1
2 **A Direct Numerical Simulation analysis of spherically expanding**
3 **turbulent flames in fuel droplet-mists for an overall equivalence**
4 **ratio of unity**

5
6 Gulcan Ozel Erol¹, Josef Hasslberger², Markus Klein², Nilanjan Chakraborty¹

7
8 ¹*School of Engineering, Newcastle University, Newcastle-Upon-Tyne, NE17RU, United*
9 *Kingdom.*

10 ²*Universität der Bundeswehr München, Fakultät für Luft- und Raumfahrttechnik, LRT1,*
11 *Werner-Heisenberg-Weg 39, 85577 Neubiberg, Germany*

ABSTRACT

Three-dimensional Direct Numerical Simulations (DNS) with a modified single-step Arrhenius chemistry have been used to analyse spherically expanding n-heptane flames propagating into mono-sized fuel droplet mists for different droplet diameters and an overall equivalence ratio of unity. The evolutions of flame surface area and burned gas volume for both laminar and turbulent spherically expanding droplet flames have been compared to the corresponding gaseous stoichiometric premixed spherically expanding flames with the same initial burned gas radius. It has been found that the initial droplet diameter significantly affects the burned gas volume and flame area generation, which increase with decreasing droplet diameter for both laminar and turbulent cases. The droplet-flame interaction plays a key role in determining flame wrinkling under laminar conditions, which is reflected in a range of local curvatures for a given reaction progress variable isosurface, whereas each progress variable isosurface in spherically expanding laminar premixed flames exhibits a single value of curvature. The effect of droplet-induced curvature becomes less distinguishable from turbulence-induced wrinkling for the turbulent cases considered here but the reaction progress variable isosurfaces in droplet cases exhibit wider curvature probability density functions than in the corresponding turbulent premixed flame cases. It has been found that heat release rate arises principally from premixed mode in small droplet cases, whereas the contribution of the non-premixed mode to the overall heat release rate increases with increasing droplet diameter and turbulence intensity.

Keywords: Droplet combustion, spherical expanding flame, flame surface area, curvature, Direct Numerical Simulations.

1. INTRODUCTION

Fuel is introduced as dispersed liquid droplets in many engineering devices such as Internal Combustion (IC) engines (e.g. Direct Injection (DI) and Compression Ignition (CI) engines) and gas turbines¹⁻³. Although turbulent combustion of droplet-laden mixtures has significant practical interests, it remains as one of the most challenging topics in thermo-fluid mechanics due to the involvement of complex interaction of fuel droplet evaporation, heat and mass transfer, fluid dynamics, and combustion thermo-chemistry. Therefore, gaining a thorough understanding for the turbulent spray combustion is of pivotal importance for the purpose of designing and developing fuel-efficient and low-emission combustion devices. Over the past decades, a number of experimental,⁴⁻¹³ analytical¹⁴⁻¹⁶ and Direct Numerical Simulation (DNS)¹⁷⁻³³ studies have focused on spray combustion. However, relatively limited effort^{6,7,13,24,30,31} has been directed to the analysis of the spherical flame propagation into droplet-laden mixtures in spite of its importance in automotive engines and explosion hazards.

Flame structure and propagation in droplet-laden mixtures have been analysed by several experimental studies. The pioneering experimental analysis by Burgoyne and Cohen⁴ indicated that the burning velocity and flammability limits of tetralin aerosols depend significantly on the droplet diameter. Suspensions of small droplets (diameter of 10 μm and below) behave similar to a gaseous mixture, whereas large droplets (with diameter of 40 μm and above) burn individually and can ignite other droplets around them. Ballal and Lefebvre⁸ experimentally demonstrated that the burning velocities for iso-octane droplet-mist for both overall fuel-lean and stoichiometric combustion increase with decreasing droplet diameters. They also indicated that the evaporation rate influences the flame propagation rate in the case of large droplets more than those of small droplets. Hayashi *et al.*⁷ experimentally observed that small droplet sizes of about 7 μm diameter create a smooth and continuous flame front but for the large droplet

sizes of about 20 μm diameter, the flame front is rugged, undulated and thickened because of the locally heterogeneous mixture fraction and burning rate. They suggested that the large fuel droplets of n-octane can augment the burning velocity under some conditions and these effects are negligible for small droplet sizes. Evaporation characteristic of the large propane droplets with a wide range of size (400-1000 μm diameter) was investigated as a function of mass fraction by Szekely and Faeth¹⁰ and Faeth¹¹ in a turbulent environment. Mizutani and co-workers^{5,6} reported that in spherically expanding flames, droplets may affect the rate of burning differently from the open burner flames. Lawes and Saat¹³ carried out a series of experiments to reveal the difference in the flame structure and burning rates between iso-octane spray and premixed combustion. It was observed by Lawes and Saat¹³ that the spray flame is more wrinkled than the gaseous flame under low turbulence intensity but no significant difference between gaseous and spray flame structure have been found under high turbulence intensity. All the aforementioned experimental investigations have indicated that the flame speed of spray flames depends on the complex interaction of droplet diameter, equivalence ratio and turbulence intensity.

The flame-droplet interaction was also analytically addressed by Polymeropoulos¹⁴ who found that the burning velocity assumes a maximum value at a certain droplet diameter. Silverman *et al.*¹⁵ developed an expression for laminar burning velocity of polydisperse spray flames by separating the combustion structure into a number of distinct zones. They also observed that initial Sauter mean diameter (SMD) is not enough to characterise polydisperse sprays because flames in different initial distributions with the same SMD behave differently. Greenberg *et al.*¹⁶ proposed an analytical model in order to capture the enhancement of burning velocity due to the distortion created by droplets on the flame front. Flame stretch was modelled for spherically expanding polydisperse spray flames for the first time by Greenberg and Kalma³⁴

who also accounted for the effects of finite-rate evaporation and droplet drag.³⁵ It was indicated that droplet evaporation rate plays an important role on the flame existence.

With the advancement of high-performance computing several researchers have used the carrier phase DNS to investigate the combustion of spray flames.^{17–33} Miller and Bellan¹⁷ carried out DNS to investigate a three-dimensional planar gas mixing layer growth and development for different initial liquid mass loading ratios, initial Stokes numbers and initial droplet temperatures. They revealed that the initial Stokes number has no significant influence on the evolution of the mixing layer and an increase in the mass loading ratio of the droplets leads to an attenuation of the growth rate of the mixing layer. Wang and Rutland¹⁹ and Schroll *et al.*²⁵ analysed the autoignition of n-heptane fuel droplets using DNS and the ignition delay has been found to increase with increasing equivalence ratio. Reveillon and Vervisch,¹⁸ Reveillon and Demoulin,²² Sreedhara and Huh²³ and Luo *et al.*²⁸ used the carrier phase DNS to explore the interactions between turbulent motion and spray combustion, which was subsequently used for different aspects of modelling of turbulent spray flames. Wandel *et al.*,²⁴ Wandel,³⁰ Neophytou *et al.*^{27,31} applied DNS to analyse spark ignition in turbulent droplet-laden mixing layers and confirmed previous experimental observations by Ballal and Lefebvre⁸ that large droplet diameters adversely affect the chance of successful ignition due to the lack of availability of gaseous fuel arising from slow evaporation rate. Wandel and co-workers^{24,30} analysed spark ignition of droplet-laden mixtures using DNS and indicated that both number and size of droplets influence the probability of successful ignition in turbulent environment. A comprehensive DNS investigation of the influences of equivalence ratio, fuel droplet size and radiation on jet spray flames has been conducted by Fujita *et al.*²⁹ It was found that the high temperature region moves from the jet centre to the outer jet edges as equivalence ratio increases, and an increase in droplet diameter leads to a decrease in the gas temperature in the

central downstream region. Wandel³⁰ performed a three-dimensional DNS based analysis of localised forced ignition of n-heptane liquid fuel droplets and suggested that the dissipation rate of reaction progress variable c can be a quantitative indicator to determine whether a flame will globally extinguish after being ignited by the spark. Neophytou *et al.*³¹ demonstrated that the structure of spray kernel flames can be very different from that of a pure gaseous flame. They carried out a parametric investigation on spark ignition of n-heptane turbulent sprays by three-dimensional detailed chemistry DNS. Recently, Wacks and co-workers^{32,33} have analysed the effects of initial droplet diameter, overall equivalence ratio, and turbulence intensity on statistically planar turbulent flame propagation into fuel-droplet mists. It has been found that the gaseous phase combustion takes place predominantly under fuel-lean mode even for fuel-rich overall equivalence ratios, and this trend strengthens with increasing droplet diameter because of slow evaporation of large droplets. Flame propagation in spray combustion has been analysed in terms of displacement speed statistics in Refs. 32 and 33 and it was indicated that the mean density-weighted displacement speed of turbulent spray flames remains smaller than the unstrained laminar burning velocity of a stoichiometric mixture due to the predominance of the fuel-lean mode of combustion.³² Furthermore, spray flames under turbulent conditions show a thicker flame structure than the corresponding turbulent stoichiometric premixed flames and this tendency strengthens with increasing droplet diameter³³.

To date, most DNS analyses on flame propagation in droplet-laden mixtures have been conducted for configurations without any global mean curvature.^{17,18,20,22,23,25,26,28,29,32,33} However, in many applications (e.g. automotive engines, accidental explosions) the flame is initiated as a spherical kernel, and it has been found that the evolution of spherically expanding premixed flames is fundamentally different from statistically planar turbulent premixed

flames³⁶⁻³⁹. A similar conclusion was also drawn based on experimental analysis of turbulent spray flames.⁶ DNS of spherically expanding flames in droplet mists is yet to be reported in the existing literature. In this paper, DNS of spherically expanding spray flames propagating into fuel-droplet mists have been conducted for a range of different initial droplet diameters ($a_d/\delta_{th} = 0.04, 0.05, 0.06$) and turbulence intensities ($u'/S_{b(\phi_g=1)} = 0.0, 4.0, 8.0$) at an overall equivalence ratio of $\phi_{ov} = 1.0$ (i.e. $\phi_{ov} = \phi_{liq} + \phi_g = 1.0$ considering fuel both in liquid and gaseous phases) in order to analyse the influences of droplet size and turbulence intensity on flame wrinkling and the burned gas volume in turbulent spherically expanding spray flames.

In this respect, the main objectives of this paper are:

- (a) To analyse the flame wrinkling induced by flame-droplet interaction for different droplet sizes and turbulence intensities in statistically spherical turbulent spray flames with $\phi_{ov} = 1.0$.
- (b) To investigate and explain the influences of droplet size and turbulence intensity on the flame structure and mode of combustion in turbulent spherically expanding spray flames with $\phi_{ov} = 1.0$.
- (c) To indicate the implications of the above information on the evolution of flame surface area and volume of burned gas in statistically spherical spray flames and their relative magnitudes in comparison to the corresponding values in statistically spherical stoichiometric turbulent premixed flames under similar turbulent flow conditions.

The rest of the paper is organised as follows. The mathematical background and numerical implementation related to this analysis are presented in the next two sections. Following that, the results will be presented and subsequently discussed. The main findings will be summarised and the conclusions are drawn in the final section of this paper.

