# Impact of direct integration of Analytically Reduced Chemistry in LES of a sooting swirled non-premixed combustor

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## Abstract

Large-eddy simulation (LES) of a swirl-stabilized non-premixed ethylene/air aero-engine combustor experimentally studied at DLR is performed, with direct integration of Analytically Reduced Chemistry (ARC). Combined with the Dynamic Thickened Flame model (DTFLES), the ARC-LES approach does not require specific flame modeling assumptions and naturally adapts to any flow or geometrical complexity. To demonstrate the added value of the ARC methodology for the prediction of flame structures in various combustion regimes, including formation of intermediate species and pollutants, it is compared to a standard tabulation method (FPI). Comparisons with available measurements show an overall good agreement with both chemistry approaches, for the velocity and temperature fields. However, the flame structure is shown to be much improved by the inclusion of explicitly resolved chemistry with ARC. In particular, the ability of ARC to respond to strain and curvature, and to intrinsically contain  $CO/O_2$  chemistry greatly influences the flame shape and position, as well as important species production and consumption throughout the combustion chamber. Additionally, since both chemistry descriptions are able to account for intermediate species such as OH and  $C_2H_2$ , soot formation is also investigated using a two-equations empirical soot model with  $C_2H_2$ as the sole precursor. It is found that, in the present configuration, this precursor is strongly impacted by differential diffusion and partial premixing, not included in the FPI approach. This leads to a strong under-prediction of soot levels by about one order of magnitude with FPI, while ARC recovers the correct measured soot concentrations.

Keywords: Large Eddy Simulation, Chemical kinetics, Reduced chemistry, Gas Turbines, Soot

#### 1. Introduction

Recent implementation of emission control regulations has resulted in a considerable demand from industry to improve the efficiency while minimizing the consumption and pollutant emissions of the next generation aero-engine combustors. Less expensive than Direct Numerical Simulations (DNS), Large Eddy Simulation (LES) is an attractive tool to address these issues with high accuracy at a reasonable computing cost, and is nowadays widely employed for the simulation of turbulent combustion in both academic and applied research (see, e. g., reviews of Pitsch [1] and Gicquel *et al.* [2], and references therein).

Accurate pollutant predictions rely heavily upon the fidelity of the chemistry description; however, the computation of combustion chemistry and its coupling with turbulent flows, a recent review of which can be found in Fiorina *et al.* [3], remains challenging in LES. One main reason is that fuel pyrolysis and oxidation proceed through complex and highly non-linear mechanisms involving hundreds of different chemical species over a wide range of characteristic length and time scales. As pointed out by Lu & Law [4] in their extensive review on the subject, the direct integration of such detailed chemistry in CFD applications like LES is not a viable path, because of excessive computational demands and numerical stiffness. Employing overly detailed chemical schemes for a specific application may not be desirable either, as this introduces a large number of reaction parameters, which individually contribute very little to the global flame behaviour, while introducing possibly large uncertainties [5].

In practice, chemical kinetics in LES today is often taken into account through pre-tabulated laminar flame solutions based on detailed chemistry. As discussed by Peters [6], this method assumes that thermo-

chemical evolutions in the composition/temperature space can be parametrized by a reduced set of variables. Usually, these include the mixture fraction, characterizing the degree of fuel and oxidizer mixing, and the progress variable, monitoring the progress of reaction towards chemical equilibrium. This approach is very attractive in that it does not require much CPU while enabling to retrieve virtually any thermochemical information of interest. Some recent techniques falling into that category include the flame-generated manifold (FGM) [7], the Flame Prolongation of ILDM (FPI) [8] or the Flamelet Progress Variable (FPV) [9] which have all been successfully coupled to various turbulence-chemistry interaction models to perform LES of complex geometries [10, 11, 12, 13]. On the downside, simulations using tabulation are obviously very much dependent upon the type of canonical configurations chosen to build the look-up table (either premixed, partially-premixed or non-premixed) [14, 15], even if recent efforts addressing this issue should be acknowledged [16, 17]. Another main disadvantage of this approach is that the interactions between the flame and the flow are most often oversimplified. Indeed, taking into account complex phenomena such as turbulent, multi-species transport, dilution, heat losses or slow pollutant chemistry requires additional modeling efforts that can be far from trivial. Very often, additional parametrization variables are introduced, for which transport equations must be solved [18, 19, 20], resulting in additional unclosed terms and in an increase of the look-up table dimensionality. It is especially true in the context of soot modeling. Mueller et al. [21], for example, used a statistical Method of Moments in conjunction with a tabulated gas-phase chemistry description to investigate soot production in a real aeronautical burner, and their formulation required specific treatments to account for the loss of soot precursors from the gas-phase chemistry, and to include thermal radiation effects.

Introduced for example in Goussis *et al.* [22], another promising approach to model chemistry in LES is Analytically Reduced Chemistry (ARC). ARC is directly derived from detailed mechanisms, and accurately describes combustion phenomena by retaining only the main competing pathways and associated reactions. Typically, from 10 to 30 species and up to 500 reactions are considered, depending upon the hydrocarbon. ARC is nowadays easily derived to meet any type of requirements (combustion regime, operating range, stiffness), thanks to automated reduction tools [23, 24] implementing efficient reduction techniques [25, 26, 27, 28]. Associated with the ever increasing computational power, the use of ARC in LES has become very attractive. Jones *et al.* [29] were amongst the first to use ARC to investigate the flame structure in a relatively complex gaseous premixed burner, using a transported sub-grid PDF approach to model turbulent combustion. More recently, Bulat *et al.* [30] applied the same strategy to investigate CO and NO<sub>x</sub> formation in an industrial gas turbine combustor. It is however of particular interest to combine ARC with a geometrical combustion model such as the Dynamically Thickened Flame (DTFLES [31]) approach, so as to benefit from the direct integration of the chemistry. A first successful attempt was made by Jaravel *et al.* [32] who employed ARC with DTFLES in the same configuration as Bulat *et al.* [30]. The authors also recently applied the same methodology in a two-phase flow burner, and obtained encouraging results [33].

To further assess the capacity of the ARC-DTFLES approach to accurately predict complex flame structures, it is used in the present study to investigate partial-premixing and gaseous chemistry involved in soot production. The configuration is a sooting swirled non-premixed gas turbine combustor burning ethylene with air, experimentally studied by Geigle *et al.* [34]. The objective is to perform a comprehensive investigation of the predictive capabilities offered by the ARC-DTFLES approach in a complex geometry where dilution and various combustion regimes are present, and to show how it may be used to predict soot levels. The LES is also performed with a standard tabulated approach (FPI [8]) in order to provide a comparison with a widely employed chemistry model. Finally, the ARC-DTFLES is applied to a second operating point in order to assess the flexibility of the approach.

The paper is organized as follows: the modeling approaches used for chemistry in this study are first reviewed in Section 2, and evaluated on relevant canonical one-dimensional problems. Next, the soot modeling strategy is described in Section 3. The experimental configuration is then introduced in Section 4 and the numerical strategy is presented in Section 5. Finally, in Section 6, the flow-field and flame structure obtained with both chemistry descriptions (ARC and FPI) are compared and discussed with respect to the experimental results. The analysis is completed with a discussion on soot precursors and soot predictions.

# 2. Chemistry models

#### 2.1. Development of the ARC mechanism

As discussed in the introduction, advanced reduction techniques have been developed to preserve the relevant chemical information of a detailed mechanism [22]. These techniques often complement each other and are therefore gathered in so-called "multi-step" reduction tools like YARC [24], employed in this study. The reduction with YARC is carried out in three steps, briefly presented hereafter.

First, a set of accuracy-preserving targets (flame speed  $s_l$ , auto-ignition time  $\tau_{ig}$ ) associated with a set of canonical zero- or one-dimensional configurations is prescribed. Next, based upon the results of these canonical cases, a skeletal reduction is performed where, following the formalism of Turanyi [35], unimportant species and reactions are removed from the original detailed mechanism. This step relies on the Directed Relation Graph with Error Propagation method (DRGEP). Finally, Quasi Steady State Approximation (QSSA) is formulated for a subset of species exhibiting small characteristic timescales, as identified by the Level of Importance (LOI) criterion [36]. This last step allows a considerable reduction of the stiffness from the mechanism. A full description of this procedure can be found in Pepiot *et al.* [37]. Note that to simplify the evaluation process, no quadratic coupling is allowed between QSS candidates.

