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| **Authors**           | Federica Ferraro, Sandro Gierth, Steffen Salenbauch, Wang Han and Christian Hasse |
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SOOT PARTICLE SIZE DISTRIBUTION RECONSTRUCTION IN A TURBULENT SOOTING FLAME WITH THE SPLIT-BASED EXTENDED QUADRATURE METHOD OF MOMENTS

FEDERICA FERRARO\textsuperscript{1,*}, SANDRO GIERTH\textsuperscript{1}, STEFFEN SALENBACH, WANG HAN\textsuperscript{2}, CHRISTIAN HASSE\textsuperscript{1}

\textsuperscript{1}Technical University of Darmstadt, Department of Mechanical Engineering, Simulation of reactive Thermo-Fluid Systems, Otto-Berndt-Straße 2, Darmstadt 64287, Germany
\textsuperscript{2}School of Engineering, University of Edinburgh, Edinburgh EH8 3JL, Scotland, UK

ABSTRACT. The Method of Moments (MOM) has largely been applied to investigate sooting laminar and turbulent flames. However, the classical MOM is not able to characterize a continuous particle size distribution (PSD). Without access to information on the PSD, it is difficult to accurately take into account particle oxidation, which is crucial for shrinking and eliminating soot particles. Recently, the Split-based Extended Quadrature Method of Moments (S-EQMOM) has been proposed as a numerically robust alternative to overcome this issue (Salenbauch et al., 2019). The main advantage is that a continuous particle number density function can be reconstructed by superimposing kernel density functions (KDF). Moreover, the S-EQMOM primary nodes are determined individually for each KDF, improving the moment realizability.

In this work, the S-EQMOM is combined with a Large Eddy Simulation/presumed-PDF flamelet/progress variable approach for predicting soot formation in the Delft Adelaide Flame III. The target flame features low/high sooting propensity/intermittency and comprehensive flow/scalar/soot data are available for model validation. Simulation results are compared with the experimental data for both the gas phase and the particulate phase. A good quantitative agreement has been obtained especially in terms of the soot volume fraction. The reconstructed PSD reveals predominantly unimodal/bimodal distributions in the first/downstream portion of this flame, with particle diameters smaller than 100 nm. By investigating the instantaneous and statistical sooting behavior at the flame tip, it has been found that the experimentally observed soot intermittency is linked to mixture fraction fluctuations around its stoichiometric value that exhibit a bimodal probability density function.

1. INTRODUCTION

Soot formation is an undesired phenomenon in the combustion of hydrocarbon fuels. Its harmful effects on climate change and human health call for strict regulations in terms of both the soot volume fraction and the particle size distributions emitted by engines and combustors. Accurately predicting the particle formation processes and particle size distribution (PSD) is therefore fundamental when designing sustainable combustion devices such as gas turbines or internal combustion engines with reduced levels of pollutants. The strong coupling between the turbulent flow field, gas-phase chemical reactions especially involving soot precursors, and the solid phase yields a highly complex physico-chemical problem. The particle evolution is typically obtained by solving the population balance equation (PBE) that describes the spatial and temporal transport of the particle number density function (NDF). Due to the high dimensionality of the PBE, sectional methods (SMs) \cite{1-6} and the Method of Moments (MOM) \cite{7-11} are the most widely used approaches to characterize the particle NDF in turbulent reacting flows. Specifically, sectional methods are based on the discretization of the particle size distribution in several sections representing particles in a defined interval of the particle property space, e.g., particle volume. One or more scalars are transported to represent the particles in one section, yielding a set of transport equations. Therefore, the SM naturally provides local information on the NDF. On the other hand, the method of moments does not solve the NDF directly, but provides an approximation of the NDF solving only transport equations for its low-order moments. This yields a computationally efficient strategy to evaluate the particle population that is suitable to be combined in reacting Large Eddy Simulations (LESs) and Direct Numerical simulations (DNSs). Other
approaches have been recently developed that are able to predict the particle size distribution. Examples are an LES filtered density function approach extended by including the particle property distribution [12, 13] and an hybrid stochastic/fixed-sectional method proposed to solve the population balance equation [14, 15].

In recent years, LESs [3, 5, 9, 13, 16] and DNSs [8, 17–20] of turbulent sooting flames have improved soot prediction, yielding a deeper understanding of the interaction between turbulent reacting flows and soot formation. Reviews on numerical studies can be found in [21–23]. In [8, 19], a DNS of a turbulent non-premixed n-heptane/air flame was performed to analyze the interactions of turbulent mixing, detailed chemical reactions, and soot formation. The Hybrid Method of Moments (HMOM) [24] was applied to evaluate the soot particle population. Soot precursors were observed to be extremely sensitive to the local scalar dissipation rate, which correlates to soot particle formation and growth. In [20] it was found that when the differential diffusion of gas-phase species is neglected, the soot number density is not altered significantly, while the soot mass yield decreases by a factor of two. In [17], non-premixed counterflow flames were calculated and the effect of the turbulence-chemistry interaction on the soot evolution was studied. It was found that turbulence can raise the soot volume fraction by increasing the flame volume, or reduce it by transporting the soot pockets out of the high-temperature regions. Several studies focus on LESs of turbulent flames exploring different soot models, combustion and turbulence-combustion closures, e.g., [3, 5, 9, 13, 16, 25]. However, uncertainties remain, related to the kinetics, particle dynamics, and their interaction with the turbulence [23]. Few works have numerically investigated soot formation in complex configurations such as gas turbines [11, 26–32].

Quadrature-based Methods of Moments (QbMOM) represent an alternative closure for moment transport equations [33–37]. Here, the unknown soot particle NDF is approximated either by a set of Dirac delta functions as in the Quadrature Method of Moments (QMOM) [38] or by kernel density functions (KDFs) as in the Extended Quadrature Method of Moments (EQMOM) [33, 39]. Of the different quadrature-based moment closures, the EQMOM approach [33] has proven particularly suitable for the prediction of soot particle formation, growth, and oxidation [36], since it provides a continuous reconstruction of the NDF. Pointwise information on the NDF is indeed necessary to describe the complete oxidation of the smallest soot particles [36, 40]. However, the EQMOM algorithm presented some numerical issues [41, 42] that make the approach hard to integrate into complex three-dimensional reacting simulations.

