

# **A Direct Numerical Simulation investigation of spherically expanding flames propagating in fuel droplet-mists for different droplet diameters and overall equivalence ratios**

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## **ABSTRACT**

Laminar and turbulent spherically expanding n-heptane flames in mono-sized fuel droplet mists have been simulated for a range of different overall equivalence ratios and droplet diameters using three-dimensional Direct Numerical Simulations (DNS). Flame wrinkling and the evolutions of flame surface area and burned gas volume have been investigated for spherically expanding spray and gaseous premixed flames with the same initial burned gas radius and overall equivalence ratios. It has been found that droplet-induced wrinkling for laminar flame kernels strengthens with increasing overall equivalence ratio and droplet diameter. However, the effects of droplet-induced flame wrinkling are masked by wrinkling induced by fluid motion in turbulent spherically expanding spray flames. The gaseous phase mixture within the flame has been found to have smaller equivalence ratios (predominantly fuel-lean) in comparison to the overall equivalence ratio for globally stoichiometric and fuel-rich droplet cases and this tendency strengthens with increasing droplet diameter. By contrast, it is possible to obtain higher local equivalence ratio values than the overall equivalence ratio in globally fuel-lean spray flames. The presence of droplets in the globally fuel-lean cases enhances the growth of flame surface area under laminar and turbulent conditions. However, for the laminar globally stoichiometric spray flame, flame surface area for small droplets grows faster than the corresponding laminar premixed flame and this tendency is observed also for turbulent globally fuel-rich spray flames. It has been found that the burned gas mass increases for large (small) droplets for overall fuel-lean (fuel-rich) mixtures for flame propagation in droplet-laden mixtures, which is in qualitative agreement with previous experimental findings.

**Keywords:** Droplet combustion, spherically expanding flame, Direct Numerical Simulations.

## 1. INTRODUCTION

Liquid fuels, used commonly in gas turbines, liquid rocket engines, industrial furnaces and Internal Combustion (IC) engines (e.g. Direct Injection (DI) and Compression Ignition (CI) engines), are still expected to meet the majority of worldwide energy demand in the next decades (Kuo and Acharya, 2012; U.S. Energy Information Administration, 2016). Thus, the improvement in combustion performance of liquid fuel sprays is a necessity for improved energy-efficiency and environmental friendliness (McGowan et al., 2010). Turbulent combustion of sprays involves evaporation of liquid droplets, mixing of fuel vapour with the surrounding air and interaction of droplets with the flame and flow field, differently from turbulent premixed combustion. The interaction of these complex processes needs a detailed understanding of underlying physics, which motivated several experimental, analytical and numerical investigations. Various spray flame configurations including statistically planar (Burgoyne and Cohen L, 1954; Greenberg et al., 1998; Miller and Bellan, 1999; Reveillon and Demoulin, 2007; Reveillon and Vervisch, 2000; Sreedhara and Huh, 2007; Wacks and Chakraborty, 2016a, 2016b; Wang and Rutland, 2005), spherically expanding (Hayashi et al., 1977; Lawes et al., 2006; Mizutani and Nakajima, 1973; Neophytou *et al.*, 2012; Ozel-Erol et al., 2018) and jet flames (Fujita et al., 2013; Nakamura et al., 2005; Onuma et al., 1977, 1974; Schroll et al., 2009; Wandel, 2014; Wandel et al., 2009) have been investigated by several authors.

The effects of droplet size and vapour concentration on the burning velocity have been investigated experimentally by Ballal and Lefebvre (1981) for iso-octane droplet-mists for the overall (i.e. liquid + gaseous phase) equivalence ratios of  $\phi_{ov} = 0.65$  and 1.0, and they revealed that decreasing droplet size clearly promotes the flame propagation. Hayashi et al. (1977) compared the propagation of spherically expanding spray flames with those of the spherical premixed

flames, under the same overall fuel-air ratio conditions. It was reported that flame speed can be enhanced by the presence of droplets under some conditions. Evaporation characteristics of droplets with diameters ranging from 400  $\mu\text{m}$  to 1000  $\mu\text{m}$  have been investigated for different equivalence ratios by Szekely and Faeth (1983) and Faeth (1987) for turbulent spray jet flames. Mizutani and Nakajima (1973) observed that the overall equivalence ratio has significant influences on the extent of burning and propagation speed for spray flames based on their experimental analyses. They further indicated that supplement kerosene droplets enhance the burning velocity of propane-air mixtures and these effects are more prominent for leaner mixtures than richer mixtures. It was pointed out by Lawes and Saat (2011) that the growth rate of spherical spray flame kernels is a result of the complex interaction of droplet diameter, equivalence ratio and turbulence intensity. They observed that the flame speed increases with increasing droplet size under laminar conditions for  $\phi_{ov} = 0.9$  but an increase in droplet diameter may have a detrimental effect on flame speed under weak turbulence intensities. A similar qualitative trend has been observed for laminar cases with  $\phi_{ov} = 1.2$  by Lawes and Saat (2011) but an enhancement of flame speed due to large droplets has been obtained for small values of turbulence intensity for this overall equivalence ratio.

Polymeropoulos (1984) analytically calculated the burning speed for different liquid fuels and verified his model for fuel-lean and stoichiometric mixtures by comparing the analytical results with experimental findings by Burgoyne and Cohen (1954) and Ballal and Lefebvre (1981), respectively. Silverman et al. (1993) analytically derived an expression of laminar burning velocity for polydisperse laminar spray flames, and revealed that the sauter mean diameter (SMD) is not sufficient to determine the burning rate of polydisperse sprays and the distribution of droplet diameters plays a key role in determining the laminar burning velocity. Greenberg et al.

(1998) proposed an analytical model to explain the enhancement of flame speed in spray flames due to flame distortion induced by droplets. Greenberg and Kalma (2000) examined the spherical flame propagation in fuel-droplet mists for the first time for overall fuel-rich mixtures. Additionally, Greenberg (2007) analysed the effects of finite rate evaporation and droplet drag. It was noted that drag force leads to higher evaporation rate for droplets due to the longer residence time at a given location.

The advancements in high-performance computing have made it possible to carry out carrier phase Direct Numerical Simulations (DNS) of turbulent combustion of droplet-laden mixtures where the droplets are tracked in a Lagrangian sense, whereas the gaseous phase is treated in the standard Eulerian frame of reference. Miller and Bellan (1999) analysed evaporation of droplets in a mixing layer configuration and indicated that the initial Stokes number did not have a major effect on the mixing layer growth. The autoignition process of n-heptane fuel droplets for a variety of initial temperature and overall equivalence ratios has been analysed by Wang and Rutland (2005) and Schroll et al. (2009). It has been found that ignition delay time increases with the enhancement of initial equivalence ratio due to the decrease in gas temperature during the evaporation process. The carrier phase DNS was utilised by Reveillon and Vervisch (2000), Reveillon and Demoulin (2007), Sreedhara and Huh (2007) and Xia and Luo (2010) to analyse modelling of different aspects of turbulent combustion of droplet-laden mixtures (e.g. variance of mixture fraction in turbulent spray flames). Wandel et al. (2009) and Wandel (2014) used compressible DNS with single-step Arrhenius type chemistry to analyse the influences of equivalence ratio and droplet size on the success of localised ignition of mono-dispersed fuel droplets, and revealed that the increases in droplet diameter and scalar dissipation rate of progress variable are detrimental to the successful ignition. Neophytou et al. (2012)

extended the analysis by Wandel et al. (2009) by carrying out three-dimensional detailed chemistry DNS of forced ignition of n-heptane droplet-air mixtures and the simulation data has been utilised to analyse the flame structure and edge flame propagation statistics. Luo et al. (2011) analysed an n-heptane turbulent spray flame in a swirl flow configuration using DNS with one-step irreversible reaction and proposed a new parameterisation of the probability density function of mixture fraction based a-priori analysis of DNS data. Wacks et al. (2016) and Wacks and Chakraborty (2016a) carried out three-dimensional simple chemistry DNS of statistically planar flames propagating into mono-sized droplet mist for different droplet diameters and overall equivalence ratios. This revealed that a significant proportion of heat release rate in these flames arises due to premixed mode of combustion and combustion predominantly takes place in fuel-lean mode for either overall stoichiometric or overall fuel-rich mixtures. These findings are also consistent with the findings of Fujita et al. (2013) based on two-dimensional DNS of spray jet flames. Wacks et al. (2016) and Wacks and Chakraborty (2016a,b) used DNS data to analyse the statistical behaviours of displacement speed and gradients of reaction progress variable and mixture fraction for flames propagating into turbulent droplet-laden mixtures.

To date, DNS studies on spherically expanding flames in droplet-laden mixtures have mostly been performed in the context of localised ignition (Neophytou et al., 2012; Schroll et al., 2009; Wang and Rutland, 2005; Wandel, 2014) and only a relatively limited number of analyses (Greenberg, 2007; Ozel Erol et al., 2018) focussed on the influences of droplet size and equivalence ratio on flame propagation in droplet-laden mixtures, which have been reported by several experimental investigations (Ballal and Lefebvre, 1981; Hayashi et al., 1977; Lawes and Saat, 2011) in the past. However, spherically expanding flames are of fundamental importance in Internal Combustion (IC) engines and accidental explosions. Because of the fundamental

importance of spherically expanding flames, this configuration is often used as a canonical configuration for laboratory-scale experiments and numerical simulations. Furthermore, it was reported by Mizutani and Nishimoto (1972) that burning characteristics in spherically expanding spray flames are different from the corresponding open burner flames. Moreover, previous analyses (Chakraborty and Klein, 2009; Chakraborty et al., 2007, 2011) on premixed flames revealed that the flame propagation and reactive scalar gradient statistics and their stretch rate dependence for spherically expanding flames can be considerably different from statistically planar flames. Recently, Ozel-Erol et al. (2018) carried out three-dimensional carrier phase DNS with modified single step chemistry to analyse spherically expanding flames propagating into droplet-mists for different droplet diameters and turbulence intensities for an overall equivalence ratio of unity (i.e.  $\phi_{ov} = 1.0$ ). The DNS data by Ozel-Erol et al. (2018) demonstrated the influences of droplet-induced flame wrinkling under laminar conditions, and also for small turbulence intensities. However, these effects have been shown to be masked by flow-induced flame wrinkling for large turbulence intensities, which is consistent with experimental findings of Hayashi et al. (1977) and Laws and Saat (2011). Furthermore, it has been demonstrated by Ozel-Erol et al. (2018) that combustion takes place predominantly under fuel-lean premixed mode even for  $\phi_{ov} = 1.0$ , which is consistent with previous findings by Wacks et al. (2016) and Wacks and Chakraborty (2016a,b). The current analysis extends the analysis by Ozel-Erol et al. (2018) by carrying out carrier phase three-dimensional compressible DNS for spherically expanding spray flames for different overall equivalence ratios (i.e.  $\phi_{ov} = 0.8, 1.0$  and  $1.2$ ) for a range of different initial droplet diameters and turbulence intensities. In this respect, the main objectives of this study are:

(a) To reveal the influences of overall equivalence ratio on the flame structure under laminar and turbulent flow conditions in spherically expanding spray flames for a range of different

droplet diameters.

(b) To compare the evolution of flame surface area and volume of burned gas in statistically spherical spray flames to the corresponding values in statistically spherical premixed flames for a given overall equivalence ratio.

The rest of this paper will be organised in the following manner. The next two sections will focus upon the mathematical background and numerical implementation related to this analysis. This will be followed up by the presentation of the results and their discussion. The summary of the main findings along with the conclusions are provided in the final section of this paper.