Following this approach, an ARC has been derived for ethylene oxidation, starting from the detailed mechanism of Narayanaswamy *et al.* [38], originally composed of 158 species and 1804 irreversible reactions. The reduction is based on one-dimensional laminar premixed flames and zero-dimensional auto-ignition computations, in conditions representative of the studied configuration ( $T_f = 300$  K and P = 3 bar). Targets consist of auto-ignition time ( $\tau_{ig}$ ), burnt gas temperature ( $T_b$ ), laminar flame speed ( $s_l$ ), main species equilibrium values, and specific intermediate species profiles (OH and C<sub>2</sub>H<sub>2</sub>). The resulting ARC is labelled ARC\_18\_C2H4NARA in what follows, and is comprised of 18 transported species and 11 QSS species, listed in Table 1.

Transported species	QSS species		
$N_2 H H_2 HO_2 H_2O H_2O_2 O O_2 OH CO CO_2$	S-CH <sub>2</sub> CH C HCO C <sub>2</sub> H <sub>5</sub> O HCCO		
$CH_2O CH_2CO CH_3 CH_4 C_2H_2 C_2H_4 C_2H_6$	$T-CH_2 C_2H_3 CH_2CHO C_2H_5 CH_3CHO$		

Table 1: Species contained in the ARC\_18\_C2H4NARA scheme.

#### 2.2. Construction of the FPI table

The FPI-TTC tabulation method adapted to compressible solvers by Vicquelin *et al.* [39] is used. The look-up table is constructed from a collection of one-dimensional unstrained premixed laminar flames at the conditions of the target application ( $T_f = 300$  K and P = 3 bar), computed with the solver Cantera [40] by assuming unity Lewis number for all species. The same detailed mechanism of Narayanaswamy *et al.* [38] employed for the ARC derivation is used. The flammability limits considered for the construction of the table are  $0.4 < \phi < 3.0$ . The table is parametrized with the mixture fraction  $Y_z$  based on the Bilger definition [41], and the normalized progress variable *c* based on the mass fraction of CO and CO<sub>2</sub>. Both quantities follow standard transport equations, which are computed in the LES solver. The look-up table is discretized by 200 x 200 points in  $Y_z$  and *c*, respectively. The  $Y_z$  direction is refined around stoichiometry ( $z_{st} = 0.063$ ), and stays inside the flammability limits, while points are uniformly distributed in the *c* direction. A linear interpolation is used between table points.

Assuming unity Lewis number for all species can have many consequences, a major one being an underestimation of the laminar flame speed  $s_l$ , especially for near-stoichiometric mixtures. To correct this behavior, based on simplified asymptotic analysis, a correcting factor  $F_{sl,corr} = (s_l/s_{l_{Le=1}})$  is applied to the source term of the progress variable c retrieved in the look-up table [42]. The validity of such an approach will be demonstrated below.

#### 2.3. Assessment of the chemistry models on canonical configurations

The performances of the ARC and FPI chemistry descriptions are evaluated in relevant canonical configurations: first, in one-dimensional laminar unstrained premixed flames (UPF), corresponding to the test cases used to derive the ARC; second, in counterflow strained diffusion flames (SDF), as non-premixed combustion may occur in the target application. Validations are performed at both 3 and 5 bar, to be consistent with the operating points of interest, while the fresh gas temperature is systematically set to 300 K.

Computations are performed with the solver Cantera [40]. For ARC the same transport model as in the LES solver (see Section 5) is used, based on constant values of species Schmidt numbers  $(Sc_k)$  and Prandtl number (Pr) evaluated in the flame zone of a prior complex transport calculation. For FPI all species have a unity Lewis number. Results are compared to the detailed mechanism [38] used to build both the FPI table and the ARC mechanism, associated to either a complex transport formulation (Mix) or unity Lewis numbers  $(Le_k = 1)$ . For validation of the detailed mechanism, the reader is referred to published materials [43, 38].

This Section summarizes the main results of the validations performed at 3 bar. Additional information, as well as validations at 5 bar (leading to the same conclusions), can be found in the Supplementary Materials.

#### 2.3.1. Unstrained Premixed Flames (UPF)

The burnt gas temperature as well as the laminar flame speed are plotted versus equivalence ratio in Figs. 1 (a)&(b), respectively. Both ARC and FPI performances are excellent, with a relative error on  $s_l$  below 4% in the targeted range of equivalence ratio ( $0.5 < \phi < 1.5$ ) and never exceeding 10% over the entire flammability range. As expected, unity Lewis numbers lead to an under-prediction of  $s_l$  by up to 25% around stoichiometry, demonstrating the necessity and validity of the correction factor  $F_{sl,corr}$  that allows FPI to retrieve the correct  $s_l$ .

The total CO production, integrated through the flame as:

$$\dot{\omega}_{CO}^{tot} = \int_{c<0.98} \dot{\omega}_{CO} dx \tag{1}$$

where the integration is limited to c < 0.98 to illustrate the effects of chemical kinetics, is presented on Fig. 1 (c). Here also, an excellent agreement is observed between the detailed mechanism and the ARC\_18\_C2H4NARA results. The global error computed over the entire range of equivalence ratio is below 2%. On the contrary, FPI under-predicts the total CO production, as does the  $Le_k = 1$  detailed mechanism. In rich conditions ( $\phi > 1.3$ ) the relative error decreases down to less than 20%. Note that the FPI and  $Le_k = 1$  detailed mechanism results do not perfectly match, either due to the flame speed correction factor or tabulation interpolation error. Results are indeed very sensitive to the refinement in the c direction, especially for species with a strong gradient in the phase space.

Finally, the evolution of the species of interest -namely, CO,  $C_2H_2$  and OH- across the flame front is investigated. Maximum values are plotted versus mixture fraction in Figs. 1 (d)-(f). Overall, an excellent agreement is again observed between the detailed mechanism and the ARC mechanism, with only slight discrepancies at highest equivalence ratio. The maximum of  $C_2H_2$  is best predicted in moderately rich conditions (1.1 <  $\phi$  < 1.6). Outside of this range, the relative error can reach up to 10%, as shown in Fig. 1 (f). Similarly to what was observed for CO total production, FPI gives results very close to the  $Le_k = 1$ detailed chemistry, under-predicting peak levels of  $C_2H_2$  by about 20% (Fig. 1 (f)), and predicting higher peaks of OH (Fig. 1 (e)), indicating higher heat release rates. CO and CO<sub>2</sub> (not shown) maximum levels are otherwise consistent with the reference computations, with a maximum relative error below 7%. Note that all the simulations evolve towards the same burnt gas state. In particular, although using a reduced number of species, ARC predicts equilibrium values with relative errors below 0.5% for the temperature, 2% for OH, and 4% for CO and CO<sub>2</sub> over the entire range of equivalence ratio. An example of spatial profile for a stoichiometric flame can be found in the Supplementary Materials (Fig. S1).

#### 2.3.2. Strained Diffusion Flames (SDF)

A series of counterflow diffusion flames, for a large range of strain rates, is now considered. The oxidizer inlet is composed of air while the fuel inlet is composed of pure ethylene. The response to strain is assessed in



Figure 1: a) Burnt gas temperature, b) laminar flame speed, c) total CO production, and maximum of d) CO, e) OH, and f) C2H2 versus equivalence ratio in UPF. Initial temperature is 300 K, P = 3 bar. Detailed mechanism - Mix (solid line), ARC\_18\_C2H4NARA (o), Detailed mechanism - Le=1 ( $\Delta$ ), FPI (x).

terms of total fuel and CO consumption, as well as maximum temperature and major species mass fractions, as shown in Fig. 2. A global strain rate is used:

$$a = \frac{u_o + u_f}{d} \tag{2}$$

where  $u_o$  and  $u_f$  are the oxidizer and fuel inlet velocities, respectively, and d is the distance between the two inlets.



Figure 2: a) Maximum temperature, b) total  $C_2H_4$  and c) total CO production and maximum of d) CO, e) OH, and f) C2H2 versus strain rate in SDF. Fresh gas temperature is 300 K, P = 3 bar. Detailed mechanism - Mix (solid line), ARC\_18\_C2H4NARA (o), FPI (+).

