On the generalisation of the mixture fraction to a monotonic mixing-describing variable for the flamelet formulation of spray flames

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Spray flames are complex combustion configurations that require the consideration of competing processes between evaporation, mixing and chemical reactions. The classical mixture-fraction formulation, commonly employed for the representation of gaseous diffusion flames, cannot be used for spray flames owing to its non-monotonicity. This is a consequence of the presence of an evaporation source term in the corresponding conservation equation. By addressing this issue, a new mixing-describing variable, called the effective composition variable η, is introduced to enable the general analysis of spray-flame structures in composition space. This quantity combines the gaseous mixture fraction \(Z_g\) and the liquid-to-gas mass ratio \(Z_l\), and is defined as \(d\eta = \sqrt{(dZ_g)^2 + (dZ_l)^2}\). This new expression reduces to the classical mixture-fraction definition for gaseous systems, thereby ensuring consistency. The versatility of this new expression is demonstrated in application to the analysis of counterflow spray flames. Following this analysis, this effective composition variable is employed for the derivation of a spray-flamelet formulation. The consistent representation in both effective composition space and physical space is guaranteed by construction and the feasibility of solving the resulting spray-flamelet equations in this newly defined composition space is demonstrated numerically. A model for the scalar dissipation rate is proposed to close the derived spray-flamelet equations. The laminar one-dimensional counterflow spray-flamelet equations are numerically solved in \(\eta\)-space and compared to the physical-space solutions. It is shown that the hysteresis and bifurcation characterising the flame structure response to variations of droplet diameter and strain rate are correctly reproduced by the proposed composition-space formulation.

Keywords: laminar counterflow spray flame; flamelet formulation; mixture fraction; effective composition; bifurcation

Nomenclature

\[D\] Diffusivity of gas phase
\[L\] Length of the domain in physical space
\[L_v\] Latent heat of vaporisation
\[T\] Gas temperature
\[T_b\] Liquid boiling temperature
\[T_l\] Liquid temperature
\[W\] Molecular weight of the gas mixture
\[W_k\] Molecular weight of species \(k\)
\[Y_k\] Mass fraction of species \(k\)

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1. Introduction
Motivated by the utilisation of liquid fuels for transportation and propulsion systems, considerable progress has been made on the analysis of spray flames [1–6]. While gaseous diffusion flames are characterised by the competition between scalar mixing and chemistry, spray flames require the continuous supply of gaseous fuel via evaporation and transport to the reaction zone to sustain combustion. Because of this complexity, the investigation of spray flames in canonical combustion configurations, such as mixing layers, coflow and...
counterflow flames, represents a viable approach for obtaining physical insight into the behaviour of spray flames [7–11].

Counterflow spray flames have been the subject of intensive research, and considerable numerical and experimental studies have been performed by considering laminar conditions [7,12–17]. Theoretical investigations provided understanding about the underlying physical processes, flame stabilisation and extinction processes of spray flames [11,18–20]. Experiments in counterflow flames have been performed to examine extinction behaviour of mono- and polydisperse spray flames through strain variation and vortex interaction [21,22]. More recently, bistable flame structures of laminar flames were considered for examining the bifurcation in three-dimensional turbulent counterflow spray flames [23]. As such, these studies demonstrated that the structure of spray flames is of fundamental relevance for a wide range of operating regimes.

In the context of laminar gaseous diffusion flames, the flame structure is typically examined in composition space by introducing the gaseous mixture fraction $Z_g$ as an independent variable [24]. For a given strain rate, the flame structure is then fully parameterised in terms of the gaseous mixture composition, providing a unique mapping between physical and composition space. This mixture-fraction formulation is also used in turbulent combustion models, enabling the representation of the turbulence–chemistry interaction through presumed probability density function models [25,26]. Another significant advantage of a mixture-fraction representation is that it enables a computationally more efficient solution in composition space compared to the physical-space solution. Therefore, extending the mixture-fraction concept to spray flames is desirable and enables the utilisation of analysis tools that have been developed for gaseous flames.

Unfortunately, this extension is non-trivial, since the classical gaseous mixture-fraction definition loses its monotonicity owing to evaporation [27,28]. With the exception of pre-vaporised flames and other simplifying assumptions, the structure of spray flames cannot be studied in the classical mixture-fraction space.

Previous works have dealt with the extension of the mixture-fraction definition to spray flames. Sirignano [29] and Bilger [30] have investigated the definition of mixing-describing variables for two-phase combustion. Their works apply to the characterisation of the mixture evolution from the droplet (or ligament) surface to the far field. This approach is only applicable if the diffusive layer around each droplet is small compared to the droplet interspacing. In cases where the droplet interspacing is too small compared to the flame and diffusive scales, a mesoscopic point of view should be adopted and a continuum representation is required with regard to the mixture-fraction field [3]. Although Bilger’s approach is able to recover this mesoscopic limit, the detailed representation of these scales is computationally expensive. In this scenario, extending the mixture-fraction concept to spray flames is not straightforward. This issue was mentioned in [31], and a total mixture fraction was introduced to account for both gas and liquid contributions. Luo, Jianren, and Cen [32] extended the classical mixture-fraction flamelet transformation to spray flames, but only for pre-vaporised conditions that serve the definition of the boundary conditions for the gaseous flamelet equations. Olguin and Gutheil [27,33], Greenberg and Sarig [18], Dvorjetski and Greenberg [19,20], Lerman and Greenberg [11] and Maionchi and Fachini [34] directly solved the spray-flame equations in physical space and subsequently represented the flame structure in $Z_g$-space, for example by separating the purely gaseous region of the flame from the evaporation zone [27,33]. However, due to the non-monotonicity, the classical gaseous definition cannot be used to solve the spray-flamelet equations in composition space.

By addressing these issues, this work proposes a new composition-space variable that enables the description of spray flames. The key idea for this formulation consists in
identifying a monotonic representation of a mixing-describing coordinate for spray flames. This new coordinate, referred to as the effective composition variable $\eta$, is both useful for analysing the flame structure and for solving the corresponding spray-flamelet equations effectively. In addition, the effective composition variable $\eta$ is defined in such a way that it extends the classical flamelet formulation for gaseous diffusion systems [24,35–37], thereby ensuring consistency. Compared to a non-monotonic definition, the use of the proposed effective composition variable for spray flames exhibits the following advantages:

- it allows a mathematical well-posed definition of the transformation from physical to composition space, thereby providing a theoretical foundation for one-dimensional laminar spray-flamelet formulations;
- it enables the representation of the system in composition space, eliminating the explicit dependence on the spatial coordinate, thereby providing a computationally more efficient solution;
- it allows the analysis of spray flames in analogy to the work on gaseous flames based on a mixture-fraction formulation [36,38];
- it provides direct insight into the flame structure without any additional postprocessing that would otherwise be required, for example when using the classical gaseous $Z_g$-space.

The remainder of this paper is organised as follows. The spray-flamelet equations in physical space and composition space are presented in Section 2. The effective composition formulation and its mathematical properties are discussed in Section 3. The versatility of this effective composition space formulation is demonstrated by considering two applications. The first application (Section 4) is concerned with the analysis of the spray-flame structure in composition space. The second application concerns the use of $\eta$ for the direct solution of the spray-flame system in composition space. For this, the spray-flamelet equations in $\eta$-space are formulated in Section 5, and a closure model for the scalar dissipation rate is proposed in Appendix C. Comparisons of simulation results with solutions obtained in physical space are performed and different levels of model approximations are assessed. It is shown that the proposed formulation is able to reproduce bifurcation and hysteresis characterising the flame-structure response to strain-rate and droplet-diameter variations. The paper finishes by offering conclusions and perspectives.

2. Governing equations

In the present work, we consider a mono-disperse spray flame in a counterflow configuration, and the governing equations are formulated in an Eulerian framework. In this configuration, fresh air is injected against a stream consisting of a fuel spray and pure air. Consistent with the classical analysis of gaseous flames, the following assumptions are invoked [24,35,37]:

1. Steady-state solution and low-Mach number limit.
2. Unconfined flame and constant thermodynamic pressure.
4. Unity Lewis number. Equal but not necessarily constant diffusivities are assumed for all chemical species and temperature: $D_k = D_{th} = \lambda/(\rho c_p) \equiv D$. Fick’s law without velocity correction is used for diffusion velocities [37].
5. Calorically perfect gas: $c_{p,k} = c_p = \text{constant}$. 
In this context, it is noted that the composition variable and formulation proposed in this paper are not restricted to these assumptions and can equally be extended in analogy to the theory for gaseous flames; guidance on the extension to non-unity Lewis numbers is provided in Appendix E.

Under these assumptions, the transport equations for the gaseous phase and the liquid phase in physical space are introduced next. From this, we derive the general spray-flamelet formulation, which serves as foundation for the following analysis.

### 2.1. Spray-flame equations in physical space

#### Gas-phase equations

The gaseous phase is described by the transport equations for momentum, species mass fractions, temperature and gaseous mixture fraction $Z_g$:

\[
\rho u_i \frac{\partial u_j}{\partial x_i} = \frac{\partial}{\partial x_i} \left( \mu \frac{\partial u_j}{\partial x_i} \right) - \frac{\partial p}{\partial x_j} + (u_j - u_{l,j}) \dot{m} - f_j,
\]

\[
\rho u_i \frac{\partial Y_k}{\partial x_i} = \frac{\partial}{\partial x_i} \left( \rho D \frac{\partial Y_k}{\partial x_i} \right) + \dot{\omega}_k + \delta_k F - Y_k \dot{m}, \quad \text{for } k = 1, \ldots, N_s,
\]

\[
\rho u_i \frac{\partial T}{\partial x_i} = \frac{\partial}{\partial x_i} \left( \rho D \frac{\partial T}{\partial x_i} \right) + \dot{\omega}_T + \dot{m} \left( T_l - T - \frac{q}{c_p} \right),
\]

\[
\rho u_i \frac{\partial Z_g}{\partial x_i} = \frac{\partial}{\partial x_i} \left( \rho D \frac{\partial Z_g}{\partial x_i} \right) + (1 - Z_g) \dot{m},
\]

where $\rho$ is the density, $p$ is the pressure and $u_i$ is the $j$th component of the velocity vector. The production rate of species $k$ is denoted by $\dot{\omega}_k$; $\dot{\omega}_T = - \sum_{k=1}^{N_s} \dot{\omega}_k W_k h_k / c_p$ is the heat-release rate; $W_k$ is the molecular weight of species $k$; $h_k$ is the sensible and chemical enthalpy of species $k$; $c_p$ is the heat capacity of the gaseous mixture; $q$ is the ratio between the heat transfer and mass transfer rates from the gas to each droplet; $\delta_k$ is the Kronecker delta; and $N_s$ is the total number of species. The total mass vaporisation rate is $\dot{m}$; $T$ is the temperature; $\mu$ is the dynamic viscosity of the gas mixture; and $f_j$ is the $j$th component of the drag force, which is here modelled by Stokes law [39]. Subscript $l$ is used to identify quantities of the liquid phase and the subscript $F$ refers to the fuel. The gaseous non-normalised mixture fraction is here formulated with respect to the carbon-containing species [40]:

\[
Z_g = \frac{W_F}{n_{CF} W_C} \sum_{k=1}^{N_s} n_{C,k} \frac{Y_k W_C}{W_k},
\]

where $Y_k$ is the mass fraction of species $k$, $n_{C,k}$ is the number of carbon atoms in species $k$ and $W_C$ is the carbon molecular weight.

