergence from a very small spatial region from which flamelets propagate radially outwards before extinguishing, or occasionally, giving rise to ‘secondary events’. The propensity of the flamelets to propagate upstream and cause ‘flashback’ was found to weaken at higher bulk velocities, smaller injector diameters, lower turbulence intensities and larger turbulent lengthscales. In agreement with hydrogen, increasing the bulk (air and fuel) velocity while maintaining the lengthscale constant increases the autoignition length. For the same turbulence intensity the autoignition length increases when a larger injector diameter is used or when the turbulent lengthscale is reduced. A simultaneous decrease in turbulence intensity and increase in lengthscale causes a reduction in autoignition length. Furthermore, the frequency of appearance of the autoignition spots was measured, and was found to increase when autoignition occurred closer to the injector and also at higher air velocities. The observed trends are consistent with expectations from the dependence of the mixture fraction and scalar dissipation on geometrical and flow parameters. The results can serve as a test-bed for model development and validation.

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Tables

Table 1: Experimental investigation envelope – geometric variables. ‘Original’ geometry in bold. The data in the last two columns are based on hot wire measurements, as described in Section 3.1.

Case Study #	CTHC Apparatus Configuration	D [mm]	M [mm]	s %	x [mm]	d [mm]	x/M [-]	d/M [-]	u'/U [-]	L_{turb}/M [-]
CS1 (Effect of d)	‘Small injector’ ‘Original’	25.0	3.0	45	66	1.1 2.2	22	0.37 0.73	0.11-0.13	1.0-1.5
CS2 (Effect of x)	‘Small injector’ ‘Long injector’				66 105	1.1 35	22 35	0.37	0.11-0.13 0.09-0.10	1.0-1.5 1.2-1.7
CS3 (Effect of M)	‘Original’ ‘Small grid’		3.0 1.2	45 55	66 53	2.2	22 44	0.73 1.8	0.11-0.13	1.0-1.5 4.2-5.8

Table 2: Range of autoignition experiment conditions. Rows correspond to entries in Table 1.

Case Study #	T_{air} [K]	U_{air} [m/s]	F_{fuel} [-]	Y_{fuel} [-]	T_{fuel} [K]	U_{fuel} [m/s]	Re_M [-]	δ_{fuel} [-]	v_{fuel} [-]
CS1	864-912	11.5-29.8	C_2H_2/N_2	0.65 ± 0.05	788-902	13.5-35.4	360-880	1.3 ± 0.2	1.0 ± 0.2
	806-904	10.5-28.4			595-826	9.4-28.4	360-850		
CS2	864-912	11.5-29.8			788-902	13.5-35.4	360-880		
	854-910	14.3-32.5			827-884	17.6-38.2	450-970		
CS3	806-904	10.5-28.4			595-826	9.4-28.4	360-850		
	784-861	10.5-22.8			758-798	9.8-21.6	200-380		

Table 3: Comparison of autoignition results for acetylene from the current study in the ‘original’ geometry configuration, with former experimental results for hydrogen [11] and n-heptane [12] in the same geometry.

Fuel [Ref. #]	T_{air} [K]	U_{air} [m/s]	Y_{fuel} [-]	T_{fuel} [K]	U_{fuel} [m/s]	L_{MIN} [mm]
Acetylene (C_2H_2) [Current study]	806-904	10.5-28.4	0.65 ± 0.05	595-826	9.4-28.4	26-88
Hydrogen (H_2) [11]	941-966	18.0-27.3	0.20 ± 0.10	847-926	19.4-29.7	18-63
N-Heptane ($n-C_7H_{16}$) [12]	1104-1138	13.8-17.8	0.95 ± 0.05	1027-1043	14.3-21.3	34-103

Figures

Figure 2: Acetylene ‘random spots’ autoignition in CTHC. Direct RGB photography with $T_{air} = 877$ K, $U_{air} = 24.8$ m/s, $Y_{fuel} = 0.65$ and 160 ms exposure (far left); $T_{air} = 846$ K, $U_{air} = 12.1$ m/s, $Y_{fuel} = 0.70$ and 100 ms exposure (left); and two examples of normalized emission spectra with 100 s exposure (right)