Recently, following on from the idea proposed by Megaridis and Dobbins [43] of considering the NDF as a sum of KDFs interacting via the coagulation term, Salenbauch et al. [40] have shown that the numerical difficulties of the EQMOM can be overcome. The so-called Split-based Extended QMOM approach (S-EQMOM) represents an alternative Extended QMOM, where transport equations are only solved for the lower order moments of coupled KDFs (or sub-NDFs as in [40]). While in the EQMOM, an iterative and non-unique procedure [33, 41, 42] is applied to invert the moments, from low and high order moments of the entire NDF, in S-EQMOM one-node EQMOM systems are solved for each sub-NDF yielding a unique solution [40]. Therefore, the S-EQMOM provides a numerically robust algorithm to reconstruct the soot particle NDF. The validity of the method has been demonstrated in [40] by simulating laminar premixed flames with unimodal and bimodal soot particle distributions and a two-stage burner flame configuration [44] under highly soot oxidizing conditions.

Nevertheless, the performance of S-EQMOM in modeling soot formation and PSD in turbulent flames has not been examined. With this background, the objective of this work is threefold: (i) to extend the S-EQMOM approach in the context of LES; (ii) to examine the reconstructed particle size distribution by the S-EQMOM in a turbulent flame for the first time; and (iii) to analyze the dynamics of soot formation, growth and oxidation using the detailed results from the LES.

The feasibility and accuracy of the S-EQMOM in turbulent sooting flames is first proven, then its capability to reconstruct the soot PSD is exploited to gain further insights into the physical processes affecting the soot evolution.

Here, the S-EQMOM is combined with a tabulated flamelet/progress variable (FPV) approach and a presumed Beta-PDF, which provides the turbulence-chemistry interaction closure. The developed numerical framework is applied to simulate a turbulent natural gas/air flame, the Delft-Adelaide Flame III, which is one of the target flames of the ‘International Sooting Flame Workshop’ (ISF) [45, 46]. This flame exhibits low sooting propensity with high intermittency, therefore it represents a challenging benchmark for the soot
model. The gas-phase flow field and soot properties are discussed and compared with the experimental data \cite{45,47}. Subsequently, the PSD is reconstructed and analyzed at different positions along the flame. Finally, the soot dynamics is examined with respect to the mixture fraction field.

The extension of the S-EQMOM approach to the LES and the PSD reconstruction in turbulent flames constitute one of the novelties of this work in the context of soot modeling. Furthermore, by exploiting the detailed information from the LES, special emphasis is given to the investigation of the soot intermittency, which has a strong impact on the soot statistics in this slightly sooting flame. In particular, it is found that the bimodal probability density function (PDF) of the maximum soot volume fraction at the flame tip, observed in the experiments \cite{46}, is linked to a bimodal distribution of the mixture fraction PDF.

2. **Numerical Model**

2.1. **Method of Moments.** The evolution of the soot NDF \( n(t, x; \xi) \) is governed by the population balance equation (PBE)

\[
\frac{\partial n(t, x; \xi)}{\partial t} + \frac{\partial u n(t, x; \xi)}{\partial x} - \frac{\partial}{\partial x} \left( 0.55 \frac{\nu}{T} \frac{\partial T}{\partial x} n(t, x; \xi) \right) - \frac{\partial}{\partial x} \left( \Gamma_m \frac{\partial n(t, x; \xi)}{\partial x} \right) = \dot{m}_{\text{nuc}} + R_{\text{cond}} + R_{\text{sg}} + R_{\text{coag}} + R_{\text{ox}},
\]

with \( u \) the velocity vector, \( \nu \) the kinematic viscosity, \( T \) the temperature, \( \Gamma_m \) the Brownian diffusivity and \( \xi \) the vector of the internal coordinates. The source terms \( R_Y \) with \( Y \in \{ \text{nuc}, \text{cond}, \text{sg}, \text{ox}, \text{coag} \} \) take into account the physical and chemical process of particle nucleation \( R_{\text{nuc}} \), PAH condensation \( R_{\text{cond}} \), surface growth \( R_{\text{sg}} \) by the HACA mechanism, particle coagulation \( R_{\text{coag}} \) and oxidation \( R_{\text{ox}} \). The fourth term on the left-hand side represents the molecular diffusion transport, which is negligible due to the high Schmidt number of soot particles, as shown by \cite{19}, and is therefore not taken into consideration in this study. The vector of the internal coordinates \( \xi \) contains the properties to characterize the soot particles. In this work, soot particles are considered to be spherical and are characterized only by their volume \( V, \xi = |V| \).

Due to the high dimensionality of the PBE, it is not feasible to solve it directly. Here, the Method of Moments (MOM) is used, which only solves the evolution of low-order moments of the NDF. The moment of order \( k \) of the univariate NDF is defined as

\[
m_k = \int_0^\infty V^k n(t, x; V) dV.
\]

Applying Eq. \( \ref{eq:moment} \) to Eq. \( \ref{eq:pbe} \), the transport equation for the \( k \)-th moment is obtained

\[
\frac{\partial m_k}{\partial t} + \frac{\partial u m_k}{\partial x} - \frac{\partial}{\partial x} \left( 0.55 \frac{\nu}{T} \frac{\partial T}{\partial x} m_k \right) = \dot{m}_k.
\]

The source terms \( \dot{m}_k \) contain the corresponding contributions of the physico-chemical processes of particle formation, condensation, surface growth, coagulation and oxidation. Closures are required to determine the moment source terms, which depend on the unknown NDF. Here, the Split-based Extended Quadrature Method of Moments (S-EQMOM) formulated in \cite{40} is employed to achieve the closure.

2.2. **Split-based Extended QMOM.** In the standard QMOM method \cite{48}, the unknown NDF is approximated by a linear combination of \( N_V \) Dirac delta functions

\[
n(V) \approx \sum_{i=1}^{N_V} w_i \delta(V - V_i),
\]

where \( w_i \) and \( V_i \) are the Gaussian quadrature weights and nodes, respectively. The determination of these parameters, the so-called inversion of the moments, is performed using suitable algorithms available in the literature, such as the Wheeler algorithm \cite{49}.