2. MATHEMATICAL BACKGROUND

A modified single-step Arrhenius-type irreversible chemical reaction is chosen for the current

analysis for the purpose of computational economy because three-dimensional DNS simulations with detailed chemistry are extremely expensive for a parametric analysis involving three initial droplet diameters and three different flow conditions as carried out in this paper. For the present analysis, the following single-step irreversible reaction has been considered:

where s is the mass of the oxidizer consumed per unit mass of fuel consumption. Considering the modified Arrhenius-type chemical mechanism, the fuel reaction rate is given by:^{24,30,32,33,40}

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

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

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

Here B is the pre-exponential factor, E_{ac} is the activation energy, R is the universal gas constant, $T_{ad}(\phi_g=1)$ is the adiabatic flame temperature for the stoichiometric mixture, $\hat{T}$ is the instantaneous dimensional temperature, T_0 is the unburned gas temperature and $\tau = (T_{ad}(\phi_g=1) - T_0)/T_0$ is a heat release parameter. Considering the suggestion of Tarrazo *et al.*,⁴⁰ the activation energy, E_{ac} , and the heat of combustion have been taken to be functions of the gaseous equivalence ratio, ϕ_g which provides a realistic equivalence ratio ϕ_g dependence of the unstrained laminar burning velocity $S_{b(\phi_g)}$ in hydrocarbon-air flames. According to Tarrazo *et al.*,⁴⁰ the Zel'dovich number, β , can be written as $\beta = 6f(\phi_g)$ where $f(\phi_g)$ is expressed based on the gaseous phase equivalence ratio ϕ_g as:⁴⁰

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

The heat release per unit mass of fuel $H_{\phi_g} = \left[(T_{ad}(\phi_g) - T_0) C_p \right] / [Y_{F0}(\phi_g) - Y_{Fb}(\phi_g)]$ 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 fraction in the unburned and burned gases, respectively for a premixed flame of equivalence ratio ϕ_g .⁴⁰ The modified single-step chemical mechanism used for this analysis provides realistic equivalence ratio dependence of unstrained laminar burning velocity, especially for fuel-rich mixtures for hydrocarbon fuels⁴⁰. It has been shown in Fig. 1 in Tarrazo *et al.*⁴⁰ that this mechanism compares favourably with both experiments and detailed chemistry simulations. The interested readers are also referred to Fig. 1a of Malkeson and Chakraborty.⁴¹ where it is shown that the modified single step chemistry by Tarrazo *et al.*⁴⁰ captures the realistic variation of laminar burning velocity with gaseous equivalence ratio ϕ_g in the context of the thermo-chemistry used in this analysis. A recent analysis by Haruki *et al.*⁴² followed a similar practice for calibrating a two-step chemical mechanism for a DNS analysis involving droplet combustion. There have been several previous studies^{22–24,29,30,32} where a single-step simple chemical mechanism was used to analyse spray combustion. It is worthwhile to indicate that, most three-dimensional DNS analyses on droplet combustion used single step simple chemistry and only a few studies^{27,31,43} have been performed in the presence of detailed chemical mechanisms.

Several previous DNS analyses on gaseous turbulent premixed flames demonstrated that the statistical behaviours of reaction rate of progress variable and the reaction progress variable gradient can be adequately captured by single-step chemistry (see comparison between

Chakraborty and Cant⁴⁴ and Chakraborty *et al.*⁴⁵ for reaction rate; comparison between Chakraborty *et al.*⁴⁴ and Peters *et al.*⁴⁶, Echekki and Chen⁴⁷ for flame propagation; comparison between Chakraborty and Klein⁴⁸ and Chakraborty *et al.*⁴⁵, Chakraborty *et al.*⁴⁹, Lai *et al.*⁵⁰ for scalar gradient statistics). As the current analysis principally focuses on the reaction rate and reaction progress variable statistics, it can be expected that the present findings will at least be qualitatively valid in the context of a detailed chemistry based analysis.

The Lewis numbers ($Le_i = \alpha_i/D_i$ where α_i is the thermal diffusivity and D_i is the species diffusivity of i^{th} species) of all species are taken to be equal to unity. In this analysis, all species in the gaseous phase are taken to be perfect gases and thus in the context of compressible flow DNS, the pressure p is evaluated using density and temperature as $p = \rho R \hat{T}$ where R is the gas constant. Standard values have been taken 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 μ , is the dynamic viscosity and λ is the thermal conductivity of the gaseous phase).

The individual droplets are tracked in a Lagrangian sense, whereas compressible Navier-Stokes equations are solved in the Eulerian frame for the carrier gaseous phase. The evolution of the position, $\vec{x}_d$, velocity, $\vec{u}_d$, diameter $\vec{a}_d$, and temperature $\vec{T}_d$ is expressed in the following manner according to Reveillon and Vervisch:¹⁸

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

where L_v is the latent heat of vaporization, and τ_d^u, τ_d^p and τ_d^T are relaxation time scales associated with droplet velocity, diameter and temperature, respectively which are expressed as:^{18,22,24,30,32,33}

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

where ρ_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 defined as:^{18,22,24,30,32,33}

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

where Y_F^s is the fuel mass fraction at the surface of the droplet and given by: $Y_F^s = (1 + W_o/W_F [p(\vec{x}_d, t)/p_F^s - 1])^{-1}$ where W_o and W_F are the molecular weights of oxidiser and fuel, respectively. Here p_F^s represents the partial pressure of the fuel vapor at the droplet surface, and is calculated using 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. Recently, Haruki et al.⁴², compared the performance of an evaporation model based on a Level-Set approach with the point source assumption for evaporating droplets and a good agreement between these approaches has been reported.

236

For this analysis, the drag coefficient is corrected ($C_u = 1 + Re_d^{2/3}/6$) for large values of the droplet Reynolds number $Re_d = \rho |\vec{u}(\vec{x}_d, t) - \vec{u}_d| a_d / \mu$ (i.e. $Re_d \gg 1$). The same expression was used in many previous analysis for spray combustion simulations.^{18,19,22,23,26} Here, 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 is not expected to have a major significant influence on the simulation results. Accordingly, a simple drag coefficient correction has been considered in this analysis. Alternative empirical drag coefficients, including the blowing velocity at the droplet surface, were preferred in some previous analyses^{17,20,21,51,52} but the exact

nature of empiricism is likely to have higher order effects for small values of Re_d when DNS is carried out for the carrier phase and the droplets are considered 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 given in a general format:^{18,22,24,30,32,33}

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

where $\psi = \{1, u_j, e, Y_F, Y_O\}$ and $\psi_1 = \{1, u_j, \hat{T}, Y_F, Y_O\}$ for the conservation equations of mass, momentum, energy, and mass fractions, respectively and $\Gamma_\psi = \rho \nu / \sigma_\psi$ for $\psi = \{1, u_j, Y_F, Y_O\}$ and $\Gamma_\psi = \lambda$ for $\psi = e$, respectively. Here, u_j represents the velocity in the j^{th} direction. 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 due to droplet evaporation, which is tri-linearly interpolated from the droplet's sub-grid position, $\vec{x}_d$, to the eight surrounding nodes. Other variables are ν , kinematic viscosity, and σ_ψ , an appropriate Schmidt number corresponding to ψ . The droplet source term for any variable ψ is expressed as:^{24,25,30-33}

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

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 defined as a function of oxygen mass fraction, Y_O and mixture fraction, ξ and takes values between 0 (in unburned reactants) and 1 (in burned products):^{24,30-33}

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

where $Y_{O\infty} = 0.233$ is the oxygen mass fraction in air, $Y_{F\infty} = 1.0$ is the fuel mass fraction in the pure fuel stream and ξ is the mixture fraction which is defined as:⁵³

$$\xi = \frac{(Y_F - Y_O/s + Y_{O\infty}/s)}{(Y_{F\infty} + Y_{O\infty}/s)} \quad (11)$$

where 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

A three dimensional compressible DNS code SENGAs^{24,25,30–33} which solves the standard conservation equations of mass, momentum, energy and species of the gaseous phase in non-dimensional form, has been used for this analysis. The spatial differentiation is carried out using a 10th order central difference scheme for the internal grid points, but the order of differentiation gradually decreases to a one-sided 2nd order scheme at the non-periodic boundaries. The time advancement is carried out using an explicit low-storage 3rd order Runge-Kutta scheme⁵⁴. 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 α_{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.⁵⁵ A uniform computational grid of $512 \times 512 \times 512$ is used to discretize the domain, which ensures about 10 grid points within $\delta_{th} = (T_{ad(\phi_g=1)} - T_0)/\max|\nabla T|_L$, which is the unstrained thermal laminar flame thickness of the stoichiometric mixture.

For the purpose of initialization, 1D steady-state laminar spray flame solutions are generated for three different initial values of droplet diameter a_d (i.e. $a_d/\delta_{th} = 0.04, 0.05$ and 0.06) for an overall equivalence ratio ϕ_{ov} of unity (i.e. $\phi_{ov} = 1.0$) using COSILAB⁵⁶ following Neophytou and Mastorakos.⁵⁷ These 1D steady state solutions are then specified in the radial direction from the centre of the domain. This initialised scalar field is allowed to evolve in such

290 a manner in a quiescent environment that the spherical flame can propagate and adjust itself.
 291 This phase has been continued until the radius of the fully burned gas region (i.e. the region
 292 corresponding to reaction progress variable c value greater than 0.99) r_0 reaches $2\delta_{th}$ (i.e.
 293 $r_0/\delta_{th} = 2.0$). It is recognised that the energy content for a stoichiometric premixed spherical
 294 flame with radius $r_0 = 2\delta_{th}$ is different from a spherical droplet flame with same burned gas
 295 radius (i.e. $c \geq 0.99$). However, for the purpose of comparison the initial burned gas (i.e. $c \geq$
 296 0.99) radius for all cases considered here is taken to be $r_0/\delta_{th} = 2.0$.

297

298 For turbulent simulations, the turbulent velocity field is superimposed on the laminar spherical
 299 flames with $r_0/\delta_{th} = 2.0$. A standard pseudo-spectral method⁵⁸ has been used to initialize
 300 turbulent velocity fluctuations by an incompressible homogeneous isotropic velocity field and
 301 the unburned gas temperature T_0 is taken to be 300 K. This yields a heat release parameter $\tau =$
 302 $(T_{ad(\phi_g=1)} - T_0)/T_0$ of 6.54 (i.e. $\tau = 6.54$). The turbulent flame simulations are performed for
 303 initial values of normalised root-mean-square (rms) turbulent velocities $u'/S_{b(\phi_g=1)} = 4.0$ and
 304 8.0 with a non-dimensional longitudinal integral length-scale of $L_{11}/\delta_{th}=2.5$, and have been
 305 continued for at least 2.1 chemical time scale (i.e. $2.1t_{chem} = 2.1\alpha_{T0}/S_{b(\phi_g=1)}^2$), which
 306 corresponds to about 2.0 and 4.0 initial eddy turnover times (i.e. $2L_{11}/\sqrt{k} - 4L_{11}/\sqrt{k}$ where
 307 k is the kinetic energy evaluated over the whole domain) for $u'/S_{b(\phi_g=1)} = 4.0$ and 8.0 cases
 308 respectively. The simulation time used in this analysis remains comparable to a number of
 309 recent DNS analyses,^{24,27,30–33,36–39,59–62} which significantly contributed to the fundamental
 310 understanding of turbulent combustion. The turbulent kinetic energy evaluated over the whole
 311 domain does not vary rapidly with time when the statistics were extracted. The rms velocity
 312 fluctuation evaluated over the whole domain decayed by about 30% and 50% for
 313 $u'/S_{b(\phi_g=1)}=4.0$ and 8.0 cases respectively when the statistics were extracted.