## 2. MATHEMATICAL BACKGROUND

A modified single-step Arrhenius-type irreversible chemical reaction (Tarrazo et al., 2006) is used for the current analysis for the purpose of a detailed parametric analysis:



where  $s$  is the mass of the oxygen consumed per unit mass of fuel consumption. Based on the modified Arrhenius-type chemical mechanism, the fuel reaction rate can be expressed as (Wacks et al., 2016; Wacks and Chakraborty, 2016a,b; Wandel, 2014; Wandel et al., 2009):

$$\dot{w}_F = -\rho B^* Y_F Y_O \exp\left(-\frac{\beta(1-T)}{1-\alpha(1-T)}\right) \quad (2)$$

where  $\rho$  is the gas density,  $Y_F$  and  $Y_O$  are the fuel and oxygen mass fractions, respectively. The heat release parameter  $\alpha$ , the normalised pre-exponential factor  $B^*$ , the non-dimensional temperature  $T$  and the Zel'dovich number  $\beta$ , can be given as follows:

$$\alpha = \frac{\tau}{\tau+1}, B^* = B \exp\left(-\frac{\beta}{\alpha}\right), T = \frac{\hat{T}-T_0}{T_{ad(\phi_g=1)}-T_0}, \beta = \frac{E_{ac}(T_{ad(\phi_g=1)}-T_0)}{RT_{ad(\phi_g=1)}^2} \quad (3)$$

Here  $\tau = (T_{ad(\phi_g=1)} - T_0)/T_0$  is a heat release parameter,  $B$  is the pre-exponential factor,  $\hat{T}$  is the instantaneous dimensional temperature,  $T_0$  is the unburned gas temperature,  $T_{ad(\phi_g=1)}$

is the adiabatic flame temperature for the stoichiometric mixture,  $E_{ac}$  is the activation energy and  $R$  is the universal gas constant. According to the model of Tarrazo et al. (2006), the activation energy,  $E_{ac}$ , and the heat of combustion can be calculated based on the gaseous equivalence ratio,  $\phi_g$  which ensures a reasonable equivalence ratio  $\phi_g$  dependence of the unstrained laminar burning velocity  $S_{b(\phi_g)}$  in hydrocarbon-air flames. According to Tarrazo et al. (2006) the Zel'dovich number,  $\beta$ , can be expressed as:  $\beta = 6f(\phi_g)$  where  $f(\phi_g)$  is defined depending on the gaseous phase equivalence ratio  $\phi_g$  as:

$$f(\phi_g) = \begin{cases} 1.0 + 8.250(\phi_g - 0.64)^2 & , \phi_g \leq 0.64 \\ 1.0 & , 0.64 < \phi_g < 1.07 \\ 1.0 + 1.443(\phi_g - 1.07)^2 & , \phi_g \geq 1.07 \end{cases} \quad (4)$$

The heat release per unit mass of fuel  $H_{\phi_g} = [(T_{ad(\phi_g)} - T_0) C_p] / [Y_{F0(\phi_g)} - Y_{Fb(\phi_g)}]$  is given by  $H_{\phi_g}/H_{\phi_g=1} = 1$  for  $\phi_g \leq 1$  and  $H_{\phi_g}/H_{\phi_g=1} = 1 - \alpha_H(\phi_g - 1)$  for  $\phi_g > 1$ , where  $\alpha_H=0.18$ ,  $C_p$  is the specific heat at constant pressure,  $Y_{F0(\phi_g)}$  and  $Y_{Fb(\phi_g)}$  are the fuel mass fractions in the unburned and burned gases, respectively for a premixed flame of equivalence ratio  $\phi_g$  (Tarrazo et al., 2006). The modified single-step chemical mechanism applied for this analysis provides realistic equivalence ratio dependence of unstrained laminar burning velocity, specifically for fuel-rich mixtures for hydrocarbon fuels and interested readers are referred to Tarrazo et al. (2006) and Malkeson and Chakraborty (2010) for the change of the laminar burning velocity  $S_{b(\phi_g)}$  with equivalence ratio  $\phi_g$  for methane-air mixtures. However, it has been demonstrated by Swaminathan and Bray (2011) that normalised laminar burning velocity  $S_{b(\phi_g)} / \{S_{b(\phi_g)}\}_{max}$  dependence of equivalence ratio  $\phi_g$  is not sensitive to the fuel type for hydrocarbon-air mixtures but it is affected strongly by the unburned gas temperature. The variations of  $S_{b(\phi_g)} / \{S_{b(\phi_g)}\}_{max}$  and  $T_{ad(\phi_g)}$  with  $\phi_g$  for the present thermo-

chemistry have been found to be in good agreement with experimental findings (Kumar et al., 2007) and the results obtained from a detailed chemical mechanism (Chaos et al., 2007).

The initialisation of spherical flames with non-unity Lewis number ( $Le_i = \alpha_i/D_i$  where  $\alpha_i$  is the thermal diffusivity and  $D_i$  is the species diffusivity of  $i^{th}$  species) is not straightforward and computationally expensive. Moreover, spherical flames with  $Le < 1$  expand rapidly due to thermo-diffusive instabilities so keeping the flame within the computational domain for a meaningful timespan becomes extremely computationally expensive. In addition, a non-unity value of Lewis number complicates the expression used for the corrected Sherwood number  $Sh_c$ , which is used for the dispersed phase modelling (see Eq. 7 later in the paper). Furthermore, the thermo-diffusive effects induced by non-unity Lewis number significantly affect the burning rate and flame wrinkling statistics, and thus it becomes difficult to isolate the effects of flame wrinkling induced by droplets and fluid turbulence from the ones due to differential diffusion of heat and mass. Thus, all the species are considered to be of unity Lewis number for this analysis following several previous analyses (Reveillon and Demoulin, 2007; Reveillon and Vervisch, 2000; Wandel, 2014; Wandel et al., 2009; Wang et al., 2005) for the purpose of simplicity so that the effects of droplet-induced flame wrinkling effects can be analysed in absence of the additional complexities induced by differential diffusion of heat and mass. In spite of the unity Lewis number assumption, the present thermo-chemistry captures the variations of normalised unstrained laminar burning velocity  $S_{b(\phi_g)}/\{S_{b(\phi_g)}\}_{max}$  and adiabatic flame temperature  $T_{b(\phi_g)} = (T_{ad(\phi_g)} - T_0)/(T_{ad(\phi_g=1)} - T_0)$  with gaseous phase equivalence ratio  $\phi_g$  reasonably accurately when compared to experimental data (Kumar et al., 2007) and with the results obtained from a detailed chemical mechanism (Chaos et al., 2007).

In this analysis, all species in the gaseous phase are assumed to be perfect gases. Standard values have been used for the ratio of specific heats ( $\gamma = C_p^g/C_v^g = 1.4$ , where  $C_p^g$  and  $C_v^g$  are the gaseous specific heats at constant pressure and volume respectively) and Prandtl number ( $Pr = \mu C_p^g/\lambda = 0.7$  where  $\mu$ , is the dynamic viscosity and  $\lambda$  is the thermal conductivity of the gaseous phase).

A Lagrangian approach is used to simulate droplets, whereas the Eulerian approach is adopted to solve compressible Navier-Stokes equations for the carrier gaseous phase. The liquid phase equations for the position,  $\vec{x}_d$ , velocity,  $\vec{u}_d$ , diameter  $a_d$ , and temperature  $T_d$ , can be written following the approach proposed by Reveillon and Vervisch (2000):

$$\frac{d\vec{x}_d}{dt} = \vec{u}_d; \quad \frac{d\vec{u}_d}{dt} = \frac{\vec{u}(\vec{x}_d, t) - \vec{u}_d}{\tau_d^u}; \quad \frac{da_d^2}{dt} = \frac{a_d^2}{\tau_d^p} \quad \text{and} \quad \frac{dT_d}{dt} = \frac{\hat{T}(\vec{x}_d, t) - T_d - B_d L_v / C_p^g}{\tau_d^T} \quad (5)$$

where  $L_v$  is the latent heat of vaporization, and  $\tau_d^u$ ,  $\tau_d^p$  and  $\tau_d^T$  are relaxation time scales associated with droplet velocity, diameter and temperature, respectively which are defined as (Reveillon and Demoulin, 2007; Reveillon and Vervisch, 2000; Wandel, 2014; Wandel et al., 2009):

$$\tau_d^u = \frac{\rho_d a_d^2}{18 C_u \mu}; \quad \tau_d^p = \frac{\rho_d a_d^2}{4 \mu} \frac{Sc}{Sh_c} \frac{1}{\ln(1+B_d)}; \quad \tau_d^T = \frac{\rho_d a_d^2}{6 \mu} \frac{Pr}{Nu_c} \frac{B_d}{\ln(1+B_d)} \frac{C_p^L}{C_p^g} \quad (6)$$

where  $\rho_d$  is the droplet density,  $C_p^L$  is the specific heat for the liquid phase,  $C_u = 1 + Re_d^{2/3}/6$  is the corrected drag coefficient. Here,  $Re_d$  is the droplet Reynolds number,  $Sc$  is the Schmidt number,  $B_d$  is the Spalding mass transfer number,  $Sh_c$  is the corrected Sherwood number and  $Nu_c$  is the corrected Nusselt number, which are expressed as (Reveillon and Demoulin, 2007; Reveillon and Vervisch, 2000b; Wacks et al., 2016; Wacks and Chakraborty, 2016a; Wandel, 2014; Wandel et al., 2009):

$$Re_d = \frac{\rho |\vec{u}(\vec{x}_d, t) - \vec{u}_d| a_d}{\mu}; B_d = \frac{Y_F^s - Y_F(\vec{x}_d, t)}{1 - Y_F^s}; Sh_c = Nu_c = 2 + \frac{0.555 Re_d Sc}{(1.232 + Re_d Sc^{4/3})^{1/2}} \quad (7)$$

where  $Y_F^s$  denotes the fuel mass fraction at the surface of the droplet and can be written as:  $Y_F^s = (1 + W_{air}/W_F [p(\vec{x}_d, t)/p_F^s - 1])^{-1}$  where  $W_{air}$  and  $W_F$  are the molecular weights of air and fuel, respectively. Here  $p_F^s$  indicates the partial pressure of the fuel vapor at the droplet surface, and can be defined based on the Clausius–Clapeyron equation:  $p_F^s = p_{ref} \exp(L_v/R [1/T_{ref}^s - 1/T_d^s])$  where  $T_{ref}^s$  is the boiling point of the fuel at pressure  $p_{ref}$  and  $T_d^s$  is the droplet surface temperature.

For large values of the droplet Reynolds number (i.e.  $Re_d \gg 1$ ) drag coefficient is corrected as  $C_u = 1 + Re_d^{2/3}/6$  in this study. The same expression was employed in many previous analysis for spray combustion simulations (Reveillon and Demoulin, 2007; Reveillon and Vervisch, 2000b; Sreedhara and Huh, 2007; Wang and Rutland, 2005; Xia and Luo, 2010). In the simulations considered in this analysis, the droplet Reynolds number  $Re_d$  remains either of the order of unity or smaller than unity (i.e.  $Re_d \leq 1$ ) in most (~95%) locations (especially in the vicinity of the flame) and therefore the exact nature of the corrections to the drag coefficient may not have a major influence on the simulation results. Accordingly, a simple drag coefficient correction is assumed in this analysis. Alternative empirical drag coefficients, considering the blowing velocity at the droplet surface, were adopted in some previous analyses (Baba and Kurose, 2008; Miller and Bellan, 1999; Nakamura et al., 2005; Watanabe et al., 2008, 2007) but the exact nature of empiricism is likely to have higher order effects for small values of  $Re_d$  when DNS is performed for the carrier phase and the droplets are treated as sub-grid point sources.

The coupling between Lagrangian and Eulerian phases is obtained by the additional source

terms in the gaseous phase transport equations which can be expressed in the following generic form (Reveillon and Vervisch, 2000; Wandel, 2014; Wandel et al., 2009):

$$\frac{\partial \rho \psi}{\partial t} + \frac{\partial \rho u_j \psi}{\partial x_j} = \frac{\partial}{\partial x_j} \left( \Gamma_\psi \frac{\partial \psi_1}{\partial x_j} \right) + \dot{w}_\psi + \dot{S}_g + \dot{S}_\psi \quad (8)$$

where  $\psi = \{1, u_i, e, Y_F, Y_O\}$  and  $\psi_1 = \{1, u_i, \hat{T}, Y_F, Y_O\}$  for the conservation equations of mass, momentum, energy, and mass fractions, respectively and  $\Gamma_\psi = \rho \nu / \sigma_\psi$  for  $\psi = \{1, u_i, Y_F, Y_O\}$  and  $\Gamma_\psi = \lambda$  for  $\psi = e$ , respectively. Here,  $u_i$  and  $e$  denote the velocity in the  $i^{th}$  direction and the specific stagnation internal energy, respectively. The  $\dot{w}_\psi$  term arises due to chemical reaction rate,  $\dot{S}_g$  is an appropriate source/sink term in the gaseous phase and  $\dot{S}_\psi$  is the appropriate source term associated with droplet evaporation, which is tri-linearly interpolated from the droplet's sub-grid position,  $\vec{x}_d$ , to the eight surrounding nodes. Kinematic viscosity is  $\nu$  and an appropriate Schmidt number corresponding to  $\psi$  is represented by  $\sigma_\psi$ . The droplet source term for any variable  $\psi$  is defined as (Neophytou et al., 2012; Schroll et al., 2009; Wandel, 2014; Wandel et al., 2009):

$$\dot{S}_\psi = -\frac{1}{V} \sum_d \frac{d(m_d \psi_d)}{dt} \quad (9)$$

where,  $V$  is the cell volume,  $m_d = \rho_d (1/6) \pi a_d^3$  is the droplet mass.

A reaction progress variable,  $c$ , can be expressed based on the oxygen mass fraction,  $Y_O$  and mixture fraction,  $\xi = (Y_F - Y_O/s + Y_{O\infty}/s)/(Y_{F\infty} + Y_{O\infty}/s)$  in such a manner that it increases from 0 (in unburned reactants) to 1 (in burned products) (Neophytou et al., 2012; Wacks et al., 2016; Wacks and Chakraborty, 2016a; Wandel, 2014; Wandel et al., 2009):

$$c = \frac{(1-\xi)Y_{O\infty} - Y_O}{(1-\xi)Y_{O\infty} - \max(0, [\xi_{st} - \xi]/\xi_{st})Y_{O\infty}} \quad (10)$$

where  $Y_{O\infty} = 0.233$  is the oxygen mass fraction in air and  $Y_{F\infty} = 1.0$  is the fuel mass fraction

in the pure fuel stream  $s$  is given by:  $s = 3.52$  for n-heptane,  $C_7H_{16}$ , which leads to stoichiometric fuel mass fraction and mixture fraction values as  $Y_{Fst} = \xi_{st} = 0.0621$ .