#### Liquid-phase equations

As we are considering spray combustion, the liquid phase is composed of a set of droplets. The following assumptions are made.

- Monodisperse/Monokinetic/Mono-temperature spray: all the droplets in the same vicinity have the same diameter, velocity and temperature.
• Dilute spray: the spray volume fraction is negligible compared to that of the gas phase. Consequently, the gas-phase volume fraction is assumed to be unity in the gas-phase equations.
• The only external force acting on the particle trajectory is the drag force.
• One-way coupling and no droplet/droplet interaction or secondary break-up are considered.

Consequently, the balance equations for the total liquid mass, the individual droplet mass, the liquid momentum, and the enthalpy of the liquid phase \( dh_l = c_l dT_l \) read as [7]:

\[
\frac{\partial (\rho_l \alpha_l u_{l,i})}{\partial x_i} = -\dot{m}, \quad (3a)
\]
\[
\frac{n_l u_{l,i} \partial m_d}{\partial x_i} = -\dot{m}, \quad (3b)
\]
\[
\frac{\partial (\rho_l \alpha_l u_{l,i} u_{l,j})}{\partial x_i} = -f_j - \dot{m} u_{l,j}, \quad (3c)
\]
\[
\frac{\partial (\rho_l \alpha_l u_{l,i} h_l)}{\partial x_i} = -\dot{m} (h_l - q + L_v), \quad (3d)
\]

where \( \alpha_l = n_l \pi d^3 / 6 \) is the liquid volume fraction, \( m_d = \rho_l \pi d^3 / 6 \) is the individual droplet mass, \( \rho_l \) is the liquid density, \( d \) is the droplet diameter, \( n_l \) is the liquid droplet number density, \( c_l \) is the liquid heat capacity, and \( L_v \) is the latent heat of evaporation. By introducing the liquid-to-gas mass ratio:

\[
Z_l = \frac{\alpha_l \rho_l}{(1 - \alpha_l) \rho} \approx \frac{\alpha_l \rho_l}{\rho}, \quad (4)
\]

Equations (3) can be written in non-conservative form:

\[
\rho u_i \frac{\partial Z_l}{\partial x_i} = \frac{\partial [\rho (u_i - u_{l,i}) Z_l]}{\partial x_i} - \dot{m} (1 + Z_l), \quad (5a)
\]
\[
\rho u_i \frac{\partial m_d}{\partial x_i} = -\frac{\rho}{n_l} \dot{m} + \frac{\partial [\rho (u_i - u_{l,i}) m_d]}{\partial x_i}, \quad (5b)
\]
\[
\rho u_i \frac{\partial (u_{l,j} Z_l)}{\partial x_i} = \frac{\partial [\rho u_{l,j} (u_i - u_{l,i}) Z_l]}{\partial x_i} - f_j - \dot{m} u_{l,j} (1 + Z_l), \quad (5c)
\]
\[
\rho u_i \frac{\partial (Z_l h_l)}{\partial x_i} = \frac{\partial [\rho h_l (u_i - u_{l,i}) Z_l]}{\partial x_i} - \dot{m} (1 + Z_l) h_l + \dot{m} (L_v - q). \quad (5d)
\]

In the following, Equations (1) and (5) are used to derive the spray-flamelet equations.

### 2.2. General spray-flamelet formulation

The general spray-flamelet equations can be derived in analogy to the analysis for counterflow gaseous flames [24]. The physical coordinate along the flame-normal direction can be expressed in terms of a generic variable \( \zeta \), which is assumed to increase monotonically from the oxidiser side to the spray injection side. By introducing the transformation from
physical space to composition space, \((x_1, x_2, x_3) \rightarrow (\xi(x_i), \xi_2, \xi_3)\), all spatial derivatives can be written as

\[
\frac{\partial}{\partial x_1} = \frac{\partial \xi}{\partial x_1} \frac{\partial}{\partial \xi},
\]

\[
\frac{\partial}{\partial x_i} = \frac{\partial}{\partial \xi_i} + \frac{\partial \xi}{\partial x_i} \frac{\partial}{\partial \xi} \quad \text{for} \quad i = 2, 3.
\]

It is important to note that the strict monotonicity of the quantity \(\xi\) is essential to guarantee the mathematical well-posedness of the transformation in Equations (6).

Peters assumed [24] that derivatives along the \(\xi\)-direction are much larger compared to those along the tangential directions \((\xi_2, \xi_3)\). By neglecting these high-order contributions, the following expressions are obtained:

\[
\rho u_i \frac{\partial \phi}{\partial x_i} = \Xi_\xi \frac{\partial \phi}{\partial \xi},
\]

\[
\frac{\partial}{\partial x_i} \left( \rho D \frac{\partial \phi}{\partial x_i} \right) = \Xi_\xi \frac{\partial \phi}{\partial \xi} \left[ \frac{\rho D}{2} \frac{\partial}{\partial \xi} \left( \frac{\chi \xi}{2D} \right) + \frac{\chi \xi D}{2} \frac{\partial \rho D}{\partial \xi} \right] + \frac{\rho \chi \xi}{2} \frac{\partial^2 \phi}{\partial \xi^2},
\]

\[
\frac{\partial}{\partial x_i} \left( \mu \frac{\partial \phi}{\partial x_i} \right) = \Xi_\xi \frac{\partial \phi}{\partial \xi} \left[ \frac{\mu}{2} \frac{\partial}{\partial \xi} \left( \frac{\chi \xi}{2D} \right) + \frac{\chi \xi}{2D} \frac{\partial \mu}{\partial \xi} \right] + \frac{\mu \chi \xi}{D} \frac{\partial^2 \phi}{\partial \xi^2},
\]

where

\[
\Xi_\xi = \rho u_i \frac{\partial \xi}{\partial x_i}
\]

is the material derivative of \(\xi\), and \(\chi \xi\) is the scalar dissipation of the variable \(\xi\):

\[
\chi \xi = 2D \frac{\partial \xi}{\partial x_i} \frac{\partial \xi}{\partial x_i}.
\]

With this, the equations for the gas phase, Equations (1), can be rewritten as

\[
\frac{d\xi}{d\zeta} \left( \Xi_\xi - \frac{\mu}{2} \frac{d \xi}{d \zeta} \left( \frac{\chi \xi}{2D} \right) - \frac{\chi \xi}{2D} \frac{d \mu}{d \zeta} \right) = \frac{\mu \chi \xi}{D} \frac{d^2 \xi}{d \zeta^2} + \left( u_j - u_{i,j} \right) \dot{m}_j - f_j + J_j \frac{dp}{d \zeta},
\]

\[
\frac{dY_k}{d\zeta} \left( \Xi_\xi - \frac{\rho D}{2} \frac{d \xi}{d \zeta} \left( \frac{\chi \xi}{2D} \right) - \frac{\chi \xi}{2D} \frac{d(\rho D)}{d \zeta} \right) = \frac{\rho \chi \xi}{2} \frac{d^2 Y_k}{d \zeta^2} + \left( \delta_{kF} - Y_k \right) \dot{m} + \dot{\omega}_k.
\]

\[
\frac{dT}{d\zeta} \left( \Xi_\xi - \frac{\rho D}{2} \frac{d \xi}{d \zeta} \left( \frac{\chi \xi}{2D} \right) - \frac{\chi \xi}{2D} \frac{d(\rho D)}{d \zeta} \right) = \frac{\rho \chi \xi}{2} \frac{d^2 T}{d \zeta^2} + \dot{m} \left( T_l - T - \frac{q}{c_p} \right) + \dot{\omega}_T.
\]

\[
\frac{dZ_g}{d\zeta} \left( \Xi_\xi - \frac{\rho D}{2} \frac{d \xi}{d \zeta} \left( \frac{\chi \xi}{2D} \right) - \frac{\chi \xi}{2D} \frac{d(\rho D)}{d \zeta} \right) = \frac{\rho \chi \xi}{2} \frac{d^2 Z_g}{d \zeta^2} + \left( 1 - Z_g \right) \dot{m},
\]

where \(J_j = -\partial \xi / \partial x_j\). The equations for the liquid phase are
where $\Psi[\phi]$ is defined as the contribution to the slip velocity due to the drag force:

$$

\Psi[\phi] = \frac{\partial \xi}{\partial x_i} \frac{\partial}{\partial \xi} \left[ \rho \phi (u_i - u_{t,i}) \right].

$$

In accordance with Peters’s theory for gaseous flames, the flamelet transformation assumes that the flame structure is locally one-dimensional. The formulation of an appropriate mixing-describing variable $\xi$ is discussed in the following section.

3. Composition-space definition for counterflow spray flames

The spray-flamelet equations, Equations (10) and (11), are derived by invoking two assumptions, namely the presence of a one-dimensional flame structure and the strict monotonicity of $\xi$ with respect to the spatial coordinate. The last constraint is required to guarantee the existence of the derivative and that the solution remains single-valued. Identifying an appropriate definition of $\xi$ that meets this last criterion is the central focus of this paper. Before introducing this variable, we will review previously suggested formulations from the literature.