In the EQMOM approach \cite{33,48}, the NDF is approximated by a weighted sum of overlaying kernel density functions \( \delta_\sigma \), instead of delta functions,

\[
n(\hat{V}) \approx \sum_{i=1}^{N_{\text{df}}} w_i \delta_\sigma (\hat{V} - \hat{V}_i),
\]

where the parameter \( \sigma \), representing the scale of the KDFs, additionally needs to be determined. \( \delta_\sigma (\hat{V} - \hat{V}_i) \) are the \( N_{\text{df}} \) continuous KDFs, positioned at \( \hat{V}_i \) and weighted by \( w_i \), and are approximated by kernel density
functions of a known shape, e.g., gamma or lognormal distributions. Gamma distributions are commonly used to approximate the soot particle NDF \cite{36, 40}. A change of variable

$$\hat{V} = V - V_{\text{min}}, \quad \hat{V} \in [0, \infty),$$

(6)
is applied, since the physical soot distribution is defined in the interval \(V \in [V_{\text{min}}, \infty)\), with \(V_{\text{min}}\) denoting the volume of a newly nucleated particle. The nodes \(V_i\), the weights \(w_i\), and the shape parameter \(\sigma\) are determined from \(2N_{\text{df}} + 1\) moments using the moment-inversion algorithm proposed by Yuan et al. \cite{33}.

The S-EQMOM used in this work is a recently developed alternative formulation of the EQMOM approach \cite{40} that, while allowing a continuous PSD reconstruction similarly to the EQMOM, leads to a numerically robust moment inversion procedure. The S-EQMOM indeed avoids the problems related to uniqueness and realizability discussed in \cite{41, 42} by solving for multiple coupled sub-NDFs rather than for the entire NDF and was validated in laminar flames in \cite{40}. This approach allows to take only the lower-order moments \(m_k^i\) of each \(N_i\) sub-NDFs \(n_{s_i}(V)\) into consideration instead of the higher order moment of the entire NDF, \(n(V)\). A generic sub-NDF moment is defined as

$$m_k^i = \int_{V_{\text{min}}}^\infty V^k n_{s_i}(V) dV,$$

(7)

where \(s_i\) is the index of the sub-NDF. The entire NDF may then be approximated as

$$n(\hat{V}) = \sum_{i=1}^{N_i} n_{s_i}(\hat{V}) \approx \sum_{i=1}^{N_i} w_{s_i} \delta_{s_i}(\hat{V}; \hat{V}_{s_i}).$$

(8)

In the S-EQMOM approach, the moment inversion procedure consists in analytically calculating the value of the nodes \(V_{s_i}\), the weights \(w_{s_i}\), and the scale parameters \(\sigma_{s_i}\) directly from the first three moments \([m_{0i}^1, m_{0i}^2, m_{0i}^3]^T\) of the \(N_i\) sub-NDFs. Note that this corresponds to the inversion of a series of one-node EQMOM systems \((N_{\text{df}} = 1)\) using the solution algorithm proposed by Yuan et al. \cite{33}. The main advantage of the S-EQMOM over the EQMOM is that the inversion procedure yields a system of equations that is solved analytically and has a unique solution \cite{40}, while in the case of the EQMOM, an iterative and non-unique procedure \cite{33, 41, 42} is applied to invert the moments from low- and high-order moments of the entire NDF. It should be noted that although the S-EQMOM and the EQMOM share similarities, the moment inversion strategy in the two approaches is different, as described above, so the positions and weights determined in the S-EQMOM do not necessarily coincide with the EQMOM values \cite{40}. The S-EQMOM formulation greatly improves the stability of the inversion algorithm, allowing a computationally efficient and robust local reconstruction of the soot particle NDF \cite{40}.

In order to capture both unimodal and bimodal particle distributions which arise from the interplay of particle nucleation and coagulation \cite{50}, a special treatment of these two source terms was formulated and validated in \cite{40}. The nucleation source term is included only in the first sub-NDF \(n_{s_1}\), which tracks small, freshly formed particles. As discussed in \cite{51}, the first particle mode consists of so-called nanoparticles, i.e. freshly nucleated particles with a diameter of less than 10 nm. The other sub-NDFs are therefore not affected by nucleation and can evolve to larger particle sizes. The coagulation source term takes into account collisions between particles not only from the same sub-NDF but also from different sub-NDFs, proving a coupling term between the sub-NDFs \cite{40}. Further, condensation, chemical surface growth (HACA), and oxidation are processes active on all sub-NDFs. The detailed formulation of the source terms is described in our previous work \cite{40} and is not repeated here for brevity.

The soot kinetics employed in this study follows our previous works \cite{34, 40} and it is here only shortly described. The particle nucleation is modeled by a dimerization reaction of two PAH molecules \cite{52}. A lumped PAH species, \(Y_{PAH}\), is defined as the sum of pyrene \(A_4\) and cyclopentapyrene \(A_4R_5\). Condensation is described as the collision of PAH molecules with soot particles \cite{52}, while the soot surface growth follows the H-abstraction-C\(_2\)H\(_2\)-addition (HACA) mechanism from Frenklach and Wang \cite{53, 54}. The rates of the HACA mechanism are taken from Appel et al. \cite{55}. The coagulation process accounts for the transition between the continuum and the free molecular regime by applying a harmonic mean interpolation of the collision kernels of the two individual regimes \cite{56}. Finally, particle oxidation is assumed to occur through reactions with \(O_2\) and \(OH\) \cite{54} with the rate parameters taken from Appel et al. (2000) \cite{55}.
2.3. Combustion Model. In the present study, the flamelet/progress variable (FPV) approach \([57, 58]\) is employed, in which flamelet solutions are generated by solving the steady-state flamelet equations \([59]\) with different stoichiometric scalar dissipation rates ranging from thermochemical equilibrium to quenching conditions.

The chemical reactions are described by the kinetic mechanism presented in \([60, 61]\). The thermochemical state is hence parametrized by the mixture fraction \(Z\) and the scalar dissipation rate \(\chi_s\), which is mapped to the progress variable \(\tilde{Y}_C\). Here, \(\tilde{Y}_C\) is defined as \(\tilde{Y}_C = Y_{\text{CO}_2} + Y_{\text{CO}} + Y_{\text{H}_2\text{O}} + Y_{\text{H}_2}\). Soot radiation effects are neglected in this study due to the very low soot volume fraction of the flame \([16]\).