### 3. NUMERICAL IMPLEMENTATION

Simulations have been conducted using a three dimensional compressible DNS code SENGAs (Neophytou et al., 2012; Schroll et al., 2009; Wacks et al., 2016; Wacks and Chakraborty, 2016a; Wandel, 2014; Wandel et al., 2009), which solves the standard conservation equations of mass, momentum, energy and species of the gaseous phase in non-dimensional form. The spatial differentiation is performed using a 10<sup>th</sup> order central difference scheme for the internal grid points, but the order of differentiation gradually decreases to a one-sided 2<sup>nd</sup> order scheme at the non-periodic boundaries. An explicit low-storage 3<sup>rd</sup> order Runge-Kutta scheme (Wray, 1990) is employed to calculate the time advancement. Partially non-reflecting boundary conditions are applied to all directions of the cubic domain of size  $84.49\delta_z \times 84.49\delta_z \times 84.49\delta_z$  where  $\delta_z = \alpha_{T0}/S_{b(\phi_g=1)}$  is the Zel'dovich flame thickness with  $\alpha_{T0}$  and  $S_{b(\phi_g=1)}$  being the thermal diffusivity in the unburned gas and unstrained laminar burning velocity for the stoichiometric mixture, respectively. The partially non-reflecting boundary conditions are specified using the Navier-Stokes Characteristic Boundary Conditions (NSCBC) technique (Poinsot and Lele, 1992). A uniform computational grid of  $512 \times 512 \times 512$  is used to discretise the domain, which ensures about 10 grid points within  $\delta_{st} = (T_{ad(\phi_g=1)} - T_0)/\max|\nabla T|_L$ , which is the unstrained thermal laminar flame thickness of the stoichiometric mixture.

The reacting flow field is initialised using COSILAB (Rotexo-Softpredict-Cosilab) following Neophytou and Mastorakos (2009) for three different initial values of droplet diameter  $a_d$  (i.e.

$a_d/\delta_{st} = 0.04, 0.05$  and  $0.06$ ) and overall equivalence ratios  $\phi_{ov}$  (i.e.  $\phi_{ov} = 0.8, 1.0$  and  $1.2$ ) as described in a previous study by Ozel-Erol et al. (2018). For the present analysis, initially mono-sized droplets have been considered, so that the effects of droplet diameter on flame-droplet interaction can be analysed in isolation. Silverman et al. (1993) suggested that polydispersity may lead to an increase or decrease in local SMD, which is different from a monotonically decreasing trend in the case of a monodisperse droplet size distribution. By contrast, Jaegle et al. (2011) reported that LES simulations with monodisperse and polydisperse droplet distributions show similar behaviours and monodisperse droplet diameter can be a good representation of the polydisperse diameter distribution in terms of droplet dynamics for the cases considered in their analysis. More analysis in this regard will be necessary but mono-sized droplets are considered here for the reasons mentioned above and also for the purpose of simplicity.

A perfectly spherical kernel flame is placed at the centre of the domain and an incompressible homogeneous isotropic velocity field (Rogallo, 1981) is superimposed on the laminar spherical flames when the radius of the fully burned gas region (i.e. the region corresponding to reaction progress variable  $c$  values greater than 0.99)  $r_0$  reaches  $2\delta_{st}$  (i.e.  $r_0/\delta_{st} = 2.0$ ). It is important to note that the energy content for a premixed spherical flame with radius  $r_0 = 2\delta_{st}$  is different from a spherical droplet flame with the same burned gas radius (i.e.  $c \geq 0.99$ ) for a given equivalence ratio. However, the same initial burned gas (i.e.  $c \geq 0.99$ ) radius (i.e.  $r_0/\delta_{st} = 2.0$ ) has been considered here for the purpose of comparison. The unburned gas temperature  $T_0$  is assumed to be 300 K and this implies a heat release parameter  $\tau = (T_{ad(\phi_g=1)} - T_0)/T_0$  of 6.54. The turbulent flame simulations are carried out for initial value

of normalised root-mean-square (rms) turbulent velocities  $u'/S_{b(\phi_g=1)} = 4.0$  with a non-dimensional longitudinal integral length-scale of  $L_{11}/\delta_{st}=2.5$ , and have been continued for  $2.52\alpha_{T0}/S_{b(\phi_g=1)}^2$ , which corresponds to about 2.0 initial eddy turnover times (i.e.  $2L_{11}/u'$ ).<sup>1</sup> The initial turbulent Reynolds number  $Re_t = \rho_0 k^2 / \mu_0 \varepsilon$  (where  $\rho_0$  and  $\mu_0$  are the unburned gas density and viscosity respectively) based on turbulent kinetic energy  $k$  and its dissipation rate  $\varepsilon$  for the turbulent cases considered here remains about 70. The simulation time (i.e.  $2.52t^+ = 2.52\alpha_{T0}/S_{b(\phi_g=1)}^2$ ) used in this analysis remains comparable to a number of recent DNS analyses (Grout, 2007; Han and Huh, 2008; Neophytou et al., 2012, 2010; Pera et al., 2013; Reddy and Abraham, 2012; Wandel, 2014; Wandel et al., 2009), which provided valuable insights into the fundamental understanding of turbulent combustion. The turbulent kinetic energy evaluated over the whole domain does not vary rapidly with time at  $2.52\alpha_{T0}/S_{b(\phi_g=1)}^2$ . The rms velocity fluctuation evaluated over the whole domain decayed by about 40% when the statistics were extracted (i.e.  $2.52t^+ = 2.52\alpha_{T0}/S_{b(\phi_g=1)}^2$ ).

The simulation parameters are summarised in Table 1. The initial droplet number density  $\rho_N$  ranges between  $1.28 \leq (\rho_N)^{1/3} \delta_{st} \leq 2.19$  in the unburned gas, and the liquid volume fraction remains well below 0.01. The droplet diameter remains smaller than the Kolmogorov length scale for all cases and the ratio of initial droplet diameter to the Kolmogorov length scale is  $a_d/\eta = 0.15, 0.19, 0.23$  for  $a_d/\delta_{st} = 0.04, 0.05, 0.06$  respectively. The Stokes number  $St = \tau_p \sqrt{k}/L_{11} = \rho_d a_d^2 \sqrt{k}/(18C_u \mu L_{11})$  (where  $\tau_p = \rho_d a_d^2 / 18C_u \mu$  is the particle time

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<sup>1</sup> It is worth noting that in order to ensure that all cases are subjected to same initial turbulence both  $u'$  and  $L_{11}$  are kept unchanged. This means that  $u'/S_{b(\phi_g)}$  and  $L_{11}/\delta_{(\phi_g)}$  values are different for  $\phi_{ov} = 0.8$  (i.e.  $u'/S_{b(\phi_g=0.8)} = 6.66$  and  $L_{11}/\delta_{(\phi_g=0.8)} = 1.47$ ) and 1.2 (i.e.  $u'/S_{b(\phi_g=1.2)} = 4.76$  and  $L_{11}/\delta_{(\phi_g=1.2)} = 2.77$ ) cases where  $\delta_{(\phi_g)}$  is the thermal flame thickness for the equivalence ratio  $\phi_g$ .

scale and  $L_{11}/\sqrt{k}$  is the turbulent time scale) for the largest droplet remains smaller than 0.1 in the turbulent cases considered here. Alternatively, the Stokes number  $St' = \tau_p S_b^2(\phi_g=1)/\alpha_{T0} = \rho_d a_d^2 S_b^2(\phi_g=1)/(18C_u \mu \alpha_{T0})$  can be calculated based on  $\alpha_{T0}/S_b^2(\phi_g=1)$  and the maximum value of  $St'$  remains smaller than  $5.0 \times 10^{-2}$  for the largest droplets considered in this analysis. The mean normalised inter-droplet distance  $s_d/\eta$  varies between 2.47 and 3.71. The ratio of the initial droplet diameter to grid spacing used here remains comparable to several previous analyses (Fujita et al., 2013; Neophytou et al., 2012, 2010; Schroll et al., 2009; Wandel, 2014; Wandel et al., 2009; Wang and Rutland, 2005). Due to the relatively high volatility of n-heptane, the droplet diameter decreases significantly by the time the droplets reach the most reacting place in the reaction zone. The droplet diameter decreases at least by 50% when it reaches the reaction zone. As a result, the flame interacts with the droplets, which are much smaller in reality than the initial size of the droplets. Thus, the assumption of sub-grid evaporation is not expected to affect the statistics of flame-droplet interaction, which is the subject of this analysis. It is worth noting that a recent analysis by Haruki et al. (2018) demonstrated that the point source assumption accurately captures the evaporation characteristics obtained from fully resolved multi-phase simulations for droplets with sizes smaller than the Kolmogorov length scale. The Eulerian approaches, which resolve both gaseous and liquid phases, also have some limitations (e.g. capturing polydispersity and droplet crossing). de Chaisemartin *et al.* (2009) compared combustion of polydisperse droplets in a two-dimensional free jet using carrier phase DNS with point source and fully Eulerian phase-DNS. The results by de Chaisemartin *et al.* (2009) showed a good agreement between gaseous fuel mass fraction fields obtained from these two approaches. These authors also reported that simulations with the full Eulerian approach were 10 times more expensive than those with the point source approach and a huge parallelisation capability for 3D simulations is needed for the Eulerian phase DNS for

sprays. Moreover, combustion simulations involving fully resolved dispersed phase are currently in a primitive stage and need further validation to be used on a routine basis. Thus, a point source assumption for the dispersed phase (i.e. droplets) has been considered in this analysis.

## 4. RESULTS AND DISCUSSION

### 4.1 Flame-turbulence interaction

Figures 1-3 show the instantaneous distributions of reaction progress variable  $c$ , normalised gaseous fuel mass fraction  $Y_F/Y_{Fst}$ , and non-dimensional temperature  $T$ , fields, respectively, at the central  $x - y$  mid-plane at  $t = 2.52\alpha_{T0}/S_b^2(\phi_g=1)$  under laminar and turbulent flow conditions (with initial  $u'/S_b(\phi_g=1) = 4.0$ ) and for overall equivalence ratios  $\phi_{ov} = 0.8, 1.0$  and 1.2. The droplets residing on the  $x - y$  mid-plane are shown by black dots in Figs. 1-3. It is worth noting that these droplets are individually tracked in a Lagrangian manner and they are not meant to indicate individual droplet burning. The group number  $G = 3(1 + 0.276Re_d^{1/2}Sc^{1/3})LeN^{2/3}(a_d/s_d)$  (where  $Le$  and  $Sc$  are the Lewis and Schmidt numbers respectively,  $N$  is the number of droplets in a specified volume and  $s_d$  is the mean inter-droplet distance) according to Chiu and Liu (1977) remains much greater than unity for the cases considered here and thus these cases represent the external sheath combustion ( $G \gg 1.0$ ) (Reveillon and Vervisch, 2000). During the evaporation process, the droplets reduce in size and create gaseous fuel clouds in their surroundings (examples are shown in the insets of Fig. 1-3). Small droplets mostly evaporate before reaching the flame front whereas large droplets continue to evaporate across the flame, as can be seen in Figs. 1-3. For globally stoichiometric (i.e.  $\phi_{ov} = 1.0$ ) droplet cases, unburned fuel pockets can appear in the burned gas region notably for large droplets. However, these unburned fuel pockets are obtained for all droplet sizes

for  $\phi_{ov} = 1.2$  under both laminar and turbulent conditions. Unburned fuel is mostly found ahead of the flame for droplet cases with  $\phi_{ov} = 0.8$  due to small number density of droplets. The latent heat of evaporation induces local temperature drop in the burned gas region for  $\phi_{ov} = 1.0$  and  $\phi_{ov} = 1.2$  (see Figs. 1 and 3) and this tendency is particularly prominent for large droplets (e.g.  $a_d/\delta_{st} = 0.06$ ). However, the burned gas temperature increases with increasing droplet diameter for  $\phi_{ov} = 0.8$  and the burned gas temperature for the initial  $a_d/\delta_{st} = 0.04$  case is found to be similar to that in the corresponding fuel-lean premixed case under both laminar and turbulent flow conditions. It can be seen from Fig. 3 that the burned gas temperature for  $\phi_g = 0.8$  cases is smaller than that in the corresponding  $\phi_g = 1.0$  cases due to predominantly fuel-lean combustion. The burned gas temperature for  $\phi_g = 1.2$  cases is also found to be marginally smaller than but comparable to that in the corresponding  $\phi_g = 1.0$  cases. This can be explained from the Burke-Schumann relation for the burned gas temperature<sup>2</sup>, which indicates that the drop of the burned gas temperature for  $\phi_g = 0.8$  is expected to be greater than that in case of  $\phi_g = 1.2$ . A comparison between Figs. 1 and 2 reveals that the fuel mass fraction in the unburned gas remains smaller than the corresponding value in the premixed flame case, which indicates that the evaporation of droplets is not sufficient to supply as much fuel in the gaseous phase as in the corresponding premixed flame case. Thus, the gaseous phase combustion in droplet cases takes place mostly under leaner conditions than in the corresponding premixed flame cases.