314

315 The initial droplet number density ρ_N varies between $1.38 \leq (\rho_N)^{1/3} \delta_{th} \leq 2.07$ in the
 316 unburned gas, and the liquid volume fraction remains well below 0.01. The droplet diameter
 317 remains smaller than the Kolmogorov length scale for all cases and the ratio of initial droplet
 318 diameter to the Kolmogorov length scale is $a_d/\eta = 0.25, 0.32, 0.38$ for the cases of $a_d/\delta_{th} =$
 319 $0.04, 0.05, 0.06$ respectively for the highest turbulence intensity (i.e. the initial
 320 $u'/S_{b(\phi_g=1)}=8.0$) case considered here. The Stokes number $= \tau_p \sqrt{k}/L_{11} =$
 321 $\rho_d a_d^2 \sqrt{k}/(18 C_u \mu L_{11})$ (where $\tau_p = \rho_d a_d^2/18 C_u \mu$ is the particle time scale and $L_{11}/\sqrt{k}$ is the
 322 turbulent time scale) for the largest droplet remains smaller than 0.1 in the turbulent cases
 323 considered here. An alternative Stokes number $St' = \tau_p S_{b(\phi_g=1)}^2/\alpha_{T0} =$
 324 $\rho_d a_d^2 S_{b(\phi_g=1)}^2/(18 C_u \mu \alpha_{T0})$ can be defined based on the chemical timescale t_{chem} and the
 325 maximum value of St' remains smaller than 5.0×10^{-2} for the largest droplet considered in
 326 this analysis. The mean normalised inter-droplet distance s_d/η ranges between 4.17 and 6.25
 327 for the highest turbulence intensity (i.e. the initial $u'/S_{b(\phi_g=1)}=8.0$) case considered here. The
 328 ratio of the initial droplet diameter to grid spacing used here remains comparable to several
 329 previous analyses.^{19,24,25,27,29–33} Moreover, the evaporation of droplets due to the relatively high
 330 volatility of n-heptane, leads to a decrease in droplet diameter by the time they reach the most
 331 reacting place in the reaction zone. The droplet diameter decreases at least by 50% when it
 332 reaches the reaction zone. Thus, the droplets which interact with the flame front are much
 333 smaller in reality than the initial size of the droplets. Accordingly, the assumption of sub-grid
 334 evaporation is not expected to significantly affect the statistics of flame-droplet interaction,
 335 which is the main focus of the analysis. As the droplets, which interact with the flame front,
 336 are in reality much smaller than the initial size of the droplets, the modelling of sub-grid
 337 evaporation is not expected to significantly affect the statistics of flame-droplet interaction. For

this reason, it can be expected that the qualitative nature of the findings is not sensitive to the evaporation modelling used in this analysis.

4. RESULTS AND DISCUSSION

4.1 Flame-turbulence interaction

FIG. 1. (Left to right) Distribution of T , Y_F/Y_{st} (magenta lines show $c = 0.1, 0.5, 0.9$ contours from outer to inner periphery) and 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 flames. Black dots show the droplets residing on the plane (not to the scale). All figures correspond to $t = 2.1 t_{chem}$.

FIG. 2. (Left to right) Distribution of T , Y_F/Y_{st} (magenta lines show $c = 0.1, 0.5, 0.9$ contours from outer to inner periphery) and 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 turbulent flames with initial $u'/S_{b(\phi_g=1)} = 4.0$ at $t = 2.1t_{chem}$. Black dots show the droplets residing on the plane (not to the scale).

344

345 The instantaneous non-dimensional temperature T , normalized gaseous fuel mass fraction,

346 Y_F/Y_{st} and reaction progress variable c fields at the central $x - y$ mid-plane at $t = 2.1t_{chem}$

347 are shown in Figs. 1-3 for both laminar and turbulent flow conditions, respectively. It can be

seen from Fig. 1 that the contours of c remain perfect circles for the laminar stoichiometric premixed flame case indicating that the flame grows as a perfect sphere. By contrast, the contours of c for laminar droplet kernels are not perfectly circular because these flames do not remain perfectly spherical due to flame-droplet interaction. The above observations are consistent with the previous experimental findings of Hayashi *et al.*⁷ and Lawes and Saat¹³. It can be seen from Fig. 2 and 3 that the droplet-induced flame wrinkling cannot be distinguished from turbulence-induced flame deformation for both premixed and droplet cases.

The droplets residing on the $x - y$ mid-plane are shown as black dots in Figs. 1-3. Chiu and Liu^{63,64} defined a group number as: $G = 3(1 + 0.276Re_d^{1/2}Sc^{1/3})LeN^{2/3}(a_d/s_d)$ (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) in order to distinguish between individually burning droplets ($G \ll 1.0$) and external sheath combustion ($G \gg 1.0$). All droplet cases considered here come under the category of external sheath combustion (i.e. have values of G much greater than unity). Although Figs. 1-3 may give an impression of individually burning droplets, these droplets are indeed sub-grid particles which reside on the plane shown in the figure. Interested readers are referred to Reveillon and Vervisch¹⁸ for further discussion in this regard.

In both laminar and turbulent cases, the droplets reduce in size due to evaporation as they approach the flame and eventually disappear as a result of complete evaporation (examples are shown in the insets of Fig. 1-3, see 1st column and 2nd row). During the evaporation process, droplets extract latent heat from the surrounding gaseous phase and this contributes to the local reduction in the temperature of the burned gas region in the droplet cases in comparison to the corresponding gaseous premixed flame cases, and this effect is prominent for large droplets

373 (see 1st column of Figs. 1-3). However, the evaporation rate was not sufficient to provide
374 stoichiometric mixture ahead of the flame in all the droplet cases considered here (see 2nd
375 column of Figs. 1-3) and thus the combustion of predominantly fuel-lean mixture ($Y_F/Y_{Fst} <$
376 1.0) also acts to bring down the burned gas temperature in comparison to that in the
377 stoichiometric premixed flame. For the initial $a_d/\delta_{th} = 0.04$ case, the evaporation rate is greater
378 than in the other droplet cases and thus droplets evaporate either mostly ahead of or within the
379 flame in this case but for the initial $a_d/\delta_{th} = 0.05$ and 0.06 cases droplets penetrate the flame
380 and continue to evaporate in the burned gas region and unburned fuel pockets can be found
381 within the burned gas kernel in these cases (significantly fuel-rich pockets in the burned gas
382 can be discerned in the $a_d/\delta_{th} = 0.06$ case in Figs. 1-3).

FIG. 3. (Left to right) Distribution of T , Y_F/Y_{st} (magenta lines show $c = 0.1, 0.5, 0.9$ contours from outer to inner periphery) and 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 turbulent flames with initial $u'/S_{b(\phi_g=1)} = 8.0$ at $t = 2.1t_{chem}$. Black dots show the droplets residing on the plane (not to the scale).

Under high turbulence intensity, the flames become severely wrinkled and move away from the centre of the domain and the out of plane movement leads to broken contours of T , Y_F/Y_{st} and c in Fig. 3 for both premixed and droplet cases. The strong turbulent motion also carries the droplets across the flame and increases the availability of droplets in the burned gas side.

This tendency is particularly dominant for large droplet cases. The effects of droplets on c -contour lines (see 3rd column of Figs. 1-3) are evident for both laminar and turbulent cases and the flames exhibit instances of local flame thickening for large droplet cases. This tendency is particularly evident under high turbulence intensity cases.

The initial values of Damköhler and Karlovitz numbers (i.e. $Da_{(\phi_g=1)} = L_{11}S_{b(\phi_g=1)}^2/u'\alpha_{T0}$ and $Ka_{(\phi_g=1)} = (u'/S_{b(\phi_g=1)})^{1.5} (L_{11}S_{b(\phi_g=1)}/\alpha_{T0})^{-0.5}$) for the premixed case with initial $u'/S_{b(\phi_g=1)} = 4.0$ are 1.31 and 3.49 respectively. The values of $Da_{(\phi_g=1)}$ and $Ka_{(\phi_g=1)}$ are 0.65 and 9.88 respectively for the premixed case with initial $u'/S_{b(\phi_g=1)} = 8.0$. The value of Damköhler (Karlovitz) number for the droplet cases is likely to be smaller (greater) than the corresponding premixed flame cases because of predominantly fuel-lean burning (which implies that the chemical time scale is likely to be larger than $\alpha_{T0}/S_{b(\phi_g=1)}^2$ because of smaller burning velocity). These Damköhler and Karlovitz number values are representative of the distributed burning regime and thus all the cases 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 influences of the droplets on the flame surface are evident from Fig. 4 where the instantaneous $c = 0.5$ isosurfaces are coloured by local values of flame curvature $\kappa_m \times \delta_{th}$. The local curvature κ_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, the flame normal vector points towards the reactants and an element of flame surface, which is convex (concave) to the reactants, has a positive (negative) curvature. Fig. 4 shows dimples (i.e. droplet-induced curvatures) on $c = 0.5$ isosurfaces in both laminar (1st row) and turbulent (2nd and 3rd row) droplet cases, which

are completely absent in the laminar premixed flame.

Fig. 4. Instantaneous view of $c = 0.5$ isosurface coloured with local values of $\kappa_m \times \delta_{th}$ for laminar (1st row) and turbulent (2nd and 3rd row) flames at $t = 2.1t_{chem}$.

413

414 The droplet-induced flame deformation can be quantified with the help of the Probability
 415 Density Functions (PDFs) of κ_m shown in Fig. 5 for $c = 0.1, 0.5$ and 0.9 isosurfaces. The
 416 curvature PDFs of the laminar droplet cases show a spread of curvature values due to flame-
 417 droplet interaction, whereas the PDF of κ_m is a delta function for the laminar premixed flame
 418 owing to its perfect spherical shape. The PDFs in the turbulent cases show higher probability
 419 of finding positive values (i.e. flame surface convex to the reactants) than obtaining negative
 420 curvatures (i.e. flame surface concave to the reactants) but the droplet cases exhibit wider κ_m
 421 distributions than the premixed case because of the droplet-induced curvatures. The widening
 422 of curvature PDFs is particularly prominent for larger droplets especially in the burned gas
 423 region for both laminar and turbulent flames. The large droplets can survive longer than others

due to their slow evaporation and as a result they can interact with the flame towards the burned gas side of the flame front. The same tendency was also experimentally observed^{7,13} but these effects were more magnified in Lawes and Saat¹³ due to the presence of thermo-diffusive and hydro-dynamic instabilities.

