



A Numerical Study on Soot Formation and Evolution in Pressurized Turbulent Sooting Flames

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Understandings on soot formation and evolution in pressurized flames are of significant interest due to the increasing operating pressures in different combustors and the accompanying increased soot emissions. In this study, a series of pressurized turbulent sooting flames at 1 bar, 3 bar and 5 bar, are simulated to study the pressure effect on the soot formation and evolution. The inflow conditions are chosen such that the Reynolds number at different pressures keep constant. Via a Radiation Flamelet Progress Variable (RFPV) approach with a conditional soot sub-filter Probability Density Function (PDF) to consider the turbulence-chemistry-soot interaction, quantitatively good agreements (e.g., maximum discrepancy within one order of magnitude) are achieved for soot volume fraction predictions compared with the experimental data at different pressures. Soot volume fraction source terms are then discussed to show the pressure effect on nucleation, condensation, surface growth and oxidation at different axial positions in these flames.

I. Introduction

To achieve higher efficiency, combustion is nowadays pushed to extremely high pressure limits, such as the supercritical combustion in sCO₂ combustors and high-pressure combustion in pressure gain engines. As such, it is fundamentally needed to understand the pressure effect on combustion. There exists a few computational works investigating the pressure effect on turbulent non-premixed flames. Tabet et al. [1] studied turbulent non-premixed hydrogen-air flame structure in pressure range of 1-10 atm, in which, the pressure is found to increase the gas density and thus change the flame structure. Other studies such as Hong et al. [2] studied the pressure effect on a non-premixed turbulent flame, reporting a more focused flame with high pressures. Under high pressures conditions, one major concern is the dramatically increased soot emissions, as experimentally observed in many pressurized laminar and turbulent sooting flames [3-6]. Soot is the major pollutants in the air, as the form of particulate matter. In addition, soot is also the major agent responsible for climate change. It is hence vital to understand the mechanisms of soot formation and evolution in high pressure combustion.

To understand the pressure effect on soot formation and evolution, many numerical studies on pressurized laminar flames have been conducted to show that the change of polycyclic aromatic hydrocarbons (PAH) concentration, scalar dissipation rate, and the different chemical pathways are the major issues for the increasing soot at higher pressures [7, 8]. However, there lack studies on the soot formation and evolution in pressurized turbulent flames due to two reasons. First of all, it is well recognized that developing predictive models for turbulent sooting flames is very challenging due to the complicated interactions among turbulence, detailed chemistry and soot. Secondly, measurements of key parameters such as temperature and major species in turbulent flames is significant for the comparison and validation of computational models. Although there are a variety of measurement of soot quantities in atmospheric turbulent sooting flames, the measurement of these parameters are not easy for pressurized flames, due to the high soot volume fraction under elevated pressures and thus the interfered combustion zone. The successful measurement in a swirl gas turbine model combustor (DLR) with elevated pressures, to the best knowledge of the authors, is the only available data

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for pressurized turbulent sooting flame data [6]. The computational study in Chong *et al.* [9] studied the pressure and dilution-jet effect on soot formation in this aircraft swirl combustor. However, it is seen that in this numerical study the soot prediction has over one order of magnitude error when comparing with experimental data at high pressures. It is reported in this study that the discrepancy is due to the chemical/physical models uncertainty, which however, is not discussed in detail. To obtain more fundamental understanding on the pressure effect on soot formation and evolution in pressurized turbulent sooting flames, a simpler and well-understood configuration without too complex geometry and flow (e.g., turbulent jet flames) is needed.

Recently, Boyette *et al.* [10] established a high-pressure turbulent sooting flame configuration (KEN flames) by diluting the fuel with N₂, allowing a non-sooting region for temperature and major species measurements. In this study, we for the first time simulated the KEN turbulent jet sooting flames with the pressure ranges from 1 -5 bar. A recently proposed conditional soot sub-filter probability density function (PDF) [11] is used in this study to account for the turbulence-soot-chemistry interaction. Under different pressures, prediction accuracy within one order of magnitude is achieved. Pressure effect on soot formation and evolution is then analyzed and discussed in this study.