It can further be seen from Figs. 1-3 that the laminar premixed flames remain perfectly spherical

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<sup>2</sup>  $T_b(\xi) = T_{F\infty}\xi + T_{O\infty}(1 - \xi) + (1 - \xi)/(1 - \xi_{st})$  for  $\xi > \xi_{st}$  and  $T_b = T_{F\infty}\xi + T_{O\infty}(1 - \xi) + \xi/\xi_{st}$  for  $\xi \leq \xi_{st}$  where  $T_b$ ,  $T_{F\infty}$  and  $T_{O\infty}$  are non-dimensional burned gas temperature, pure fuel stream temperature and pure air stream temperature, respectively

but laminar spray flames exhibit weakly wrinkled  $c$  isosurfaces especially for large droplets (i.e.  $a_d/\delta_{st} = 0.06$ ). This tendency strengthens with increasing  $\phi_{ov}$ . For laminar globally stoichiometric (i.e.  $\phi_{ov} = 1.0$ ) spray flames, flame-droplet interaction can be seen from imperfectly spherical  $c$  isosurfaces. This is prominently evident in the cases with  $\phi_{ov} = 1.2$  because of the availability of larger number of droplets. Moreover, it can be discerned from the  $c$  contours in Figs. 1 and 2 that all cases exhibit instances of local flame thickening and this tendency is particularly prevalent for droplet cases especially for  $\phi_{ov} = 0.8$ . The initial values of Damköhler number (i.e.  $Da_{(\phi_g)} = L_{11}S_b^2(\phi_g)/u'\alpha_{T0}$ ) for the premixed flames with  $\phi_g = 0.8, 1.0$  and  $1.2$  with initial  $u'/S_b(\phi_g=1) = 4.0$  are 0.473, 1.31 and 0.93 respectively. The corresponding values of the Karlovitz number (i.e.  $Ka_{(\phi_g)} = (u'/S_b(\phi_g))^{1.5} (L_{11}S_b(\phi_g)/\alpha_{T0})^{-0.5}$ ) are 9.70, 3.50 and 4.85 for  $\phi_g = 0.8, 1.0$  and  $1.2$  respectively. The value of Damköhler (Karlovitz) number at least for the globally stoichiometric and fuel-rich droplet cases is likely to be smaller (greater) than the corresponding premixed flame cases because of the availability of leaner fuel-air mixture (which implies that the chemical time scale in droplet cases is likely to be larger than  $\alpha_{T0}/S_b^2(\phi_g)$  because of smaller burning velocity). The Damköhler and Karlovitz number values are representative of the distributed burning regime for all cases considered here, and thus they exhibit local flame thickening and these tendencies are particularly strong for droplet cases due to small (large) values of  $Da$  ( $Ka$ ).

## 4.2 Effects of droplets on flame wrinkling

The flame-droplet interaction can be discerned from Fig. 4 where the instantaneous  $c = 0.5$  isosurfaces are coloured by local values of flame curvature  $\kappa_m \times \delta_{st}$  for the cases considered here. The local curvature  $\kappa_m$  is defined as  $\kappa_m = \nabla \cdot \vec{N}/2$  where  $\vec{N} = -\nabla c/|\nabla c|$  is the local

flame normal vector. According to these definitions, an element of the flame surface, which is convex (concave) to the reactants, has a positive (negative) curvature and the flame normal vector points towards the reactants. Dimples can be seen from Fig. 4 for  $a_d/\delta_{st} = 0.05$  and  $0.06$  droplet cases under laminar conditions for  $\phi_{ov} = 0.8$ , whereas the  $c = 0.5$  isosurface is a smooth sphere in the corresponding premixed flame case. However, the  $c = 0.5$  isosurface for the laminar droplet case with initial  $a_d/\delta_{st} = 0.04$  for  $\phi_{ov} = 0.8$  does not show dimples but also does not remain spherical. A similar qualitative trend is observed for the turbulent droplet cases for  $\phi_{ov} = 0.8$  where the droplet-induced flame wrinkling increases with increasing droplet diameter. For  $\phi_{ov} = 1.0$  and  $1.2$ , dimples on the  $c = 0.5$  isosurface due to flame-droplet interaction can be seen for both laminar and turbulent cases. A comparison between laminar  $\phi_{ov} = 1.0$  and  $\phi_{ov} = 1.2$  cases reveals that evaporation of clustered droplets creates large distributed dimples for the  $\phi_{ov} = 1.2$  cases in contrast to small densely packed dimples in the  $\phi_{ov} = 1.0$  cases. This difference is eclipsed by flow-induced flame wrinkling in turbulent  $\phi_{ov} = 1.0$  and  $1.2$  cases. The droplet-induced flame wrinkling can be quantified in terms of the Probability Density Functions (PDFs) of  $\kappa_m$ . The PDFs of  $\kappa_m \times \delta_{st}$  for  $c = 0.1, 0.5$  and  $0.9$  isosurfaces for all cases considered here are shown in Fig. 5. The perfect spherical shape of laminar premixed flames leads to delta functions for the curvature PDFs but laminar droplet cases exhibit a distribution of curvature values, which is indicative of droplet-induced flame wrinkling. The extent of droplet-induced flame wrinkling is the smallest (largest) for the initial droplet size of  $a_d/\delta_{st} = 0.04$  ( $a_d/\delta_{st} = 0.06$ ) for  $\phi_{ov} = 0.8$  ( $\phi_{ov} = 1.2$ ). The combination of small number density of droplets and rapid evaporation of small droplets gives rise to a weak laminar flame front deformation for  $a_d/\delta_{st} = 0.04$  in the case of  $\phi_{ov} = 0.8$ . The number density of droplets increases with increasing  $\phi_{ov}$  for a given value of  $a_d/\delta_{st}$  and accordingly the extent of droplet-induced deformation of laminar flame increases with an increase in  $\phi_{ov}$ .

This is reflected in the widening of the PDF of  $\kappa_m \times \delta_{st}$  with increasing  $\phi_{ov}$  for a given value of  $a_d/\delta_{th}$  in the laminar droplet cases. The evaporation rate decreases with increasing  $a_d/\delta_{st}$  and thus the large droplets survive relatively longer within the flame, and thus the laminar flame front deformation is greater in extent for larger values of  $a_d/\delta_{st}$  for a given value of  $\phi_{ov}$ . This can be substantiated by the narrower PDFs of  $\kappa_m \times \delta_{st}$  in droplet cases for smaller values of  $a_d/\delta_{st}$  for a given value of  $\phi_{ov}$ . The signature of droplet-induced flame wrinkling is less obvious from the curvature PDFs for turbulent flames although the turbulent droplet cases show wider curvature PDFs than in the corresponding turbulent premixed cases for  $\phi_{ov} = 1.0$  and 1.2 but the PDFs of  $\kappa_m \times \delta_{st}$  for droplet cases with  $\phi_{ov} = 0.8$  are comparable to the curvature PDFs for the corresponding turbulent premixed flame case. The masking of droplet-induced flame wrinkling with increasing  $u'$  is consistent with experimental findings of Hayashi et al. (1977) and Lawes and Saat (2011).

The extent of the departure from the perfectly spherical shape can be quantified in terms of the magnitude of the inner product of the local flame normal vector  $\vec{N}$  and the unit radial vector  $\vec{r}$  from the centre of the mass of the flame kernel. The angle between these two unit vectors ( $\vec{N}$  and  $\vec{r}$ ) is equal to zero (or  $|\vec{N} \cdot \vec{r}|$  equals to  $|\cos\theta| = |\vec{N} \cdot \vec{r}| = 1.0$ ) for a perfectly spherical flame. The extent of the deviation of the projection  $|\vec{N} \cdot \vec{r}|$  from 1.0 provides the measure of the departure from the perfect spherical shape. The PDFs of  $|\vec{N} \cdot \vec{r}|$  are shown for  $c = 0.1, 0.5$  and 0.9 isosurfaces in Fig. 6. The PDFs of  $|\vec{N} \cdot \vec{r}|$  are represented by delta functions at  $|\vec{N} \cdot \vec{r}| = 1.0$  for laminar premixed flames irrespective of the equivalence ratio values. Although the PDFs of  $|\vec{N} \cdot \vec{r}|$  for laminar droplet cases show peak values at 1.0, these cases show finite probability of finding  $|\vec{N} \cdot \vec{r}| < 1.0$  which is an indication of an imperfect spherical shape. A further

investigation for laminar spray flames shows that the extent of departure from the perfectly spherical shape increases with the increasing droplet size and also with increasing overall equivalence ratio. The extent of departure from a perfectly spherical shape in turbulent cases is greater than laminar flames for both premixed and droplet cases. The probability of finding a perfectly spherical flame surface (i.e.  $|\vec{N} \cdot \vec{r}| = 1.0$ ) in turbulent droplet cases is smaller than the corresponding turbulent premixed flame case for  $\phi_{ov} = 1.2$ , whereas the probability of finding  $|\vec{N} \cdot \vec{r}| = 1.0$  is comparable for turbulent premixed and droplet cases for  $\phi_{ov} = 0.8$  and 1.0. It has been discussed in the context of Fig. 4 that the flame front deformation due to droplets is relatively stronger for  $\phi_{ov} = 1.2$  than for  $\phi_{ov} = 0.8$  and 1.0. As a result of this, the flow-induced flame deformation does not completely eclipse the droplet-induced wrinkling effects in  $\phi_{ov} = 1.2$  droplet cases and some of the effects of flame wrinkling induced by droplets can be discerned for initial  $u' = 4S_b(\phi_g=1)$  but for this turbulence intensity droplet-induced wrinkling effects are masked by flame wrinkling due to turbulence for  $\phi_{ov} = 0.8$  and 1.0. It has been found that the PDFs of  $|\vec{N} \cdot \vec{r}|$  remain mostly insensitive to the value of  $a_d/\delta_{st}$  for a given value of  $\phi_{ov}$  in turbulent droplet cases considered here.

The extent of flame wrinkling plays a key role in determining the growth of flame surface area. Figure 7a shows the temporal evolutions of flame surface area normalised by its initial value  $A/A_0$  (where  $A$  is evaluated by the volume-integral  $A = \int_V |\nabla c| dV$ ) for all cases with different overall equivalence ratios  $\phi_{ov} = 0.8, 1.0$  and 1.2. The value of  $A/A_0$  decreases with increasing  $a_d/\delta_{st}$  for the droplet cases with  $\phi_{ov} = 1.0$  and 1.2 under both laminar and turbulent flow conditions but the droplet cases with  $\phi_{ov} = 0.8$  behave in the opposite manner. Under laminar conditions,  $A/A_0$  in droplet cases increases faster than the corresponding gaseous premixed flame for  $\phi_{ov} = 0.8$  and 1.0. By contrast,  $A/A_0$  for the laminar droplet case with

initial  $a_d/\delta_{st} = 0.04$  remains comparable to that for the corresponding laminar premixed flame for  $\phi_{ov} = 1.2$ , whereas the growth rates of  $A/A_0$  for the laminar droplet cases with initial  $a_d/\delta_{st} = 0.05$  and  $0.06$  have been found to be smaller than that in the corresponding laminar premixed flame for all values of  $\phi_{ov}$  considered here. Under turbulent condition,  $A/A_0$  values for droplet cases with initial  $a_d/\delta_{st} = 0.05$  and  $0.06$  have been found to be smaller than the corresponding gaseous premixed flame for  $\phi_{ov} = 1.0$  and  $1.2$ , whereas  $A/A_0$  in droplet cases assumes higher values than the corresponding turbulent premixed flame for initial  $a_d/\delta_{st} = 0.04$  for  $\phi_{ov} = 1.0$  and  $1.2$ , and for all droplet diameters considered here for  $\phi_{ov} = 0.8$ . The difference in  $A/A_0$  values between droplet and premixed gaseous flames decreases under turbulent conditions for a given set of values of  $\phi_{ov}$  and  $a_d/\delta_{st}$ . This is consistent with the observations from Figs. 5 and 6, which indicate that the effects of flame wrinkling due to droplets play a marginal role in determining the overall flame front corrugation and flame area generation under turbulent conditions.