The first observation is that ARC prediction is excellent, especially considering the fact that non-premixed flames were not targeted in the reduction. The relative error on the maximum temperature never exceeds 1% for all strain rates, and extinction occurs at  $a_{ext} \approx 2320 \ s^{-1}$ , consistently with the prediction of the detailed mechanism. CO and OH maximum levels are equally well predicted, with a relative error below 5%, except for small strain rates ( $a < 50 \ s^{-1}$ ) where the relative error on the maximum of OH reaches up to 10%. Levels of C<sub>2</sub>H<sub>2</sub> are less accurately reproduced, and over-predicted with a relative error ranging from 10% to 30% for small strain rates ( $a < 75 \ s^{-1}$ ). A flame structure in phase space ( $a = 50 \ s^{-1}$ ), illustrating the discussed discrepancies, is given in the Supplementary Materials (Fig. S2).

Results obtained with the FPI table reveal a different response to strain. Figures 2 (a)-(c) show that global tendencies are correctly captured: the maximum temperature decreases with strain rate while the total fuel consumption increases. The predicted strain rate of extinction is 2105  $s^{-1}$ , which is not far from the expected value of  $a_{ext} \approx 2320 \ s^{-1}$ . However, Figs. 2 (d)-(f) show that maximum levels of species of interest are either over- or under- predicted. Maximum levels of C<sub>2</sub>H<sub>2</sub>, in particular, are about one third of the reference values. Moreover, maximum levels of CO decrease with strain rate, while they should increase. These results are consistent with previous observations [15] in similar cases, and are due to the fact that premixed flames were considered for the tabulation (see discussion about Fig. S2 in the Supplementary Materials). The unity Lewis assumption can not be invoked here.

The poor FPI results on SDF should be kept in mind in the analysis of the target configuration.

#### 3. Soot modelling

Soot formation is a highly intermittent and complex process depending upon the flow time history and proceeding in stages. Starting from nucleation, which is the inception of a new soot particle from its gaseous precursors, the process continues through various surface reactions and with particle agglomeration [44, 45]. At the heart of these processes, aromatic species such as  $C_6H_6$  and intermediate species such as  $C_2H_4$ ,  $C_2H_2$  and OH, play a key role. Obviously, an accurate gas-phase chemistry description is thus a pre-requisite to any accurate soot modeling [45]. As a first step towards detailed soot chemistry, the simple two-equations empirical model proposed by Leung *et al.* [46] is employed here, based on  $C_2H_2$  as the sole soot precursor, known to yield good estimates and trends [47, 48, 49]. This choice is made to demonstrate, at a reasonable CPU cost, how the link between gaseous chemistry and soot formation can be taken into account in LES, and how sensitive is soot formation to combustion chemistry.

In the Leung model, transport equations for the number density of soot particles  $n_s$  and the soot mass fraction  $Y_s$  are solved:

$$\frac{\partial \rho n_s}{\partial t} + \frac{\partial \rho u_i n_s}{\partial x_i} = k_T \frac{\partial}{\partial x_i} \left( \frac{\rho \nu n_s}{T} \frac{\partial T}{\partial x_i} \right) + \dot{\omega}_{n_s},\tag{3}$$

$$\frac{\partial \rho Y_s}{\partial t} + \frac{\partial \rho u_i Y_s}{\partial x_i} = k_T \frac{\partial}{\partial x_i} \left( \frac{\rho \nu Y_s}{T} \frac{\partial T}{\partial x_i} \right) + \dot{\omega}_{Y_s} \tag{4}$$

where  $k_T = 0.54$  is a constant and  $\nu$  is the gas kinematic viscosity. The source terms on the right-hand-side of Eqs.(3) & (4) are given by:

$$\dot{\omega}_{n_s} = \frac{2N_A k_1(T)}{C_{min}} [C_2 H_2] - 2C_a \left(\frac{6M_s}{\pi \rho_s}\right)^{1/6} \left(\frac{6\kappa T}{\rho_s}\right)^{1/2} (\rho n_s)^{11/6} \left(\frac{\rho Y_s}{M_s}\right)^{1/6},\tag{5}$$

$$\dot{\omega}_{Y_s} = M_s \left[ k_1(T) [C_2 H_2] + k_2(T) [C_2 H_2] S^{1/2} - k_3(T) S[O_2] \right]$$
(6)

where  $C_{min} = 100$  is the number of carbon atoms in a nascent soot particle and  $C_a = 9$  is an agglomeration rate constant;  $N_A$  and  $\kappa$  are the Avogadro and Boltzmann constants, respectively, and [X] designs the molar concentration of species X.  $M_s = 12.011$  kg.kmol<sup>-1</sup> and  $\rho_s = 2000$  kg.m<sup>-3</sup> are the carbon molar mass and density, respectively; S is the soot volume surface per unit volume of gas, based on the evaluation of a mean particle size [46].

In essence, this model postulates that the soot formation and growth occur in four main stages:  $Y_s$  evolution is governed by nucleation, surface growth, and oxidation, while  $n_s$  evolves according to nucleation and coagulation. These stages are described by a set of reaction rates  $k_i$  of the Arrhenius type. In such empirical models, results are much sensitive to the rate constants which must be carefully calibrated on representative canonical laminar test cases or more complex turbulent flames [12]. In the present study, the calibration led to the final set of parameters reported in Table 2. Note that soot oxidation only involves  $O_2$ , as prescribed in the pioneering paper of Leung *et al.* [46].

	$A_i$	$n_i$	$T_i$	units
$k_1$ (nucleation)	$2.0 \ 10^4$	0	21100	$[s^{-1}]$
$k_2$ (growth)	$1.2 \ 10^4$	0	12100	$[m^{3/2}m_{soot}^{-1}s^{-1}]$
$k_3$ (oxidation $O_2$ )	$1.0 \ 10^2$	0.5	19680	$[m^3 m_{soot}^{-2} s^{-1}]$

Table 2: Modified Arrhenius constants of the empirical soot model of Leung et al. [46].

# 4. Experimental configuration

# 4.1. Presentation of the experiment

The experimental configuration studied in this work is installed at DLR and was presented and investigated in several recent publications [34, 50, 51, 52]. This burner was designed to study soot formation in gas turbine combustors under elevated pressure, with or without secondary air dilution. To this end, the burner features a non-premixed swirled injection system consisting of three concentric nozzles and two radial swirlers (see Fig. 3 (a)). Air at 300 K is supplied to the flame through both the central (diameter 12.3 mm) and annular (inner diameter 14.4 mm, outer diameter 19.8 mm) nozzle. The air flows are fed from separate plenums and go through radial swirlers consisting of 8 channels for the central nozzle (width 4.2 mm, height 5.4 mm, swirl number 0.82) and 12 channels for the annular nozzle (width 3.2 mm, height 4.5 mm, swirl number 0.79). Gaseous fuel ( $C_2H_4$ ) is injected at 300 K between both air flows through 60 straight channels, forming a concentric ring. The fuel injection mimics the behavior of an atomizing lip observed in air-blast liquid atomizer [53]. All nozzle exit planes are located at the level of the combustion chamber dump plane. The combustion chamber measures 120 mm in height, has a square section of  $68 \times 68 \text{ mm}^2$ , and features large optical access from all 4 sides. To enhance soot oxidation, additional air ducts (5 mm diameter) inject secondary air into the combustor 80 mm downstream the combustion chamber inlet.



Figure 3: a) Sketch of the experimental configuration from Geigle *et al.* [50]. b) Computational domain. c) Focus on the mesh in the first half of the combustion chamber with a close-up near the very thin fuel injection nozzle.

Amongst the various operating points experimentally studied [34], the two cases (A and B) chosen for the present study are summarized in Table 3. They differ by the operating pressure, which has been shown to have a strong effect on the soot production [34, 54]. Even though the burner is operated under overall lean conditions ( $\phi = 0.86$ ) the "primary" combustion zone localized near the main injection system (PZ, see Fig. 5) is characterized by an overall rich equivalence ratio ( $\phi_{PZ} = 1.2$ ).

	Pressure	$Q_{air,central}$	$Q_{air,radial}$	$Q_{air,oxy}$	$Q_{fuel}$	$\phi$	$\phi_{PZ}$	$P_{global}$
	[Bar]	$[kg.s^{-1}]$	$[kg.s^{-1}]$	$[kg.s^{-1}]$	$[kg.s^{-1}]$	[-]	[-]	[kW]
Case A	3.0	$3.089 \times 10^{-3}$	$7.049 \times 10^{-3}$	$4.016 \times 10^{-3}$	$8.191\times10^{-4}$	0.86	1.2	38.6
Case B	5.0	$5.025\times10^{-3}$	$1.172\times 10^{-2}$	$6.697\times10^{-3}$	$1.364\times10^{-3}$	0.86	1.2	64.4

Table 3: Experimental operating conditions.