II. Methodology

A. Turbulent combustion modeling

In this study, the Radiation Flamelet/Progress Variable (RFPV) [12, 13] is used to decouple the detailed flame structure from the computation of flow field. The non-premixed flame structure *a priori* computed in the mixture fraction space, forming a series of steady non-premixed flamelet solutions with different radiation heat losses and mixture fraction dissipation rates. Radiation is modeled with an optically thin gray gas approach, where the absorption coefficients for CO₂, H₂O, CH₄, and CO are obtained from Barlow *et al.* [14]. The KM2 chemical reaction model [15] is used in this study when computing the non-premixed flamelet solutions.

B. Soot modeling

A bivariate parameterization (volume and surface area) of the Number Density Function (NDF) is employed to model detailed aggregation of soot. To avoid the computationally expensive solution of every soot number density function and yet capture the global quantities of soot formation and evolution, the method of moments (MOM) is used. In MOM, the joint moments of the NDF are given by

$$M_{x,y} = \sum_{i=0}^{\infty} V_i^x S_i^y N_i, \quad (1)$$

where summation over i implies summation over the entire two-dimensional state space and N_i is then the number density of particles of volume V_i and surface area S_i . The subscripts x and y denote the order of the moment in volume and surface area, respectively. In this study, four moments are transported while their source terms are closed with the Hybrid Method of Moments (HMOM) [16].

C. Transport equations

In this study, large eddy simulations (LES) are used, which apply filtering operations to the conservation equations for momentum and scalars. More specifically, the filtered transport equations are given by

$$\frac{\partial \bar{\rho} \tilde{Z}}{\partial t} + \frac{\partial \bar{\rho} \tilde{u}_i \tilde{Z}}{\partial x_i} = \frac{\partial}{\partial x_i} \left(\bar{\rho} \tilde{u}_i \tilde{Z} - \bar{\rho} \widetilde{u_i Z} \right) + \frac{\partial}{\partial x_i} \left(\bar{\rho} \tilde{D}_Z \frac{\partial \tilde{Z}}{\partial x_i} \right) + \bar{m}_Z, \quad (2)$$

$$\frac{\partial \bar{\rho} \tilde{C}}{\partial t} + \frac{\partial \bar{\rho} \tilde{u}_i \tilde{C}}{\partial x_i} = \frac{\partial}{\partial x_i} \left(\bar{\rho} \tilde{u}_i \tilde{C} - \bar{\rho} \widetilde{u_i C} \right) + \frac{\partial}{\partial x_i} \left(\bar{\rho} \tilde{D}_C \frac{\partial \tilde{C}}{\partial x_i} \right) + \bar{m}_C, \quad (3)$$

$$\frac{\partial \bar{\rho} \tilde{H}}{\partial t} + \frac{\partial \bar{\rho} \tilde{u}_i \tilde{H}}{\partial x_i} = \frac{\partial}{\partial x_i} \left(\bar{\rho} \tilde{u}_i \tilde{H} - \bar{\rho} \widetilde{u_i H} \right) + \frac{\partial}{\partial x_i} \left(\bar{\rho} \tilde{D}_H \frac{\partial \tilde{H}}{\partial x_i} \right) + \bar{\rho} \tilde{H} + \bar{q}_{\text{RAD}}, \quad (4)$$

$$\frac{\partial \bar{\rho} \tilde{Y}_{\text{PAH}}}{\partial t} + \frac{\partial \bar{\rho} \tilde{u}_i \tilde{Y}_{\text{PAH}}}{\partial x_i} = \frac{\partial}{\partial x_i} \left(\bar{\rho} \tilde{u}_i \tilde{Y}_{\text{PAH}} - \bar{\rho} \widetilde{u_i Y_{\text{PAH}}} \right) + \frac{\partial}{\partial x_i} \left(\bar{\rho} \tilde{D}_{\text{PAH}} \frac{\partial \tilde{Y}_{\text{PAH}}}{\partial x_i} \right) + \bar{m}_{\text{PAH}},$$