It is important to understand that the initial value of flame surface area  $A_0$  (i.e.  $\int_V |\nabla c| dV$ ) for spherical flames with the burned gas radius  $r_0 = 2\delta_{st}$  are not the same for all cases considered here. As the distribution of  $|\nabla c|$  (i.e. the magnitude of  $|\nabla c|$  and the volume over which  $|\nabla c|$  assumes non-zero values) varies from one case to another, the values of  $A_0$  for these cases have been found to be different in spite of having the same burned gas radius  $r_0 = 2\delta_{st}$ . In order to provide a comparison between the actual flame surface areas of different cases, the temporal evolutions of flame surface area  $A$  normalised by the initial value for the stoichiometric laminar premixed flame kernel  $(A_{0,pre})_{st}$  (i.e.  $A/(A_{0,pre})_{st}$ ) for all cases considered here are shown in Fig. 7b. It can be clearly seen from Fig. 7b that  $A/(A_{0,pre})_{st}$  for the premixed stoichiometric flame assumes the highest value among the cases considered here under

both laminar and turbulent conditions. Figure 7b shows that  $A/(A_{0,pre})_{st}$  values for the droplet case with initial  $a_d/\delta_{st} = 0.04$  are marginally smaller than that in the gaseous premixed flame for  $\phi_{ov} = 0.8$  under both laminar and turbulent conditions. This is consistent with the earlier observations from Figs. 5 and 6, which demonstrated that the flame front distortion for the  $\phi_{ov} = 0.8$  droplet case with initial  $a_d/\delta_{st} = 0.04$  is comparable to the corresponding premixed flame. However,  $A/(A_{0,pre})_{st}$  values for  $\phi_{ov} = 0.8$  droplet cases with initial  $a_d/\delta_{st} = 0.05$  and  $0.06$  have been found to be greater than the corresponding premixed flame cases under both laminar and turbulent conditions. Under both laminar and turbulent conditions, the  $\phi_{ov} = 1.0$  droplet cases exhibit smaller values of  $A/(A_{0,pre})_{st}$  than the corresponding premixed flame cases and the value of  $A/(A_{0,pre})_{st}$  decreases with increasing droplet size. In the case of  $\phi_{ov} = 1.2$ , the laminar and turbulent droplet cases with initial  $a_d/\delta_{st} = 0.04$  show greater value of  $A/(A_{0,pre})_{st}$  than the corresponding premixed flames with an equivalence ratio of 1.2. The values of  $A/(A_{0,pre})_{st}$  for the laminar and turbulent  $\phi_{ov} = 1.2$  droplet cases with initial  $a_d/\delta_{st} = 0.05$  and  $0.06$  have been found to be comparable to the values obtained for the corresponding premixed flames. The  $A/(A_{0,pre})_{st}$  values for turbulent flames are greater than the values obtained in the corresponding laminar flames due to flame surface area generation under turbulence.

A flame speed based on the flame surface area calculations can be defined as  $S_A = dr_A/dt$  where the equivalent radius  $r_A$  is defined as:  $r_A = \sqrt{A/4\pi}$ . The value of  $S_A$  is estimated by using the slope of the linear part of the temporal evolution of  $r_A$  and this slope remains unchanged since halfway through the simulation for all cases considered here. The variation of  $S_A$  with droplet diameter and overall equivalence ratio is shown in Table 2, which indicates that the presence of droplets enhances the normalised flame speed  $S_A/S_b(\phi_{ov})$  for  $\phi_{ov} = 0.8$

except for the initial  $a_d/\delta_{st} = 0.04$  case under turbulent conditions. However, for  $\phi_{ov} = 1.0$ , only the small droplets with initial  $a_d/\delta_{st} = 0.04$  under laminar conditions demonstrate higher  $S_A/S_{b(\phi_{ov})}$  than the corresponding laminar premixed flame and this tendency is evident for both laminar and turbulent conditions for  $\phi_{ov} = 1.2$ . Normalised flame speed  $S_A/S_{b(\phi_{ov})}$  increases with increasing droplet diameter for  $\phi_{ov} = 0.8$ , whereas it shows just the opposite trend for  $\phi_{ov} = 1.0$  and 1.2. These findings are consistent with the observations made from Fig. 7 but Table 2 provides quantitative measure of the effects of  $\phi_{ov}$  and  $a_d$  on the flame surface area evolution in spherically expanding turbulent spray flames.

It is instructive to look into the statistical behaviours of the equivalence ratio in gaseous phase  $\phi_g$  and the magnitude of reaction progress variable gradient  $|\nabla c|$  in order to explain the  $A/(A_{0,pre})_{st}$  and  $S_A/S_{b(\phi_{ov})}$  behaviours shown in Fig. 7 and Table 2 respectively.

### 4.3 Reaction zone structure in spherically expanding droplet flames

The PDFs of gaseous phase equivalence ratio  $\phi_g$  within the region given by  $0.01 \leq c \leq 0.99$  for all cases considered here are shown in Fig. 8. Although a peak of the  $\phi_g$ -PDF can be discerned at  $\phi_g \approx \phi_{ov}$  in Fig. 8, there is a significant probability of finding fuel-air mixtures with  $\phi_g < \phi_{ov}$  and  $\phi_g > \phi_{ov}$ . However, the probability of finding  $\phi_g < \phi_{ov}$  supersedes the probability of obtaining  $\phi_g > \phi_{ov}$  for all values of  $\phi_{ov}$  and  $a_d/\delta_{st}$ , which is consistent with the observations made from Fig. 2. The evaporation rate is slower for larger droplets and thus the probability of finding  $\phi_g < \phi_{ov}$  increases with increasing  $a_d/\delta_{st}$ . Moreover, evaporation of droplets produce localised fuel-rich pockets, which are more frequent for larger droplets due to slower evaporation. Thus, the width of the PDF increases with increasing droplet

diameter for both laminar and turbulent cases. In turbulent flows, the evaporated fuel is transported to the locations far from the evaporation sites by the dispersion process due to turbulent fluid motion, and this increases the probability of finding  $\phi_g < \phi_{ov}$  in turbulent droplet cases. It can especially be seen from Fig. 8 that combustion takes place predominantly in fuel-lean mode for the turbulent cases for all values of  $\phi_{ov}$  and  $a_d/\delta_{st}$  considered here.

It can be seen from Fig. 8 that a mild peak of the  $\phi_g$ -PDF is obtained at  $\phi_g = 1.0$  for  $\phi_{ov} = 1.0$  and 1.2 cases. This can be explained in terms of the mode of combustion, which can be characterised by the flame index  $FI = (\nabla Y_F \cdot \nabla Y_O) / (|\nabla Y_F| |\nabla Y_O|)$  (Yamashita et al., 1996). A positive (negative) flame index FI indicates premixed (non-premixed) mode of combustion. The percentages of heat release rate arising from premixed and non-premixed modes of combustion are shown in Fig. 9 for laminar and turbulent droplet cases. Based on the heat release rate per unit mass of fuel  $H_\phi$  proposed by Tarrazo et al. (2006), the total heat release rate is calculated as  $HR_{total} = \int_V H_\phi |\dot{w}_F| dV$  (Wacks et al., 2016). For laminar  $\phi_{ov} = 0.8$  droplet cases the total heat release rate arises due to premixed mode of combustion. For laminar  $\phi_{ov} = 1.0$  and 1.2 droplet cases heat release rate arises also predominantly due to premixed mode of combustion but a non-negligible percentage of total heat release rate is obtained from non-premixed mode of combustion. The contribution of non-premixed mode of combustion to the overall heat release drops with increasing droplet diameter. Moreover, the contribution of non-premixed mode of combustion is relatively smaller in the laminar  $\phi_{ov} = 1.2$  case than in the laminar  $\phi_{ov} = 1.0$  case. The slower evaporation of droplets induces greater mixture inhomogeneity for cases with larger droplets, which eventually gives rise to a greater extent of non-premixed combustion contribution to the overall heat release. The non-premixed flame is formed at the stoichiometric mixture (i.e.  $\phi_g = 1.0$ ) and the likelihood of obtaining  $\phi_g = 1.0$

is greater in the laminar  $\phi_{ov} = 1.0$  case than in the laminar  $\phi_{ov} = 1.2$  case (see Fig. 8). Thus, the percentage contribution of non-premixed combustion to the total heat release rate is greater in the  $\phi_{ov} = 1.0$  cases than in the  $\phi_{ov} = 1.2$  cases.

The total heat release rate arises due to premixed mode of combustion for the  $\phi_{ov} = 0.8$  droplet case for small droplets with initial  $a_d/\delta_{st} = 0.04$ . However, a non-negligible contribution to the overall heat release rate in the  $\phi_{ov} = 0.8$  droplet cases comes from non-premixed mode of combustion for droplet diameters with initial  $a_d/\delta_{st} = 0.05$  and  $0.06$ . Droplets with  $\phi_{ov} = 0.8$  mostly complete their evaporation on the unburned gas side (Fig. 2) and create fuel-lean mixture ahead of the flame under laminar flow conditions. However, under turbulent condition some droplets penetrate into the burned gas side without complete evaporation, where they evaporate relatively readily and create unburned fuel pockets. The evaporated fuel vapour diffuses back from the burned gas side and mixes with the excess unused air to form the pockets of stoichiometric mixture where non-premixed flame can be obtained. Turbulence promotes and increases the contribution of a non-premixed combustion for droplet cases with  $\phi_{ov} = 1.0$  and  $1.2$ . The turbulent fluid motion disperses evaporated fuel from the droplet sites to the surrounding gaseous mixture, which increases the mixing rate between the evaporated fuel and the surrounding air and the probability to obtain locally stoichiometric mixture to support non-premixed combustion.

The non-premixed mode of combustion is responsible for the peak of the gaseous equivalence ratio PDF at  $\phi_g = 1.0$  for all droplet cases with  $\phi_{ov} = 1.0$  and  $1.2$ . Interested readers are referred to Ozel-Erol et al. (2018) for the variation of FI with  $\xi$  and  $c$  for the droplet cases with  $\phi_{ov} = 1.0$ . The statistical behaviours of the chemical reaction rate  $\dot{\omega}_c$  of the reaction

progress variable<sup>3</sup> for both premixed and non-premixed modes of combustion for droplet cases with  $\phi_{ov} = 1.0$  have been presented in Ozel-Erol et al. (2018) and they are not repeated here for the sake of conciseness as these behaviours are qualitatively similar for  $\phi_{ov} = 0.8$  and 1.2.

The distributions of  $\phi_g$  and  $c$  determine the statistical behaviours of  $|\nabla c|$  as the flame thickness  $\delta_{(\phi_g)}$  scales as:  $\delta_{(\phi_g)} \sim 1/(\max|\nabla c|_L) \sim \alpha_{T0}/S_b(\phi_g)$ . As the laminar burning velocity  $S_b(\phi_g)$  attains its maximum value (i.e. a value slightly greater than  $S_b(\phi_g=1)$ ) for  $\phi_g \approx 1.10$  for the present thermo-chemistry (Tarrazo et al., 2006), it can be expected that the flame thickness is greater than the stoichiometric premixed flame for  $\phi_g < 1$  and  $\phi_g \gg 1.10$ . Thus,  $|\nabla c|$  distribution with  $c$  (e.g. the value of  $\max|\nabla c|_L$  and the value of  $c$  at which this maximum value is obtained) is expected to be different depending on  $\phi_g$ . The PDFs of  $|\nabla c| \times \delta_{st}$  for  $c = 0.1, 0.5$  and  $0.9$  isosurfaces for all laminar and turbulent cases are shown in Figs. 10a and 10b respectively. It can be seen from Fig. 10a that the droplet cases with  $\phi_{ov} = 0.8$  show greater values of  $|\nabla c|$  than the corresponding premixed flame. The laminar  $\phi_{ov} = 1.0$  and 1.2 droplet cases with initial  $a_d/\delta_{st} = 0.04$  show higher probability of finding greater values of  $|\nabla c|$  than that in the corresponding premixed case. A similar qualitative trend has been observed for  $c = 0.1$  and  $0.9$  in the laminar  $\phi_{ov} = 1.0$  droplet case with initial  $a_d/\delta_{st} = 0.05$ . The likelihood of having both greater and smaller values of  $|\nabla c|$  than that in the corresponding premixed case is almost the same at  $c = 0.5$  in the  $\phi_{ov} = 1.0$  and 1.2 laminar droplet cases with initial  $a_d/\delta_{st} = 0.05$ . By contrast,  $|\nabla c|$  for the laminar  $\phi_{ov} = 1.0$  and 1.2 droplet cases with initial  $a_d/\delta_{st} = 0.06$  assumes smaller values than that in the corresponding premixed flame case for  $c = 0.5$  and  $0.9$  but the converse is true for  $c = 0.1$ . It can be seen from

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<sup>3</sup>  $\dot{\omega}_c$  is the reaction rate of progress variable, which is given by  $\dot{\omega}_c = -\xi_{st}\dot{\omega}_O/[\xi(1-\xi_{st})Y_{O\infty}]$  ( $\dot{\omega}_c = -\dot{\omega}_O/[(1-\xi)Y_{O\infty}]$ ) for  $\xi \leq \xi_{st}$  ( $\xi > \xi_{st}$ ) (Wacks et al., 2016).