# 4.2. Available data

Comprehensive data obtained by several laser diagnostics are available for each operating point. Velocity component statistics at several positions downstream of the dump plane were obtained from Stereo - Particle Image Velocimetry (Stereo-PIV). Due to the high level of soot luminosity, two different detection schemes were employed, referred to as the Field of View (FoV) and the sum-of-correlation (SoC) (see [55] for definitions). These two sets of velocity data will be employed in the following analysis, although it is noted that Geigle *et al.* [55] describe the FoV as being more reliable. For flame analysis, temperature measurements at several locations in the combustor were obtained by Coherent Anti-Stokes Raman scattering (CARS), and Laser Induced Fluorescence (LIF) was used to provide a qualitative estimate of the OH radical distribution. Finally, Planar Laser-Induced Incandescence (LII) was used to measure the soot volume fraction.

# 5. Numerical setup

#### 5.1. Computational domain and numerical strategy

The computational domain is displayed in Fig. 3 (b). It includes the channels of both air inlets, the combustion chamber, secondary air ducts and part of the outside atmosphere (not shown). The 60 straight channels for the fuel inlet are modeled by a continuous annular nozzle. The domain is discretized into a fully unstructured mesh using  $\approx 40$ M tetrahedral elements, with a cell size of about 0.07 mm in the very thin (0.4 mm) fuel injection nozzle and in the primary mixing region. A picture of the mesh with focus on the fuel injection can be seen in Fig. 3 (c). The axial direction is referred to as the z-axis, corresponding to the main flow direction, while the x-axis and y-axis denote the transverse directions.

All simulations are performed with the LES solver AVBP [56, 57], an explicit cell-vertex massively-parallel code solving the compressible multi-species reacting Navier-Stokes equations. A third-order accurate in space and time Taylor-Galerkin finite-element scheme with low-dissipation [58] is used for the discretization of the convective terms, while a second order Galerkin scheme is used for diffusion terms. A full description of the filtered LES equations can be found, e.g., in the review of Gicquel *et al.* [2], and only the main features are recalled hereafter.

All filtered viscous terms are modelled via a gradient assumption. The diffusive fluxes of each species rely on the Hirschfelder and Curtis approximation [59] with the definition of constant but non necessarily equal Schmidt numbers for each species. Likewise, the heat flux rely on the definition of a constant Prandtl number. With the ARC\_18\_C2H4NARA mechanism, this leads to the consideration of constant but nonunity Lewis numbers for each species. The viscosity follows a classical power law. The subgrid-scale (SGS) stress tensor is modelled following the WALE approach [60], while the SGS diffusive heat and species fluxes are modelled similarly to the filtered fluxes: assuming constant turbulent Prandtl and Schmidt numbers  $(Pr^t = Sc_k^t = 0.6)$ . Modelling of turbulence-chemistry interactions is detailed in Section 5.2. Inlet and outlet boundary conditions are treated according to the Navier-Stokes Characteristic Boundary Conditions formulation [61], and all walls are considered adiabatic non-slipping.

# 5.2. Chemistry-turbulence interaction

Two LES simulations have been performed using either the FPI tabulation or the ARC\_18\_C2H4NARA mechanism. In both cases, the Dynamically Thickened Flame (DTFLES) model [31] was employed, where a thickening factor  $\mathcal{F}$  is calculated based on the ratio of the laminar flame thickness  $\delta_l^0$  to the local mesh size  $\Delta x$ :

$$\frac{F\delta_l^0}{\Delta x} = 5\tag{7}$$

to ensure at least 5 points in the flame front. An efficiency function  $\mathcal{E}$  based on a SGS equilibrium assumption between turbulence and flame surface [62, 31] is used to account for the SGS flame wrinkling. Note that in partially premixed conditions a wide range of equivalence ratios is encountered, requiring the use of local values of  $\delta_l^0$  and  $s_l$  consistent with the local mixture fraction.

To leave the non-reacting regions unperturbed, a dynamic sensor S detects regions of high heat release where the real thickening factor  $\mathcal{F} = 1 + (F - 1)S$  is then applied to the filtered equations. S is based on the source term of fuel in the ARC simulation while it uses the gradient of the progress variable in the FPI simulation. It has been verified, however, that the resulting sensor shape is similar in both cases. Figure 4 displays instantaneous snapshots of efficiency and thickening fields, with superimposed heat release rate iso-contours to localize the flame front. It is observed that the spatial distribution with respect to the flame front and maximum levels exhibited by both quantities are fairly similar in both simulations.



Figure 4: Instantaneous fields of a) efficiency  $\mathcal{E}$  and b) thickening factor  $\mathcal{F}$  in ARC (top) and FPI (bottom) - LES of the target configuration (zoom near the injector).

Accordingly with the DTFLES intent, which is to allow the flame resolution directly on the mesh, all turbulent SGS models are deactivated, in both the energy and species equations, when the thickening is activated.

# 6. Results and discussion

Flow statistics have been collected during 30 ms, i.e., about 2 flow through times of the combustor, for all simulations. The discussion and detailed analysis are first focused on Case A, having many more available experimental data than Case B, with both the ARC approach (ARC-LES) and the FPI table (FPI-LES). Differences induced by increasing the operating pressure on the flame dynamics and structure are then discussed with regards to their effect on species and soot production, with the ARC mechanism only.

# 6.1. Flow and flame features

#### 6.1.1. Structure of the turbulent flame

Figure 5 shows instantaneous fields of temperature as well as OH and CO mass fractions, in a central cut plane of the combustor, for both LES. The main flow structures are characteristic of swirled stabilized burners where a large inner recirculation zone (IRZ) is located in the center of the combustion chamber, induced by the radial expansion of the swirled jets. This IRZ is clearly visible on all instantaneous fields, enabling fresh air from the dilution jets to be convected upstream and fill the core of the combustion chamber. The temperature is thus lowered in this region, and the mixture fraction is globally lean. The IRZ most upstream point is located within the central air injection nozzle in both LES, and is subjected to strong oscillations: as observed in the experiment, the burner is prone to instabilities and flame stabilization is difficult to reach. This is especially true in the case of the FPI-LES, where the main flame can intermittently reach all the way up into the central air injection nozzle. Small outer recirculation zones (ORZ) are also observed in both simulations, but an investigation of the OH levels in this region (Fig. 5 (b)) suggests very different dynamics, resulting in a less homogeneous OH distribution in the FPI-LES.

A highly turbulent "V" shape flame is observed near the injection with both chemistry models, as inferred from the white heat release iso-contours on the temperature fields displayed in Fig. 5 (a). Also, note that the main flame extent obtained with the FPI-LES appears to be significantly longer. However, the flame



Figure 5: Instantaneous fields of a) temperature and b) OH mass fraction with heat release rate iso-contours in white to localize the reaction zone and stoichiometric iso-contours in grey. Top: ARC-LES, bottom: FPI-LES.

surface, calculated as the integral of the flame surface density  $(\int_V |\nabla c| dV$  where c is a transported variable in the FPI-LES and is computed a *posteriori* in the ARC-LES) gives a total flame surface of about 70 cm<sup>2</sup> with FPI and of about 85 cm<sup>2</sup> with ARC. This indicates that the flame front in the FPI-LES is both more lifted and fragmented, consistently with the highly corrugated stoichiometric iso-contours observed in Fig. 5 (bottom). This suggests a faster and more efficient mixing process in the main flame region of the ARC-LES.

Downstream the V-shape flame and aligned with the stoichiometric line, intermittent secondary reaction zones with relatively high heat release rate and OH levels are observed in both LES. However these structures appear to have higher heat release rate levels in the FPI-LES. Furthermore, similar structures also appear as isolated burning pockets in leaner and colder regions near the main axis in the FPI-LES, while they are never seen in the ARC-LES.

This brief analysis clearly indicates the presence of two different reaction fronts, one in the vicinity of the main central injection and one in the vicinity of the dilution jets. Following these observations, the domain is divided into two zones (see Fig. 5): the Primary combustion Zone (PZ) and the secondary Dilution Zone (DZ), which are of different nature and will be studied separately, in Sections 6.3.2 and 6.3.3, respectively.