2.4. Coupled LES-FPV approach. The flow field is described by the Favre-filtered Navier-Stokes equations. The turbulence closure is achieved using the eddy viscosity hypothesis with the \(\sigma\) model \([62]\). The model constant is determined using the dynamic procedure \([63]\). The suitability of the LES-FPV numerical framework for turbulent jet flames was previously shown in \([64-67]\). The Favre-filtered transport equations of the mixture fraction and progress variable are given by

\[
\frac{\partial}{\partial t} \left( \bar{\rho} \tilde{Z} \right) + \frac{\partial}{\partial x} \left( \bar{\rho} \tilde{u} \tilde{Z} \right) = \frac{\partial}{\partial x} \left[ \tilde{\rho} \left( D_Z + \frac{\nu_l}{Sc_i} \right) \frac{\partial \tilde{Z}}{\partial x} \right],
\]

\[
\frac{\partial}{\partial t} \left( \bar{\rho} \tilde{Y}_C \right) + \frac{\partial}{\partial x} \left( \bar{\rho} \tilde{u} \tilde{Y}_C \right) = \frac{\partial}{\partial x} \left[ \tilde{\rho} \left( D_{Y_C} + \frac{\nu_l}{Sc_i} \right) \frac{\partial \tilde{Y}_C}{\partial x} \right] + \tilde{\omega}_Y.
\]

Here, \(D_Z\) and \(D_{Y_C}\) denote the diffusion coefficient of the mixture fraction and progress variable, respectively, which are evaluated under the assumption of a unity Lewis number as \(D_Z = D_{Y_C} = D = \alpha\), with the thermal diffusivity \(\alpha\) retrieved from the flamelet table. Furthermore, \(\nu_l\) represents the turbulent viscosity, \(Sc_i\) is the turbulent Schmidt number set equal to 0.7 \([68]\) and \(\tilde{\omega}_Y\) is the filtered source term of the progress variable, which is also obtained from the flamelet table. The variance of the mixture fraction \(\tilde{Z}^2\) is calculated using an algebraic equation following \([57]\).

The thermochemical state is retrieved using a normalized progress variable \(\tilde{C}\) \([69]\), defined as

\[
\tilde{C} = \frac{\tilde{Y}_C - \tilde{Y}_{C_{\text{min}}} \left( \tilde{Z} \right)}{\tilde{Y}_{C_{\text{max}}} \left( \tilde{Z} \right) - \tilde{Y}_{C_{\text{min}}} \left( \tilde{Z} \right)}.
\]

Here, the minimum and maximum progress variable values are functions of the mixture fraction and are determined from the tabulated flamelet solutions. To take into account non-resolved fluctuations, a \(\beta\)-shaped filtered density function (FDF) is assumed for the mixture fraction, whereas a \(\delta\)-FDF is applied for the progress variable. Hence, the Favre-filtered thermochemical state \(\tilde{\phi}\) is parametrized as \(\tilde{\phi}(\tilde{Z}, \tilde{Z}^2, \tilde{C})\).

To model the slow PAH chemistry and the mass transfer from the gas to the solid phase due to soot particle formation, a transport equation for the Favre-filtered PAH mass fraction is solved following \([9]\),

\[
\frac{\partial \bar{\rho} \tilde{Y}_{\text{PAH}}}{\partial t} + \frac{\partial \bar{\rho} \tilde{u} \tilde{Y}_{\text{PAH}}}{\partial x} = \frac{\partial}{\partial x} \left[ \tilde{\rho} \left( D + \frac{\nu_l}{Sc_i} \right) \frac{\partial \tilde{Y}_{\text{PAH}}}{\partial x} \right] + \tilde{\omega}_{\text{PAH}}.
\]

Here \(\tilde{Y}_{\text{PAH}}\) is the sum of the PAH soot precursors considered (pyrene \(A_4\) and cyclopentapyrene \(A_4R_3\)). Following \([9, 70]\) the filtered source term \(\tilde{\omega}_{\text{PAH}}\) is decomposed into three components: a chemical production term \(\tilde{\omega}_{\text{PAH}}^\chi\), that is independent of the PAH species concentration, a chemical consumption term \(\tilde{\omega}_{\text{PAH}}^\gamma\), that is a linear function of the species concentration, and a consumption term representing the mass transfer rate from the gas phase to soot \(\tilde{\omega}_k\), which is quadratic with the species concentration,

\[
\tilde{\omega}_{\text{PAH}} = \tilde{\omega}_{\text{PAH}}^\chi + \tilde{\omega}_{\text{PAH}}^\gamma + \tilde{\omega}_k.
\]

The filtered source term is then decomposed as

\[
\tilde{\omega}_{\text{PAH}} = \tilde{\omega}_{\text{PAH}}^\chi + \tilde{\omega}_{\text{PAH}}^\gamma \left( \frac{\tilde{Y}_{\text{PAH}}}{\tilde{Y}_{\text{PAH}}^\chi} \right) + \tilde{\omega}_k \left( \frac{\tilde{Y}_{\text{PAH}}}{\tilde{Y}_{\text{PAH}}^\gamma} \right)^2,
\]

where the superscript \(T\) indicates the value obtained from the flamelet table. It is important to note that the rate \(\tilde{\omega}_k\) can also be determined in the laminar flamelet calculation since it is only dependent on the gas-phase soot precursor concentration, therefore it is obtained here along with the thermochemical state.
3. APPLICATION TO A TURBULENT FLAME

3.1. Experimental configuration. The experimental setup under investigation is the Delft-Adelaide Flame III, a benchmark case of the International Sooting Flame workshop [71]. The burner was first described in [45] and consists of a central main fuel jet with a diameter of 6 mm. This is surrounded by a rim containing 12 pilot flames stabilizing the main flame, a primary air annular coflow with inner and outer diameters equal to 15 mm and 45 mm, respectively, and a secondary coflow. The fuel jet has a bulk velocity of 21.94 m/s and the coflow streams have a velocity of 4.4 m/s and 0.3 m/s, respectively. The fuel used in the experiments was commercial natural gas. Its composition varied slightly between the experiments performed on this burner in different laboratories. However, the adiabatic flame temperature variations were small with respect to the variation in fuel composition [72]. In this study, the natural gas composition used for the soot particle measurements [46] was chosen. Laser-Doppler anemometry (LDA) data are available for the flow field [47], while Raman-Rayleigh-LIF measurements provide the temperature, major species mass fractions, and mixture fraction [72]. Soot measurements were performed by [46] using planar laser-induced incandescence (LII).