3.1. Review of previously suggested composition-space formulations

Gaseous mixture fraction

The first candidate is the classical gaseous non-normalised mixture fraction,

$$

\zeta = Z_g.

$$

which is defined in Equation (2) and the corresponding conservation equation is given by Equation (1d). This definition was used previously to parameterise the spray-flamelet equations [27]. As discussed in [27,28,41], the presence of a source term results in a non-conserved quantity for counterflow spray flames. Furthermore, due to competing effects between evaporation and mixing, $Z_g$ becomes non-monotonic, resulting in the multi-valued representation of the flame structure in $Z_g$-space. While this prevents the direct solution of the spray-flamelet equations in composition space, $Z_g$ has been used for the parameterisation of spray flames using two different approaches:

- separating the spray zone and the purely gaseous zone to identify two distinct regions where $Z_g$ is monotonic as done in [8]. However, it will be shown in the subsequent
section that this approach does not always guarantee monotonicity in these phase-separated regions when diffusion and evaporation are not spatially separated; separating the flame structure at the tangent point $d, Z_g = 0$ [27]. Although this method provides a valid representation of the flame structure, the location of this inflection point is not known a priori and can therefore not be used in a straightforward manner as a separation indicator.

**Total mixture fraction**
An alternative to using $Z_g$ as the describing composition variable is to consider also the contribution from the two-phase region in the definition of $\zeta$. A possible definition of such a quantity was first proposed in [31], and further investigated in [28,41], as

$$\zeta = Z_t = Z_g + Z_l.$$  \hfill (14)

This definition can be considered as an extension of Equation (2). In this context it is noted that the consistency of this formulation is guaranteed by the fact that $Z_g \equiv Y_F$ for pure fuel. The conservation equation in physical space is given by [28,41]:

$$\rho u_i \frac{\partial Z_t}{\partial x_i} = \frac{\partial}{\partial x_i} \left( \rho D \frac{\partial Z_g}{\partial x_i} + \rho(u_i - u_{l,i})Z_l \right) - Z_t \dot{m}.  \hfill (15)$$

The evaporation source term in this equation is negative, leading to a decreasing $Z_t$ along the material derivative. Consequently, this term will not affect the monotonicity. However, due to differential diffusion between liquid and gaseous phases and the presence of the slip velocity, the monotonicity of $Z_t$ is not guaranteed. This issue was discussed in [28,41] and demonstrated in [28].

**Conserved mixture fraction**
Another definition of a mixture fraction can be obtained by eliminating the evaporation source term, which is achieved through the following definition:

$$\zeta = Z_c = \frac{Z_g + Z_l}{1 + Z_l},$$  \hfill (16)

and the corresponding conservation equation in physical space:

$$\rho u_i \frac{\partial Z_c}{\partial x_i} = \frac{1}{1 + Z_l} \frac{\partial}{\partial x_i} \left( \rho D \frac{\partial Z_g}{\partial x_i} \right) + \frac{1 - Z_c}{1 + Z_l} \frac{\partial}{\partial x_i} \left[ \rho(u_i - u_{l,i})Z_l \right].  \hfill (17)$$

This definition also suffers from contributions by slip velocity and differential diffusion between the gaseous and liquid phases.

**3.2. Effective composition variable**
A composition variable that is strictly monotonic for counterflow spray flames can be obtained by restricting the two-dimensional space $(Z_g, Z_l)$ to the 1D manifold to which the
solution belongs. By doing so, one can define a composition space variable \( \eta \) as the metric of the 1D manifold, corresponding to its tangent in the \((Z_g, Z_l)\)-space (see Figure 1):

\[
(d\zeta)^2 = (d\eta)^2 = (dZ_g)^2 + (dZ_l)^2.
\]

(18)

from which follows\(^2\)

\[
d\eta = \sqrt{(dZ_g)^2 + (dZ_l)^2}.
\]

(19)

This expression defines the arc-length of the spray-flamelet along the 2D state-space that is defined by \(Z_g\) and \(Z_l\). This must be contrasted with the expressions (13), (14) and (16), which contract the 2D composition space through linear or nonlinear projection.

By combining Equations (19) and (5a) with Equation (1d), the transport equation for \( \eta \) can be written as

\[
\rho u_i \frac{\partial \eta}{\partial x_i} = \text{sgn}(u_\eta) \sqrt{\left( \rho u_i \frac{\partial Z_g}{\partial x_i} \right)^2 + \left( \rho u_i \frac{\partial Z_l}{\partial x_i} \right)^2}
\]

\[
= \text{sgn}(u_\eta) \sqrt{\left( \frac{\partial}{\partial x_i} \left( \rho D \frac{\partial Z_g}{\partial x_i} \right) + (1 - Z_g)\dot{m} \right)^2 + \left( \frac{\partial}{\partial x_i} \left( \rho(u_i - u_{l,i})Z_l \right) - \dot{m}(1 + Z_l) \right)^2},
\]

(20)

where \( u_\eta = u_i \frac{\partial x_i}{\sqrt{\left( \partial x_i \frac{\partial x_i}{\partial x_i} \right)^2} } \) is the gas velocity projected along the gradient of \( \eta \). Note that this definition of \( \eta \) reduces to the classical gaseous mixture-fraction expression in the absence of a liquid phase, guaranteeing consistency with the single-phase flamelet formulation [24]. This follows by imposing the condition

\[
\text{sgn}(u_\eta) = \text{sgn} \left( u_i \frac{\partial x_i}{\sqrt{\left( \partial x_i \frac{\partial x_i}{\partial x_i} \right)^2} } \right) \quad \text{if} \quad Z_l = 0.
\]

(21)

The particular advantage of definition (19) is that it enables a direct solution of the flamelet equations in composition space. Further, with regard to application to tabulation
methods, this definition overcomes the ambiguity that is associated with the construction of different chemistry libraries to represent gaseous and two-phase zones. It has to be noted that the evaporation process contributes twice to the evolution of $\eta$, as it acts both on $Z_g$ and $Z_l$. This double contribution is necessary for cases where the evaporation process does not happen in the mixing layer. This situation occurs for instance if a premixed two-phase flame propagates towards the fuel injection, if the liquid fuel vaporises prior to injection, or if preferential concentration occurs before the mixing layer. In this context it is also noted that $\eta$ contains a source term and is therefore not a conserved scalar. Moreover, $\eta$, as defined in Equation (19), is non-normalised. However, this does not represent an issue for numerical simulations since the resulting flamelet equations are numerically well behaved. In fact, this property is strictly not necessary for the correct identification of the flame-normal direction, which only requires monotonic increase from the oxidiser side to the spray injection side (or vice versa). The maximum value of $\eta$, found for the limiting case with separated mixing and evaporation zones, as provided in Appendix B, could be used to normalise this quantity, if deemed necessary.

4. Analysis of spray-flame structure

This work considers a counterflow configuration, which consists of two opposed injection slots that are separated by a distance $L = 0.02$ m along the $x_1$-direction, see Figure 2. On the fuel side, a mono-disperse kerosene ($C_{10}H_{20}$) spray is injected with air. On the oxidiser side, pure air is injected. Similar to the works by Dvorjetski and Greenberg [19] and Lerman and Greenberg [11], the gaseous flow field is assumed to be described by a constant strain rate: $u_1 = -ax_1$ and $u_2 = ax_2$. Compared to gaseous flames, the boundary conditions are not imposed at infinity in order to take into account the effect of evaporation on the mixing and reaction. The following gaseous boundary conditions are imposed at both sides: $T^0 = 600$ K, $Y_{O_2}^0 = 0.233$, $Y_{N_2}^0 = 0.767$. For the liquid phase at the spray side, the liquid-to-gas mass ratio is $Z_l^0 = 0.2$. In the present study, we examine the effects of the droplet diameter of the injected spray, $d_0$, and the strain rate, $a$, on the flame structure. To focus on the coupling between mass transfer, mixing and reaction processes, approximations of the evaporation model, the liquid velocity and the temperature have been invoked for numerical solutions.
of the spray-flame equations. These assumptions and the resulting system of equations are presented in Appendix A. The reaction chemistry developed in [42] for kerosene/air flames is used in the following.

4.1. Choice of composition-space variable

The solution of the counterflow spray flame at atmospheric pressure for \( d^0 = 40 \, \mu m \) and \( a = 100 \, s^{-1} \) in physical space is shown in Figure 3. The gaseous fuel from the droplet evaporation is consumed in the reaction zone, which is characterised by the high temperature region and high product concentration. As a result of the fuel-rich injection condition, all oxygen that is injected at the fuel side is consumed.

The excess fuel is eventually consumed in the diffusion region, where it reacts with the oxygen that is provided from the oxidiser stream. In the following, the evaporation zone \( (Z_l > 0) \) identifies the spray side of the flame, and the gas side of the flame coincides with the region where \( Z_l = 0 \).

The different definitions for mixture fraction are evaluated and compared in Figure 3(b). This comparison shows that gaseous \( (Z_g) \), total \( (Z_t) \) and conserved \( (Z_c) \) mixture fractions are not monotonic, which is a result of the slip velocity, the evaporation and differential diffusion effects\(^5\) between the liquid and gaseous phases. It is noted that this non-monotonic character is not due to the constant strain rate assumption, and the same effect has been observed for variable strain-rate spray flames in [28].

As shown in Figure 4(a), the spray-flame structure cannot be easily studied in the classical mixture-fraction space. The potential of representing the spray-flame structure in \( Z_g \)-space is assessed by separating the solution into two parts following two distinct approaches: by distinguishing between gas and spray regions [8] or by using the maximum value of \( Z_g \) as a separation threshold [27]. However, as shown in Figure 4(b), representing the flame structure in the \( Z_g \)-space by separating the solution into gas and spray regions is not adequate since the solution is not necessarily unique due to the non-monotonicity of \( Z_g \) in the spray region. The second strategy circumvents this issue (cf. Figure 4(c)), but unfortunately, the \textit{a priori} evaluation of the maximum value of \( Z_g \) is not possible, so that this separation strategy cannot be used in a straightforward manner.