Fig. 10b that the turbulent droplet cases with  $\phi_{ov} = 1.0$  and 1.2 exhibit greater likelihood of smaller magnitudes of  $|\nabla c|$  than the corresponding premixed turbulent flame and this tendency becomes more prevalent for larger droplet diameters due to the predominant availability of fuel-lean mixtures as a result of slower evaporation of droplets. However, the PDFs of  $|\nabla c|$  for the turbulent  $\phi_{ov} = 0.8$  droplet cases with initial  $a_d/\delta_{st} = 0.04$  are comparable to the corresponding premixed flame, and this remains valid for  $c = 0.1$  in the cases with initial  $a_d/\delta_{st} = 0.05$  and 0.06. However, the  $\phi_{ov} = 0.8$  droplet cases with initial  $a_d/\delta_{st} = 0.05$  and 0.06 exhibit wider PDFs for  $c = 0.5$  and 0.9 with greater likelihood of obtaining greater values of  $|\nabla c|$  than the corresponding premixed flame cases.

The integral, which yields the flame surface area,  $A$  is influenced by the magnitude of  $|\nabla c|$  and the volume over which  $|\nabla c|$  assumes non-zero values. For  $\phi_{ov} = 0.8$ , mostly higher magnitudes of  $|\nabla c|$  of the droplet cases with initial  $a_d/\delta_{st} = 0.05$  and 0.06 are responsible for yielding greater values of flame surface area  $A$  than in the corresponding premixed flames. However, the smaller volume of the region where large magnitudes of  $|\nabla c|$  are obtained in the  $\phi_{ov} = 0.8$  droplet case with initial  $a_d/\delta_{st} = 0.04$  is responsible for yielding slightly smaller value of flame surface area  $A$  than that in the corresponding premixed flames.

The predominance of smaller magnitudes of  $|\nabla c|$  in the laminar  $\phi_{ov} = 1.0$  droplet cases with initial  $a_d/\delta_{st} = 0.05$  and 0.06 in comparison to those of the corresponding premixed case are principally responsible for yielding smaller values of  $A_0$  than  $A_{0,pre}$  in spite of all cases having same initial burned gas radius (i.e.  $r_0 = 2\delta_{st}$ ). Figures 10a and b show that the  $\phi_{ov} = 1.0$  droplet cases exhibit greater probability of finding small values of  $|\nabla c|$  than the corresponding premixed flame cases except for the laminar droplet flame case with  $a_d/\delta_{st} = 0.04$ , and the

probability of finding smaller values of  $|\nabla c|$  is greater for initial  $a_d/\delta_{st} = 0.06$  droplet cases than in the cases with initial  $a_d/\delta_{st} = 0.05$ . This along with smaller volume over which  $|\nabla c|$  assumes non-zero values (see Fig. 1 for the region with  $0.01 \leq c \leq 0.99$ ) gives rise to smaller  $A/(A_{0,pre})_{st}$  in the  $\phi_{ov} = 1.0$  droplet cases with  $a_d/\delta_{st} = 0.05$  and  $0.06$  in comparison to the corresponding premixed case. Thicker flame (i.e. larger flame volume) in the  $\phi_{ov} = 1.0$  laminar droplet cases with initial  $a_d/\delta_{st} = 0.05$  and  $0.06$  in comparison to the corresponding droplet case with initial  $a_d/\delta_{st} = 0.04$  gives rise to greater values of  $A_0$  for larger droplet diameters in spite of higher probability of obtaining smaller values of  $|\nabla c|$  and identical values of  $r_0$ . In the  $\phi_{ov} = 1.0$  laminar droplet cases with initial  $a_d/\delta_{st} = 0.05$  and  $0.06$ , the smaller magnitude of  $|\nabla c|$  is principally responsible for yielding smaller value of  $A_0$  than  $(A_{0,pre})_{st}$  in spite of all cases having same initial burned gas radius  $r_0 = 2\delta_{st}$ . Although the probability of finding smaller values of  $|\nabla c|$  in the  $\phi_{ov} = 1.2$  droplet cases with initial  $a_d/\delta_{st} = 0.05$  and  $0.06$  is greater than the corresponding premixed flame cases, the thicker droplet flames yield flame surface areas  $A$ , which are comparable to the values obtained for the  $\phi_{ov} = 1.2$  premixed flame cases subjected to similar flow conditions. Higher probability of finding greater values of  $|\nabla c|$  in the  $\phi_{ov} = 1.2$  droplet cases with  $a_d/\delta_{st} = 0.04$  than that in the corresponding premixed cases is principally responsible for giving rise to greater values of  $A$  than in the corresponding premixed flame cases.

The probability of finding local high values of  $|\nabla c|$  is greater in turbulent cases than in the corresponding laminar case for both droplet and premixed flames, which is consistent with higher values of  $A/(A_{0,pre})_{st}$  in turbulent flames. It can be seen from Fig. 10 that the probability of finding greater magnitude of  $|\nabla c|$  increases with increasing  $a_d$  for droplet cases for  $\phi_{ov} = 0.8$  because of the greater availability of more reactive mixture than  $\phi_g \leq 0.8$  (see Fig.

8). This is principally responsible for an increase in flame surface area with increasing droplet diameter for  $\phi_{ov} = 0.8$ . By contrast, slower evaporation of larger droplets for  $\phi_g = 1.0$  and 1.2 gives rise to higher probability of fuel-lean mixtures and smaller magnitudes of  $|\nabla c|$ . This in turn is principally responsible for a decrease in flame surface area with increasing droplet diameter for  $\phi_g = 1.0$  and 1.2.

#### 4.4 Extent of burning in spherically expanding flames

The effects of  $\phi_{ov}$  and  $a_d$  on evaporation characteristics affect not only the mixture composition in the gaseous phase and flame surface area but also influences the extent of burning. This can be substantiated from Fig. 11a where the temporal evolutions of the volume-integrated fuel reaction rate magnitude  $\Omega_F = \int_V |\dot{w}_F| dV$  normalised by its initial value (i.e.  $\Omega_F/\Omega_{F0} = \int_V |\dot{w}_F| dV / [\int_V |\dot{w}_F| dV]_{t=0}$ ) are shown for all cases considered here.<sup>4</sup> It can be seen from Fig. 11a that  $\Omega_F/\Omega_{F0}$  increases rapidly with time for all cases considered here. The  $\phi_{ov} = 0.8$  droplet cases exhibit smaller values of  $\Omega_F/\Omega_{F0}$  than in the corresponding fuel-lean premixed flames for both laminar and turbulent conditions. The value of  $\Omega_F/\Omega_{F0}$  decreases with increasing droplet diameter under both laminar and turbulent conditions for  $\phi_{ov} = 1.0$ . However,  $\Omega_F/\Omega_{F0}$  in the laminar  $\phi_{ov} = 1.0$  droplet cases has been found to be greater than in the corresponding premixed flame, whereas  $\Omega_F/\Omega_{F0}$  values for turbulent droplet cases have been found to be smaller than the corresponding premixed flames. The values of  $\Omega_F/\Omega_{F0}$  increase with decreasing droplet size for  $\phi_{ov} = 1.2$  and  $\Omega_F/\Omega_{F0}$  values for droplet cases remain smaller than the corresponding premixed flames under both laminar and turbulent conditions except for the cases with initial  $a_d/\delta_{st} = 0.04$ . Since initial values of volume integrated fuel reaction

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<sup>4</sup> The temporal evolution of  $\Omega_c = \int_V \dot{\omega}_c dV$  and  $\Omega_c/\Omega_{c0}$  are qualitatively similar to  $\Omega_F$  and  $\Omega_F/\Omega_{F0}$  respectively

rate  $\Omega_{F0}$  are different for all cases, the temporal evolutions of  $\Omega_F/(\Omega_{F0,pre})_{st}$  are presented in Fig. 11b to gain a better understanding of the relative magnitudes of  $\Omega_F$ . It can be seen from Fig. 11b that  $\Omega_F/(\Omega_{F0,pre})_{st}$  for all the  $\phi_{ov} = 1.0$  droplet cases is smaller than the corresponding stoichiometric premixed flames. The presence of droplets enhances the value of  $\Omega_F/(\Omega_{F0,pre})_{st}$  in comparison to premixed flame for  $\phi_{ov} = 0.8$  in the droplet cases with initial  $a_d/\delta_{st} = 0.05$  and  $0.06$  for both laminar and turbulent conditions but the values of  $\Omega_F/(\Omega_{F0,pre})_{st}$  for the droplet case with initial  $a_d/\delta_{st} = 0.04$  have been found to be comparable to the values obtained for the corresponding premixed flames. The enhancement of  $\Omega_F/(\Omega_{F0,pre})_{st}$  due to droplets for  $\phi_{ov} = 1.2$  has been found for both laminar and turbulent cases with  $a_d/\delta_{st} = 0.04$  but just the opposite trend is observed for droplet cases with initial  $a_d/\delta_{st} = 0.05$  and  $0.06$ .

The dependences of  $\Omega_F/(\Omega_{F0,pre})_{st}$  on  $\phi_{ov}$  and  $a_d$  are determined by their influences on flame surface area and the volume-integrated reaction rate of progress variable to the flame surface area. The influences of  $\phi_{ov}$  and  $a_d$  on flame surface area  $A$  have already been shown in Fig. 7b and thus the temporal evolutions of the volume-integrated reaction rate of progress variable to the flame surface area  $S = [\rho_0 S_b(\phi_g)]^{-1} \int_V \dot{\omega}_c dV / \int_V |\nabla c| dV$  for all cases considered here are shown in Fig. 12a. In all cases  $S$  deviates from unity due to the stretch rate induced by mean positive curvature of statistically spherical flames, which has been discussed elsewhere (Klein et al., 2006; Chakraborty and Klein, 2009) and will not be elaborated here. Figure 12a shows that  $S$  for the droplet cases assumes greater values than the corresponding premixed flames for  $\phi_{ov} = 0.8$  and the values of  $S$  for droplet cases with initial  $a_d/\delta_{st} = 0.05$  and  $0.06$  are found to be greater than the cases with initial  $a_d/\delta_{st} = 0.04$ . The possibility of obtaining more reactive (i.e.  $\phi_g > \phi_{ov} = 0.8$ ) fuel-air mixtures is relatively greater for

cases with initial  $a_d/\delta_{st} = 0.05$  and  $0.06$  than in the cases with initial  $a_d/\delta_{st} = 0.04$  (see Fig. 8) and this contributes to the higher values of the volume-integrated reaction rate of progress variable per unit flame surface area for cases with initial  $a_d/\delta_{st} = 0.05$  and  $0.06$ . For  $\phi_{ov} = 1.0$  and  $1.2$ , the values of  $S$  for the initial  $a_d/\delta_{st} = 0.04$  cases remain comparable to that in the corresponding premixed flames but  $S$  decreases with increasing initial droplet size. The probability of finding less reactive fuel-lean mixture for  $\phi_{ov} = 1.0$  and  $1.2$  droplet cases increases with increasing  $a_d/\delta_{st}$  due to slower evaporation of larger droplets. This contributes to the smaller values of  $S$  in the  $\phi_{ov} = 1.0$  and  $1.2$  droplet cases and also decreasing trend of  $S$  with increasing  $a_d/\delta_{st}$ .

The combined influences of  $\phi_{ov}$  and  $a_d/\delta_{st}$  on  $A$  and  $S$  ultimately determine the extent of burning which can be quantified in terms of the volume of the burned gas with  $c \geq 0.99$  (i.e.  $V_b$ ). The temporal evolution of the normalised burned gas volume  $V_b/(4\pi r_0^3/3)$  for all cases considered here is shown in Fig. 12b. The combination of the larger values of  $A$  and  $S$  than the premixed flame for  $\phi_{ov} = 0.8$  gives rise to an increase in burned gas volume in comparison to the corresponding premixed flame for the droplet cases with initial  $a_d/\delta_{st} = 0.05$  and  $0.06$ . The burned gas volume for the  $\phi_{ov} = 0.8$  droplet cases with initial  $a_d/\delta_{st} = 0.04$  remains comparable to the corresponding premixed flame cases. The smaller values of  $S$  and  $A$  yield smaller burned gas volume in the  $\phi_{ov} = 1.0$  and  $1.2$  droplet cases with initial  $a_d/\delta_{st} = 0.05$  and  $0.06$  in comparison to that in the corresponding premixed flames. The smaller value of  $S$  in the  $\phi_{ov} = 1.0$  droplet cases with initial  $a_d/\delta_{st} = 0.04$  than in the corresponding premixed flames dominates over greater flame surface area for the droplet cases to yield a smaller burned gas volume in comparison to that in the premixed flame cases. The combination of the larger values of  $A$  and  $S$  in the laminar  $\phi_{ov} = 1.2$  droplet cases with initial

$a_d/\delta_{st} = 0.04$  than the corresponding laminar premixed flame gives rise to an increase in burned gas volume in comparison to the corresponding premixed flame for the droplet cases. Thus, the burned gas volume drops with increasing  $a_d/\delta_{st}$  for  $\phi_{ov} = 1.0$  and 1.2. It can be seen from Fig. 12b that the largest burned gas volume is obtained for the stoichiometric premixed flame case for both laminar and turbulent conditions analysed in this investigation. Moreover, it can be seen from Fig. 12b that the burned gas volume increases under turbulent conditions for  $\phi_{ov} = 1.0$  and 1.2 irrespective of  $a_d/\delta_{st}$  values principally due to greater amount of flame area generation under turbulence than in laminar conditions. The value of  $S$  under turbulent condition decreases in comparison to the laminar flame value for  $\phi_{ov} = 0.8$  because the stretch rate effects due to curvature are expected to be stronger in these cases due to high Karlovitz number values (Peters, 2000). The small values of  $S$  dominate over increased  $A$  under turbulent conditions to give rise to a reduction in the burned gas mass under turbulent conditions for  $\phi_{ov} = 0.8$  cases except for the droplet case with initial  $a_d/\delta_{st} = 0.06$  where the enhancement of flame surface area dominates over reduced  $S$  to yield an enhanced burned gas volume under the turbulent condition analysed here.