FIG. 5. PDFs of $\kappa_m \times \delta_{th}$ for $c =$ (a,d,g) 0.1, (b,e,h) 0.5 and (c,f,i) 0.9 isosurfaces for the premixed stoichiometric flame (—) and for all droplet cases with initial $a_d/\delta_{th}=0.04$ (—), 0.05 (—), 0.06 (—) for laminar (dashed line) and turbulent (solid lines). Same colour keys are used in Figs. 6-8,11-13,15-17,19.

The magnitude of inner product of the local flame normal $\vec{N}$ and the unit radial vector $\vec{r}$ from the centre of the mass of the flame kernel can be used to quantify the extent of flame wrinkling. The angle between these two unit vectors ($\vec{N}$ and $\vec{r}$) is equal to zero (or the projection $|\vec{N} \cdot \vec{r}|$ equals to $|\cos\theta| = |\vec{N} \cdot \vec{r}| = 1.0$) for a perfectly spherical flame. Greater extent of flame wrinkling leads to higher extent of the deviation of the projection $|\vec{N} \cdot \vec{r}|$ from 1.0. The PDFs of $|\vec{N} \cdot \vec{r}|$ are shown for $c = 0.1, 0.5$ and 0.9 isosurfaces in Fig. 6. It can be seen from Figs. 6a-c that PDFs of $|\vec{N} \cdot \vec{r}|$ become a delta function at $|\vec{N} \cdot \vec{r}| = 1.0$ for the laminar premixed flame case

436 due to its perfect spherical shape. The PDFs of $|\vec{N} \cdot \vec{r}|$ for the laminar droplet flames assume
 437 peak value at 1.0 but they also show finite probability of having $|\vec{N} \cdot \vec{r}| < 1.0$ indicating
 438 imperfect spherical shape as apparent from Fig. 4 due to droplet-induced flame wrinkling. A
 439 careful examination of the PDFs of $|\vec{N} \cdot \vec{r}|$ for laminar cases reveals that the extent of deviation
 440 of $|\vec{N} \cdot \vec{r}|$ from 1.0 is the highest for $a_d/\delta_{th} = 0.06$ amongst all the droplet cases considered
 441 here. However, there is no significant difference between the extent of flame wrinkling of the
 442 premixed and droplet turbulent flames with the initial $u'/S_{b(\phi_g=1)}=4.0$ where the wrinkling by
 443 turbulence eclipses the wrinkling due to the flame-droplet interaction, which is consistent with
 444 previous experimental findings^{7,13}. Additionally, increasing turbulence intensity leads to a
 445 significant increase in the extent of wrinkling for premixed flame, and this is reflected in the
 446 greater extent of deviation of $|\vec{N} \cdot \vec{r}|$ from 1.0 for the premixed case than the droplet cases at
 447 high values of $u'/S_{b(\phi_g=1)}$. This behaviour originates due to the differences in flame surface
 448 area between the premixed and droplet flame cases.

FIG. 6. PDFs of $|\vec{N} \cdot \vec{r}|$ on $c = 0.1$ (a,d,g), 0.5 (b,e,h) and 0.9 (c,f,i) isosurfaces. See Fig. 5 caption for the colour keys.

FIG.7. (a-c) Temporal evolution of normalised flame surface area A/A_0 ; (d-f) Temporal evolution of flame surface area normalized by initial value of premixed flame $A/A_{0,pre}$; (g-i) PDF of $|\nabla c| \times \delta_{th}$ on $c = 0.5$. See Fig. 5 caption for the colour keys.

is around 5 for laminar premixed flame. However, the turbulent droplet case with initial
 $a_d/\delta_{th} = 0.04$ and $u'/S_{b(\phi_g=1)} = 4$ (see Fig. 7b) shows marginally higher value of A/A_0 than
 the turbulent stoichiometric premixed flame, whereas turbulent droplet cases with initial
 $a_d/\delta_{th} = 0.05$ and 0.06 exhibit A/A_0 values smaller than the corresponding premixed flame
 case for $u'/S_{b(\phi_g=1)} = 4$. The value of A/A_0 in the premixed flame is greater than all droplet
 cases for initial turbulence intensity of $u'/S_{b(\phi_g=1)} = 8$ (see Fig. 7c). Moreover, A/A_0
 decreases with increasing droplet diameter for both laminar and turbulent cases. However, it is
 worth noting that A_0 values are different for the cases considered here and A_0 (i.e. flame surface
 area $A = \int_V |\nabla c| dV$ of a spherical laminar flame of initial burned gas radius $r_0 = 2\delta_{th}$) for
 $a_d/\delta_{th} = 0.04, 0.05$ and 0.06 cases have been found to be 0.53, 0.576, 0.66 times the value for
 the stoichiometric laminar premixed flame $A_{0,pre}$, respectively. The temporal evolution of
 $A/A_{0,pre}$ for all the laminar and turbulent cases are shown in Figs. 7d-f which indicate that
 $A/A_{0,pre}$ in droplet cases remains smaller than the stoichiometric premixed flame case for both
 laminar and turbulent conditions. In order to explain this difference in $A/A_{0,pre}$ values between
 the cases considered here, the PDFs of $|\nabla c| \times \delta_{th}$ are exemplarily shown in Fig. 7g-i for $c =$
 0.5 isosurface (same qualitative trend is observed for other c isosurfaces) for all cases
 considered here. Figures 7g-i show that the droplet cases exhibit greater probability of finding
 small values of $|\nabla c|$ than the corresponding premixed flame cases except for the laminar case
 with $a_d/\delta_{th} = 0.04$, and the probability of finding smaller values of $|\nabla c|$ is greater for
 $a_d/\delta_{th} = 0.06$ cases than in $a_d/\delta_{th} = 0.05$ cases. 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 $A/A_{0,pre}$ in the droplet cases with $a_d/\delta_{th} = 0.05$ and 0.06 in comparison to the
 corresponding premixed case. It is worth noting that the volume over which $|\nabla c|$ assumes non-
 zero values is smaller in the laminar droplet flame with $a_d/\delta_{th} = 0.04$ than in the

corresponding laminar premixed flame. This leads to smaller values of $A/A_{0,pre}$ and A_0 in the laminar droplet flame with $a_d/\delta_{th} = 0.04$ than in the corresponding laminar premixed flame in spite of exhibiting greater probability of obtaining higher $|\nabla c|$ value in comparison to the premixed laminar flame. The integral that yields A_0 value is influenced by the magnitude of $|\nabla c|$ and the volume over which $|\nabla c|$ assumes non-zero values. The smaller magnitudes of $|\nabla c|$ in laminar droplet cases with initial $a_d/\delta_{th} = 0.05$ and 0.06 in comparison to those in 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 $r_0 = 2\delta_{th}$. Thicker flame (i.e. larger flame volume) in the laminar droplet cases with initial $a_d/\delta_{th} = 0.05$ and 0.06 , in comparison to the corresponding droplet case with initial $a_d/\delta_{th} = 0.04$ gives rise to greater values of A_0 than the corresponding value in the $a_d/\delta_{th} = 0.04$ case in spite of exhibiting higher probability of showing smaller values of $|\nabla c|$ and identical values of r_0 . In the laminar droplet cases with initial $a_d/\delta_{th} = 0.05$ and 0.06 the combination of smaller magnitude of $|\nabla c|$ is principally responsible for yielding smaller value of A_0 than $A_{0,pre}$ in spite of all cases having same initial burned gas radius $r_0 = 2\delta_{th}$. 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 $A/A_{0,pre}$ under turbulent conditions in Figs. 7d-f.

Table 1. Normalised flame speed $S_A/S_{b(\phi_g=1)}$, which quantifies the growth rate of flame surface area A .

	<i>Laminar</i>	$u'/S_{b(\phi_g=1)} = 4$	$u'/S_{b(\phi_g=1)} = 8$
$a_d/\delta_{th}=0.04$	5.460	7.956	9.154
$a_d/\delta_{th}=0.05$	4.871	6.928	7.228
$a_d/\delta_{th}=0.06$	4.687	6.777	7.312
Premixed	5.423	10.445	16.000

The rate of flame surface area growth can be estimated by a flame speed S_A , which is defined

504 as $S_A = dr_A/dt$ where $r_A = \sqrt{A/4\pi}$ is an equivalent radius based on flame surface area. The
 505 value of S_A is estimated by using the slope of the linear part of temporal evolution of r_A and
 506 this slope remains unchanged since halfway through the simulation. The evaluated values of
 507 S_A are summarised in Table 1, which indicates that the laminar droplet case with initial
 508 $a_d/\delta_{th} = 0.04$ grows slightly faster than the spherically expanding stoichiometric laminar
 509 premixed flame but S_A for laminar droplet cases with initial $a_d/\delta_{th} = 0.05$ and 0.06 are found
 510 to be smaller than that in the premixed case. However, turbulent stoichiometric premixed cases
 511 exhibit greater values of S_A than the droplet cases. The value of S_A tends to decrease with
 512 increasing a_d/δ_{th} for droplet cases. Furthermore, S_A increases with increasing $u'/S_{b(\phi_g=1)}$.
 513 All the above conclusions are consistent with the observations made from Figs. 7a-c but Table
 514 1 provides quantifications of the effects of droplet size and turbulence intensity on the growth
 515 rate of flame surface area.

FIG. 8. (a-c) PDF of ϕ_g in the region corresponding to $0.01 \leq c \leq 0.99$; (d-f) PDF of non-dimensional temperature T in the region corresponding to $c \geq 0.99$.

516

517 A comparison between Y_F/Y_{Fst} fields in Fig. 1-3 (2nd column) reveals that combustion
 518 predominantly takes place under fuel-lean mode (i.e. $Y_F/Y_{Fst} < 1$) for all laminar and turbulent

droplet cases in spite of having an overall equivalence ratio of $\phi_{ov} = 1.0$. The PDFs of gaseous phase equivalence ratio ϕ_g for $0.01 \leq c \leq 0.99$ are shown in Fig. 8a-c, which shows a considerable variation of ϕ_g within the flame. Although the PDF of ϕ_g in Fig. 8b peaks at 1.0 for the $a_d/\delta_{th} = 0.04$ case under turbulence conditions with initial $u'/S_{b(\phi_g=1)} = 4$, the probability of finding fuel-lean (i.e. $\phi_g < 1$) mixture overwhelms the availability of $\phi_g \geq 1$ mixtures. This tendency strengthens further for the $a_d/\delta_{th} = 0.05$ and 0.06 cases. Moreover, it is evident from Fig. 8c that an increase in turbulence intensity enhances the likelihood of finding fuel-lean mixtures. The turbulent mixing strengthens with increasing u' which enables turbulent transport of gaseous fuel from the evaporation sites to distant locations which would have not been possible for either laminar or smaller values of u' . This reduces the probability of finding localised fuel-rich pockets and increases the possibility of obtaining fuel-lean mixtures in the droplet cases for an initial turbulence intensity of $u'/S_{b(\phi_g=1)} = 8.0$ in comparison to the corresponding laminar and $u'/S_{b(\phi_g=1)} = 4.0$ droplet cases.