The newly proposed composition variable \( \eta \) addresses both issues, and the flame structure as a function of \( \eta \) is shown in Figure 4(d). Compared to the mixture-fraction parameterisation with respect to \( Z_g \) and \( Z_t \), the solution is guaranteed to have a unique value for any given \( \eta \). Moreover, compared to the two-zone separation, this parameterisation eliminates
the need for a separation criterion. The flame structure on the spray side can be correctly represented when working in physical space or in $\eta$-space.

4.2. Flame structure in effective composition space

The counterflow spray-flame equations (A3) are solved in physical space and the effective composition variable $\eta$ is used to analyze the flame structure for different values of $d^0$ and $a$. The solutions for $Z_g$ and $Z_l$ are compared with results from an asymptotic analysis. The derivation of the analytical solution is provided in Appendix B, and is obtained under the assumption that evaporation and diffusion occur in two distinct regions. The analytic solutions for $Z_g$ and $Z_l$ present piecewise linear behaviors with respect to $\eta$ when the evaporation is completed without interaction with the diffusion process. The gaseous mixture fraction reaches its maximum value $Z_g^* = Z_l^0/(1 + Z_l^0) = 0.166$ at $Z_l = 0$. The spray side is then located at $\eta > Z_g^*$ and is mainly governed by evaporation. In contrast, the gas side ($\eta \leq Z_g^*$) is characterized by diffusion. By construction, $\eta$ coincides with $Z_g$ on the gas side, thereby retaining consistency with the mixture-fraction formulation for purely gaseous flames.

Results for different initial droplet diameters and strain rates are illustrated in Figures 5 and 6, showing the solution in physical space (left) and in effective composition space (middle). The location separating the evaporation and mixing regions is indicated by the vertical line. To assess the significance of the diffusion process at the spray side, a budget
Figure 5. Flame structure obtained from the solution in physical space for \(a = 100 \text{ s}^{-1}\) as a function of different initial droplet diameters \(d^0\): solution in \(x\)-space (left), \(\eta\)-space (middle), and budget analysis (right) of the \(Z_g\)-conservation equation (1d); the grey area corresponds to the diffusion zone; the vertical line separates the spray side from the gas side. For comparison, asymptotic solutions for \(Z_g\) and \(Z_l\) are shown by symbols.

analysis of the \(Z_g\)-transport equation (1d) is performed. In this budget analysis, the contribution of each term appearing in Equation (1d), i.e. advection, diffusion and evaporation, is evaluated. Compared to the work of [33], the contribution of the evaporation to the budget of \(Z_g\) is not split, since both terms in Equation (1d) relate to the sole evaporation process.

These results are presented in the right column of Figures 5 and 6. The comparison of the results with the asymptotic solutions also allows one to quantify the diffusion contribution at the spray side without looking at the budget analysis. Discrepancies between the asymptotic solutions will occur when the diffusion and evaporation zones overlap. Indeed, diffusion contributions in the spray region are apparent in Figures 5 and 6 as deviation from the linear behaviour of \(Z_g\) with respect to \(\eta\) on the spray side. The region where diffusion affects the results is then presented in grey in all figures based on the \(Z_g\)-profiles. The vertical line separates the spray side from the gas side based on the \(Z_l\)-profiles.

4.2.1. Effects of droplet diameter on spray-flame structure

Results for a constant strain rate of \(a = 100 \text{ s}^{-1}\) and three different initial droplet diameters of \(d^0 = \{20, 40, 80\} \mu\text{m}\) are presented in Figure 5. For \(d^0 = 20 \mu\text{m}\) (Figure 5(a)), the liquid
Figure 6. Flame structure obtained from the solution in physical space for $d^0 = 40 \, \mu m$ as a function of different strain rates: solution in $x$-space (left), $\eta$-space (middle), and budget analysis (right) of the $Z_g$-conservation equation (1d); the grey area corresponds to the diffusion zone; the vertical line separates the spray side from the gas side. For comparison, asymptotic solutions for $Z_g$ and $Z_l$ are shown by symbols.

Fuel fully evaporates before reaching the flame reaction zone, and the high temperature region is confined to the gas region of the flame. By considering the budget analysis, it can be seen that the diffusion contribution on the spray side is negligible for small droplet diameters. This is further confirmed by comparisons with the asymptotic solution for the gaseous mixture fraction (shown by symbols), which is in very good agreement with the simulation results.

By increasing the initial droplet diameter to $d^0 = 40 \, \mu m$, shown in Figure 5(b), it can be seen that a small amount of liquid fuel reaches the preheat zone of the flame. The evaporation is not separated anymore from the diffusion region: as shown in the right panel of Figure 5(b), the diffusive part of the budget can no longer be neglected close to the maximum value of $Z_g$. This may also be recognised by comparing the numerical results with the asymptotic profiles. Here, the maximum values for $\eta$ and $Z_g$ are small compared to the analytic solution, demonstrating that the underlying modelling hypothesis of distinct evaporation and mixing zones is inadequate.

For the case with $d^0 = 80 \, \mu m$ (Figure 5(c)), liquid fuel is penetrating into the reaction zone, and a high temperature region and a second heat-release region on the spray side can
be observed. This complex flame structure is clearly visible in the η-space. Moreover, as evidenced by the overlap between the grey region and the liquid volume fraction $Z_l$, as well as by the budget analysis, both diffusive and evaporative contributions are mixed. These interacting processes are not represented by the asymptotic solution, which relies on the spatial separation between both processes.

Considering the η-space, the effect of the droplet diameter on the flame structure is clearly identified. For all three cases considered, the first temperature peak is located on the gas side under stoichiometric conditions. However, with increasing initial droplet diameter, a second temperature peak is formed on the spray side, which identifies the transition from a single-reaction to a double-reaction flame structure for large droplets, as observed in [16,23]. Moreover, by comparing the profiles of $Z_g$ and $Z_l$ with the analytic solution, the diffusive contribution on the spray side can be clearly recognised. By increasing the droplet diameter, diffusion effects become increasingly important in the spray region, and the diffusive processes overlap with evaporation. These effects are not reproduced by the analytic solution that is derived in Appendix B.

4.2.2. Effects of strain rate on spray-flame structure

Results for different strain rates $a = \{200, 400, 600\}$ s$^{-1}$ and a fixed initial droplet diameter of $d_0 = 40$ μm are presented in Figure 6. Compared to the results in physical space for a strain rate of $a = 100$ s$^{-1}$ (Figure 5(b)), the flame structure in Figure 6(a) is confined to a narrow region for $a = 200$ s$^{-1}$. However, the representation of the flame structure with respect to the effective composition variable η provides a clear description of the different regions that are associated with heat release and diffusion. The comparison with the analytic profiles provides an assessment of competing effects between diffusion, advection and evaporation.

The flame structure for a strain rate of $a = 400$ s$^{-1}$ is shown in Figure 6(b). For this condition, a double-flame structure is observed in which the primary heat-release zone is formed on the spray side and the unburned vaporised fuel is consumed in a secondary reaction zone on the gaseous side of the flame. This result is similar to that presented in [28], but has the opposite behaviour compared to the findings of [33], for which a double-flame structure is observed for low strain rates. Since, however, investigations in [33] used methanol or ethanol, for which the latent heat is twice that of kerosene used here and in [28], there is no contradiction between the three studies. The different reaction zones are conveniently identified in composition space, and the budget analysis provides a clear description of the contributions arising from a balance between diffusion and advection in the absence of evaporation effects.

By further increasing the strain rate to a value of $a = 600$ s$^{-1}$ a high-temperature region is observed on the spray side (Figure 6(c)). However, compared to the case with $a = 400$ s$^{-1}$ the two heat-release zones are closer without exhibiting a significant reduction in temperature. At this condition, the flame on the gas side is highly strained, leading to a reduction of the maximum temperature (from 2400 to 2000 K) and both temperature peaks are located on the spray side. In comparison, the maximum temperature on the spray side is less affected by variations in strain rate.

5. Derivation of spray-flamelet equations in effective composition space

One of the main motivations for introducing the monotonic composition-space variable η is to enable the direct solution of Equations (10) and (11) in composition space.
Rewriting Equation (8) by introducing the effective composition-space variable $\eta$ and the transformation operators (7), the term $\Xi_\eta$ (corresponding to the advection term in Equation 20) can be written as

$$\Xi_\eta = \text{sgn}(u_\eta) \sqrt{\left( \rho u_i \frac{\partial Z_g}{\partial x_i} \right)^2 + \left( \rho u_i \frac{\partial Z_l}{\partial x_i} \right)^2} \left( \frac{\rho}{2} \frac{d}{d\eta} \left( \frac{\chi_\eta}{2D} + \frac{\chi_\eta}{2D} \frac{d(\rho D)}{d\eta} \right) + \frac{\rho \chi_\eta}{2} \frac{d^2 Z_g}{d\eta^2} + (1 - Z_g) \dot{m} \right)^2$$