$$\frac{\partial \bar{M}_j}{\partial t} + \frac{\partial \bar{u}_i^* \bar{M}_j}{\partial x_i} = \frac{\partial}{\partial x_i} \left(\bar{u}_i^* \bar{M}_j - \bar{u}_i^* \bar{M}_j \right) + \bar{M}_j, \quad (5)$$

where the overline indicates the filtering operation, and the tilde implies a density-weighted filtering. For each equation, the first term on the right-hand side is known as the subfilter scalar flux and contains the effects of unresolved turbulent transport; the second term is the filtered molecular diffusion fluxes (except for the soot moment equations). The models to close those terms are well-established in the literature and thus not discussed here in detail. The last term in these equations is the filtered source term, including the filtered reaction source term and moments source term, which are discussed in the following section.

D. Turbulence-chemistry-soot interaction

To model the subfilter soot-turbulence-chemistry interactions, the filtered source term $\bar{Q}$ is usually closed through convolution with a density-weighted joint subfilter PDF $\tilde{P}(\xi_i, M_j)$, which is a function of the vector of thermochemical variables ξ_i and the vector of soot variables M_j .

$$\bar{Q}(\tilde{\xi}_i, \bar{M}_j) = \iint \dot{Q}(\xi_i, M_j) \tilde{P}(\xi_i, M_j) d\xi_i dM_j. \quad (6)$$

The joint subfilter PDF $\tilde{P}(\xi_i, M_j)$ is unknown and must be modeled using transported PDF or presumed PDF approaches. The presumed PDF approach will be adopted in this work. Following the recently proposed method in [11]. The soot-chemistry-turbulence interaction is considered by a conditional soot sub-filter PDF, which only activates the sooting mode at fuel-rich mixture fractions when oxidation is slower than surface growth.

E. Experimental and numerical configuration

KEN flames are turbulent piloted jet flames. The fuel jet is composed of ethylene diluted with 65% (by volume) nitrogen. The fuel central inner inlet diameter is $D = 3.4$ mm. The flame is stabilized by a pilot providing 18% of the heat release of the jet. The piloted flames are pre-mixtures of $\text{C}_2\text{H}_4/\text{air}$ with equivalence ratio 0.9. The surrounding co-flow is air and the co-flow diameter is 19.05 mm. The jet velocity (U_j), air co-flow velocity (U_c) and Reynolds number (Re) of the simulated cases are listed in Table 1.

Table 1 Operating conditions of the simulated cases, following the experimental details in [10].

Case	Pressure (bar)	Re	U_j (m/s)	U_c (m/s)
1	1	10000	36.6	0.6
2	3	10000	12.2	0.2
3	5	10000	7.32	0.12

To simulate KEN flames with LES, a cylindrical computational domain with $300D$ in the axial direction is discretized into $192 \times 96 \times 32$ in the axial, radial and azimuthal direction, respectively. The grid points are concentrated near the

mixing and combustion regions by stretching the grid in the downstream direction axially and keeping uniform spaced grid in the circumferential direction, with 0.1 mm as the minimum grid size. To account for the turbulent inlet velocity profile, a separate periodic pipe flow simulation is firstly conducted with the mean axial velocity values as indicated in Table 1 and then enforced as the boundary conditions for the turbulent flame simulations. The simulations are run over 10 flow through time to reach statistically stationary status and then another over 10 flow through time is run to collect statistics.

III. Results and Discussion

A. Soot volume fraction

We firstly compared our predicted soot volume fraction with the experimental data to show the good accuracy of our numerical models and framework. Figure 1 shows the comparison of the predicted (time averaged) and measured soot volume fraction at the flame centerline. As seen for all the three pressures, the predicted time averaged soot volume fractions capture the measured data very well (i.e., the discrepancy is within one order of magnitude). More specifically, our model underpredicts 4 times at 1 bar, while overpredicts 4 times at 5 bar.