The predominant fuel-lean mode of combustion and the extraction of latent heat by droplets reaching the burned gas act to reduce the burned gas temperature, and this can be substantiated from the PDFs of T , shown for the region corresponding to $c \geq 0.99$ in Fig. 8d-f, which indicate that there is a considerable probability of finding $T < 1.0$ in the burned gas for the droplet cases whereas the corresponding PDF for the stoichiometric premixed flames (both laminar and turbulent) is a delta function at $T = 1.0$. However, the non-dimensional burned gas temperature for the droplet cases also shows small finite probabilities of obtaining $T \geq 1.0$ even when the peak value of the gaseous equivalence ratio PDF is obtained at $\phi_g < 1$ (e.g. see $a_d/\delta_{th} = 0.05$ and 0.06 cases under turbulent conditions in Fig. 8a-c). In order to explain this behaviour, it is necessary to understand the reaction zone structure in the spherically expanding droplet flames.

4.3 Reaction zone structure in spherically expanding droplet flames

The mode of combustion can be distinguished with the help of the flame index FI, which is defined as $FI = (\nabla Y_F \cdot \nabla Y_O) / (|\nabla Y_F| |\nabla Y_O|)^{0.5}$. The flame index FI assumes positive (negative) values for 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. The total heat release rate is calculated with $HR_{total} = \int_V H_\phi |\dot{w}_F| dV$ where H_ϕ indicates the heat release per unit mass of fuel and is defined using the approach developed by Tarrazo *et al.*

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 $a_d/\delta_{th}=0.04$ ($\triangle$), 0.05 ($\circ$), 0.06 ($\times$).

It is evident from Fig. 9 that the cases with small droplets show that heat release predominantly arises from the premixed mode of combustion. The contribution of premixed (non-premixed) mode of combustion to the overall heat release rate decreases (increases) with increasing droplet diameter due to slow evaporation rates. Furthermore, turbulent cases show a higher contribution of non-premixed combustion mode to total heat release rate than the corresponding laminar droplet cases. Turbulence not only mixes the gaseous fuel with the surrounding air but also transports droplets. As evident from Figs. 2 and 3, more droplets can penetrate towards the burned gas side of the flame under turbulent conditions than in the

corresponding laminar cases and this effect is particularly prevalent for large droplets. These droplets evaporate and release gaseous fuel in the burned side and some of that fuel diffuses back to the unburned gas side to form local diffusion (i.e. non-premixed) flames, which also contribute to the overall heat release. This trend can be seen from the c isosurfaces coloured by FI for $a_d/\delta_{th}=0.06$ under laminar and high turbulence intensity conditions in Fig.10. For $c = 0.5$ and 0.9 isosurfaces, the flame index FI predominantly assumes positive values indicating the dominance of premixed mode but local islands of non-premixed mode ($FI < 0$) can also be discerned. However, this behaviour is reversed on the unburned gas side (e.g. $c = 0.1$) of the flame front. In this configuration, the fuel in the gaseous phase originates from evaporation of droplets and the evaporated fuel mixes with air as it burns on the unburned gas side of the flame front and thus the flame index FI assumes predominantly negative values.

FIG. 10. Distribution of flame index (FI) on different c isosurfaces ($c = 0.9, 0.5$ and 0.1 left to right) for $a_d/\delta_{th}=0.06$ case for laminar flow and initial $u'/S_{b(\phi_g=1)}=8.0$ conditions.

For a detailed investigation on flame index and reaction rate of c across the flame, the mean values of FI and the normalised reaction rate of progress variable $\dot{w}_c \times \delta_{th}/\rho_0 S_{b(\phi_g=1)}$ (with

577 ρ_0 being the unburned gas density) conditional on c are shown in Fig. 11 where $\dot{w}_c$ is given
 578 as:^{24,27,30–33}

$$\dot{w}_c = \begin{cases} -\frac{\xi_{st}\dot{w}_O}{[\xi(1-\xi_{st})Y_{O\infty}]} & , \xi \leq \xi_{st} \\ -\frac{\dot{w}_O}{[(1-\xi)Y_{O\infty}]} & , \xi > \xi_{st} \end{cases} \quad (12)$$

579 where $\dot{w}_O = s\dot{w}_F$ is the oxygen reaction rate. It is evident from Figs. 11a-c that low values of
 580 c are associated with negative values of FI, whereas the flame index is predominantly positive
 581 for $c \geq 0.3$. However, the mode of combustion becomes predominantly non-premixed (FI <
 582 0) around $c \approx 1.0$. This non-premixed mode of combustion originates from the diffusion of
 583 oxidiser from the unburned gas side and diffusion of gaseous fuel from post-flame evaporation
 584 for the droplets which escape through the flame on the burned gas side. The transport and
 585 mixing of evaporated fuel from the droplet sites to the surrounding gaseous mixture strengthens
 586 with increasing turbulence intensity and thus the extent of non-premixed combustion increases
 587 with increasing $u'/S_{b(\phi_g=1)}$. A comparison between FI and $\dot{w}_c$ distributions in Fig. 12 indicates
 588 that the premixed mode of combustion (i.e. FI > 0) is obtained in the reaction zone where the
 589 mean value of $\dot{w}_c$ conditional upon c assumes large values.

FIG. 11. Variations of the mean values of flame index FI and normalised reaction rate of reaction

progress variable $\dot{w}_c \times \delta_{th}/\rho_0 S_{b(\phi_g=1)}$ conditional upon c .

The probability of obtaining fuel-lean mixture increases for increasing a_d/δ_{th} due to decreasing evaporation rates (see Fig. 8). This leads to decreases in the peak mean value of $\dot{w}_c$ conditional upon c with increasing droplet size for both laminar and turbulent cases. The mean value of $\dot{w}_c$ conditional upon c peaks approximately at $c = 0.75$ for all premixed cases but this location for droplet cases moves towards the burned gas side for all droplet cases with increasing turbulence intensity and the peak value decreases with increasing droplet diameter. For the present thermo-chemistry the activation energy (or Zel'dovich number β , see eq. 3) does not change and remains same as the stoichiometric mixture for fuel-rich mixtures in the equivalence ratio range of $1 < \phi_g < 1.07$ (see eq. 4)⁴⁰. Thus, the considerable availability of fuel-rich mixture with $1 < \phi_g < 1.07$ in the laminar droplet case with initial $a_d/\delta_{th} = 0.04$ (see Fig. 8) leads to a higher peak mean value of $\dot{w}_c$ conditional upon c than in the stoichiometric laminar premixed flame case. However, the probability of finding less reactive mixtures corresponding to $\phi_g < 1.0$ and $\phi_g > 1.07$ increases with increasing $u'/S_{b(\phi_g)}$. This leads to a decrease in the peak mean value of $\dot{w}_c$ conditional upon c in the turbulent droplet cases in comparison to that in the corresponding turbulent stoichiometric spherical premixed flame.

It can be seen from Fig. 11d-f that the peak mean value of $\dot{w}_c$ for the droplet cases decreases with increasing $u'/S_{b(\phi_g=1)}$. This behaviour can be explained with the help of the variations of the mean normalized heat release rate $HR^* = H_\phi |\dot{w}_F| \times \delta_{th}/(C_p T_0 \rho_0 S_{b(\phi_g=1)})$ conditional upon c for the droplet cases, which are shown in Fig. 12. It can be seen from Fig. 12 that the mean value of HR^* arising from premixed mode of combustion remains greater than that from non-premixed mode of combustion for all droplet cases and this tendency is particularly strong

for the cases with small droplet diameter. This is consistent with the observations made earlier from Fig. 9. The peak mean value of HR^* for the premixed mode of combustion takes place close to $c = 0.75$, whereas the peak mean value of HR^* for non-premixed mode of combustion takes place towards the burned gas side of the flame. The maximum heat release for non-premixed combustion takes place at the stoichiometric mixture (i.e. $\xi = \xi_{st}$) which occurs for a reaction progress variable value close to unity (i.e. $c \approx 1.0$), which is consistent with previous findings by Wacks *et al.*³². The value of reaction progress variable, where the peak location of HR^* is obtained, changes depending on the contribution of the non-premixed mode of combustion to the total HR^* .

FIG. 12. Variation of normalised mean heat release, $HR^* = H_\phi |\dot{w}_F| \times \delta_{th} / (C_p T_0 \rho_0 S_b(\phi_g=1))$ with c for droplet cases. Total mean heat release (—), mean heat release arising from premixed (FI>0) combustion mode (—) and non-premixed (FI<0) combustion mode (—).

Figure 13 shows the variation of mean values of flame index FI and reaction rate $\dot{w}_c$ conditional on mixture fraction ξ in the region corresponding to $0.01 \leq c \leq 0.99$. It can be seen from

Fig.13a-c that FI is predominantly negative for $\xi < \xi_{st}$. In this configuration, the fuel in gaseous form is obtained only upon evaporation of droplets. Thus, the bulk of the fuel-lean mixture is obtained when fuel vapour diffuses to surrounding air from evaporation sites. The evaporation rate is high for small droplets and there are more evaporation sites for small values of a_d for a given value of ϕ_{ov} . The combination of these effects leads to an increased probability of having positive values of FI for small droplet cases in the gaseous mixture where some fuel is already present. Accordingly, mean FI values remain positive for $\xi > 0.05$ in the case of $a_d/\delta_{th} = 0.04$ irrespective of flow conditions. A similar behaviour has been observed for the laminar droplet case with $a_d/\delta_{th} = 0.05$. However, the mean value of FI assumes negative value close to $\xi \approx \xi_{st}$ for the laminar case with $a_d/\delta_{th} = 0.06$ and also for the turbulent cases considered here for $a_d/\delta_{th} = 0.05$ and 0.06 because of the diffusion flame formed at the stoichiometric mixture. The signature of the diffusion flame at $\xi = \xi_{st}$ can be seen as a kink at $\xi = \xi_{st}$ in the profiles of the mean values of $\dot{w}_c$ conditional upon ξ , which are presented in Figs. 12d-f. A comparison between Figs. 13a-c with Figs. 13d-f reveals that high mean values of $\dot{w}_c$ conditional on ξ originate due to the rich premixed mode of combustion. It has been found that the mean value of $\dot{w}_c$ conditional on ξ is obtained at around $\xi \approx 0.07$ for all flow conditions. It is worth noting that $\xi \approx 0.07$ corresponds to a gaseous phase equivalence ratio $\phi_g = \xi(1 - \xi_{st})/[(1 - \xi)\xi_{st}] \approx 1.07$ for which the highest laminar burning velocity $S_{b(\phi_g)}$ is obtained for this thermo-chemistry.⁴⁰ A decrease in droplet size for a given ϕ_{ov} gives rise to an increase in the number of droplets and these small droplets evaporate more readily than large droplets owing to their high surface to volume ratio. The extraction of latent heat at the evaporation sites tends to reduce the gas temperature, which in turn acts to increase the number of samples with small values of c for a given mixture fraction ξ value with decreasing a_d/δ_{th} (not shown here but the mean value of c conditional upon ξ decreases with decreasing a_d/δ_{th}), and this leads to a reduction of mean value of $\dot{w}_c$ conditional upon ξ . This effect is

more prominent for large values of $u'/S_{b(\phi_g=1)}$ (e.g. initial $u'/S_{b(\phi_g=1)} = 8.0$ case) because the enhanced mixing of fuel from the evaporation sites increases the Spalding number B_d , which in turn increases evaporation rate of droplets. It is worth noting that the profile of the mean value of HR^* conditional on ξ is qualitatively similar to the variation of mean reaction rate $\dot{w}_c$ conditional on mixture fraction and thus is not shown here for the sake of conciseness. A comparison between Figs. 12 and 13d-f confirms that non-premixed mode of combustion occurs at $c \approx 1.0$ where mixture fraction is close to its stoichiometric value $\xi \approx \xi_{st}$.