#### 6.1.2. Unsteady flame mechanisms

In both LES, the PZ flame is found to oscillate at a given frequency corresponding to the experimentally observed precessing vortex core (PVC) frequency (500 Hz [51]). The interaction with the PVC wrinkles the flame front located in the inner shear layer (ISL) and induces the intermittent formation of rich pockets of hot gas mixture at the tip of the V-shaped flame, which are convected along the walls towards the exit of the combustor. This behavior is illustrated on Fig. 6, showing instantaneous fields of CO mass fraction in black and white, with an additional field of  $C_2H_2$  mass fractions superimposed (iso-volume). Fuel and temperature iso-contours are appended, in black and red, respectively. In the flame core, these rich pockets are mainly composed of fuel and  $C_2H_2$  while the conversion to CO and further oxidation of CO into CO<sub>2</sub> is a post-flame process. If high CO levels are retrieved throughout the combustion chamber in both simulations (see the solid black arrows in Fig. 6),  $C_2H_2$  in particular is exclusively localized in the PZ flame region in the FPI-LES (Fig. 6 bottom) when high levels can be seen also in the post-flame region in the ARC-LES (see the dashed arrow in Fig. 6 top). In this case, the different species and temperature diffusion velocities impact the mixing of the rich pockets with the hot gas, which then deviates from the reference flame state and leads to different chemical behaviors. In particular, the fact that  $C_2H_2$  can be found in higher temperature regions eventually alters its production rate, as will be seen in Section 6.4. In the case of FPI-LES the strict correlation of c with the temperature and gas composition (prescribed by the unity Lewis numbers) does not allow any deviation from a reference flame state, such that  $C_2H_2$  for example can never be observed in



Figure 6: Instantaneous fields of CO mass fraction with superimposed  $C_2H_2$  iso-volume: ARC-LES (top), FPI-LES (bottom). Black iso-contours localize pockets of  $C_2H_4 > 0.1$ ; red iso-contours correspond to T = 1550 K.

high temperature regions. This might be a reason for the more fragmented flame described in the previous Section and the less homogeneous CO field observed in Fig. 6 (bottom).

Note finally that  $C_2H_2$  is exclusively produced in the PZ flame region in both LES, which is also where pure ethylene flames are observed. The secondary reaction zones of the DZ are not ethylene flames. The structure of these reaction fronts will be further analyzed in Section 6.3.3.



Figure 7: Instantaneous fields of temperature, from the ARC-LES. Heat release rate iso-contours in white localize the reaction zone while grey iso-contours localize the stoichiometry.

Another interesting intermittent behavior of the flame is observed in the central region of the PZ, near the dump plane. As mentioned previously, the flame is prone to strong oscillations. In particular, this maintains a low frequency motion of the IRZ in the axial direction, giving rise to different instantaneous flow-field configurations. As illustrated in Fig. 7 (a), most of the time the IRZ (2) containing recirculated gases is separated from the fresh inflow feeding the PZ (1) by a weak reaction front and a small layer of hot burnt gases. However, intermittently, this flame locally quenches and the fresh air inflowing from the injection system interacts with the fresh air inflowing from the dilution jets, leading to a dramatic temperature drop in the PZ region, as seen on Fig. 7 (b). Fortunately, the recurrent interaction of the inner shear layer (ISL) with the PVC brings back reacting fronts and flammable premixed pockets toward the central axis (as illustrated by the black arrows in Fig. 7), allowing flame re-ignition and the re-establishment of a configuration of the type displayed in Fig. 7 (a). This intermittent behavior is mainly observed in the ARC-LES, while the FPI-LES mostly exhibits a flow configuration of the type displayed in Fig. 7 (a), with a main flame stabilized further upstream.

# 6.2. Validation by comparison with experiment

#### 6.2.1. Velocity and temperature statistics

Figure 8 (a) provides a comparison of mean axial velocity fields obtained with both chemistry descriptions. The white iso-contours delimit large IRZ and small ORZ. Figure 8 (b) displays mean axial temperature fields obtained in both LES in a mid-cut plane through the dilution holes. White heat release rate iso-contours identify the main reacting region. As was already observed in the instantaneous fields, similar flow structures are observed in both LES, despite a radial expansion of the IRZ slightly more pronounced in the ARC-LES. The IRZ most upstream point is located within the central air injection nozzle in both LES, but in the case of the FPI-LES, consistently with a main flame reaching further upstream, the mean IRZ extends all the way up into the main injection system. This IRZ extends downstream to z = 90 mm for both LES, where it interacts with the dilution air injected at z = 80 mm, resulting in hot gas cooling and entrainment of part of this low temperature gas upstream towards the PZ (Fig. 8 (b)). As discussed in the previous Section, this motion leads to intermittent temperature drops in the PZ region, which appear to be more frequent with the ARC mechanism. Radially also, this region of low temperature appears broader in ARC-LES, which is due to a different chemical behavior in the DZ, as will be shown in Section 6.3.3. Note that the secondary reaction zones of the DZ do not appear on the average fields, as they are much more intermittent, and therefore weaker in average than the main PZ flame.



Figure 8: (a) Time-averaged axial mean velocity fields, with white iso-contours corresponding to zero axial velocity. ARC (top half) and FPI (bottom half). (b) Mean temperature fields, with heat release rate iso-contours in white to localize the reaction zone. ARC-LES (top half) and FPI-LES (bottom half).

In Fig 9, numerical axial, radial and tangential velocity profiles are validated against experimental data at 4 axial positions of interest in the combustion chamber: z = 15 mm and 18 mm are located in the PZ, z = 95 mm is located near the secondary air injection while z = 38 mm sits in between. These positions are materialized by white solid lines on Fig. 8 (a). Note that both sets of experimental data discussed in Section 4.2 are presented. A very reasonable agreement is reached in both ARC-LES and FPI-LES at the first 3 axial positions. The width and magnitude of the IRZ near the injector, in particular, is well captured. Larger discrepancies are observed at z = 95 mm, where the tangential velocity is under predicted, while an over prediction of axial velocity near the central axis is observed. This axial position is located just downstream the meeting zone of the 4 dilution jets, which were found quite intermittent with important variations of the jets positions and associated stagnation point.

In Fig. 10 (a), the mean temperature profiles along the combustor axis obtained with both LES are compared to experimental data. Results are much satisfying over most of the combustion chamber, lying in particular within the range of experimental standard deviation. A large over prediction of the mean



Figure 9: Mean axial, radial and tangential velocity profiles extracted in the mid-plane at z = 15, 18, 38 and 95 mm, ARC 18 C2H4NARA (solid line), FPI (dotted line), experiment: FoV( $\blacktriangle$ ) and SoC(o).

temperature is however observed close to injection (in the range 0 < z < 10 mm) with FPI-LES, in a region where both experiment and ARC-LES exhibit a sharp mean temperature gradient. This is to be related with the PZ flame stabilizing further upstream with FPI-LES, as was already observed in previous Sections. This is also revealed by the temperature histogram at location z = 3 mm and x = 0 mm (probe P1 in Fig. 8 (b)) displayed in Fig. 10 (c) top, confirming the frequent occurrence of burnt gases in this region while only fresh gases are found with ARC-LES and in the experiment. Consistently with Fig. 10 (a), other temperature histograms along the central axis (probes P2 to P4) show a good agreement between both LES and the experiment, with no occurrence of fresh gases in P2 or P3, and a visible cooling effect of the dilution jets at P4.

As was observed for the IRZ, a closer inspection of Fig. 8 (b) reveals that the mean opening angle of the V-shape flame is slightly wider with the ARC mechanism. This is clearly reflected on the first two radial profiles of temperature provided in Fig. 10 (b), at locations materialized by dashed white lines in Fig 8 (b). These two radial profiles also show that the FPI-LES gives lower minimum temperature, closer to the experiment than the ARC-LES. Examining the corresponding temperature histograms (for example probe R2: z = 18 mm and x = 8 mm) reveals indeed the occurrence of low temperature gas in the FPI-LES, originating from the aforementioned more fragmented flame. This trend is also present in the experiment, but less marked, and is absent in the ARC-LES. From all histograms displayed in Fig. 10 (c), it is evident that the ARC-LES leads to a more efficient mixing than the experiment, as deduced from the narrower distributions. The flame obtained with ARC-LES is therefore more compact, as clearly seen in Fig 8 (b) (top), and ends around z = 18 mm.

Looking now at mid chamber and the DZ, probes P3, P4 and R3 in Fig. 10 (c) along with the radial temperature profiles at these axial locations (not shown) suggest that the broader radial extent of the diluted low temperature gas region obtained with the ARC-LES is more in agreement with the experiment.