By assuming a constant pressure along the $\eta$-direction, we obtain the complete spray-flamelet equations:

$$\Xi^{\ast}_\eta \frac{d\mu_j}{d\eta} = \frac{\mu_\eta}{D} \frac{d^2 \mu_j}{d\eta^2} + (u_j - u_{i,j}) \dot{m} - f_j,$$

$$\Xi^\dagger_\eta \frac{dY_k}{d\eta} = \frac{\rho \chi_\eta}{2} \frac{d^2 Y_k}{d\eta^2} + (\delta_k - Y_k) \dot{m} + \dot{\omega}_k,$$

$$\Xi^\dagger_\eta \frac{dT}{d\eta} = \frac{\rho \chi_\eta}{2} \frac{d^2 T}{d\eta^2} + \dot{m} \left( T_i - T - \frac{q}{c_p} \right) + \dot{\omega}_T,$$

$$\Xi^\dagger_\eta \frac{dZ_g}{d\eta} = \frac{\rho \chi_\eta}{2} \frac{d^2 Z_g}{d\eta^2} + (1 - Z_g) \dot{m},$$

$$\Xi^\dagger_\eta \frac{dZ_l}{d\eta} = -\dot{m} (1 + Z_i) + \Psi [Z_i],$$

$$\Xi^\dagger_\eta \frac{dm_d}{d\eta} = -\dot{m} \frac{\rho}{n_i} + \Psi [m_d],$$

$$\Xi^\dagger_\eta \frac{d(u_{i,j} Z_l)}{d\eta} = -f_j - \dot{m} u_{i,j} (1 + Z_l) + \Psi [u_{i,j} Z_l],$$

$$\Xi^\dagger_\eta \frac{d(Z_l h_l)}{d\eta} = -\dot{m} h_l (1 + Z_i) + \dot{m} (L_v - q) + \Psi [h_l Z_i],$$

where the following quantities are introduced:

$$\Xi^{\ast}_\eta = \Xi_\eta - \left[ \frac{\mu}{2} \frac{d}{d\eta} \left( \frac{\chi_\eta}{2D} \right) + \frac{\chi_\eta}{2D} \frac{d\mu}{d\eta} \right],$$

$$\Xi^\dagger_\eta = \Xi_\eta - \left[ \rho D \frac{d}{d\eta} \left( \frac{\chi_\eta}{2D} \right) + \frac{\chi_\eta}{2D} \frac{d(\rho D)}{d\eta} \right],$$

$$\Psi [\phi] = \frac{\partial \eta}{\partial x_i} \frac{\partial \eta}{\partial \eta} \left[ \rho \phi (u_i - u_{i,i}) \right],$$

$$\chi_\eta = 2D \left( \frac{\partial \eta}{\partial x_i} \right)^2.$$
Figure 7. Comparison of the spray-flame structure for $d_0 = 40 \mu m$ and $a = 100 s^{-1}$ obtained from the solution in physical space (symbols) and in $\eta$-composition space (solid lines). To facilitate a direct comparison, $\chi_\eta$ is extracted from the $x$-space solution.

To confirm consistency, it can be seen that the spray-flamelet formulation (24) reduces to the classical gaseous mixture-fraction formulation in the absence of a liquid phase. Moreover, its consistency is guaranteed by construction, since no assumption has been applied to rewrite the general equation system, Equations (10) and (11), into the formulation (24), except for $d_\eta p = 0$.

To solve Equations (24) in the effective composition space, closure models are required for the terms $\partial_\nu \eta$ and $(\partial_\nu \eta)^2$ that appear in the expressions for the slip velocity and the scalar dissipation rate (Equations 25c and 25d). Before discussing in Section 5.2 the validity of the closure models developed in Appendices C and D, we will first verify the feasibility of directly solving Equations (24) in composition space through direct comparisons with spray-flame solutions from physical space. For this, the spray-flamelet equations (24) are solved using expressions for $\chi_\eta$ and $\Psi$ that are directly extracted from the physical-space spray-flame solutions. In the following, the assumptions described in Appendix A will be used to simplify the numerical simulations. However, it is noted that the spray-flamelet equations (24) are general and do not rely on such assumptions.

5.1. Feasibility of $\eta$-space simulations

A spray-flamelet formulation has been proposed in $Z_\eta$-composition space in [27]. However, due to the non-monotonicity of $Z_\eta$, the system could not be directly solved in composition space. Instead, the system was solved in physical space and contributions of each term from the solution of the counterflow spray flame were post-processed in the $Z_\eta$-space.

In contrast, the introduction of $\eta$ enables the direct solution of the spray-flamelet equations in composition space. To demonstrate the consistency of the spray-flamelet formulation, one-dimensional counterflow spray flames are solved in $\eta$-space by invoking the assumptions introduced in Appendix A.

A direct comparison of the solutions obtained in physical space using 400 mesh points with adaptive refinement (solid lines) and in composition space with 100 mesh points with equidistant grid spacing (symbols) are shown in Figure 7. The operating conditions correspond to the case discussed in Section 4 ($d_0 = 40 \mu m$ and $a = 100 s^{-1}$). The excellent
agreement between both solutions confirms the validity of the newly proposed spray-flamelet formulation for providing a viable method for the flame-structure representation and as a method for solving the spray-flamelet equations in composition space.

5.2. Closure models for $\chi_\eta$ and $\Psi$

In this section, the performance of the closure model for the scalar dissipation rate $\chi_\eta$ on the simulation results is assessed. Here, we consider the formulation of $\chi_\eta$ developed in Appendix C. This closure model is based on the linearisation of the evaporation model, controlled by the constant vaporisation time $\tau_v$ and the spatial separation of evaporation and diffusion. A model for the slip-velocity term $\Psi$, under consideration of the small Stokes number limit based on the drag Stokes number $St_d = a \tau_d$, is also provided in Appendix D. Since the numerical simulation considers the limiting case of zero slip velocity, the only unclosed term is the scalar dissipation rate $\chi_\eta$. This term is essential not only to characterise the gas side of the flame structure, but also to account for the effects of advection, mixing and evaporation on the liquid spray side. To ensure consistency with the assumptions that were introduced in developing the closure for $\chi_\eta$ in Appendix C, we utilise the linearised evaporation model, which introduces a constant evaporation time $\tau_v$ (see Equation B2 in Appendix B). The solution of the spray-flame equations in physical space is then compared against the solution obtained by solving the spray-flamelet equations in $\eta$-space, for which $\chi_\eta$ is either directly extracted from the solution in physical space or from the analytical expression given by Equation (C5). Two cases are considered here: $\tau_v = 0.005$ s and $\tau_v = 0.02$ s. Comparing the flame structure with results obtained in physical space for the evaporation model of Section 4, these cases are representative for conditions of $d^0 = 20 \mu m$ and $d^0 = 60 \mu m$, respectively.

Comparison of the flame structures for $\tau_v = 0.005$ s is presented in Figure 8. The maximum value of $\eta$ is slightly overestimated when using the analytical expression for $\chi_\eta$, resulting in a small shift of the flame-structure profile in effective composition space. This can be attributed to the fact that evaporation and diffusion overlap in a small region.
However all solutions give comparable results, confirming the validity of the model for small Stokes numbers.

The flame structure for $\tau_v = 0.02$ s is analysed in Figure 9. The flame structure is substantially different from the other case, showing the presence of a double-flame and an overlap of evaporation and diffusion regions. The results in effective composition space are in good agreement with the physical-space solution, but some differences can be seen in the region where evaporation and diffusion overlap. Radicals and intermediate species are expected to be more sensitive to strain rate and, consequently, to be more sensitive to the closure model for $\chi_\eta$. This can be observed by comparing the CO mass fraction in Figures 8(b) and 9(b). For $\tau_v = 0.005$ s, the assumptions underlying the $\chi_\eta$ closure model are verified, leading to good agreement between the physical results and the two composition-space solutions. In contrast, for $\tau_v = 0.02$ s diffusion and evaporation overlap, violating the assumptions that we invoked in the development of the closure for $\chi_\eta$. Indeed, some discrepancies for the CO-profile are noted for the calculation with the analytical closure model, whereas the calculation using $\chi_\eta$ extracted from the $x$-space solution is still in good agreement with the physical-space solution. Nevertheless, the overall agreement remains satisfactory for all simulations.

The same analysis was performed using the $d^2$-evaporation model of Appendix A (Equation A1) and variable density. The results show the same trend discussed for constant $\tau_v$, and this will be examined further in the following section. Although further improvements for the closure model of $\chi_\eta$ are desirable to extend its applicability to larger values of $\tau_v$, results obtained from the $\eta$-space solution are in satisfactory agreement with the $x$-space solutions.

5.3. **Effect of droplet diameter and strain rate: bifurcation and hysteresis**

The effects of droplet diameter and strain rate on the flame structure are examined by solving the spray-flamelet equations (A6) in $\eta$-space using the analytical closure for $\chi_\eta$ and the $d^2$-evaporation model that we introduced in Appendix A. Starting from the solution
for \( d_0 = 10 \mu m \) and \( a = 100 \, \text{s}^{-1} \), the droplet diameter at injection is successively increased until \( d_0 = 80 \mu m \) in increments of 10 \( \mu m \).

Results for \( d_0 = \{20, 40, 80\} \mu m \) are presented in Figure 10. It can be seen that for small droplet diameters a single-reaction structure is observed whereas for larger droplet diameters \((d_0 > 50 \mu m)\) the flame is characterised by a double-reaction structure. Starting from the solution for \( d_0 = 80 \mu m \), the droplet diameter at injection is incrementally decreased until \( d_0 = 10 \mu m \). The double-reaction structure is retained until \( d_0 = 40 \mu m \) with a transition from double- to single-reaction structure occurring at \( d_0 = 30 \mu m \). Hence, for a droplet diameter between \( d_0 = 40 \mu m \) and \( d_0 = 60 \mu m \), depending on the initial condition, two different flame structures are found. This is shown for the case of \( d_0 = 40 \mu m \) in Figure 10(b), obtained when increasing the droplet diameter, and in Figure 10(d), corresponding to the transition from double- to single-reaction structure. The occurrence of this bifurcation was suggested by Continillo and Sirignano [7] and confirmed by Gutheil [16], and is attributed to the increased nonlinearity that is introduced through the evaporation term. Capturing this phenomenon is a confirmation of the suitability of our flamelet formulation for the description of the physics of spray flames.
Figure 11. Counterflow solution in $\eta$-space for: (a) variations in droplet diameter at a fixed strain rate of $a = 100 \, \text{s}^{-1}$; and (b) variations in strain rate for a fixed droplet diameter of $d^0 = 40 \, \mu\text{m}$. The solution from the $\eta$-space formulation is shown by open squares, and the corresponding reference solution in physical space is shown by closed circles. Arrows indicate the direction of the parametric variation.

The behaviour of the flame in response to a variation in the droplet diameter strongly depends on the evaporation model and the reaction chemistry. Vié et al. [23] identified a hysteresis for droplet diameter variations, which was characterised by a double-branch structure. Following this analysis, the mean flamelet temperature is used as a robust metric to distinguish between single- and double-reaction structures:

$$\overline{T} = \frac{1}{\max(\eta)} \int_0^{\max(\eta)} T(\eta) \, d\eta. \quad (26)$$

In the following, the mean flame temperature is normalised by the corresponding value for $d^0 = 10 \, \mu\text{m}$ and $a = 100 \, \text{s}^{-1}$. Results for variations in droplet diameter are shown in Figure 11 to represent the hysteresis loop. Results from the physical space are also included in Figure 11 for comparison. The hysteresis behaviour is captured by both formulations, and slightly higher values for the double-reaction structure are obtained from the solution in physical space.