FIG. 13. Variation of mean flame index FI and normalised reaction rate of reaction progress variable $\dot{w}_c \times \delta_{th}/\rho_0 S_{b(\phi_g=1)}$ conditional upon mixture fraction ξ in the region corresponding to $0.01 \leq c \leq 0.99$. The red dashed line shows the value of stoichiometric mixture fraction.

For a diffusion flame formed at $\phi_g \approx 1.0$ (i.e. $\xi \approx \xi_{st}$), the corresponding burned gas temperature is expected to be $T \approx 1.0$ under globally adiabatic condition with unity Lewis number for all species. Moreover, the premixed mode of combustion also takes place for $\phi_g \approx 1.0$, which yields a burned gas temperature corresponding to $T \approx 1.0$. It can be seen from Figs.

1-3 that some droplets penetrate the flame front, evaporate at the burned gas side, and generate locally fuel-rich pockets, whereas the unused oxidiser diffuses from the unburned gas side to the burned gas side from the fuel-lean zones. Instances of heat transfer to the gaseous phase from high temperature droplets give rise to a small probability of obtaining $T \geq 1.0$ upon completion of combustion of the evaporated fuel.

The reaction rate and heat release variations have implications on the statistical behaviours of the magnitude of gradient of reaction progress variable $|\nabla c|$ and also on thermal expansion due to heat release which can be quantified in terms of dilatation rate $\nabla \cdot \vec{u}$. It can be seen from Fig. 14 that both $\nabla \cdot \vec{u} \times \delta_{th}/S_L$ (see Figs. 14a-c) and $|\nabla c| \times \delta_{th}$ (see Figs. 14d-f) conditional upon c show a decreasing trend with increasing droplet size, which is qualitatively consistent with the variations of mean value of $\dot{w}_c$ conditional upon c (see Fig. 11). For the premixed flame cases, there is no considerable effect of turbulence intensity on mean values of $\nabla \cdot \vec{u} \times \delta_{th}/S_{b(\phi_g=1)}$ conditional upon c . However normalised mean dilatation rate $\nabla \cdot \vec{u} \times \delta_{th}/S_L$ in the droplet cases decreases with increasing turbulence intensity and droplet diameter because of the reduction in heat release rate (see Fig. 12 and 13d-f) as a result of predominant fuel-lean burning. The mean value of $\nabla \cdot \vec{u} \times \delta_{th}/S_{b(\phi_g=1)}$ for turbulent premixed flames remains greater than those for turbulent droplet cases and the same trend has been observed for laminar droplet cases with initial $a_d/\delta_{th} = 0.05$ and 0.06 . However, the laminar droplet case with initial $a_d/\delta_{th} = 0.04$ shows higher peak mean value of $\nabla \cdot \vec{u} \times \delta_{th}/S_{b(\phi_g=1)}$ than in the premixed flame, which is consistent with the higher mean value of $\dot{w}_c$ conditional upon c for this droplet case (see Fig. 11).

FIG. 14. Variations of mean values of (a-c) $\nabla \cdot \vec{u} \times \delta_{th}/S_L$ and (d-f) $|\nabla c| \times \delta_{th}$ conditional upon c at $t = 2.1t_{chem}$.

686

687 The variation of the mean value $|\nabla c| \times \delta_{th}$ conditional upon c exhibits a peak value close to
688 $c \approx 0.6$ for premixed cases, whereas its location shifts to $c \approx 0.65$ for droplet cases. The peak
689 mean value of $|\nabla c| \times \delta_{th}$ for the premixed flames increases with increasing turbulence
690 intensity. The peak mean value of $|\nabla c|$ can be taken as a measure of the inverse of flame
691 thickness^{41,66–68} and thus Figs. 14d-f indicate a thinning in flame thickness with increasing
692 turbulence intensity for premixed flame cases. This is consistent with findings from several
693 previous analyses and interested readers are referred to Sandeep *et al.*⁶⁷ and Chakraborty *et*
694 *al.*⁶⁸ for further discussion in this regard. However, the peak mean value of $|\nabla c| \times \delta_{th}$ decreases
695 with increasing a_d/δ_{th} and $u'/S_{b(\phi_g=1)}$ in turbulent droplet cases, which suggests that the
696 flame tends to be thicker as turbulence intensity and droplet size increase. This is consistent
697 with the predominance of fuel-lean combustion for large values of turbulence intensity and
698 droplet size, and thus the flame thickness is expected to increase with decreasing ϕ_g in fuel-
699 lean gaseous mixtures. The flame thickening in droplet cases in comparison to the

corresponding stoichiometric premixed flame is obtained for all turbulent cases, and also for laminar flames with initial $a_d/\delta_{th} = 0.05$ and 0.06 . However, the laminar droplet case with initial $a_d/\delta_{th} = 0.04$ shows higher peak mean value of $|\nabla c| \times \delta_{th}$ than in the premixed flame, which is consistent with the higher mean value of $\dot{w}_c$ conditional upon c for this droplet case (see Fig. 11) due to the considerable availability of $1 < \phi_g < 1.07$, as discussed earlier. For this thermo-chemistry the laminar burning velocity $S_{b(\phi_g)}$ and the characteristic flame thickness $\delta \sim \alpha_{T0}/S_{b(\phi_g)}$ attain their maximum and minimum values respectively for $\phi_g \approx 1.07$ ⁴⁰. Thus, the significant presence of $1 < \phi_g < 1.07$ in the flame in the laminar droplet case with initial $a_d/\delta_{th} = 0.04$ gives rise to the thinner flame than in the stoichiometric premixed spherical laminar flame. This has implications on the overall flame area $A = \int_V |\nabla c| dV$ and volume of the burned gas (i.e. volume of gas with $c \geq 0.99$) as it gives rise to smaller values of A in the droplet flame with initial $a_d/\delta_{th} = 0.04$ than in the corresponding stoichiometric premixed spherical flame even though the mean value of $|\nabla c| \times \delta_{th}$ is greater in this droplet case (see Fig. 14). This arises because the volume over which $|\nabla c|$ assumes significant values in this droplet case (i.e. $a_d/\delta_{th} = 0.04$) is smaller than the corresponding volume in the premixed flame case. The same reason is responsible for yielding smaller value of A_0 in the droplet case with initial $a_d/\delta_{th} = 0.04$ than in the laminar stoichiometric premixed spherical flame even though the burned gas radius is the same (i.e. $r_0 = 2\delta_{th}$) for both cases.

4.4 Statistical behaviour related to the burned gas volume

A combination of fuel-lean combustion and the predominant probability of obtaining reduced burned gas temperature adversely affects the growth rate of spherically expanding flames. This can be verified from Fig. 15a-c which shows temporal evolution of the volume-integrated fuel reaction rate magnitude $\Omega_F = \int_V |\dot{w}_F| dV$ normalised by its initial value (i.e. $\Omega_F/\Omega_{F0} =$

724 $\int_V |\dot{w}_F| dV / [\int_V |\dot{w}_F| dV]_{t=0}$.

FIG. 15. Temporal evolutions of (a-c) volume integrated fuel reaction rate magnitude normalised by its initial value Ω_F/Ω_{F0} ; (d-f) volume integrated fuel reaction rate magnitude normalised by Ω_{F0} value (i.e. $\Omega_{F0,pre}$) for the stoichiometric laminar premixed flame.

725

726 Figures 15a-c show that Ω_F/Ω_{F0} increases rapidly with time for all cases but under turbulent
 727 environment the growth rate is the highest for the premixed case and Ω_F/Ω_{F0} decreases with
 728 increasing droplet diameter under both laminar and turbulent conditions. However, Ω_F/Ω_{F0} in
 729 the laminar droplet cases is found to be greater than in the laminar stoichiometric premixed
 730 flame. The growth rate of Ω_F/Ω_{F0} is found to be qualitatively consistent with the evolution of
 731 A/A_0 , as shown in Fig. 7a-c. However, it is worth noting that Ω_{F0} values (i.e. Ω_F of a spherical
 732 laminar flame of initial radius $r_0 = 2\delta_{th}$) are not the same as shown in Fig. 15d-f and Ω_{F0} for
 733 $a_d/\delta_{th} = 0.04, 0.05$ and 0.06 cases have been found to be 0.564, 0.567, 0.57 times the Ω_{F0}
 734 value (i.e. $\Omega_{F0,pre}$) for the stoichiometric laminar premixed flame, respectively. Thus,
 735 $\Omega_F/\Omega_{F0,pre}$ in the droplet cases remains smaller than the stoichiometric premixed flame, and it
 736 decreases with increasing a_d under both laminar and turbulent conditions. It is worth

737 mentioning that the temporal evolution of $\Omega_c = \int_V \dot{w}_c dV$ and Ω_c/Ω_{c0} are qualitatively similar
 738 to Ω_F and Ω_F/Ω_{F0} respectively. The temporal evolutions of $S = [\rho_0 S_{b(\phi_g=1)}]^{-1} \int_V \dot{w}_c dV /$
 739 $\int_V |\nabla c| dV$ are shown in Fig. 16a-c, which shows that S remains close to unity in the premixed
 740 flame under both laminar and turbulent conditions. The departure of S from a unity value (i.e.
 741 $S < 1$) for spherical stoichiometric premixed flames takes place due to the non-zero stretch rate
 742 effects induced by non-zero mean flame curvature. This behaviour is consistent with previous
 743 findings by Klein *et al.*³⁶ for statistically spherical turbulent premixed flames and detailed
 744 explanations for this behaviour have been provided in Refs. 39-39 and thus are not repeated
 745 here. Figures 16a-b shows that the values of S for the initial $a_d/\delta_{th} = 0.04$ cases remain
 746 comparable to the stoichiometric premixed flame but S decreases with increasing initial droplet
 747 size, which is qualitatively consistent with the experimental findings of Lawes and Saat¹³.
 748 Under high turbulence intensity (e.g. initial $u'/S_{b(\phi_g=1)}=8.0$ case), S for the droplet case (Fig.
 749 16c) diminishes and stays at considerably lower values compared with other flow conditions
 750 (i.e. the initial $u'/S_{b(\phi_g=1)}=0.0$ and 4.0). The product formation rate per unit flame surface
 751 area in the large droplet cases is smaller than that in the stoichiometric premixed flame case.
 752 This can be verified from Figs. 16d-f, which show a reduction of the volume of the gas with
 753 $c \geq 0.99$ (i.e. V_b) with increasing droplet size and the highest V_b is obtained for the gaseous
 754 premixed flame. It is evident from Figs. 16d-f that the difference of V_b between premixed and
 755 spray flame widens with increasing turbulence intensity and this difference for high turbulence
 756 intensity (e.g. initial $u'/S_{b(\phi_g=1)}=8.0$ case) originates due to the smaller S in droplet flames
 757 than in the corresponding stoichiometric turbulent premixed case.