3.2. Numerical setup. The numerical simulations are performed using the software OpenFOAM (v2006) [73] to solve the filtered Navier-Stokes, reactive scalar, and moment transport equations. In-house libraries are used for the combustion and turbulence-combustion closures [74] [75] and for the S-EQMOM closure [40]. In the S-EQMOM, the NDF is described using \( N_{s} = 2 \) sub-NDFs, which is a suitable number to approximate typical bimodal soot particle distributions, as shown by Salenbauch et al. [40]. This yields six additional transport equations for solving the first three moments of each sub-NDF.

The computational domain is a cylinder that spans 150 \( D \) in the axial direction and 33.3 \( D \) in the radial direction, with \( D \) being the main fuel jet diameter. The domain is discretized by a block-structured mesh with approximately 12.8 million hexahedral cells, with 1000, 184 and 68 cells in the axial, radial and circumferential directions, respectively. In the axial and circumferential directions, a uniform mesh is employed; in the radial direction, the mesh is refined close to the main jet, pilot and inner coflow and stretched towards the lateral direction. For the generation of the turbulent inflow boundary conditions at the main jet and primary coflow, two LESs of the upstream geometry were performed separately. A convergent pipe was calculated for the main jet and a divergent annulus pipe for the primary coflow, according to the burner geometry [72]. A bulk velocity was applied at the secondary coflow. Following Mueller and Pitsch [9] and Ayache and Mastorakos [76], the pilot nozzles are modeled as an annulus ring with a bulk velocity boundary condition, which preserves the experimental mass flow rate.

An implicit second-order temporal discretization scheme is employed with a maximum Courant number of 0.03 and a second-order central difference scheme is used for the convective flux in the momentum equations. A TVD scheme using the Sweby limiter [77] is applied for the convective scalar fluxes.

The simulation results discussed in Section 4 were obtained by collecting temporal statistics over 0.15 s and by averaging in the circumferential direction. The simulations were computed on 576 cores on six Intel Xeon Platinum 9242 nodes with a total computational cost of about 0.5 million CPU hours.

4. RESULTS

In this section, the simulation results of the gas phase are first compared to the experimental data from [47] [72]. Velocity and reacting scalar data are only available in the upstream region, where no soot is formed. The soot-related quantities compared with LII data from [46] are then presented and discussed, providing a validation of the S-EQMOM approach in LES of turbulent flames. The PSD, which is not available from the experiments, is hence reconstructed and analyzed at different positions in the flame in order to investigate its spatial evolution. Finally, the soot dynamics and its relation with the gas-phase dynamics are examined to gain a further insight into the high soot intermittency of this flame.

4.1. Gas-phase statistics. The time-averaged radial distributions of the axial velocity, mixture fraction, and temperature and their root mean square (rms) are shown in Fig. 1 at three axial locations: \( x/D = 16.66, 25, \) and 41.66. The simulation results show very good agreement with the experimental data at all locations, both in terms of mean and rms values, indicating that the flow field, scalar mixing and subsequent combustion are predicted accurately. A slight overestimation is observed for time-averaged temperature
profiles. Using only steady flamelet solutions in the LES/FPV approach leads to an under-prediction of the local extinction in the first portion of the flame. More advanced LES/FPV models, which include gas-phase radiation [78] or assume an extended presumed PDF for the reactive scalars [70], have shown improved prediction in flames with a comparable amount of local extinction, e.g., the Sandia Flame E. However, the over-prediction of the temperature is found to have a minor effect on the soot formation process downstream in the flame, as discussed below.

Figure 2 shows the time-averaged radial profiles of the CO, CH$_4$, CO$_2$, H$_2$O and H$_2$ mass fraction. Similarly, the LES results indicate that the experimental species data are estimated well at all locations. This further shows that the turbulence and turbulent combustion models applied are able to correctly capture the main features of the flame structure in the gas phase.

**Figure 2.** Time-averaged radial profiles of the CO, CH$_4$, CO$_2$, H$_2$O and H$_2$ mass fraction. The LES results indicate that the experimental species data are estimated well at all locations.
4.2. Soot-phase statistics. Instantaneous contours of the temperature, PAH mass fraction, soot volume fraction, and particle number density are shown in Fig. 3. The superimposed white line represents the stoichiometric mixture fraction isoline ($Z = Z_{st} = 0.073$). It can be observed that the soot precursor mass fraction $Y_{PAH}$ is produced on the fuel-rich side of the flame. A significant amount of soot is formed downstream in the flame, for $x/D > 50$. Due to turbulent fluctuations, fuel-rich pockets sporadically detach from the main flame zone, connected to the fuel-rich core, at around $x/D = 80–100$, and are transported further downstream where they are oxidized. This flame region is characterized by high soot intermittency, as observed in the experiments in [46] and previous numerical studies [9, 16]. This phenomenon will be discussed in more detail in Section 4.4.

Time-averaged contours of the same quantities are shown in Fig. 4. The number density contour indicates the presence of a significant number of particles, $\theta(10^{11} \text{ 1/cm}^3)$, between $x/D = 30$ and 120, while the soot volume fraction is of the order of particles per billion (ppb) between $x/D = 50$ and 125, which is also beyond the time-averaged stoichiometric mixture fraction isoline.

The time-averaged volume fraction source terms are plotted in Fig. 5. Note that only nucleation and oxidation rates are plotted on the same color scale, while condensation and HACA rates are plotted on different color scales due to their different orders of magnitude. It can be observed that nucleation and condensation are the predominant processes on the fuel-rich side of the flame, although the maximum condensation rate is smaller than the nucleation rate by a factor of 5. The HACA process is instead one order of magnitude smaller than the condensation, as also numerically observed in [9]. It takes place in regions with a locally rich mixture, reaching a maximum in the region between $x/D = 80$ and 100, close to the flame tip, and also, with minor intensity, around the stoichiometric isoline. The oxidation rate is of the same order of magnitude as the nucleation and takes place around the stoichiometric isoline and downstream in the lean flame region, where most of the soot particles are oxidized.