The effect of the strain rate is investigated further. Starting from the solution for $d^0 = 40 \, \mu\text{m}$ and $a = 100 \, \text{s}^{-1}$ at the lower branch in Figure 11(a), the strain rate is initially increased in increments of $\Delta a = 50 \, \text{s}^{-1}$ until $a = 600 \, \text{s}^{-1}$. Results for $a = \{200, 400, 600\} \, \text{s}^{-1}$ are illustrated in Figure 12. These results reproduce the behaviour of the $x$-space solution from Section 4, with a transition from a single- to a double-reaction structure at $a = 350 \, \text{s}^{-1}$. However, when starting from a double-reaction solution for $a > 350 \, \text{s}^{-1}$ and decreasing the strain rate, the flame retains its double-reaction structure. Moreover, it has been verified that when starting from the double-reaction solution for $d^0 = 40 \, \mu\text{m}$ and $a = 100 \, \text{s}^{-1}$, the double-reaction structure is retrieved both by increasing and by decreasing the droplet diameter. Consequently, a stable branch is identified for which the flame structure is of double-reaction type, whereas the solution stays on the lower single-reaction structure branch of Figure 11(b) as long as the strain rate remains below 350 $\text{s}^{-1}$. This type of bifurcation was also observed in [23], where two branches were identified without the occurrence of hysteresis. It may also be noted that the temperature is overestimated for
Figure 12. Counterflow flame structure in $\eta$-space for $d^0 = 40 \mu m$ and increasing strain rates of: (a) $a = 200$ s$^{-1}$; (b) $a = 400$ s$^{-1}$; and (c) $a = 600$ s$^{-1}$. The solution was obtained in $\eta$-space using the closure for $\chi_\eta$ developed in Appendix C. The grey area corresponds to the diffusion zone; the vertical line separates the spray side from the gas side.

the highest values of the strain rate when solving the system in $\eta$-space. This is due to the fact that the assumptions underlying the closure for $\chi_\eta$ are not valid for high strain rate values, as discussed in Section 4.2. However, the proposed closure for $\chi_\eta$ is a first attempt to model the scalar dissipation rate of spray flames. Despite its shortcomings, the proposed $\eta$-space formulation is able to reproduce the effects of droplet diameter and strain rate on the spray-flame structure. This capability of the spray-flamelet formulation was further demonstrated by showing that it captures the hysteresis process.

6. Conclusions

An effective composition variable $\eta$ was proposed to study the structure of spray flames in composition space in analogy with the classical theory for purely gaseous diffusion flames. Unlike previous attempts [27,32] that have been used to describe a mixture-fraction variable, the newly proposed effective composition variable is monotonic, thereby enabling the solution of spray flames in composition space. Furthermore, since this new definition is also based on the liquid-to-gas mass ratio, it can capture the evolution of the disperse phase even if no evaporation occurs, which is not the case for purely gaseous-based definitions.

This new composition space was used to analyse counterflow spray flames that were simulated in physical space, showing its ability to represent the spray-flame structure. Subsequently, a flamelet formulation was derived and solved, showing the feasibility of directly evaluating the resulting spray-flamelet equations in $\eta$-space. From these flamelet equations arises the necessity of closures for the scalar dissipation rate and the slip velocity. A simplified model was proposed and the potential of the closure for $\chi_\eta$ was verified against solutions in physical space. The complete flamelet formulation was used to investigate the effects of strain rate and droplet diameter on the flame behaviour, reproducing the bifurcation and hysteresis of the flame structure.

The proposed spray-flamelet formulation represents a theoretical tool for the asymptotic analysis of spray flames [11] in composition space. Formulation in an Eulerian form can be extended to polydisperse flow fields, by using for instance a multifluid formulation [43] for the droplet phase. This enables the consideration of the liquid mixture fraction as the sum of all liquid size volume fractions, where the polydispersity only acts on the overall vaporisation rate. Another interesting extension could be to take into account large Stokes number effects such as droplet velocity reversal [6], which can be done by introducing additional droplet classes and adding each droplet class contribution to the
mixture-fraction definition. This work is also a first step towards the development of spray-flamelet based turbulent models, which will require development of subgrid scale models for the composition-space variable \( \eta \) as well as evaporation source terms.

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Notes
1. Since the definition of mixture fraction is reserved for a conserved quantity, \( Z_f \) from Equation (1d) does not strictly represent a mixture fraction. However, for reasons of consistency with previous works, we follow this convention.
2. The sign of \( d\eta \) is chosen to be positive in order to derive a monotonically increasing coordinate from the oxidiser side to the fuel side.
3. Despite the fact that this assumption is not exact for variable-density flows, it reduces the computational complexity of the counterflow while retaining the main physics. This approximation is often used as a simplified model for two-phase flame analysis.
4. For \( L \to \infty \), the pre-evaporated case is retrieved.
5. The liquid phase does not have a diffusion term, and is therefore characterised by an infinite Lewis number.
6. To take into account the variability of the evaporation time, the vaporisation Stokes number is approximated by \( \text{St}_V = \alpha \tau_{v,\text{ref}} (d/d_{\text{ref}})^2 \) where \( \tau_{v,\text{ref}} = 0.04 \) s and \( d_{\text{ref}} = 40 \) \( \mu \)m.
7. It is noted that the flame transition from single- to double-reaction and vice versa is sensitive to the numerical procedure that is used to vary the strain rate and droplet diameter.
8. The assumption of constant liquid temperature is not valid for real applications [3], the transient heating time being of primary importance. However, since the main concern about the definition of a composition space is the effect of the vaporisation rate, this assumption has no consequence for the suitability of our methodology when liquid temperature variations are taken into account.
9. It is worth mentioning that this assumption could be relaxed to take into account density effects on the flow structure, by using the Howarth–Dorodnitzyn approximation under the classical boundary layer approximation [35].

References


Appendix A. One-dimensional counterflow spray-flame equations

A.1. Modelling approach

The counterflow spray-flame equations are solved on the axis of symmetry $x_2 = 0$, from the fuel to the oxidiser side. To focus on the coupling between mass transfer, mixing and reaction, the following simplifying assumptions are invoked for numerical solution of the governing equations.

- A constant strain rate is assumed [19,20]: $u_1 = -ax_1$ and $u_2 = ax_2$.
- For evaporation, a simplified $d^2$-model is considered by fixing the droplet temperature at $T_1 = T_b$, where $T_b$ is the boiling temperature of the fuel species. Consequently, the evaporation model reads as [6]

$$
\dot{m} = 2\pi n_d \rho D \ln \left[ \frac{c_p(T - T_1)}{L_v} \right] \mathcal{H}(T - T_1),
$$

(A1)

$$
q = L_v,
$$

(A2)

where $\mathcal{H}(\cdot)$ is the Heaviside function. The liquid fuel properties for kerosene are $T_b = 478$ K and $L_v = 289.9$ kJ/kg.
The liquid velocity is assumed to be the same as that of the gas velocity. This assumption is valid for small Stokes number droplets based on the gaseous flow strain rate \( St_d = \rho \tau_d \sigma / 18 \mu \) (where \( \tau_d = \rho_i d_p^2 / 18 \mu \) is the particle relaxation time \([39]\)). It has to be noted that such a system cannot capture droplets with a Stokes number greater than \( 1/4 \), which could potentially cross the stagnation and exhibit velocity reversal. Capturing such a behaviour should be handled by using more velocity moments \([44]\) or by introducing additional droplet classes \([6]\).

Constant thermo-diffusive properties \( \rho D = 2 \times 10^{-5} \text{kg/m·s} \) and \( c_p = 1300 \text{J/kg·K} \).

With these assumptions, the system of equations that is solved in physical space takes the following form:

\[
-ax \frac{dY_k}{dx} = \rho D \frac{d^2 Y_k}{dx^2} + \dot{m} (\delta_{kF} - 1) + \dot{\omega}_k, \tag{A3a}
\]

\[
-ax \frac{dT}{dx} = \rho D \frac{d^2 T}{dx^2} + \dot{m} \left( T_l - T - \frac{L_v}{c_p} \right) + \dot{\omega}_T, \tag{A3b}
\]

\[
-ax \frac{dZ_l}{dx} = -\dot{m} (Z_l + 1), \tag{A3c}
\]

\[
-ax \frac{dm_d}{dx} = -\dot{m} \rho / n_l, \tag{A3d}
\]

where the density is calculated from the species mass fractions, the temperature and the constant thermodynamic pressure using the ideal gas law.

In this configuration, the equation for \( \eta \) is

\[
-ax \frac{d\eta}{dx} = \text{sgn}(u_\eta) \sqrt{\left( -ax \frac{dZ_g}{dx} \right)^2 + \left( -ax \frac{dZ_l}{dx} \right)^2}. \tag{A4}
\]

To construct a monotonic composition space, we thus impose

\[
-ax \frac{d\eta}{dx} = \text{sgn}(-ax) \sqrt{\left( -ax \frac{dZ_g}{dx} \right)^2 + \left( -ax \frac{dZ_l}{dx} \right)^2}. \tag{A5}
\]

The corresponding spray-flamelet system in composition space reads as

\[
\Xi^Y_\eta \frac{dY_k}{d\eta} = \frac{\rho \chi_\eta}{2} \frac{d^2 Y_k}{d\eta^2} + (\delta_{kF} - Y_k) \dot{m} + \dot{\omega}_k, \tag{A6a}
\]

\[
\Xi^Y_\eta \frac{dT}{d\eta} = \frac{\rho \chi_\eta}{2} \frac{d^2 T}{d\eta^2} + \dot{m} \left( T_l - T - \frac{L_v}{c_p} \right) + \dot{\omega}_T, \tag{A6b}
\]

\[
\Xi^Z_\eta \frac{dZ_l}{d\eta} = -\dot{m} (1 + Z_l), \tag{A6c}
\]

\[
\Xi^\rho_\eta \frac{dm_d}{d\eta} = -\dot{m} \rho / n_l, \tag{A6d}
\]
with

$$
\Xi_{\eta} = \left[ 1 - 2\mathcal{H} \left( Z_g - \frac{Z_g^*}{2} \right) \right] \left\{ \left[ \frac{dZ_g}{d\eta} \frac{\rho D}{2} \frac{d}{d\eta} \left( \frac{\chi_{\eta}}{2D} \right) + \frac{\rho \chi_{\eta}}{2} \frac{d^2 Z_g}{d\eta^2} + \left( 1 - Z_g \right) \dot{m} \right]^2 \right. \\
+ \left. \left[ \dot{m} \left( 1 + Z_l \right) \right]^2 \right\}^{1/2},
$$

(A7a)

$$
\Xi_{\eta}^\dagger = \Xi_{\eta} - \frac{\rho D}{2} \frac{d}{d\eta} \left( \frac{\chi_{\eta}}{2D} \right),
$$

(A7b)

and \( 1 - 2\mathcal{H} \left( Z_g - \frac{Z_g^*}{2} \right) \) is introduced to model \( \text{sgn}(u_{\eta}) = -\text{sgn}(x) \) as shown in Appendix B. It is noted that \( \Xi_{\eta}^\dagger \) is equal to zero on the gas side.