# 6.2.2. Qualitative OH comparison

Experimental OH PLIF data allow to perform a qualitative comparison between measurements and LES. At this point it is interesting also to compare the qualitative behaviors of Cases A and B (only ARC was employed to compute Case B). Overall, the major flame structures are well retrieved by both chemistry



Figure 10: a) Mean axial temperature profiles with experimental standard deviation (shaded area), b) mean radial evolution of temperature extracted in mid-plane at z = 12, 18 and 95 mm, ARC\_18\_C2H4NARA (solid line), FPI (dotted line), experiment (symbols) and c) Probability Density Functions of temperature at various locations (see probes location in Fig. 8), ARC\_18\_C2H4NARA (solid line), FPI (dotted line), experiment (bars).

descriptions, and for both operating pressure, as shown in Fig. 11. No major differences can be seen in terms of instantaneous flame shape or position between Cases A and B, and differences appear only in concentration levels, which are lower for Case B (higher pressure) in and right behind the primary flame front. This trend is well reproduced by the ARC-LES, and is in good accordance with one-dimensional premixed laminar flame behavior. Note that computational results have been rescaled to match the minimum/maximum values of the FPI-LES (Case A) computation, while experiments are normalized by the maximum value of Case A, to allow a direct comparison.

These instantaneous OH images confirm, in particular, the presence of the secondary reaction zones, originating from the PZ and extending downstream toward the DZ, responsible for maintaining significantly high OH levels throughout the combustion chamber. A major difference between LES and experiments is however observed in the center of the upstream part of the IRZ, where significant OH levels are seen in the experiments but not retrieved in the LES. The origin of this discrepancy is unclear, but could be due to the detection of additional species also excited in the OH measurements. Indeed, no reaction is expected within the IRZ.

Looking now at time-averaged OH fields (Fig. 12), the difference of levels between both operating pressure is striking. The main observations previously made on instantaneous fields still holds: high OH levels are found in the outer IRZ limit throughout the combustion chamber and both LES exhibit the right trends except in the upstream part of the IRZ. Note that in average, higher OH levels are detected in the ORZ in the LES, particularly with FPI. This could be due to heat losses not taken into account in the LES, to longer residence times resulting from too large predicted ORZ or even to the limit of the experimental window.

# 6.3. Analysis of the local instantaneous flame structure

# 6.3.1. Identification of the combustion regimes

First introduced by Yamashita *et al.* [63], the local Takeno Index (TI) is used to distinguish premixed from non-premixed reaction fronts:

$$TI = \frac{\nabla Y_F \cdot \nabla Y_{O_2}}{|\nabla Y_F \cdot \nabla Y_{O_2}|} \tag{8}$$



Figure 11: Instantaneous snapshots of OH mass fraction (top left and bottom left: ARC-LES; top right : FPI-LES) and OH-PLIF (top center and bottom right). Top: Case A, bottom: Case B.



Figure 12: Time-averaged fields of OH mass fraction (ARC-LES and FPI-LES, left half) and OH-PLIF (experiment, right half) for Cases A and B. LES values are normalized by the extremum values of the FPI-LES while experiments are normalized by the maximum value of Case A.

Alignment of fuel (F) and oxidizer (O) gradients reveal a premixed flame front, while opposite gradients indicate a diffusion front. Several improvements to the TI have been proposed in the literature. Fiorina *et al.* [15] use different evaluations of the gradient of oxidizer to better distinguish diffusion driven fronts apparently premixed burning. Knudsen *et al.* [17] evaluate the relative contributions of premixed and nonpremixed terms to the total progress variable source term budget. These authors further acknowledge that, by focusing solely on the fuel, the classical formulation has limitations when complex kinetic mechanisms are employed. In particular the combustion of long hydrocarbons proceeds through smaller components, which are the true fuel species undergoing oxidation through the flame. This is indeed what happens in the present case, where  $C_2H_4$  may dissociate into  $C_2H_2$  and CO (see Section 6.1.2) without involving  $O_2$  directly, and the secondary reaction zones effectively turn to be CO oxidation fronts. In order to characterize all flames, two TIs are therefore employed, based either on  $C_2H_4$  (TI<sub>C2H4</sub>) or CO (TI<sub>CO</sub>) as the fuel in Eq. 8. Note that both TIs are conditioned on the source term of the considered fuel, so that TI = 0 in non-reacting regions.

With both ARC and FPI, the  $TI_{C2H4}$  shown in Fig. 13 (a) reveals that the PZ exhibits a premixed flame in the wake of the swirled jets.  $C_2H_4$  diffusion flames are not observed, indicating that this fuel has sufficient time to mix with air before burning. The premixed flame based FPI table is thus expected to accurately describe the flame structure in this region. On the contrary, the  $TI_{CO}$  displayed in Fig. 13 (b)



Figure 13: Takeno Index computed based on either (a)  $C_2H_4$  or (b) CO as a fuel, with overlaid grey stoichiometric iso-contour and  $Y_Z$  iso-volume. Top: ARC-LES, bottom: FPI-LES. The red frame identifies the DZ center (see Section 6.3.3).

shows mostly non-premixed combustion, which occurs in some peripheral regions of the premixed ethylene flame in the PZ, and in the secondary reaction zones extending downstream to the DZ. Note that these secondary combustion fronts are aligned with the  $C_2H_4$ -based, Bilger formulation of the stoichiometric isocontour. This could be surprising as  $C_2H_4$  is not any more present in these zones. It can be demonstrated however, as developed in A, that the local equivalence ratio of any intermediate species I can be expressed as a function of the fuel-based, Bilger mixture fraction and that its stoichiometry then coincides with fuel stoichiometry. As a consequence, the alignment of the secondary reaction zones with the ethylene-based stoichiometric line indicates a true CO diffusion flame structure, whereas the non-premixed reacting zones around the PZ premixed flame correspond to post-combustion, falsely identified as diffusion flames by  $TI_{CO}$ due to aligned species gradients resulting from CO production in this region.

Again note that the FPI-LES flame appears much fragmented but CO oxidation follows the same trends even in isolated flame fragments. A main difference with the ARC-LES, however, appears in the region where the IRZ mixes with the dilution jets, and labelled hereafter as the DZ center (red frame in Fig. 13): a few isolated structures exhibiting relatively high heat release rate and already mentioned in Section 6.1.1 are identified there, and will be discussed in Section 6.3.3.

# 6.3.2. Primary Zone (PZ)

As found in Section 6.3.1, the PZ exhibits a detached, premixed ethylene-air flame front, with however strong mixture fraction non-homogeneities. As was previously observed, the mixing prior to combustion differs between the ARC-LES and the FPI-LES, resulting in a different distribution of equivalence ratio in the PZ. The ARC-LES gives a nearly Gaussian distribution in the reacting zone (i.e., source term of fuel < -10 kg.m<sup>-3</sup>.s<sup>-1</sup>), with a most probable value around  $\phi = 1.2$  corresponding to the global equivalence ratio of the PZ (without the dilution jets, see Table 3). In contrast, the FPI-LES distribution exhibits a bi-modal shape with peaks around  $\phi = 0.8$  and  $\phi = 1.6$ . Additionally, the distribution peaks at the global equivalence ratio, and then rapidly goes back to the bi-modal rich and lean burning. As a result, nonnegligible differences in the levels of species produced in the main PZ flame can be seen, and stratification effects may be important. The transport of small radical species with very high diffusivities -such as H or H<sub>2</sub>- in the ARC-LES further enhances the mixing discrepancies between both simulations. Indeed, a direct consequence is a smoothing of the mixture fraction field, also leading to a less wrinkled and slightly thicker flame front.