Figure 3: L_{MIN} and its relation to $\langle L_{TRUE} \rangle$ for acetylene over a range of conditions. Lines on the left are PDFs of true autoignition lengths L_{TRUE} from individual autoignition spots captured by the high-speed CMOS camera. On the right are PDFs of autoignition lengths L_{IGN} measured with the ICCD camera in the same experiments. The horizontal axis is a normalised distance from the injector. Shown at the bottom are the reported measures of L_{IGN} in this paper, i.e. L_{MIN} from the ICCD (in red), and the true mean $\langle L_{TRUE} \rangle$ from the CMOS (in black)

Figure 4: Mean mixture fraction along the centreline in both geometries associated with CS1 for various Re_M (left) and in both geometries associated with CS3 for various Re_M (right)

Figure 5: Normalized radial profiles of mean mixture fraction plotted such that Gaussian dispersion appears as a straight line (left) and radial spreading as a function of distance from injection such that Gaussian dispersion appears horizontal (right)

Figure 6: RMS of mixture fraction fluctuations (left) and two-dimensional scalar dissipation rate (right) along the centreline for various Re_M in geometries associated with CS1

Figure 7: Minimum autoignition length as a function of bulk air temperature for CS1

Figure 8: Minimum autoignition length as a function of bulk air temperature for CS₂

Figure 9: Minimum autoignition length as a function of bulk air temperature for CS3

Figure 10: (a) Typical global OH* timeseries in the original geometry. Many superposed individual autoignition events (time-translated to commence at $t = 0$ s). Conditions: $T_{air} = 807$ K, $T_{fuel} = 762$ K, $U_{air} = 10.9$ m/s and $Y_{fuel} = 0.64$. (b) Averaged OH* profiles compiled over many events such as those in (a), one for each different condition. Also showing (bottom) fast camera images taken simultaneously during one of the events in (a) at 10 kHz. Flow direction upwards

Figure 11: Single autoignition spot starting from dark background, growing radially, giving rise to 'secondary events' and extinguishing, taken at 13.5 kHz. Conditions same as in Figure 10. The dashed arrow shows a 'secondary event'. Flow direction upwards

Figure 12: Typical PMT OH* signals in the original geometry. ‘Slow spotting’ (left) and ‘fast spotting’ regimes (right), at conditions: $T_{air} = 834$ K, $U_{air} = 16.1$ m/s; and $T_{air} = 843$ K, $U_{air} = 12.9$ m/s respectively

Figure 13: Sequential images of random spots with the small injector taken at 7 kHz. Conditions: $T_{air} = 885$ K and $U_{air} = 20.6$ m/s. Imaged RoI corresponds to an area of about 21 x 70 mm. The white arrows show internal reflections of the light emitted from the reacting regions. The dashed arrow in Frame 16 shows a ‘secondary event’ that follows from the emergence of the circled ‘primary event’ in Frame 15. Qualitatively identical phenomena were observed for all injectors, and conditions in the random spots regime. Flow direction from left to right

Figure 14: Mean of global chemiluminescence intensity as a function of L_{MIN} . Original geometry (left) with conditions: $U_{air} = 10.9$ m/s (circles) and $U_{air} = 17.3$ m/s (squares), with T_{air} in the range 806 – 825 K and 850 – 879 K respectively. Small injector geometry (right) with conditions: $U_{air} = 15.8$ m/s (circles), $U_{air} = 19.5$ m/s (squares) and $U_{air} = 29.7$ m/s (diamonds), with T_{air} in the range 869 – 884 K, 870 – 885 K and 885 – 920 K respectively

Figure 15: PDFs of Δt_{IGN} (left) and f_{IGN} (right) in the original geometry, spanning all conditions as the data in Figure 14 (left). The inset on the left shows six examples of PDFs shown in the main figure, but with the x-axis normalized by the corresponding mean $\langle \Delta t_{IGN} \rangle$ for each run

Figure 16: PDFs of Δt_{IGN} (left) and f_{IGN} (right) in the small injector geometry, spanning all conditions as the data in Figure 14 (right). The inset on the left shows the same PDFs, but with the x-axis normalized by the corresponding mean $\langle \Delta t_{IGN} \rangle$ for each run

Figure 17: $1/\langle\Delta t_{IGN}\rangle$ as a function of L_{MIN} in the original (left) and small injector (right) geometries.

Conditions as in the legend of Figure 14

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