Figure 2. Radial distributions of time-averaged CO, CH$_4$, CO$_2$, H$_2$O and H$_2$ mass fractions at three axial locations above the burner. Solid line: LES; symbols: experimental data.
Figure 3. Instantaneous contours of temperature $T$, PAH mass fraction $Y_{\text{PAH}}$, soot volume fraction $f_V$ and particle number density $N$. The white line represents the stoichiometric mixture fraction isoline $Z = Z_{st} = 0.073$. 
Figure 4. Time-averaged contours of temperature $T$, PAH mass fraction $Y_{PAH}$, soot volume fraction $f_V$ and particle number density $N$. The white line represents the stoichiometric mixture fraction isoline $Z = Z_{st} = 0.073$. 
Figure 5. Time-averaged contours of volume fraction source terms (ppm/s). The white line represents the stoichiometric mixture fraction isoline $Z = Z_{st} = 0.073$. Note that different color scales are used.
A quantitative comparison is shown in Fig. 6a, where the evolution of the time-averaged experimental and numerical soot volume fraction along the flame centerline is plotted. It can be observed that the maximum value is slightly overpredicted and is located at $x/D = 97$, upstream compared to the maximum position from the experimental data at $x/D \approx 115$. The complete oxidation of soot particles is also predicted upstream, at $x/D = 125$, against $x/D = 140$ in the experiments. However, compared to previously published results for the same flame using different modeling approaches [13, 25, 79], the present results show a significant improvement in the prediction of the peak value and location of the experimental soot volume fraction. A similar level of agreement with the results reported in [9, 16] is instead observed. As pointed out by Mueller and Pitsch [9], the discrepancy in the location of soot onset between simulation results and experiments may be due to the significant uncertainty in PAH chemistry, which becomes particularly relevant in methane/air flames with low sooting tendency.

Furthermore, minor dependency of the particle formation is observed concerning the temperature history along the flame. Comparing the LES/FPV results presented here with those obtained from an LES sparse multi mapping conditioning [80] or from an LES transported PDF [16], which can capture significant amount of local extinction [81, 82] (see also Sec. 4.1), an earlier onset of soot particle formation is similarly predicted.

Figure 6b shows the time-averaged soot volume fraction source terms on the centerline. The curves clearly indicate that nucleation and condensation are the predominant processes contributing to the soot particle formation and growth in this flame. Further, the oxidation source term plays an important role in the reduction of the soot volume fraction, being of the same order of magnitude as the nucleation process between $x/D = 60$ and 100. Downstream, for $x/D > 100$, oxidation is the primary process occurring on soot particles, which is consistent with the qualitative observation discussed above.

Figure 7 shows the evolution of the time-averaged soot volume fraction and the mixture fraction along the radial coordinate (top), the time-averaged soot volume fraction source terms (center), and the time-averaged number density source terms (bottom). The profiles in Fig. 7a indicate that the soot volume fraction increases between $x/D = 65$ and 85, while it is almost zero at $x/D = 125$, where the particles are almost completely oxidized. At $x/D = 85$, close to the position of the maximum soot volume fraction on the centerline (see Fig. 6a), the profile reaches a maximum at $r/D = 2$, where nucleation, condensation and HACA are concurrent processes, and the nucleation is close to its maximum value, as shown in Fig. 7b. The coagulation source term in Fig. 7c also reaches at $x/D = 85$ its maximum value. At $x/D = 105$, the oxidation rate is close to its maximum value and is the dominant process at all radial locations.

Furthermore, in all the plots in Fig. 7a, the stoichiometric mixture fraction is indicated by the horizontal dashed line. It is shown that at all axial locations, time-averaged soot volume fraction is present at radial locations where the time-averaged mixture fraction is below the stoichiometric value.

In summary, the results presented here are in good agreement with the experimental data and comparable with the state-of-art soot simulations for this turbulent flame [9, 16] for the time-averaged values. The good
Figure 7. (a): Radial profiles of the time-averaged soot volume fraction and mixture fraction at four axial locations. The horizontal dashed line represents the stoichiometric mixture fraction \( Z = Z_{st} = 0.073 \). (b): Radial profiles of the time-averaged soot volume fraction source terms at four axial locations. The HACA source term is multiplied by 10. (c): Radial profiles of the time-averaged particle number source terms for nucleation and coagulation at four axial locations.

The soot quantities are now investigated in mixture fraction space at different positions along the flame. Figure 8 shows the conditional scatter plots of the soot volume fraction and PAH mass fraction versus the mixture fraction. The samples represent instantaneous values collected over several instants in time, colored according to their temperature. The conditional means are also shown by the black solid line. Furthermore, the dashed black and blue lines indicate the stoichiometric mixture fraction \( Z = Z_{st} = 0.073 \) and \( Z = 2Z_{st} = 0.146 \), respectively. The soot volume fraction and PAH mass fraction are seen to exhibit a similar dependency on the mixture fraction. High values of soot volume fraction and PAH mass fraction are obtained at mixture fraction values between \( Z_{st} \) and \( 2Z_{st} \) and high temperatures. Conditional soot volume fraction prediction of the experimental soot quantities is a prerequisite of the soot model to provide further insights into the undergoing physico-chemical processes. Therefore, in the following, the detailed simulation results are used to better understand the formation and evolution of the soot particles along the flame and their relationship with the gas-phase mixture.
fraction and $Y_{\text{PAH}}$ profiles similarly reach a maximum in this mixture fraction range at all the axial locations examined. Most of the samples are located on the rich mixture fraction side; the PAH mass fraction rapidly decreases on the lean mixture fraction side, similarly to the soot volume fraction. From $x/D = 65$ to $x/D = 105$, samples with a higher soot volume fraction are detected, but they span a smaller range on the rich mixture fraction side, similarly to the PAH mass fraction. At $x/D = 125$, downstream of the location of maximum oxidation, only few samples are detected with high volume fraction values and temperatures at mixture fractions close to stoichiometry.

Furthermore, although the time-averaged contours and profiles in Figs. 4 and 7a indicated the presence of soot in regions with a lean time-averaged mixture fraction, the conditional scatter plots show that instantaneous samples with a significant soot volume fraction are found only for rich mixture fraction.