FIG. 16. Temporal evolutions of (a-c) the ratio of volume-integrated product formation rate to the flame surface area S ; (d-e) volume $V_b/(4\pi r_0^3/3)$ of the region with $c \geq 0.99$.

758

759 The rate of growth of the burned gas volume can be estimated by a flame speed S_V , which is
760 defined as $S_V = dr_V/dt$ where $r_V = (3V_b/4\pi)^{1/3}$ is an equivalent radius based on burned gas
761 volume. The value of S_V is estimated by using the slope of the linear part of temporal evolution
762 of r_V and this slope remains unchanged since halfway through the simulation. The evaluated
763 values of S_V are summarised in Table 2, which reveals that the growth of burned gas volume
764 for the stoichiometric premixed flame and the droplet case with initial $a_d/\delta_{th} = 0.04$
765 increases with increasing turbulence intensity. However, the values of $S_V/S_{b(\phi_g=1)}$ for the
766 droplet cases with initial $a_d/\delta_{th} = 0.05$ and 0.06 remain smaller than the corresponding
767 laminar case values for initial $u'/S_{b(\phi_g=1)}=4.0$, whereas $S_V/S_{b(\phi_g=1)}$ under initial
768 $u'/S_{b(\phi_g=1)}=8.0$ in these droplet cases assumes values comparable to that under the laminar
769 condition. Although the ratio of volume-integrated product formation rate to the flame surface
770 area S decreases with increasing $u'/S_{b(\phi_g=1)}$ for the $a_d/\delta_{th} = 0.05$ and 0.06 (see Figs. 18a-
771 c), this effect is countered by the higher growth rate of flame surface area (see Table 1) with

increasing $u'/S_{b(\phi_g=1)}$. These two counter-acting effects lead to a non-monotonic variation of S_V with increasing $u'/S_{b(\phi_g=1)}$ for droplet cases with initial $a_d/\delta_{th} = 0.05$ and 0.06 .

Table 2. Normalised flame speed $S_B/S_{b(\phi_g=1)}$, which quantifies the growth rate of the burned gas volume V_b .

	<i>Laminar</i>	$u'/S_{b(\phi_g=1)} = 4$	$u'/S_{b(\phi_g=1)} = 8$
$a_d/\delta_{th}=0.04$	5.336	5.703	5.770
$a_d/\delta_{th}=0.05$	4.899	4.338	4.589
$a_d/\delta_{th}=0.06$	4.906	3.733	4.077
Premixed	5.496	6.626	9.951

5. CONCLUSIONS

The evolution of flame surface area, burned gas volume and flame structure in spherically expanding n-heptane flames propagating into fuel droplet mists has been analysed for different droplet diameters for an overall equivalence ratio of unity using three-dimensional DNS under laminar and turbulent conditions. In order to compare the droplet flame statistics with a corresponding premixed flame case, additional spherically expanding stoichiometric premixed flame cases with same initial burned gas radius and statistically similar fluid flow conditions have been considered. It has been found that turbulence intensity and droplet size have important influences on the flame structure, flame surface area and volume of burned gas. The flame-droplet interaction leads to flame wrinkling even under laminar conditions for spherically expanding flames into the droplet-mist and the effects of droplet-induced wrinkling strengthen with increasing droplet diameter. This effect is partially eclipsed by the flow-induced flame wrinkling in turbulent droplet cases but its imprint can be seen in the form of wider curvature PDFs for droplet cases in comparison to the corresponding stoichiometric statistically spherical premixed flame cases. The growth of flame area normalised by its initial value decreases with increasing droplet diameter and the turbulence enhances the normalised flame area for both premixed and droplet cases. The combustion mainly takes place under fuel-

lean mode for droplet cases and high turbulence intensity promotes the availability of fuel lean mixture. A major part of the overall heat release in droplet cases arises due to premixed mode of combustion. However, the contribution of non-premixed mode of combustion to overall heat release rate increases with high turbulence intensity and large droplet sizes. A combination of predominant fuel-lean burning and extraction of latent heat from the gaseous phase in the droplet cases leads to local reduction of burned gas temperature in comparison to the adiabatic flame temperature of the stoichiometric mixture. The turbulent droplet cases have been shown to exhibit greater propensity of obtaining smaller magnitudes of the reaction progress variable gradient, dilatation rate and product formation rate per unit flame surface area than in the corresponding stoichiometric turbulent premixed flames. These trends strengthen with increasing droplet size due to slow burning of predominant fuel-lean mixture arising from slow evaporation of large fuel droplets. Thus, an increase in initial droplet diameter and turbulence intensity adversely affects the evolution of burned gas volume and flame surface area for the parameter range considered here, which is consistent with previous experimental findings^{7,13}. It has been shown that an increase in turbulence intensity leads to flame thinning for the premixed flames considered here, whereas for droplet cases, the flame becomes thicker under increasing turbulence intensity. Detailed physical explanations have been provided for the aforementioned observed behaviours. The statistical behaviours of scalar gradient, flame index, dilatation rate and reaction rate in response to the variations of droplet diameter and turbulence intensity discussed in this paper will be valuable for the purpose of the modelling of combustion processes involving droplet-laden mixtures. Although the current analysis has been carried out using a simple chemical mechanism for moderate values of turbulent Reynolds, number several findings of this analysis are in agreement with previous experimental observations^{7,13}. Additionally, a simple sub-grid evaporation model has been chosen to account for point source droplets. However, a variety of different evaporation models are available and a number of

821 recent investigations^{69–71} focussed on their accuracies and computational performances. Thus,
822 further analyses in the presence of detailed chemical mechanism and more advanced
823 evaporation models at higher values of turbulent Reynolds number will be necessary for
824 obtaining more comprehensive physical understanding, which will form the basis of future
825 investigations.

826

827 **ACKNOWLEDGEMENTS**

828 The financial support of the Republic of Turkey Ministry of National Education and EPSRC
829 and computational support of Rocket (Newcastle University) and ARCHER are gratefully
830 acknowledged.

831

REFERENCES

- ¹ J. B. Heywood, *Internal Combustion Engine Fundamentals*, 1st ed. (1988).
- ² A.H. Lefebvre and D.R. Ballal, *Gas Turbine Combustion Alternative Fuels and Emissions*, 3rd ed. (2010).
- ³ S.K. Aggarwal, "A review of spray ignition phenomena: Present status and future research," *Prog. Energy Combust. Sci.* **24**, 565 (1998).
- ⁴ Burgoyne J. H. and Cohen L., "The Effect of Drop Size on Flame Propagation in Liquid Aerosols," **225**, 375 (1954).
- ⁵ Y. Mizutani and T. Nishimoto, "Turbulent Flame Velocities in Premixed Sprays Part I. Experimental Study," *Combust. Sci. Technol.* **6**, 1 (1972).
- ⁶ Y. Mizutani and A. Najkajima, "Combustion of Fuel Vapor-Drop-Air Systems : Part II- Spherical Flames in a Vessel," *Combust. Flame* **20**, 351 (1973).
- ⁷ S. Hayashi, S. Kumagai, and T. Sakai, "Propagation Velocity and Structure of Flames in Droplet-Vapor-Air Mixtures," *Combust. Sci. Technol. Taylor & Francis* **15**, 169 (1977).
- ⁸ D.R. Ballal and A.H. Lefebvre, "Flame propagation in heterogeneous mixtures of fuel droplets, fuel vapor and air," *Symp. Combust.* **18**, 321 (1981).
- ⁹ S.K. Aggarwal and W.A. Sirignano, "Unsteady spray flame propagation in a closed volume," *Combust. Flame* **62**, 69 (1985).
- ¹⁰ G.A. Szekely and G.M. Faeth, "Effects of envelope flames on drop gasification rates in turbulent diffusion flames," *Combust. Flame* **49**, 255 (1983).
- ¹¹ G.M. Faeth, "Mixing, transport and combustion in sprays," *Prog. Energy Combust. Sci.* **13**, 293 (1987).
- ¹² H. Nomura, M. Koyama, H. Miyamoto, Y. Ujiie, J. Sato, M. Kono, and S. Yoda, "Microgravity experiments of flame propagation in ethanol droplet-vapor-air mixture," *Proc. Combust. Inst. Elsevier* **28**, 999 (2000).

857 ¹³ M. Lawes and A. Saat, "Burning rates of turbulent iso-octane aerosol mixtures in spherical
858 flame explosions," *Proc. Combust. Inst.* **33**, 2047 (2011).

859 ¹⁴ C.E. Polymeropoulos, "Flame Propagation in Aerosols of Fuel Droplets, Fuel Vapor and
860 Air," *Combust. Sci. Technol. Taylor & Francis* **40**, 217 (1984).

861 ¹⁵ I. Silverman, J.B. Greenberg, and Y. Tambour, "Stoichiometry and polydisperse effects in
862 premixed spray flames," *Combust. Flame* **93**, 97 (1993).

863 ¹⁶ J.B. Greenberg, I. Silverman, and Y. Tambour, "On Droplet Enhancement of the Burning
864 Velocity of Laminar Premixed Spray Flames," *Combust. Flame* **113**, 271 (1998).

865 ¹⁷ R.S. Miller and J. Bellan, "Direct numerical simulation of a confined three-dimensional gas
866 mixing layer with one evaporating hydrocarbon-droplet-laden stream," *J. Fluid Mech.*
867 *Cambridge University Press* **384**, 293 (1999).

868 ¹⁸ J. Reveillon and L. Vervisch, "Spray vaporization in nonpremixed turbulent combustion
869 modeling: a single droplet model," *Combust. Flame* **121**, 75 (2000).

870 ¹⁹ Y. Wang and C.J. Rutland, "Effects of temperature and equivalence ratio on the ignition of
871 n-heptane fuel spray in turbulent flow," *Proc. Combust. Inst.* **30**, 893 (2005).

872 ²⁰ M. Nakamura, F. Akamatsu, R. Kurose, and M. Katsuki, "Combustion mechanism of liquid
873 fuel spray in a gaseous flame," *Phys. Fluids* **17**, 1 (2005).

874 ²¹ H. Watanabe, R. Kurose, S.M. Hwang, and F. Akamatsu, "Characteristics of flamelets in
875 spray flames formed in a laminar counterflow," *Combust. Flame* **148**, 234 (2007).

876 ²² J. Reveillon and F.X. Demoulin, "Evaporating droplets in turbulent reacting flows," *Proc.*
877 *Combust. Inst.* **31**, 2319 (2007).

878 ²³ S. Sreedhara and K.Y. Huh, "Conditional statistics of nonreacting and reacting sprays in
879 turbulent flows by direct numerical simulation," *Proc. Combust. Inst.* **31 II**, 2335 (2007).

880 ²⁴ A.P. Wandel, N. Chakraborty, and E. Mastorakos, "Direct numerical simulations of turbulent
881 flame expansion in fine sprays," *Proc. Combust. Inst.* **32**, 2283 (2009).