To further validate the predictability of our numerical framework, the soot volume fractions at different axial positions are shown in Fig. 2, 3 and 4 to compare the predicted and measured quantities. Again, very good agreements are achieved at 6 different axial positions, at 1, 3 and 5 bar. More specifically, it is seen that at 1 bar, the simulations underpredict soot volume fraction by 6 times. At 3 bar, the predicted soot region is generally narrower than the experimental data while the soot volume fractions are captured very well. At 5 bar, the simulation over predict the soot volume fraction by 5 times. A four times difference is observed, which is considered to be accurate, compared with other simulation result for the same case [17] (a more than 10 times difference). The discrepancy at different pressure is consistent with the observation at the centerline results (i.e., Fig. 1). With the increasing pressure, although the prediction is very accurate, the model tends to predict more soot formations. This indicates that our numerical model is correlated with pressure (i.e., pressure dependent), which merits future study to further improve the model prediction at various pressures.

B. Soot volume fraction source terms

The soot volume fraction source terms, including nucleation, condensation, surface growth and oxidation are discussed at three different axial positions in this section, for all the three pressures. These soot volume fraction source terms are plotted in the mixture fraction space. As seen in Fig. 5, the increasing pressure significantly increase the rate of nucleation and condensation, which may be attributed to the higher PAH precursor concentration at elevated pressures. It is also noted that the increasing pressure pushes the nucleation and condensation to less rich regions. In addition, at $x = 0.234$ where there are large soot volume fractions for all the three pressures, condensation is dominant over nucleation due to the large values of existing soot volume fractions at this position. At more downstream ($x = 0.314$), nucleation is then stronger than condensation due to the almost oxidized soot at this position. The normalized surface growth and oxidation rates in the mixture fraction space are shown in Fig. 6. As seen, the normalized surface growth and oxidation rates are not enhanced by the increasing pressure, suggesting that the major reason for increased surface growth and oxidation are due to the increased soot volume fraction and thus the increased surface area of soot particles.

IV. Conclusions and Future Work

In this study, detailed simulations for turbulent sooting flames at elevated pressures are conducted to analyze the soot formation and evolution at elevated pressures. A conditional soot sub-filter PDF method with RFPV and HMOM are used for turbulent combustion and soot modeling. A series of KEN flames at 1, 3 and 5 bar are studied. Our numerical framework with RFPV as the turbulent combustion model, HMOM for the soot modeling, and a conditional soot subfilter PDF for the turbulence-chemistry-soot interaction, predict the soot volume fraction at different positions for all the three pressures with very good accuracy (maximum discrepancy within one order of magnitude). In addition, the soot volume fraction source terms, including nucleation, condensation, surface growth and oxidation terms, are discussed and analyzed. It is observed that the increasing pressure could significantly increase the nucleation and condensation rate. For all the three pressures, condensation is dominant over nucleation at the position where soot volume fraction peaks. In addition, it is seen that in these turbulent jet sooting flames, the normalized surface growth and oxidation are not enhanced by the increasing pressure. For future work, we plan to further analyze the chemistry