For the limit of small Stokes numbers, all droplets evaporate before crossing the stagnation plane, which corresponds to the region of negative velocity. The assumption could be violated for larger droplets if their Stokes number \( St_d = \alpha \tau_p \) is higher than 1/4 [6], requiring a closure model that accounts for the slip velocity between the gas and liquid phases. However, it is also noted that even droplets with a high Stokes number could evaporate before reaching the stagnation plane. This is likely to occur for hydrocarbon fuels, for which the latent heat of vaporisation is small compared to those fuels that are commonly used to study droplet crossings [6,27]. Moreover, a closure model accounting for the effects of the slip velocity on the flame structure is proposed in Appendix D, under the assumption of small \( St_d \). For high values of \( St_d \), the transport equation for the liquid velocity (Equation 24f) may also be added to the system. As a result of the zero-slip velocity assumption, i.e. \( u_{\eta} = u_{l,\eta} \), \( \chi_{\eta} \) is the only unclosed term in the spray-flamelet equations (A6). This term is directly evaluated from the x-space solution in Section 5.1. Subsequently, this approximation is relaxed in Sections 5.2 and 5.3 and a model for the scalar dissipation rate is developed in Appendix C.

A.2. Numerical method

To solve Equations (A3) and (A6) in their respective physical and effective composition spaces, the following four numerical ingredients are used.

- An adaptive mesh refinement method is used based on the gradients of \( \eta \) in physical space.
- Diffusive operators, i.e. second-order derivatives, are discretised using a central finite difference scheme. Considering a non-uniform mesh spacing of elements \( \Delta x_i \), the second-order derivative of a quantity \( \Phi \) at the location \( i \) is

$$
\left. \frac{d^2 \Phi}{dx^2} \right|_i \approx \frac{\Delta x_{i-1} \Phi_{i+1} - (\Delta x_{i-1} + \Delta x_i) \Phi_i + \Delta x_i \Phi_{i-1}}{\Delta x_{i-1} + \Delta x_i \Delta x_i / 2},
$$

(A8)

- Convective operators, i.e. first-order derivatives, are discretised using an upwind finite difference scheme:

$$
\left. U \frac{d\Phi}{dx} \right|_i \approx \max(0, U_i) \frac{\Phi_i - \Phi_{i-1}}{\Delta x_{i-1}} + \min(0, U_i) \frac{\Phi_{i+1} - \Phi_i}{\Delta x_i}.
$$

(A9)

- Steady-state is reached through pseudo-time advancement with an explicit Euler scheme. Considering \( \tau \) as the increment of the pseudo-time variable and \( n \) as the time iteration:

$$
\left. \frac{d\Phi}{d\tau} \right|_i \approx \frac{\Phi^\ast_{i+1} - \Phi^\ast_i}{\tau}.
$$

(A10)
Appendix B. Analytical solution for $Z_g$, $Z_l$ and $\eta$

The analytical profiles for the gaseous and liquid-to-gas mass ratio in $\eta$-space are derived here for the 1D laminar counterflow flame described in Appendix A (Equations A3). To obtain a closed-form solution, the following assumptions are introduced.

- Consistent with the modelling of the scalar dissipation rate of gaseous flames [24], a constant density $\rho = \rho^0$ is considered so that $D = D^0$.
- Starting from a $d^2$-evaporation law, for which the evaporation rate is proportional to the droplet diameter (i.e. $d \propto Z_l^{1/3}$):

$$\dot{m} = \rho^0 \frac{\tau_v}{(Z_l^0)^{2/3}} Z_l^{1/3}, \quad (B1)$$

a linearised evaporation model at $Z_l^0$ is derived:

$$\frac{\dot{m}}{a \rho^0} = \frac{1}{3 S_{\text{ev}}} \left(2Z_l^0 + Z_l\right) = \frac{1}{\alpha} \left(2Z_l^0 + Z_l\right), \quad (B2)$$

where $\alpha = 3S_{\text{ev}}$, $S_{\text{ev}} = a \tau_v$ is the evaporation Stokes number, and $\tau_v$ is the constant evaporation time.

The equations for the liquid-to-gas mass ratio and the gaseous mixture fraction, Equations (1d) and (5a), are then given in non-dimensional form as

$$\xi \frac{dZ_l}{d\xi} = \frac{1}{\alpha} \left(2Z_l^0 + Z_l\right) \left(1 + Z_l\right), \quad (B3a)$$

$$\frac{d^2Z_g}{d\xi^2} + 2\xi \frac{dZ_g}{d\xi} = \frac{2}{\alpha} \left(2Z_l^0 + Z_l\right) \left(Z_g - 1\right), \quad (B3b)$$

where $\xi = x/\delta_D$, and $\delta_D = \sqrt{2D^0/\alpha}$ is the diffusion layer thickness. Profiles of mixture-fraction distributions are schematically illustrated in Figure B1.

An analytic solution for $Z_l$ can be obtained by solving Equation (B3a); however, we were not able to find a closed-form solution for Equation (B3b). An analytical solution can be obtained for the asymptotic limit in which the effects of evaporation and species diffusion are spatially separated. For this case, the gaseous mixture fraction increases on the spray side until reaching its maximum; once the evaporation is completed ($Z_l = 0$), diffusion becomes relevant and the spatial evolution of $Z_g$ is described by the purely gaseous mixture-fraction equation (see Figures B1(a) and B1(b)). It is important to recognise that this zonal separation is different from a pre-vaporised spray flame, in...
which liquid fuel is evaporated before diffusion and combustion occur, and combustion is confined to the gaseous side. The present formulation is not restricted to this special case and allows for the spatial superposition of evaporation and combustion. With this, the flame can be separated into the following two regions.

(1) **Spray side for** \( \xi > \xi_v \). The liquid volume fraction starts evaporating close to the injection \( \xi = L/2\delta_D \) and completely disappears at \( \xi = \xi_v \). The main contribution in this region is assumed to arise from the evaporation, so that contributions from diffusion in the \( Z_g \)-equation can be neglected:

\[
\frac{dZ_l}{d\xi} = \frac{1}{\alpha} \left( 2Z_l^0 + Z_l \right) \left( 1 + Z_l \right),
\]

\[
\frac{dZ_g}{d\xi} = \frac{1}{\alpha} \left( 2Z_l^0 + Z_l \right) \left( Z_g - 1 \right).
\]

The analytic solutions for \( Z_l \) and \( Z_g \) can be written as

\[
Z_l(\xi) = -\frac{2Z_l^0}{1 + Z_l^0} \frac{1 - (\xi/\xi_v)^\beta}{Z_l^0 - (\xi/\xi_v)^\beta}, \tag{B5a}
\]

\[
Z_g(\xi) = -\frac{1 - 2Z_l^0}{1 + Z_l^0} \frac{1}{1 - 2Z_l^0(\xi/\xi_v)^{-\beta}} + 1, \tag{B5b}
\]

where \( \beta = (2Z_l^0 - 1)/\alpha \), and the value for \( \xi_v \) is obtained by imposing the boundary condition \( Z_l(L/2\delta_D) = Z_l^0 \) in Equation (B5a):

\[
\xi_v = \frac{L}{2\delta_D} \left[ \frac{3}{2(1 + Z_l^0)} \right]^{1/\beta}. \tag{B6}
\]

The extension of this region depends on the evaporation time \( \tau_v \) through the parameter \( \beta \): increasing the evaporation time leads to a broadening of the evaporation zone, and the limiting case of this model is represented in Figures B1(b) and B1(c).

The maximum value of the gaseous mixture fraction \( Z_g^* \), found at \( \xi = \xi_v \), is calculated using Equations (B5) and (B6):

\[
Z_g^* = \frac{Z_l^0}{(1 + Z_l^0)} \equiv \frac{1}{K_1}. \tag{B7}
\]

From Equations (B5) a relation between \( Z_g \) and \( Z_l \) can be derived for the evaporation region:

\[
Z_l = Z_l^0 - (1 + Z_l^0)Z_g, \quad \frac{dZ_l}{d\xi} = -\frac{K_1}{(K_1 - 1)} \frac{dZ_g}{d\xi}. \tag{B8}
\]

(2) **Gas side for** \( \xi \leq \xi_v \). In this region, the liquid volume fraction is zero and the expression for \( Z_g \) reduces to the classical equation for gaseous flames:

\[
Z_l = 0, \quad \tag{B9a}
\]

\[
Z_g = \frac{Z_g^*}{2} \left( 1 + \text{erf}(\xi) \right), \tag{B9b}
\]
where the last equation is obtained by setting the right-hand-side of Equation (B3b) to zero and using the boundary conditions \( Z_g(-\infty) = 0 \) and \( Z_g(\xi_v) = Z_g^{+\infty} \). It is noted that \( Z_g \) asymptotically reaches the value \( Z_g^{+\infty} \) at \( \xi = \xi_v \), since our model implies \( \xi_v \geq 2\delta_D \). From Equation (B9b), it is found that the stagnation point requires one to evaluate the function \( \text{sgn}(x) = \text{sgn}(\xi) \) corresponding to \( Z_g = \frac{Z_g^{+\infty}}{2} \).