To further analyze the correlation between the mixture fraction and soot volume fraction, the conditional PDFs of the soot volume fraction on the mixture fraction, $P(f_V|Z)$, are investigated. $P(f_V|Z)$, calculated based on the same samples used in Fig. 8, is shown in Fig. 9. Three mixture fraction intervals are examined: lean, with $Z < Z_{st}$, slightly rich, with $Z_{st} < Z < 2Z_{st}$, and highly rich, with $Z > 2Z_{st}$. The plots indicate that on the lean mixture fraction side (blue line), the conditional soot volume fraction PDF has a peak at $f_V = 0$ ppb and is only not zero for values of $f_V < 1$ ppb along the whole length of the flame. This confirms that there is only a small number of sooting samples at lean condition and these disappear rapidly due to oxidation. For slightly rich mixtures, the conditional PDF has a peak at $f_V = 0$ ppb followed by a rapid decay towards zero. For highly rich mixture fraction values, the conditional PDF has a peak for $f_V > 0$ ppb and becomes broader close to $x/D = 85$. Moreover, at $x/D = 105$ the conditional PDF shows a bimodal shape, with 3 ppb and 12 ppb being the peak positions, respectively. This suggests the presence of samples containing lower/higher levels of $f_V$ that may correspond to smaller/larger particles. Indeed, it is only at very rich conditions and high temperature that small particles can undergo significant surface growth due to the concurrent condensation and HACA processes. At $x/D = 125$, only the volume-fraction-conditional PDF at the slightly rich mixture fraction ($Z_{st} < Z < 2Z_{st}$) is other than zero, and samples with a very
small soot volume fraction, $f_V < 2$ ppb, are detected at this position, while the volume fraction has been completely oxidized on the lean mixture fraction side.

In conclusion, the results presented above illustrate that soot particle formation and growth occur only on the rich mixture fraction side, while soot is rapidly oxidized near the stoichiometric mixture fraction isoline. Therefore, the soot volume fraction present in the time-averaged field beyond the stoichiometric mixture fraction isoline may be due to the high soot intermittency observed experimentally in this portion of the flame. This will be analyzed in detail in Section 4.4 while the next section discusses the reconstructed PSD obtained using the S-EQMOM.

**Figure 9.** Time-averaged soot volume fraction PDF conditioned on the mixture fraction at different axial locations along the flame.

4.3. Mean Particle Size Distribution. As stated above, the S-EQMOM method offers the significant advantage of providing information for the temporal and spatial evolution of the reconstructed particle size distribution. In this section, the time-averaged reconstructed soot PSD is investigated at different locations in the flame.

The time-averaged PSD along the flame centerline is plotted in Fig. 10. Upstream of $x/D = 25$, the PSD is not detected, which is consistent with the mean average soot volume fraction discussed in Section 4.2 (see Fig. 6a). Near $x/D = 40$ (light blue curves), the PSD appears unimodal and indicates the presence of particles smaller than 10 nm. Between $x/D = 40$ and $x/D = 70$, the number density of particles with a diameter larger than 10 nm increases, while the number density of small particles remains almost constant, revealing strong particle nucleation and surface growth in this portion of the flame. Between $x/D = 75$ and $x/D = 90$, the shape of the PSD changes from a unimodal to a bimodal distribution, with a further increase in the number density of large particles. For $x/D > 100$, the number density drops dramatically over all particle sizes due to the oxidation process, which is dominant in the lean mixture.

The time-averaged PSD evolution along the radial coordinate is shown at three axial positions $x/D = 65, 85$ and 105 in Fig. 11. The results indicate a dominant unimodal distribution at $x/D = 65$. At $x/D = 85$, the PSD presents a bimodal shape between $r/D = 2$ and 6, while the soot particles are oxidized in the case of larger radii. At this location, near the peak of the soot volume fraction, all soot processes are active and the strong coagulation observed in Fig. 7c contributes to the transition of the PSD from unimodal to bimodal. Downstream, at $x/D = 105$, the PSD presents a bimodal shape close to the centerline. Its evolution outwards in the radial direction illustrates a rapid decay of the particle density at very small diameters and a shift in the PSD towards smaller particle diameters. Furthermore, in terms of both axial and radial PSD evolution, the particle diameter remains significantly below 100 nm.

Of the previous studies investigating this flame, only two analyzed the PSD [13, 80]. In [13], a joint scalar-discrete number density PDF is solved using Eulerian stochastic fields, and unimodal distributions are predicted along the centerline. In contrast, in [80], where a sparse multiple mapping condition method with a sectional method is used, a shift to a bimodal distribution is predicted near $x/D = 50$, slightly upstream compared to the results presented in this study. The soot particle size was predicted to be smaller than 100 nm, similarly to the S-EQMOM results illustrated in this study.

Since no experimental data are available for the PSD, the general agreement with the work by Huo et al. [80] is a promising result for future applications of the S-EQMOM. Further, it is noted that the computational cost of the S-EQMOM is lower than that in [13, 80].
FIGURE 10. Time-averaged PSD of soot particles at different positions along the centerline.

FIGURE 11. Time-averaged PSD of soot particles along the radius at axial position $x/D = 65$ (top left), $x/D = 85$ (top right) and $x/D = 105$ (bottom).
4.4. Dynamics of soot formation and oxidation. As mentioned above, this flame is characterized by high soot intermittency. In the experimental work by Qamar et al. [46], the intermittency was defined as the probability of not finding any soot (or below the measurement threshold of 0.1 ppb) at a given location and at a given time. The results indicated high soot intermittency for $x/D > 80$, with a peak at $x/D = 110$ (see Fig. 14 in [46]).

In order to further understand this phenomenon and its correlation with the underlying thermo-chemical state of the mixture, the time-averaged mixture fraction and soot volume fraction are first plotted along the centerline in Fig. 12. The soot volume fraction profile from Fig. 6a is repeated here for convenience. The profiles show that soot particles do exist downstream of the axial position $x/D = 102$, where the time-averaged mixture fraction reaches its stoichiometric value (indicated by the dashed line), but they are rapidly oxidized between $x/D = 102$ and 125. This area with a lean time-averaged mixture fraction is highlighted in gray in the bottom plot of Fig. 12. The analysis in the following is therefore devoted to gaining further insights into the soot quantities and their instantaneous and statistical behavior related to the mixture fraction field.