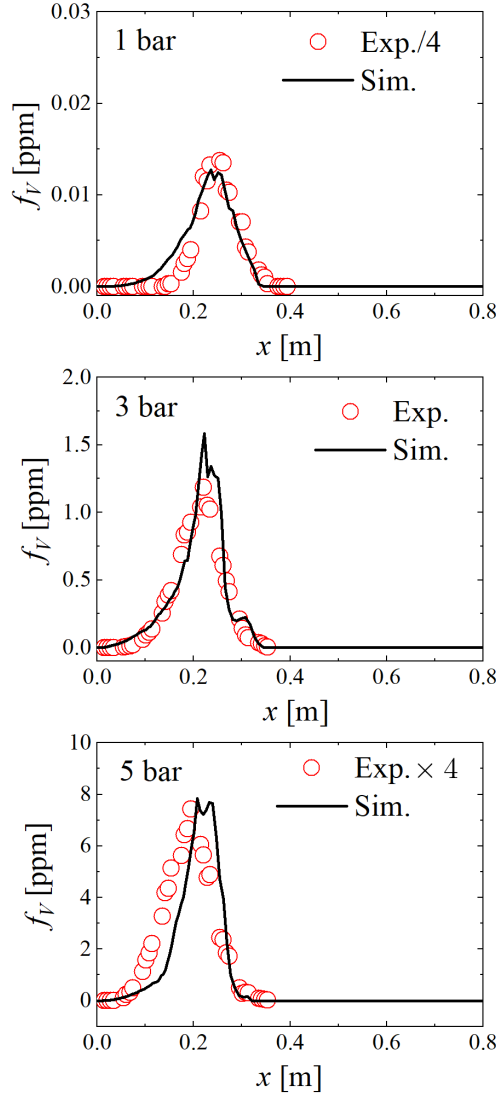


Fig. 1 Predicted and measured soot volume fractions (f_V), as a function of the axial distance (x) in the centerline.

uncertainty under high pressure in turbulent sooting flames. To conduct chemistry flux analysis for soot formation, our newly proposed method (i.e., soot-based global pathway analysis) will be used to identify the controlling reaction pathway way for soot formation and evolution.

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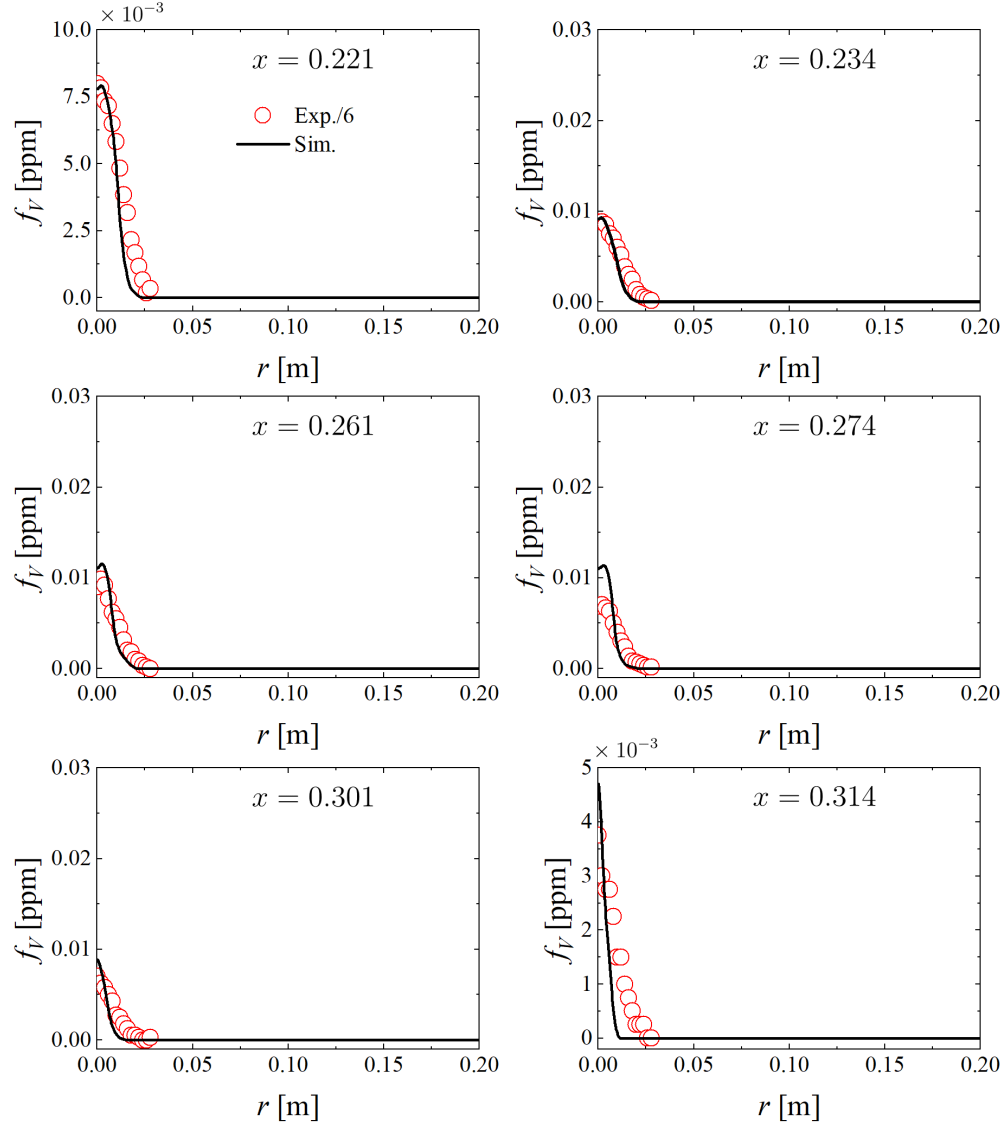