As both LES use the DTFLES model, the flame is similarly wrinkled and stretched, as seen on the left column of Fig. 14, investigating correlations between the stretch, the tangential strain rate  $a_T$  and the



Figure 14: Scatterplots of stretch versus strain rate  $a_T$  and  $S_d^*$  versus curvature  $\kappa$ , for (a) ARC-LES and (b) FPI-LES. Points are selected at  $\phi = 1.2 \pm 0.02$  and  $c = 0.5 \pm 0.02$ 

curvature  $\kappa$  in the flame front. The curvature  $\kappa = \nabla \cdot n$  is defined as the divergence of the flame surface normal  $n = -\nabla c/|\nabla c|$ , and the flame stretch is defined as the sum of the strain rate (i.e., velocity gradient along the flame front)  $a_T$  and curvature  $S_d \kappa$ , where  $S_d$  is the flame propagation speed defined as in [64]. Scatterplots are constructed from locations where  $\phi = 1.2 \pm 0.02$  and  $c = 0.5 \pm 0.02$ . The same range of strain rates  $a_T$  is observed for both LES, with positive values more likely to occur. Curvature effects are mostly associated with negative values which have a tendency to decrease the stretch rate. It seems that the FPI flame experiences slightly more curvature effects due to its more fragmented shape. The impact of curvature on the density-weighted flame displacement speed  $S_d^* = \rho S_d/\rho_f$  is shown on the right column of Fig. 14.  $S_d^*$  is negatively correlated with curvature, consistently with previous DNS studies [65, 66]. The slope of the correlation is slightly more pronounced in the ARC-LES, because of the different diffusion velocities. It is noted also that the correlation exhibits a slightly nonlinear shape for very negative curvatures in the ARC-LES, a trend not seen in the FPI-LES nor in previous studies employing global chemistries, and attributed to the diffusion of small radicals in the tangential direction. In the end, local flame discrepancies between both LES in the PZ are found to be mostly attributed to stretched induced differential diffusion effects, which are included in the ARC-LES.

# 6.3.3. Dilution Zone (DZ)

As previously mentioned, reaction fronts are detected in the DZ along the stoichiometric line, in both ARC-LES and FPI-LES (Fig. 5 (a)). In Section 6.3.1, the  $TI_{CO}$  identified them as being CO diffusion fronts (Fig. 13 (b)). To better characterize these CO burning zones in the ARC-LES, scatterplots of CO mass fraction and temperature are presented on Fig. 15 (a) and (b), coloured by the  $TI_{CO}$  and the heat release rate, respectively. As a reference, one-dimensional strained diffusion flames between a hot, rich mixture of burnt gases ( $Y_Z = 0.083$ ) and fresh air at 300 K were computed with CANTERA and the ARC mechanism, for three representative strain rates (50  $s^{-1}$ , 1200  $s^{-1}$  and 4600  $s^{-1}$ ). Profiles are superimposed to the scatterplots of the ARC-LES (green dotted lines), showing the same structure and encompassing all data points. Note that consistently with diffusion flame theory, the highest strain rate points exhibit the highest heat release rate (Fig. 15 (b)). The scatter of points in the ARC-LES thus reflects the impact of turbulent strain rate on the diffusion fronts. The FPI-LES, however, was not designed to account for such diffusion



Figure 15: Scatterplots of CO mass fraction colored by  $TI_{CO}$  and of temperature colored by the heat release rate. From the ARC-LES computation. Scatterplots are superimposed with laminar non-premixed strained CO flames in green (see text), of increasing strain rate (a).

structures. In fact, the  $TI_{CO}$  identifies premixed post-flame trajectories, where CO oxidation reactions take place, as being CO diffusion flames.



Figure 16: Instantaneous views in the plane of the dilution holes of (a) the heat release rate and (b) the CO source term. The grey iso-contour marks the stoichiometry, the white iso-contour marks the lower flammability limit, while red iso-contours delimit zones of c smaller than 0.95. Top: ARC-LES, bottom: FPI-LES.

To confirm this analysis, Fig. 16 displays a close view of the DZ center (red frame in Fig. 13). The heat release rate, and source term of CO are reported for both LES. Consistently with previous observations made about the ARC mechanism, strong CO non-premixed flames are predicted along the dilution jets (Fig. 16 (a) top), where fresh air comes into contact with the CO issued from the PZ and convected downstream along the walls, and where strain is at its maximum. In the FPI-LES, the picture is quite different, and in particular, no reacting fronts are seen around the dilution jets. Two different types of reacting regions can be distinguished, highlighted in Fig. 16 (a) (bottom). First, (B) structures closely follow the stoichiometric iso-contours. Note that these structures are not only seen in the DZ center, but also following the IRZ everywhere in the DZ, where they appeared similar to those seen in the ARC-LES (Fig. 13 (b)). (A) structures, on the other hand, are isolated strong reacting fronts that closely follow the white lower flammability limit iso-contours. They are only present in the DZ center of the FPI-LES. Remember that an inspection of the TIs in Section 6.3.1, revealed (A) type structures as being premixed-like CO flames coinciding with weak premixed-like C<sub>2</sub>H<sub>4</sub> structures (red frame in Fig. 13).

Typical examples of (A) and (B) trajectories are reported in the 2D look-up table, in Fig. 17. (A) trajectories, representative of the isolated flames, start from low c and  $Y_Z$  in the center region, due to the dilution by air. The trajectories then progress along increasing c, but stay at very low  $Y_Z$ . In fact, at low c,  $Y_Z$  belongs to the non-flammability region, so that burning is not allowed and the table returns traces of

 $C_2H_4$ . Once the flammability limit is reached ( $Y_Z = 0.028$ ), c is already high and post-flame combustion occurs, which corresponds to the isolated flames seen in the 3D simulation. This is both an artifact of the canonical configuration chosen to build the FPI table, requiring to clearly define *a-priori* the flammability limits, as well as an inherent issue of tabulation techniques, lacking the ability to adapt to other flame structures than those tabulated.



Figure 17: 2D look-up FPI table, couloured by heat release rate, with close-up (green frame). For the signification of the trajectories, see the text.

Likewise, (B) type trajectories, representative of the reacting structures following the stoichiometric iso-contours, follow trajectories in the post-flame region of the premixed-based 2D look-up table (Fig. 17). These trajectories are characterized by relatively high c (> 0.75) values, characteristic of the DZ which is rich in CO and CO<sub>2</sub>. As a consequence, the DZ reaction fronts found in the FPI-LES ((A) and (B) types) are not an entirely proper representations of reality; and in particular, species production, consumption and heat release rate predicted by FPI in this area should be regarded with caution.

# 6.4. Investigation of soot production

Figure 18 presents quantitative comparisons of time averaged fields of soot obtained in LES and experiments. Qualitatively, it is seen that soot is mostly present in the PZ. More precisely, in both operating points, high levels are found in the upstream part of the IRZ, right downstream of the post-flame of the main combustion zone. Secondary oxidation then occurs, preventing any soot from surviving near the centerline in the DZ [67]. Overall, the shape of the high soot load region is correctly predicted with LES, and is only slightly shifted towards the injection compared to experiments. This could be an artifact of the simple soot model employed in this study, where acetylene is the sole soot precursor, not necessarily produced or localized in the same flame zones than heavier PAH. Another possible explanation is suggested by the spatial discrepancies observed in the OH instantaneous and mean fields between experiment and LES, Section 6.2.2. Indeed, the upstream part of the IRZ, in the post-flame region of the PZ main flame, is characterized by a drop of OH in the experiment, which is propitious to the formation of soot. This region of OH drop is not as extended in the LES, suggesting different species distribution (not enough acetylene, too much  $O_2$ ). Note finally that on average soot is absent from the DZ in the LES of Case A, but this might simply be due to a too short averaging time, as very small levels are observed in the experiment.

Quantitatively, the FPI-LES levels seen on Fig. 18 (multiplied by 10 in the figure) are off by about one order of magnitude, while the ARC-LES recovers levels of the same order of magnitude as experiments, for both operating points. In particular the higher levels of Case B are very well predicted with ARC. Despite expected discrepancies in the main PZ flame structure where  $C_2H_2$  is exclusively formed -as discussed at



Figure 18: Comparison of time-averaged LII soot measurements with time-averaged soot mass fraction obtained: left and center: Case A with ARC-LES and FPI-LES; right: Case B with ARC-LES. Note that for comparison, soot levels in the FPI-LES have been multiplied by 10.

length in Section 6.3.2, such an important difference on soot load between ARC and FPI is surprising, since both are based upon the same detailed mechanism. This result is however consistent with preliminary work performed on the same configuration by [12]. The net source term of the soot mass fraction  $Y_s$ , plotted against temperature in Fig. 19, and colored by  $C_2H_2$  mass fraction, confirms a net soot production about twice smaller in the FPI-LES than in the ARC-LES in Case A, and clearly shows the strong correlation with the soot precursor. In particular note the absence of  $C_2H_2$  above  $T \approx 2100$ K, and the subsequent absence of soot in this high temperature zone with FPI-LES.



Figure 19: Scatterplots of soot mass fraction  $(Y_s)$  net source term for ARC-LES (Cases A and B) and FPI-LES (Case A), coloured by  $C_2H_2$  mass fraction.