For a qualitative assessment of the soot dynamics downstream of $x/D = 102$, instantaneous snapshots of the soot volume fraction, taken at 5.25 ms time intervals, are shown in Fig. 13. It is observed that pockets of rich mixture, enclosing soot particles, intermittently detach from the main jet at various axial positions for $x/D > 80$. These are transported downstream, where they mix with the air coflow streams, leading to the shrinking of the rich pockets and the complete oxidation of the soot particles. This phenomenon repeats intermittently throughout the simulation time and seems to control the presence of the volume fraction downstream of the time-averaged stoichiometric isoline.
Figure 13. Time evolution of the soot volume fraction. The snapshots are taken with a time interval $\Delta t = 5.25$ ms. The white line represents the stoichiometric mixture fraction isoline $Z = Z_{st} = 0.073$. The dotted horizontal lines denote the axial positions $x/D = 102$ and 125, respectively, and mark the gray area from Fig. [12].

To further analyze this finding, mixture fraction and soot volume fraction probes have been collected over 70 ms at $x/D = 105$ on the flame centerline (within the gray area of Fig. [12]). Their evolution over time is plotted in Fig. [14], where the time instants with a mixture fraction higher than its stoichiometric value ($Z_{st} = 0.073$) are highlighted in gray. The plots indicate strong oscillations of the mixture fraction around its stoichiometric value, mainly correlating with the presence of a soot volume fraction above the experimental threshold. No soot volume fraction is detected outside the highlighted areas, i.e. for mixture fraction below the stoichiometric value.

Extending the sample points in the neighborhood of the axial position $x/D = 105$, within 3 mm radius, makes it possible to calculate the marginal PDF of the mixture fraction, $P(Z)$, PAH mass fraction, $P(Y_{PAH})$ and soot volume fraction, $P(f_V)$, shown in Fig. [15]. Here, all three PDFs are seen to have a bimodal shape. In particular, the mixture fraction PDF has a first peak at a very lean mixture fraction value and a second, lower peak near stoichiometry. This leads to a bimodal shape for $P(Y_{PAH})$ and for $P(f_V)$. This latter result is in agreement with the experimental results presented by Qamar et al. (Fig. 8 in [46]), where the PDF of the maximum measured soot volume fraction along the centerline similarly exhibits a bimodal distribution at the flame tip.
The above analysis explains that the experimentally observed bimodal soot volume fraction PDF, namely the probability of finding non-sooting/sooting samples, which can be also represented as the soot intermittency, corresponds to a bimodal probability of finding samples of lean/rich mixture fraction at that specific location in the flame.

**Figure 14.** Instantaneous values for the mixture fraction (top) and soot volume fraction (bottom) over time at an axial position \( x/D = 105 \). The horizontal dashed and dotted lines represent the stoichiometric mixture fraction \( Z = Z_{st} = 0.073 \) and the time-averaged mixture fraction \( \langle Z \rangle = 0.061 \), respectively. The areas highlighted in gray correspond to rich mixture fraction samples.
Figure 15. Marginal PDF of the mixture fraction, PAH mass fraction and soot volume fraction in the neighborhood of the axial position $x/D = 105$. The vertical dashed line in the plot of the mixture fraction PDF represents the stoichiometric mixture fraction $Z = Z_{st} = 0.073$.

Figure 16. Soot volume fraction evolution between $t_4$ (left) and $t_6$ (right) from Fig. 13 and instantaneous PSD at two arbitrarily selected points. The white line represents the stoichiometric mixture fraction isoline $Z = Z_{st} = 0.073$. The dotted horizontal lines denote the axial positions $x/D = 102$ and 125, respectively.

Finally, the information on the spatial and temporal evolution of the reconstructed PSD obtained by the S-EQMOM can be further exploited to analyze the instantaneous PSD at a given location in the flame. As
an example, two points located in a detached rich mixture zone undergoing soot oxidation are arbitrarily
selected at two instants in time from Fig. 13, $t_{4}$ and $t_{6}$. The positions of these points and the corresponding
PSDs are shown in Fig. 16. It is observed that the particle number density decreases at all particle sizes be-
tween the two instants in time, due to the turbulent mixing with the lean mixture and the consequent particle
oxidation. This example illustrates the importance of the detailed information on the spatial and temporal
PSD evolution provided by the S-EQMOM approach as a means of correctly predicting the dynamics of
soot particle formation, growth, coagulation and oxidation.

5. CONCLUSIONS

In this study, the Split-based EQMOM soot model was integrated into an LES solver and combined with
a flamelet/progress variable (FPV) combustion model and a presumed beta-PDF for the mixture fraction.
The main advantage of the S-EQMOM is that it provides local and continuous information on the PSD
that is not accessible in standard MOM, with a feasible computational effort. The LES/FPV/S-EQMOM
was applied to simulate a benchmark turbulent sooting flame, the Delft Adelaide III flame. The gas phase
and soot phase were compared with the experimental data available. Further, a detailed analysis of the
reconstructed PSD was performed at different positions along the flame.

Numerical results for the gas phase revealed very good agreement with the experimental data available in
the lower portion of the flame, indicating that the flow field, the mixture and the flame structure are correctly
predicted by the simulation. The soot volume fraction was predicted with good agreement, comparable to
the works of Mueller and Pitsch [9] and Han et al. [16], and with a significant improvement compared to
other state-of-the-art approaches applied to simulate this flame.

Predictions of the PSD along the centerline and at different radial locations indicated that there was a
dominant unimodal distribution of the particle number density in the first portion of the flame. A transition
from unimodal to bimodal was observed between $x/D = 75$ and 90, close to the position where the soot
volume fraction reaches its maximum value. Nevertheless, small particles with a diameter lower than 100
nm are mainly formed in this flame.

The detailed information from the LES was further applied to gain insights into the soot intermittency
observed in the experiments. Both the temporal evolution and the statistical behavior of the soot volume
fraction with respect to the mixture fraction were investigated. It was found that in this slightly sooting
flame, the soot intermittency observed in the experiments is mainly correlated with oscillations of the
mixture fraction around its stoichiometric value close to the flame tip. Both marginal PDFs of the mixture
fraction and soot volume fraction indeed exhibit a bimodal shape at this location.

Finally, this work demonstrates the predictive capability of the S-EQMOM in terms of both the volume
fraction and the PSD of soot in turbulent flames. The S-EQMOM therefore appears to be a promising ap-
proach to characterize the sooting features of sustainable combustion systems that have to meet limitations
regarding mass and particle size distribution.

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reason-
able request.

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