Fig. 2 Predicted and measured soot volume fractions (f_v) at 1 bar, as a function of the radial distance (r) at different axial positions.

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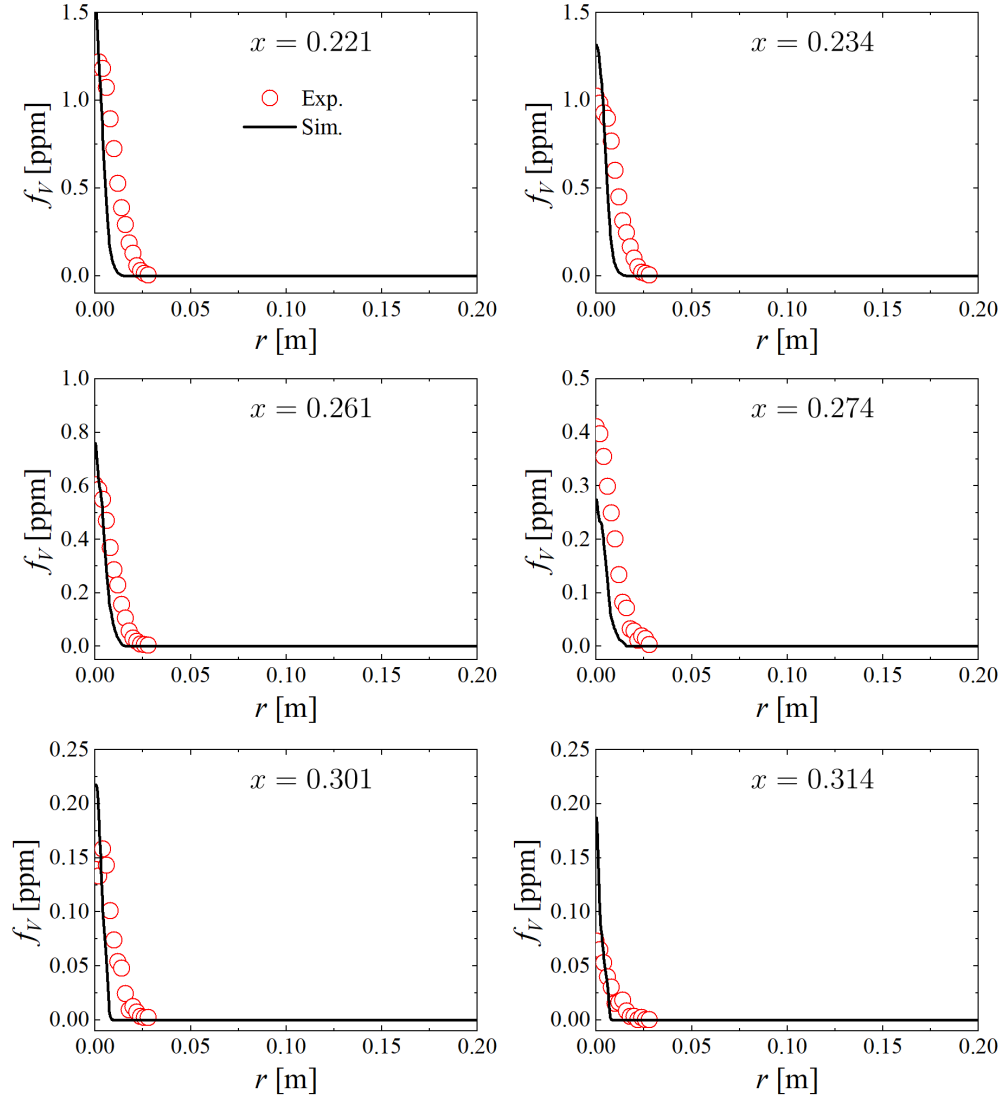


Fig. 3 Predicted and measured soot volume fractions (f_v) at 3 bar, as a function of the radial distance (r) at different axial positions.