The growth rate of the burned gas volume can be quantified by a flame speed  $S_V$ , which is defined as:  $S_V = dr_V/dt$  where  $r_V = (3V_b/4\pi)^{1/3}$  is an equivalent radius. The value of  $S_V$  is estimated by using the slope of the linear part of the temporal evolution of  $r_V$  and this slope remains unchanged since halfway through the simulation. The evaluated values of normalised flame speed  $S_V/S_{b(\phi_g)}$  are summarised in Table 3. It can be seen from Table 3 that the growth of burned gas volume for the droplet cases remains mostly smaller than the corresponding premixed flames. However, for  $\phi_{ov} = 0.8$ , large droplets with initial  $a_d/\delta_{st} = 0.05$  and 0.06 show greater  $S_V/S_{b(\phi_g)}$  values compared with the corresponding premixed flames with the

same  $\phi_{ov}$  under laminar and turbulent conditions and for  $\phi_{ov} = 1.2$ , burned gas volume can grow faster for small droplets with initial  $a_d/\delta_{st} = 0.04$  than the corresponding premixed flame under laminar conditions. Furthermore, turbulence significantly affects the normalised flame speed  $S_V/S_{b(\phi_g)}$  and increases the growth rate of burned gas volume for large droplets with  $\phi_{ov} = 0.8$  and for small droplets with  $\phi_{ov} = 1.0$  and 1.2. The enhancement of the extent of burning with increasing droplet size for overall fuel-lean mixtures is consistent with previous experimental findings by Hayashi et al. (1977) and Lawes and Saat (2011). Furthermore, the enhanced extent of burning for overall fuel-rich mixtures for small droplets is also qualitatively consistent with experimental observations by Hayashi et al. (1977) and Lawes and Saat (2011).

## 5. CONCLUSIONS

The effects of overall (liquid+gaseous) equivalence ratio (i.e.  $\phi_{ov} = 0.8, 1.0$  and 1.2) on flame wrinkling, flame surface area, and volume of the burned gas for spherically expanding n-heptane flames propagating into mono-sized droplets have been analysed using three-dimensional carrier phase DNS for different droplet diameters (i.e.  $a_d/\delta_{st} = 0.04, 0.05$  and 0.06) under both laminar and turbulent flow conditions (i.e.  $u' = 0$  and  $u' = 4S_{b(\phi_g=1)}$ ). Moreover, additional premixed gaseous flame cases have been considered in order to compare the effects of droplets on flame wrinkling and burning characteristics in terms of the evolutions of flame surface area and burned gas volume for droplet flames with the values obtained in corresponding premixed gaseous flames. It has been found that the overall equivalence ratio has major influences on flame wrinkling, flame surface area, and volume of burned gas and their responses to droplet diameter in the case of spherically expanding flames propagating into droplet-mist.

Flame-droplet interaction gives rise to droplet-induced flame wrinkling for laminar flame kernels and this strengthens with increasing overall equivalence ratio and droplet diameter. However, the effects of droplet-induced flame wrinkling cannot be readily distinguished from flame wrinkling due to fluid motion for turbulent spherically expanding spray flames. In all the cases considered here, the heat release rate takes place principally due to premixed mode of combustion, and the contribution of non-premixed combustion to overall heat release rate increases with increasing overall equivalence ratio and droplet diameter. Moreover, the contribution of non-premixed mode of combustion to overall heat release rate increases under turbulent conditions. It has been found that the gaseous phase mixture within the flame is predominantly fuel-lean in comparison to the overall equivalence ratio for  $\phi_{ov} = 1.0$  and 1.2 droplet cases, and this tendency strengthens with increasing droplet diameter due to slow evaporation of large droplets. However, mixing of evaporated fuel from droplet sites gives rise to local availability of more reactive fuel-air mixtures than a homogeneous mixture corresponding to the overall equivalence ratio of  $\phi_{ov} = 0.8$ , and this tendency strengthens with increasing droplet diameter. The droplet diameter and overall equivalence ratio have been found to influence the evolutions of flame surface area and volume of burned gas.

It has been found that the presence of droplets in the  $\phi_{ov} = 0.8$  cases enhances the growth of flame surface area except for the cases with droplet initial  $a_d/\delta_{st} = 0.04$  under turbulent conditions. However, for  $\phi_{ov} = 1.0$ , only small droplets with initial  $a_d/\delta_{st} = 0.04$  under laminar flow field can grow faster than the corresponding laminar premixed flame and for  $\phi_{ov} = 1.2$  this tendency is obvious only for the turbulent case. The growth rate of flame surface area increases with increasing droplet diameter for  $\phi_{ov} = 0.8$ , whereas an opposite behaviour has been observed for  $\phi_{ov} = 1.0$  and 1.2.

The burned gas volume for the droplet cases grows slower than for the corresponding premixed flames. For  $\phi_{ov} = 0.8$ , large droplets with initial  $a_d/\delta_{st} = 0.05$  and  $0.06$  the flames show greater growth rate of burned gas volume than with the corresponding premixed flame under laminar and turbulent conditions. For  $\phi_{ov} = 1.2$ , the burned gas volume grows faster for small droplets with initial  $a_d/\delta_{st} = 0.04$  than the corresponding premixed flame under laminar conditions. Turbulence increases the growth rate of burned gas volume for large droplets with  $\phi_{ov} = 0.8$  and for small droplets with  $\phi_{ov} = 1.0$  and  $1.2$ . The increase of the burned gas mass for large (small) droplets for overall fuel-lean (fuel-rich) mixtures for flame propagation in droplet-laden mixtures is in qualitative agreement consistent with previous experimental findings by Hayashi et al. (1977) and Lawes and Saat (2011). The above findings suggest that the modelling of turbulent spray combustion needs to explicitly account for overall equivalence ratio and droplet size.

In spite of the qualitative agreement of the present results with previous experimental observations by Hayashi et al. (1977) and Lawes and Saat (2011), it needs to be recognised that the current analysis has been carried out using a simple chemical mechanism for moderate values of turbulent Reynolds number. Thus, further analyses will be necessary so that the current findings could be validated and more comprehensive physical insights can be extracted in the presence of a detailed chemical mechanism at higher values of turbulent Reynolds number.

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## REFERENCES

- Baba, Y., Kurose, R., 2008. Analysis and flamelet modelling for spray combustion. *J. Fluid Mech.* 612, 45–79.
- Ballal, D.R., Lefebvre, A.H., 1981. Flame propagation in heterogeneous mixtures of fuel droplets, fuel vapor and air. *Symp. Combust.* 18, 321–328.
- Burgoyne J. H. and Cohen L., 1954. The Effect of Drop Size on Flame Propagation in Liquid Aerosols. *Proc. R. Soc. London. Ser. A.* 225, 375–392.
- Chakraborty, N., Klein, M., 2009. Effects of global flame curvature on surface density function transport in turbulent premixed flame kernels in the thin reaction zones regime. *Proc. Combust. Inst.* 32, 1435–1443.
- Chakraborty, N., Klein, M., Cant, R.S., 2007. Stretch rate effects on displacement speed in turbulent premixed flame kernels in the thin reaction zones regime. *Proc. Combust. Inst.* 31 I, 1385–1392.
- Chakraborty, N., Rogerson, J.W., Swaminathan, N., 2010. The scalar gradient alignment statistics of flame kernels and its modelling implications for turbulent premixed combustion. *Flow, Turbul. Combust.* 85, 25–55.
- Chaos, M., Kazakov, A., Zhao, Z., Dryer, F.L. 2007. A high-temperature chemical kinetic model for primary reference fuels. *Int. J. Chem. Kinet.* 39, 399–414
- Chiu, H.H., Kim, H.Y., Croke, E.J., 1982. Internal group combustion of liquid droplets. *Proc. Combust. Inst.* 19, 971–980.
- Chiu, H.H., Liu, T.M., 1977. Group Combustion of Liquid Droplets. *Combust. Sci. Technol.* 17, 37–41.
- de Chaisemartin, S., Fréret, L., Kah, D., Laurent, F., Fox, R., Reveillon, J., 2009. Eulerian models for turbulent spray combustion with polydispersity and droplet crossing. *C R*

- Mecanique, 337, 438-448.
- Faeth, G.M., 1987. Mixing, transport and combustion in sprays. *Prog. Energy Combust. Sci.* 13, 293–345.
- Fernández-Tarrazo, E., Sánchez, A.L., Liñán, A., Williams, F.A., 2006. A simple one-step chemistry model for partially premixed hydrocarbon combustion. *Combust. Flame* 147, 32–38.
- Fujita, A., Watanabe, H., Kurose, R., Komori, S., 2013. Two-dimensional direct numerical simulation of spray flames - Part 1: Effects of equivalence ratio, fuel droplet size and radiation, and validity of flamelet model. *Fuel* 104, 515–525.
- Greenberg, J.B., 2007. Finite-rate evaporation and droplet drag effects in spherical flame front propagation through a liquid fuel mist. *Combust. Flame* 148, 187–197.
- Greenberg, J.B., Kalma, A., 2000. A study of stretch in premixed spray flames. *Combust. Flame* 123, 421–429.
- Greenberg, J.B., Silverman, I., Tambour, Y., 1998. On Droplet Enhancement of the Burning Velocity of Laminar Premixed Spray Flames. *Combust. Flame* 113, 271–273.
- Grout, R.W., 2007. An age extended progress variable for conditioning reaction rates. *Phys. Fluids* 19, 105107.
- Han, I., Huh, K.Y., 2008. Roles of displacement speed on evolution of flame surface density for different turbulent intensities and Lewis numbers in turbulent premixed combustion. *Combust. Flame* 152, 194–205.
- Haruki, Y., Pillai, A.L., Kitano, T. and Kurose, R., 2018. Numerical investigation of flame propagation in fuel droplet arrays., *At. Sprays*, 28, 357.
- Hayashi, S., Kumagai, S., Sakai, T., 1977. Propagation Velocity and Structure of Flames in Droplet-Vapor-Air Mixtures. *Combust. Sci. Technol.* 15, 169–177.

- Jaegle, F., Senorer, J.-M., García, M., Bismes, F., Lecourt, R., Cuenot, B., Poinso, T., 2011. Eulerian and Lagrangian spray simulations of an aeronautical multipoint injector. *Proc Combust Inst*, 33, 2099-2107.
- Klein, M., Chakraborty, N., Jenkins, K.W., Cant, R.S., 2006. Effects of initial radius on the propagation of premixed flame kernels in a turbulent environment. *Phys. Fluids* 18.
- Kumar, K., Freeh, J. E., Sung, C. J. and Huang, Y., 2007. Laminar flame speeds of preheated iso-octane/O<sub>2</sub>/N<sub>2</sub> and n-heptane/O<sub>2</sub>/N<sub>2</sub> mixtures, *J. Propulsion Power* 23, 428–436.
- Kuo, K.K., Acharya, R., 2012. *Fundamentals of Turbulent Multi-Phase Combustion*, Fundamentals of Turbulent Multi-Phase Combustion.
- Lawes, M., Lee, Y., Marquez, N., 2006. Comparison of iso-octane burning rates between single-phase and two-phase combustion for small droplets. *Combust. Flame* 144, 513–525.
- Lawes, M., Saat, A., 2011. Burning rates of turbulent iso-octane aerosol mixtures in spherical flame explosions. *Proc. Combust. Inst.* 33, 2047–2054.
- Luo, K., Pitsch, H., Pai, M.G., Desjardins, O., 2011. Direct numerical simulations and analysis of three-dimensional n-heptane spray flames in a model swirl combustor. *Proc. Combust. Inst.* 33, 2143–2152.
- Malkeson, S.P., Chakraborty, N., 2010. Statistical analysis of displacement speed in turbulent stratified flames: A direct numerical simulation study. *Combust. Sci. Technol.* 182, 1841–1883.
- McGowan, E., Onysko, G., Scheer, R.M., 2010. US energy conservation and efficiency policies: Challenges and opportunities. *Energy Policy* 38, 6398–6408.
- Miller, R.S., Bellan, J., 1999. Direct numerical simulation of a confined three-dimensional gas mixing layer with one evaporating hydrocarbon-droplet-laden stream. *J. Fluid Mech.* 384, 293–338.