The analytic formulation for \( \eta \) is then obtained by combining Equations (B5) and (B9):

\[
\eta(\xi) = \begin{cases} 
\int_{-\infty}^{\xi} \sqrt{\left(\frac{dZ_g}{d\xi}\right)^2} \, d\xi = \frac{Z_g^{+\infty}}{2} \left(1 + \text{erf}(\xi)\right) & \text{if } \xi \leq \xi_v, \\
Z_g^{+\infty} + \int_{\xi_v}^{\xi} \sqrt{\left(\frac{dZ_g}{d\xi}\right)^2 + \left(\frac{dZ_l}{d\xi}\right)^2} \, d\xi = Z_g^{+\infty} + K_2 Z_l^0 - K_2(1 + Z_l^0)Z_g(\xi) & \text{if } \xi > \xi_v,
\end{cases}
\]

where \( K_2 = \sqrt{(1 - Z_g^{+\infty})^2 + 1} \). Through the spatial separation of the evaporation and diffusion regions of the flame, it can be seen from Equation (B10) that \( \eta \) is only a function of \( Z_l^0 \) and \( \text{St}_v \). The maximum value of the effective mixture fraction is evaluated as

\[
\eta_{\text{max}} = \eta(\xi = L/2\delta_D) = \frac{Z_l^0}{1 + Z_l^0} \left(1 + \sqrt{2 + 2Z_l^0 + (Z_l^0)^2}\right),
\]

which is only a function of the liquid mass fraction at injection.

Invoking the linear dependence of liquid and gaseous mixture fractions on the effective composition variable (see Figure B1(c)), \( Z_g \) and \( Z_l \) can be written as functions of \( \eta \):

\[
Z_g(\eta) = \begin{cases} 
\eta & \text{if } \eta \leq Z_g^*, \\
\frac{\eta - \eta_{\text{max}}}{1 - \eta_{\text{max}}/Z_g^*} & \text{if } \eta > Z_g^*,
\end{cases}
\]

and

\[
Z_l(\eta) = \begin{cases} 
0 & \text{if } \eta \leq Z_g^*, \\
Z_l^0 \left(\frac{\eta - Z_g^*}{\eta_{\text{max}} - Z_g^*}\right) & \text{if } \eta > Z_g^*,
\end{cases}
\]

As discussed in Section 4.2, the validity of the analytical solution relies on the assumption that mixing and evaporation occur in two distinct regions.

Appendix C. Closure model for the scalar dissipation

A closure model for the scalar dissipation rate \( \chi_\eta \) can be derived using the analytic expressions for \( Z_g \) and \( Z_l \) that were derived in the previous section. For this, we decompose Equation (25d) into liquid and gaseous contributions:

\[
\chi_\eta = \chi_Z g + \chi_Z l,
\]

and corresponding expressions directly follow from the definition of the effective composition variable.
The scalar dissipation of the liquid-to-gas mass ratio is evaluated from the analytic solution of $Z_l$ (Equation B5a):

$$\chi_{Z_l} = K_3 (2Z_l^0 + Z_l)^{2+2/\beta} (1 + Z_l)^{2-2/\beta},$$  \hspace{1cm} (C2)

where

$$K_3 = (2Z_l^0)^{-2/\beta} \frac{8D^0}{\alpha^2 L^2} \left[ \frac{2(1 + Z_l^0)}{3} \right]^{2/\beta}.$$

To derive the gaseous scalar dissipation rate $\chi_{Z_g}$, the diffusion and the evaporation contributions are considered separately: $\chi_{Z_g} = \chi_{Z_g}^{\text{evap}} + \chi_{Z_g}^{\text{mix}}$. The scalar dissipation of the gaseous mixture fraction on the gas side in the absence of a liquid volume fraction is given in analogy with a purely gaseous flame (Equation B9b):

$$\chi_{Z_g}^{\text{mix}} = \frac{a(Z_g^*)^2}{\pi} \exp \left\{ -2 \left[ \text{erf}^{-1} \left( \frac{2Z_g}{Z_g^*} - 1 \right) \right]^2 \right\}. \hspace{1cm} (C3)$$

The scalar dissipation of the gaseous mixture fraction on the spray side is obtained from Equation (B5b):

$$\chi_{Z_g}^{\text{evap}} = K_3 (2Z_l^0 + Z_l)^{2+2/\beta} (1 + Z_l)^{-2/\beta} (1 - Z_g)^2.$$

The individual contributions are combined to describe the dissipation rate of the effective composition variable in the gaseous and liquid regions of the flame:

$$\chi_{\eta} = \begin{cases} \chi_{Z_g}^{\text{mix}} & \text{if } \eta \leq Z_g^* \\ \chi_{Z_g}^{\text{evap}} + \chi_{Z_l} & \text{if } \eta > Z_g^* \end{cases} \hspace{1cm} (C5)$$

The analytical closure is compared to the $\chi_{\eta}$-profile from the solution in physical space for the case with $\tau_v = 0.005$ s from Section 5.2. The results from this comparison are shown in Figure C1.
For this case, diffusion and evaporation occur in two distinct regions and the analytical closure model is able to reproduce $\chi_\eta$ in both regions. Extending the closure model to more general cases for which evaporation and diffusion are not separated is feasible for instance by directly evaluating $\chi_\eta$ from the simulation or by combining the contributions from $\chi_{\text{mix}}^{\text{max}}, \chi_{\text{evap}}^{\text{max}}$ and $\chi_{\text{Zl}}$ in the region where diffusion and evaporation occur simultaneously. This zone may be identified by evaluating the nonlinear behaviour of $Z_g$ in $\eta$-space as discussed in Section 4.

Appendix D. Analytic model for slip velocity
The assumptions of Appendix A are here retained to derive a model for the slip velocity contribution $\Psi_d[\phi]$ for a counterflow spray flame. For this, we follow the work of Ferry and Balachandar [45] and evaluate the velocity of the liquid phase from the gaseous velocity:

$$u_{l,i} = u_i - \frac{D u_i}{D t} \tau_d + O\left(\tau_d^2\right), \quad (D1)$$

where $D/Dt$ is the material derivative of the gas phase. Under the assumption of constant strain rate and potential flow solution [11,19], the liquid velocity can be written as

$$u_{l,i} = u_i (1 - \text{St}_d) + O\left(\text{St}_d^2\right). \quad (D2)$$

By considering the limit of small Stokes number, higher-order terms are truncated, and the following expression for the slip-velocity contribution is obtained:

$$\Psi_d[\phi] = \rho u \text{St}_d \frac{\eta \phi}{\eta^2} - \frac{2 \phi \text{St}_d}{3 Z_l 1 - \text{St}_d} \dot{m}. \quad (D3)$$

This closure model can be used to take into account the effect of a slip velocity between liquid and gaseous phases on the flame structure.

The flame structure defined by the 1D spray-flamelet formulation given in Equations (24) depends on the liquid and gaseous velocities through the quantities $\Xi_\eta$ and $\Psi_d$. However, when using Equation (D2), the dependence on the velocity of both phases is eliminated and only the dependence on the droplet Stokes number is retained:

$$\Xi_\eta \frac{dY_k}{d\eta} = \frac{1}{2} \rho \chi_\eta \frac{d^2 Y_k}{d\eta^2} + (\delta_k F - Y_k) \dot{m} + \dot{\omega}_k, \quad (D4a)$$

$$\Xi_\eta \frac{dT}{d\eta} = \frac{1}{2} \rho \chi_\eta \frac{d^2 T}{d\eta^2} + \dot{\omega}_T + \dot{m} \left(T_i - T - \frac{q}{c_p}\right), \quad (D4b)$$

$$\Xi_{\eta}(1 - \text{St}_d) \frac{dZ_l}{d\eta} = -\frac{2\text{St}_d}{3(1 - \text{St}_d)} \dot{m} - \dot{m} (1 + Z_l), \quad (D4c)$$

$$\Xi_{\eta}(1 - \text{St}_d) \frac{dm_d}{d\eta} = -\frac{2\text{St}_d}{3(1 - \text{St}_d)Z_l} \dot{m} \frac{n_l}{\rho} - \dot{m} \frac{\rho}{m_d}. \quad (D4d)$$

In the limit of small Stokes number $\text{St}_d \to 0$, the liquid and gaseous velocities are identical, so that the spray-flamelet formulation simplifies to the system given by Equations (A6).

Appendix E. Non-unity Lewis number flows
In the present work, we invoked the unity Lewis assumption, which is a classical assumption for the development of flamelet methods. However it is well known that hydrocarbon liquid fuels, such as dodecane or kerosene, have a Lewis number above 2. Here we kept the unity Lewis number assumption for the sake of simplicity and clarity, since the focus of the work is on the formulation of
an effective composition space. Nevertheless, extending this formulation to non-unity Lewis-number systems is possible. For this, we recall the equation of the gaseous mixture fraction, but in the case of non-unity Lewis number

$$\rho u_i \frac{\partial Z_g}{\partial x_i} = \frac{\partial}{\partial x_i} \left( \rho D \frac{\partial Z_g}{\partial x_i} \right) + (1 - Z_g) \dot{m},$$

$$+ \frac{W_F}{n_{C,F} W_C} \sum_{k=1}^{N_s} n_{C,k} W_C \frac{\partial}{\partial x_i} \left[ \rho (D_k - D) \frac{\partial Y_k}{\partial x_i} \right],$$

(E1)

where $D$ is a mean diffusion coefficient and $D_k$ is the diffusion coefficient of species $k$. As shown for instance in [28], such a definition of the mixture fraction is not monotonic even for gaseous flames, and thus cannot be used as a proper composition-space variable. However, if we consider the purely gaseous case, and use our composition-space variable $\eta$,

$$\frac{d\eta}{dt} = \text{sgn} \left( \frac{d\eta}{dt} \right) \sqrt{\left( \frac{dZ_g}{dt} \right)^2},$$

(E2)

any variation of $Z_g$ will lead to a monotonic variation of $\eta$ on either fuel or oxidiser sides of the flow. Consequently, our $\eta$-space formulation can handle the non-unity Lewis-number assumption.

Another possible solution is to use the strategy proposed by Pitsch and Peters [46], who introduced a mixture fraction that is not linked to the species in the flow, and is by definition a passive scalar. This way, even if this formulation cannot be linked to physical quantities, it can be used as a composition-space variable.