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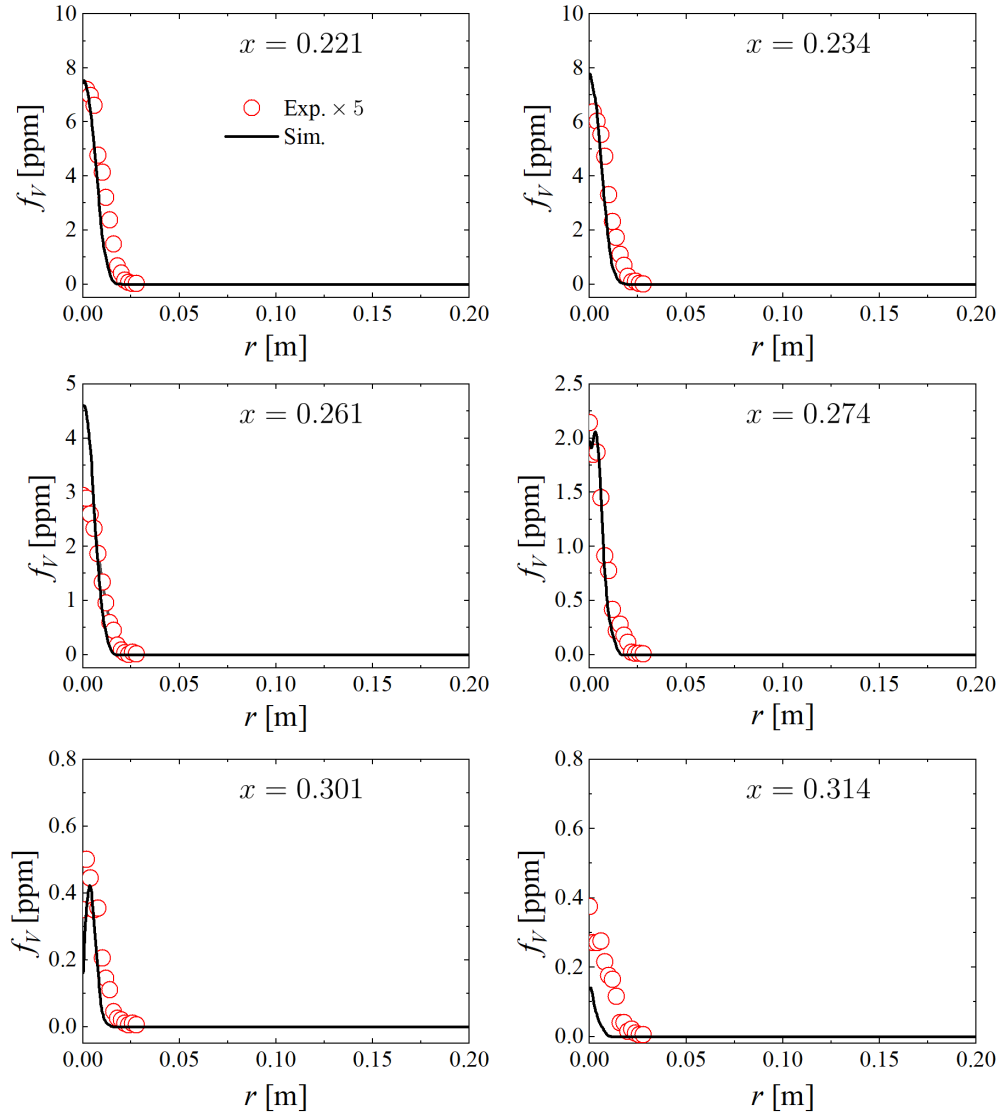


Fig. 4 Predicted and measured soot volume fractions (f_v) at 5 bar, as a function of the radial distance (r) at different axial positions.