- Mizutani, Y., Najkajima, A., 1973. Combustion of Fuel Vapor-Drop-Air Systems : Part II- Spherical Flames in a Vessel. *Combust. Flame* 20, 351–357.
- Mizutani, Y., Nishimoto, T., 1972. Turbulent Flame Velocities in Premixed Sprays Part I. Experimental Study. *Combust. Sci. Technol.* 6, 1–10.
- Nakamura, M., Akamatsu, F., Kurose, R., Katsuki, M., 2005. Combustion mechanism of liquid fuel spray in a gaseous flame. *Phys. Fluids* 17, 1–14.
- Neophytou, A., Mastorakos, E., 2009. Simulations of laminar flame propagation in droplet mists. *Combust. Flame* 156, 1627–1640.
- Neophytou, A., Mastorakos, E., Cant, R.S., 2012. The internal structure of igniting turbulent sprays as revealed by complex chemistry DNS. *Combust. Flame* 159, 641–664.
- Neophytou, A., Mastorakos, E., Cant, R.S., 2010. DNS of spark ignition and edge flame propagation in turbulent droplet-laden mixing layers. *Combust. Flame* 157, 1071–1086.
- Onuma, Y., Ogasawara, M., Inoue, T., 1977. Further Experiments on the Structure of a Spray Combustion Flame, in: Sixteenth Symposium (International) on Combustion, pp. 561–567.
- Onuma, Y., Ogasawara, M., Inoue, T., 1974. Studies of the Structure of a Spray Combustion Flame. *Symp. Combust.* 15, 453–465.
- Ozel Erol, G., Hasslberger, J., Klein, M., Chakraborty, N., 2018. A direct numerical simulation analysis of spherically expanding turbulent flames in fuel droplet-mists for an overall equivalence ratio of unity. *Phys. Fluids* 30, 086104.
- Pera, C., Chevillard, S., Reveillon, J., 2013. Effects of residual burnt gas heterogeneity on early flame propagation and on cyclic variability in spark-ignited engines. *Combust. Flame* 160, 1020–1032.
- Poinsot, T., Lele, S.K., 1992. Boundary conditions for direct simulations of compressible

- viscous flows. *J. Comput. Phys.* 101, 104–129.
- Polymeropoulos, C.E., 1984. Flame Propagation in Aerosols of Fuel Droplets, Fuel Vapor and Air. *Combust. Sci. Technol.* 40, 217–232.
- Reddy, H., Abraham, J., 2012. Two-dimensional direct numerical simulation evaluation of the flame-surface density model for flames developing from an ignition kernel in lean methane/air mixtures under engine conditions. *Phys. Fluids* 24, 105108.
- Reveillon, J., Demoulin, F.X., 2007. Evaporating droplets in turbulent reacting flows. *Proc. Combust. Inst.* 31, 2319–2326.
- Reveillon, J., Vervisch, L., 2000. Spray vaporization in nonpremixed turbulent combustion modeling: a single droplet model. *Proc. Combust. Inst.* 28, 75–90.
- Rogallo, R.S., 1981. Numerical Experiments in Homogeneous Turbulence, Report. California. Rotexo-Softpredict-Cosilab, GmbH and Co. KG Bad Zwischenahn, Germany.
- Schroll, P., Wandel, A.P., Cant, R.S., Mastorakos, E., 2009. Direct numerical simulations of autoignition in turbulent two-phase flows. *Proc. Combust. Inst.* 32 II, 2275–2282.
- Silverman, I., Greenberg, J.B., Tambour, Y., 1993. Stoichiometry and polydisperse effects in premixed spray flames. *Combust. Flame* 93, 97–118.
- Sreedhara, S., Huh, K.Y., 2007. Conditional statistics of nonreacting and reacting sprays in turbulent flows by direct numerical simulation. *Proc. Combust. Inst.* 31 II, 2335–2342.
- Swaminathan, N. and Bray, K.N.C., 2011. *Turbulent Premixed Flames*, Cambridge University Press, New York, USA, p. 5.
- Szekely, G.A., Faeth, G.M., 1983. Effects of envelope flames on drop gasification rates in turbulent diffusion flames. *Combust. Flame* 49, 255–259.
- U.S. Energy Information Administration, 2016. *International Energy Outlook 2016*, International Energy Outlook 2016.

- Wacks, D., Chakraborty, N., 2016a. Flame Structure and Propagation in Turbulent Flame-Droplet Interaction: A Direct Numerical Simulation Analysis. *Flow, Turbul. Combust.* 96, 1053–1081.
- Wacks, D., Chakraborty, N., 2016b. Statistical Analysis of the Reaction Progress Variable and Mixture Fraction Gradients in Flames Propagating into Droplet Mist: A Direct Numerical Simulation Analysis. *Combust. Sci. Technol.* 188, 2149–2177.
- Wacks, D., Chakraborty, N., Mastorakos, E., 2016. Statistical Analysis of Turbulent Flame-Droplet Interaction: A Direct Numerical Simulation Study. *Flow, Turbul. Combust.* 96, 573–607.
- Wandel, A.P., 2014. Influence of scalar dissipation on flame success in turbulent sprays with spark ignition. *Combust. Flame* 161, 2579–2600.
- Wandel, A.P., Chakraborty, N., Mastorakos, E., 2009. Direct numerical simulations of turbulent flame expansion in fine sprays. *Proc. Combust. Inst.* 32, 2283–2290.
- Wang, Y., Rutland, C.J., 2005. Effects of temperature and equivalence ratio on the ignition of n-heptane fuel spray in turbulent flow. *Proc. Combust. Inst.* 30, 893–900.
- Watanabe, H., Kurose, R., Hwang, S.M., Akamatsu, F., 2007. Characteristics of flamelets in spray flames formed in a laminar counterflow. *Combust. Flame* 148, 234–248.
- Watanabe, H., Kurose, R., Komori, S., Pitsch, H., 2008. Effects of radiation on spray flame characteristics and soot formation. *Combust. Flame* 152, 2–13.
- Wray, A.A., 1990. Minimal Storage Time Advanced Schemes for Spectral Methods, NASA Ames Research Centre. California.
- Xia, J., Luo, K.H., 2010. Direct numerical simulation of inert droplet effects on scalar dissipation rate in turbulent reacting and non-reacting shear layers. *Flow, Turbul. Combust.* 84, 397–422.

Yamashita, H., Shimada, M., Takeno, T., 1996. A numerical study on flame stability at the transition point of jet diffusion flames. *Symp. Combust.* 26, 27–34.

## TABLES

$a_d/\delta_{st}$	$\phi_{ov}$	$r_0/\delta_{st}$	$\tau$	$u'/S_{b(\phi_g=1)}$	$L_{11}/\delta_{st}$
0.04, 0.05, 0.06	0.8, 1.0, 1.2	2.0	6.54	4.0	2.5
$Re_t$	$Pr$	$Le$	$\gamma = C_p^g/C_v^g$	$Ma = S_{b(\phi_g=1)}/a_0$	$St$
70	0.7	1.0	1.4	0.014159	<0.1

**Table 1. Simulation parameters considered in the analysis.**

	$\phi_{ov} = 0.8$		$\phi_{ov} = 1.0$		$\phi_{ov} = 1.2$	
$a_d/\delta_{st}$	Laminar	$u'/S_{b(\phi_g=1)} = 4$	Laminar	$u'/S_{b(\phi_g=1)} = 4$	Laminar	$u'/S_{b(\phi_g=1)} = 4$
0.04	3.30	3.48	5.46	7.96	6.17	10.12
0.05	3.90	5.30	4.87	6.93	5.60	8.48
0.06	4.57	5.97	4.69	6.78	5.17	7.59
Premixed	3.18	4.10	5.42	10.44	5.83	8.15

**Table 2. Normalised flame speed  $S_A/S_{b(\phi_{ov})}$ , which quantifies the growth rate of flame surface area  $A$ .**

	$\phi_{ov} = 0.8$		$\phi_{ov} = 1.0$		$\phi_{ov} = 1.2$	
$a_d/\delta_{st}$	Laminar	$u'/S_{b(\phi_g=1)} = 4$	Laminar	$u'/S_{b(\phi_g=1)} = 4$	Laminar	$u'/S_{b(\phi_g=1)} = 4$
0.04	3.03	1.03	5.34	5.70	6.43	6.63
0.05	2.97	3.45	4.90	4.34	5.81	5
0.06	4.07	7.77	4.91	3.73	5.21	4.14
Premixed	3.55	2.32	5.5	6.63	6.25	7.28

**Table 3. Normalised flame speed  $S_V/S_{b(\phi_{ov})}$ , which quantifies the growth rate of the burned gas volume  $V_b$ .**

## FIGURE CAPTIONS

Fig.1: Distribution of  $c$  (white lines show  $c = 0.1, 0.5, 0.9$  contours from outer to inner periphery) on the central  $x$ - $y$  mid-plane for laminar (a) and turbulent (b) flames with  $\phi_{ov} = 0.8, 1.0$  and  $1.2$ . Black dots show the droplets residing on the plane (not to the scale). All figures correspond to  $t = 2.52\alpha_{T0}/S_{b(\phi_g=1)}^2$ .

Fig.2: Distribution of  $Y_F/Y_{Fst}$  (magenta lines show  $c = 0.1, 0.5, 0.9$  contours from outer to inner periphery) on the central  $x$ - $y$  mid-plane for laminar (a) and turbulent (b) flames with  $\phi_{ov} = 0.8, 1.0$  and  $1.2$ . Black dots show the droplets residing on the plane (not to the scale). All figures correspond to  $t = 2.52\alpha_{T0}/S_{b(\phi_g=1)}^2$ .

Fig.3: Distribution of  $T$  on the central  $x$ - $y$  mid-plane for laminar (a) and turbulent (b) flames with  $\phi_{ov} = 0.8$ ,  $\phi_{ov} = 1.0$  and  $\phi_{ov} = 1.2$ . Black dots show the droplets residing on the plane (not to the scale). All figures correspond to  $t = 2.52\alpha_{T0}/S_{b(\phi_g=1)}^2$ .

Fig. 4. Instantaneous view of  $c = 0.5$  isosurface coloured with local values of  $\kappa_m \times \delta_{st}$  for the cases with  $\phi_{ov} = 0.8$  (1<sup>st</sup> and 2<sup>nd</sup> row),  $\phi_{ov} = 1.0$  (3<sup>rd</sup> and 4<sup>th</sup> row) and  $\phi_{ov} = 1.2$  (5<sup>th</sup> and 6<sup>th</sup> row) at  $t = 2.52\alpha_{T0}/S_{b(\phi_g=1)}^2$ .

Fig. 5: PDFs of  $\kappa_m \times \delta_{st}$  for  $c = 0.1, 0.5$  and  $0.9$  isosurfaces for the premixed stoichiometric flame with  $\phi_{ov} = 0.8$  (— —),  $\phi_{ov} = 1.0$  (—)  $\phi_{ov} = 1.2$  (⊖) and for all droplet cases with initial  $\phi_{ov} = 0.8$  (—),  $\phi_{ov} = 1.0$  (—),  $\phi_{ov} = 1.2$  (—) for laminar (a) and turbulent (b) conditions. Same colour keys are used in Figs. 6-8,9-12.

Fig. 6: PDFs of  $|\vec{N} \cdot \vec{r}|$   $c = 0.1, 0.5$  and  $0.9$  isosurfaces. See Fig. 5 caption for the colour keys.

Fig.7: (a) Temporal evolution of normalised flame surface area  $A/A_0$ ; (b) Temporal evolution of flame surface area normalised by initial value of the stoichiometric premixed flame  $A/(A_{0,pre})_{st}$ . See Fig. 5 caption for the colour keys.

Fig. 8: PDF of  $\phi_g$  in the region corresponding to  $0.01 \leq c \leq 0.99$ . See Fig. 5 caption for the colour keys.

Fig. 9: Percentage of heat release arising from premixed (FI>0) and non-premixed (FI<0) modes of combustion for all droplet cases with initial  $a_d/\delta_{st}=0.04$  ( $\triangle$ ), 0.05 ( $\circ$ ), 0.06 ( $\times$ ).

Fig. 10: PDFs of  $|\nabla c| \times \delta_{st}$  on  $c = 0.1, 0.5$  and  $0.9$  isosurfaces under laminar (a) and turbulent (b) conditions. See Fig. 5 caption for the colour keys.

Fig. 11: Temporal evolutions of (a) volume integrated fuel reaction rate magnitude normalised by its initial value  $\Omega_F/\Omega_{F0}$ ; (b) volume integrated fuel reaction rate magnitude normalised by  $\Omega_{F0}$  value (i.e.  $\Omega_F/(\Omega_{F0,pre})_{st}$ ) for the stoichiometric laminar premixed flame. See Fig. 5 caption for the colour keys.

Fig. 12: Temporal evolutions of (a) the ratio of volume-integrated reaction rate of progress variable to the flame surface area  $S$  and (b) normalised volume  $V_b/(4\pi r_0^3/3)$  of the region with  $c \leq 0.99$ . See Fig. 5 caption for the colour keys.