882 ²⁵ P. Schroll, A.P. Wandel, R.S. Cant, and E. Mastorakos, "Direct numerical simulations of
883 autoignition in turbulent two-phase flows," *Proc. Combust. Inst. The Combustion Institute* **32**
884 **II**, 2275 (2009).

885 ²⁶ J. Xia and K.H. Luo, "Direct numerical simulation of inert droplet effects on scalar
886 dissipation rate in turbulent reacting and non-reacting shear layers," *Flow, Turbul. Combust.*
887 **84**, 397 (2010).

888 ²⁷ A. Neophytou, E. Mastorakos, and R.S. Cant, "DNS of spark ignition and edge flame
889 propagation in turbulent droplet-laden mixing layers," *Combust. Flame* **157**, 1071 (2010).

890 ²⁸ K. Luo, H. Pitsch, M.G. Pai, and O. Desjardins, "Direct numerical simulations and analysis
891 of three-dimensional n-heptane spray flames in a model swirl combustor," *Proc. Combust. Inst.*
892 Elsevier Inc. **33**, 2143 (2011).

893 ²⁹ A. Fujita, H. Watanabe, R. Kurose, and S. Komori, "Two-dimensional direct numerical
894 simulation of spray flames - Part 1: Effects of equivalence ratio, fuel droplet size and radiation,
895 and validity of flamelet model," *Fuel* **104**, 515 (2013).

896 ³⁰ A.P. Wandel, "Influence of scalar dissipation on flame success in turbulent sprays with spark
897 ignition," *Combust. Flame* **161**, 2579 (2014).

898 ³¹ A. Neophytou, E. Mastorakos, and R.S. Cant, "The internal structure of igniting turbulent
899 sprays as revealed by complex chemistry DNS," *Combust. Flame* **159**, 641 (2012).

900 ³² D.H. Wacks, N. Chakraborty, and E. Mastorakos, "Statistical Analysis of Turbulent Flame-
901 Droplet Interaction: A Direct Numerical Simulation Study," *Flow, Turbul. Combust.* **96**, 573
902 (2016).

903 ³³ D.H. Wacks and N. Chakraborty, "Flame Structure and Propagation in Turbulent Flame-
904 Droplet Interaction: A Direct Numerical Simulation Analysis," *Flow, Turbul. Combust.* **96**,
905 1053 (2016).

906 ³⁴ J.B. Greenberg and A. Kalma, "A study of stretch in premixed spray flames," *Combust.*

907 Flame **123**, 421 (2000).

908 ³⁵ J.B. Greenberg, "Finite-rate evaporation and droplet drag effects in spherical flame front
 909 propagation through a liquid fuel mist," Combust. Flame Elsevier **148**, 187 (2007).

910 ³⁶ M. Klein, N. Chakraborty, K.W. Jenkins, and R.S. Cant, "Effects of initial radius on the
 911 propagation of premixed flame kernels in a turbulent environment," Phys. Fluids **18**, 1 (2006).

912 ³⁷ N. Chakraborty, M. Klein, and R.S. Cant, "Stretch rate effects on displacement speed in
 913 turbulent premixed flame kernels in the thin reaction zones regime," Proc. Combust. Inst. **31 I**,
 914 1385 (2007).

915 ³⁸ N. Chakraborty and M. Klein, "Effects of global flame curvature on surface density function
 916 transport in turbulent premixed flame kernels in the thin reaction zones regime," Proc.
 917 Combust. Inst. **32 I**, 1435 (2009).

918 ³⁹ N. Chakraborty, J.W. Rogerson, and N. Swaminathan, "The scalar gradient alignment
 919 statistics of flame kernels and its modelling implications for turbulent premixed combustion,"
 920 Flow, Turbul. Combust. **85**, 25 (2010).

921 ⁴⁰ E. Fernández-Tarrazo, A.L. Sánchez, A. Liñán, and F.A. Williams, "A simple one-step
 922 chemistry model for partially premixed hydrocarbon combustion," Combust. Flame **147**, 32
 923 (2006).

924 ⁴¹ S.P. Malkeson and N. Chakraborty, "Statistical analysis of displacement speed in turbulent
 925 stratified flames: A direct numerical simulation study," Combust. Sci. Technol. **182**, 1841
 926 (2010).

927 ⁴² Y. Haruki, A.L. Pillai, T. Kitano, and R. Kurose, "Numerical investigation of flame
 928 propagation in fuel droplet arrays.," At. Sprays **28**, 357 (2018).

929 ⁴³ A. Neophytou, E. Mastorakos, and R.S. Cant, "Complex chemistry simulations of spark
 930 ignition in turbulent sprays," Proc. Combust. Inst. Elsevier Inc. **33**, 2135 (2011).

931 ⁴⁴ N. Chakraborty and R.S. Cant, "Influence of Lewis number on curvature effects in turbulent

932 premixed flame propagation in the thin reaction zones regime,” *Phys. Fluids* **17**, 105105
 933 (2005).

934 ⁴⁵ N. Chakraborty, E.R. Hawkes, J.H. Chen, and R.S. Cant, “The effects of strain rate and
 935 curvature on surface density function transport in turbulent premixed methane–air and
 936 hydrogen–air flames: A comparative study,” *Combust. Flame Elsevier* **154**, 259 (2008).

937 ⁴⁶ N. Peters, P. Terhoeven, J.H. Chen, and T. Echehki, “Statistics of flame displacement speeds
 938 from computations of 2-D unsteady methane-air flames,” *Symp. Combust. Elsevier* **27**, 833
 939 (1998).

940 ⁴⁷ T. Echehki and J.H. Chen, “Analysis of the contribution of curvature to premixed flame
 941 propagation,” *Combust. Flame Elsevier* **118**, 308 (1999).

942 ⁴⁸ N. Chakraborty and M. Klein, “Influence of Lewis number on the surface density function
 943 transport in the thin reaction zone regime for turbulent premixed flames,” *Phys. Fluids* **20**,
 944 65102 (2008).

945 ⁴⁹ N. Chakraborty, H. Kolla, R. Sankaran, E.R. Hawkes, J.H. Chen, and N. Swaminathan,
 946 “Determination of three-dimensional quantities related to scalar dissipation rate and its
 947 transport from two-dimensional measurements: Direct Numerical Simulation based
 948 validation,” *Proc. Combust. Inst. Elsevier* **34**, 1151 (2013).

949 ⁵⁰ J. Lai, M. Klein, and N. Chakraborty, “Direct numerical simulation of head-On quenching
 950 of statistically planar turbulent premixed methane-air flames using a detailed chemical
 951 mechanism,” *Flow, Turbul. Combust. Flow, Turbulence and Combustion* **1** (2018).

952 ⁵¹ H. Watanabe, R. Kurose, S. Komori, and H. Pitsch, “Effects of radiation on spray flame
 953 characteristics and soot formation,” *Combust. Flame* **152**, 2 (2008).

954 ⁵² Y. Baba and R. Kurose, “Analysis and flamelet modelling for spray combustion,” *J. Fluid
 955 Mech.* **612**, 45 (2008).

956 ⁵³ R.W. Bilger, “The structure of turbulent nonpremixed flames,” *Symp. Combust. Elsevier* **22**,

957 475 (1989).

958 ⁵⁴ A.A. Wray, "Minimal Storage Time Advanced Schemes for Spectral Methods," NASA
 959 Ames Res. Cent. California (1990).

960 ⁵⁵ T. Poinso and S.K. Lele, "Boundary conditions for direct simulations of compressible
 961 viscous flows," J. Comput. Phys. **101**, 104 (1992).

962 ⁵⁶ "Rotexo-Softpredict-Cosilab," GmbH and Co. KG Bad Zwischenahn, Germany.

963 ⁵⁷ A. Neophytou and E. Mastorakos, "Simulations of laminar flame propagation in droplet
 964 mists," Combust. Flame **156**, 1627 (2009).

965 ⁵⁸ R.S. Rogallo, "Numerical Experiments in Homogeneous Turbulence," Report California
 966 (1981).

967 ⁵⁹ R.W. Grout, "An age extended progress variable for conditioning reaction rates," Phys.
 968 Fluids **19**, 105107 (2007).

969 ⁶⁰ I. Han and K.Y. Huh, "Roles of displacement speed on evolution of flame surface density
 970 for different turbulent intensities and Lewis numbers in turbulent premixed combustion,"
 971 Combust. Flame **152**, 194 (2008).

972 ⁶¹ H. Reddy and J. Abraham, "Two-dimensional direct numerical simulation evaluation of the
 973 flame-surface density model for flames developing from an ignition kernel in lean methane/air
 974 mixtures under engine conditions," Phys. Fluids **24**, 105108 (2012).

975 ⁶² C. Pera, S. Chevillard, and J. Reveillon, "Effects of residual burnt gas heterogeneity on early
 976 flame propagation and on cyclic variability in spark-ignited engines," Combust. Flame The
 977 Combustion Institute. **160**, 1020 (2013).

978 ⁶³ H.-H. Chiu and T.M. Liu, "Group Combustion of Liquid Droplets," Combust. Sci. Technol.
 979 **17**, 37 (1977).

980 ⁶⁴ H.H. Chiu, H.Y. Kim, and E.J. Croke, "Internal group combustion of liquid droplets," Symp.
 981 Combust. **19**, 971 (1982).

982 ⁶⁵ H. Yamashita, M. Shimada, and T. Takeno, "A numerical study on flame stability at the
983 transition point of jet diffusion flames," Symp. Combust. **26**, 27 (1996).

984 ⁶⁶ N. Chakraborty, M. Klein, and R.S. Cant, "Effects of turbulent reynolds number on the
985 displacement speed statistics in the thin reaction zones regime of turbulent premixed
986 combustion," J. Combust. **2011**, 1 (2011).

987 ⁶⁷ A. Sandeep, F. Proch, A.M. Kempf, and N. Chakraborty, "Statistics of strain rates and
988 surface density function in a flame-resolved high-fidelity simulation of a turbulent premixed
989 bluff body burner," Phys. Fluids **30**, 65101 (2018).

990 ⁶⁸ N. Chakraborty, M. Klein, D. Alwazzan, and H.G. Im, "Surface Density Function statistics
991 in hydrogen- air flames for different turbulent premixed combustion regimes different turbulent
992 premixed combustion regimes," Combust. Sci. Technol. Taylor & Francis 1 (2018).

993 ⁶⁹ R.S. Miller, K. Harstad, and J. Bellan, "Evaluation of equilibrium and non-equilibrium
994 evaporation models for many-droplet gas-liquid flow simulations," Int. J. Multiph. Flow
995 Pergamon **24**, 1025 (1998).

996 ⁷⁰ S.S. Sazhin, T. Kristyadi, W.A. Abdelghaffar, and M.R. Heikal, "Models for fuel droplet
997 heating and evaporation: Comparative analysis," Fuel Elsevier **85**, 1613 (2006).

998 ⁷¹ T. Kitano, J. Nishio, R. Kurose, and S. Komori, "Effects of ambient pressure, gas temperature
999 and combustion reaction on droplet evaporation," Combust. Flame Elsevier **161**, 551 (2014).

1000