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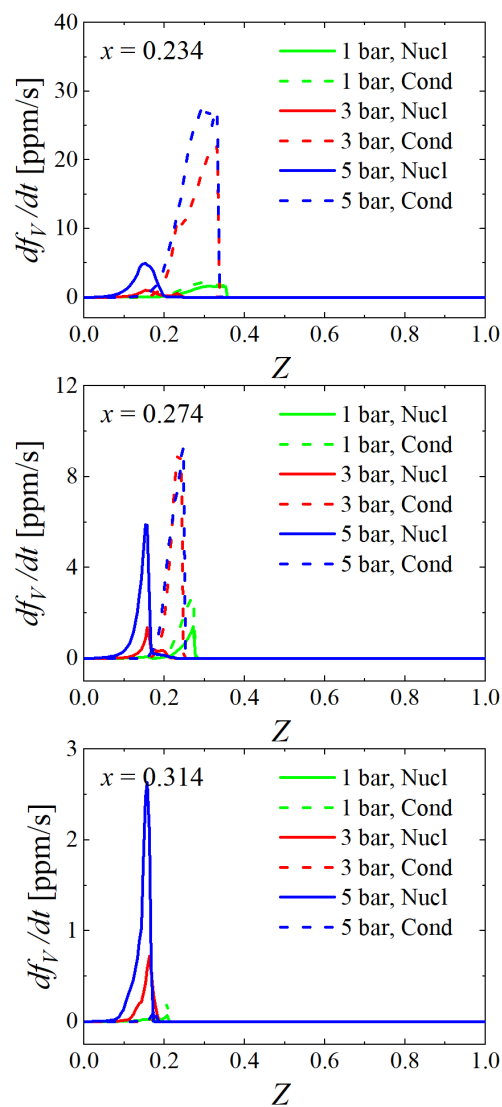


Fig. 5 Soot volume fraction nucleation (Nucl) and condensation (Cond) source terms in the mixture fraction space.

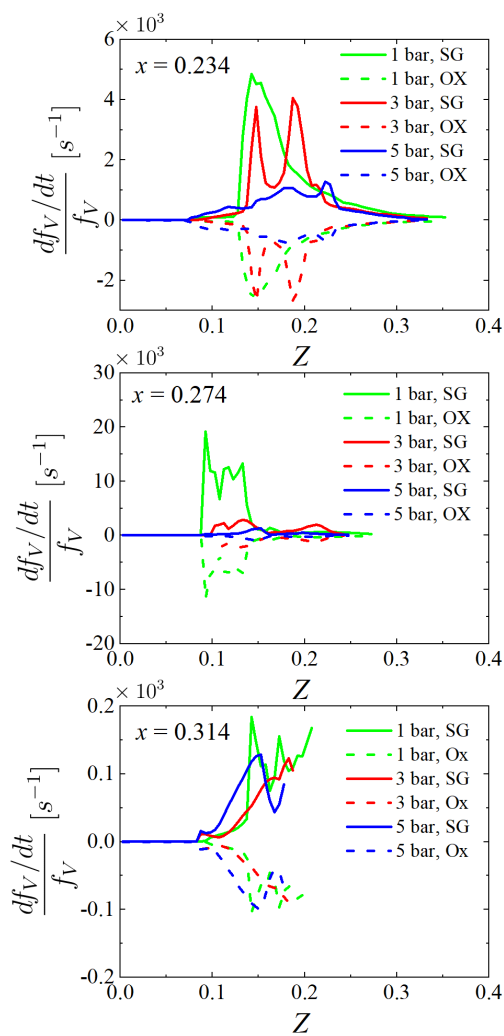


Fig. 6 Normalized soot volume fraction surface growth (SG) and oxidation (OX) source terms in the mixture fraction space.