To identify the dominant process in the production/consumption of soot, each of the terms composing  $\dot{\omega}_{Ys}$  (see Section 3) are analyzed separately. Remember that soot production is the result of nucleation and surface growth, both linearly dependent upon the levels of acetylene, and that oxidation by  $O_2$  is the only consumption source. Instantaneous fields of each of these contributions is provided in Fig. 20, for both LES of Case A. Accordingly with the simple soot model, both nucleation and surface growth are seen to be localized in the PZ flame peripheral area where acetylene levels are important and the temperature sufficiently high, while oxidation occurs around the outer limit of the IRZ. Oxidation levels are comparable in the DZ in all LES, because of fairly similar O<sub>2</sub> fields. The dominant term throughout the temperature range is found to be production by surface growth in both LES, confirming the key role of acetylene. However Fig. 20 shows strong differences in the distribution of the production source term between both LES, which is directly proportional to the distribution of acetylene already discussed in Section 6.1.2 (see Fig. 6): in the FPI-LES,  $C_2H_2$  (grey iso-contours) cannot be observed in regions of high temperature where the FPI table always returns negligible levels of this species. It is stressed that this would also be the case had the two dimensional FPI table been constructed from premixed flames computed with complex transport (see unstretched laminar profiles on Fig. 22). As a consequence, nucleation and surface growth only occur in regions of intermediate temperatures, in the flame core or following burning pockets, where the FPI table returns sufficient levels of acetylene, and exhibit smaller levels. On the other hand, the ARC-LES allows the presence of acetylene in high temperature regions, thanks to differential diffusion and other transport effects, enabling soot production to be boosted by the high temperature and to reach higher levels.



Figure 20: Instantaneous fields of nucleation, surface growth and oxidation with superimposed grey iso-contours of  $C_2H_2$  for Case A with with (a) ARC-LES and (b) FPI-LES.

Another major difference between ARC-LES and FPI-LES stems from the fact that acetylene levels are significantly smaller in the FPI-LES. Fig. 21 indeed shows average and RMS acetylene mass fraction values about twice smaller with FPI. In the case of ARC-LES, points with  $Y_{C2H2} > 10^{-2}$  are frequently observed in instantaneous solutions. Remember that  $C_2H_2$  is produced exclusively in the PZ premixed flame (see Section 6.1.2), so that FPI table values are expected to be correct, although slightly lower than that found in the ARC-LES under the same conditions, due to the unity Lewis assumption employed to compute the flames. However, it was discussed in Section 6.3.2 that the local equivalence ratio peaks at 0.8 and 1.6 in this case. As a consequence, looking at Fig. 1 (f), one would expect higher acetylene maximums in the FPI-LES than in the ARC-LES, where the mean equivalence ratio is 1.2. As the FPI table was built from unstretched premixed flames, strain and curvature effects could be invoked as possible reasons for the higher acetylene values observed in the ARC-LES. However, as illustrated on Fig. 22 for a laminar flame at  $\phi = 1.2$ , these effects appear too small to explain the LES results. Note also from Fig. 22 that if the effect of unity Lewis numbers assumption on  $C_2H_2$  levels is non negligible (especially for maximum levels), it is smaller than the observed deviations from reference flames.



Figure 21: Comparison of time-averaged C<sub>2</sub>H<sub>2</sub> mass fraction (left part) and RMS (right part), for each computation.

Another potentially non-negligible contribution to the discrepancies in predicted acetylene levels arises

from partial premixing in the PZ and the subsequent highly non-homogeneous distribution of equivalence ratio. When a flame propagates in such a stratified mixture, its inner structure is altered, possibly leading to a modification of the turbulent flame speed, pollutant production, etc. To assess the importance of this effect in the present configuration, two quantities are employed. First, the cross-scalar dissipation rate,  $\chi_{zc}$ , defined as:

$$\chi_{zc} = D_z |\nabla Y_z| |\nabla c| \tag{9}$$

giving an estimation of the local degree of simultaneous c and  $Y_z$  evolutions across the flame, and already employed, for instance, by Domingo *et al.* [68] to analyze both gaseous and spray turbulent-lifted flame structures in DNS. Second, the cross-scalar angle,  $\alpha_u$ :

$$\alpha_u = \cos^{-1} \left( \frac{\nabla Y_z \cdot \nabla c}{|\nabla Y_z| |\nabla c|} \right) \tag{10}$$

is characterizing the degree of mixture stratification. Whenever  $\alpha_u = 90^\circ$ , reaction progresses along an iso-Y<sub>z</sub> level and a perfectly premixed flame structure is expected. On the contrary,  $\alpha_u = 180^\circ$  means that the flame propagates towards rich mixtures, and  $\alpha_u = 0^\circ$  towards leaner mixtures. Stratification effects are therefore expected whenever  $\chi_{zc} >> 0$  and  $\alpha_u \neq 90^\circ$ .



Figure 22: Scatterplots of C<sub>2</sub>H<sub>2</sub> mass fraction in the ARC-LES (Case A), (a) coloured by  $\chi_{ZC}$  and (b) coloured by  $\alpha_u$ . Points are chosen in the range  $\phi = 1.2 \pm 0.01$ . Superimposed are stretched and unstretched 1D laminar profiles at  $\phi = 1.2$  computed with the ARC\_18\_C2H4NARA mechanism. Scales for  $\chi_{ZC}$  and  $\alpha_u$  are resp. from 0 to 100 s<sup>-1</sup> and from 0 to 180°.

Figures 22 (a) and (b) display  $C_2H_2$  scatterplots constructed from points with  $\phi = 1.2 \pm 0.01$  (ARC-LES of Case A), coloured by  $\chi_{zc}$  and  $\alpha_u$ , respectively. One-dimensional laminar profiles are superimposed as a reference, and the conditional mean is also reported. High acetylene levels are mostly identified as being points with  $\alpha_u = 180^\circ$ , also characterized by moderate to high  $\chi_{zc}$  values. This proves that stratification effects are also responsible, to a non negligible extent, for the higher acetylene levels observed in the ARC-LES, and in turn for the higher predicted soot levels.

# 7. Conclusions

LES of a swirled non-premixed sooting flame have been performed with two different chemistry models, based either on a tabulated (FPI) or on an analytically reduced (ARC) approach. If both chemistry models, combined with the DTFLES turbulent combustion model, give very satisfactory and similar results for the overall flame features, important discrepancies appear when it comes to the detailed flame structure and pollutant emissions. By construction, the FPI table cannot correctly reproduce complex effects such as hot gas re-burning after dilution, differential diffusion or equivalence ratio stratification. This leads in the present case to an inaccurate flame structure after dilution by fresh air and to a strong under-prediction of the sole soot precursor, leading in turn to an under-estimation of soot concentration. Of course, it would be possible to improve these results by resorting to a more sophisticated tabulation technique, particularly by incorporating differential diffusion effects, but it is not always straightforward to identify which phenomena will impact the simulation to a leading order. On the contrary, the ARC-LES results put forward the flexibility and capability of the ARC-DTFLES methodology to account for the diversity of flame regimes encountered in realistic burners; resulting, in the present case, in an accurate prediction of the complex flame structure and pollutant emissions without the need to formulate *a-priori* simplifying assumptions about the nature of the flow. Indeed, the chosen approach relies on the direct integration of the chemistry, thus allowing a natural adaptation to the problem at hands.

	Case A - ARC	Case A - FPI	Case B - ARC
Navier-Stokes	5	5	5
Transported species	18	2	18
Soot model	2	2	2
Total transported variables	<b>25</b>	9	<b>25</b>
CPUh for 1 ms	12500	6200	12500

Table 4: Summary of computational requirements for all LES.

More generally, the accuracy of the ARC-DTFLES approach is clearly dependent upon the accuracy of the ARC mechanism, which should contain a minimum number of species and reactions to minimize the CPU cost. Additionally, the control of chemical stiffness is critical to keep a reasonable time step for the explicit integration of the equations. A summary of CPU requirements for each LES presented in this paper is given in table 4, using Intel Haswell nodes (E5-2680v3) cadenced at 2.5 GHz. It is found that the CPU cost of ARC-LES is only twice that of FPI-LES, while the number of variables to integrate is multiplied by  $\approx 3$ . This leads to two important remarks: first, the CPU cost of ARC-LES is proportional to the number of transported quantities, taking into account the fact that, in the present simulation, no particular efforts were made to optimize the handling of the two-dimensional table; second, the direct integration of chemical source terms does not add any CPU cost. This means that the ARC-DTFLES methodology will be  $\approx 2$  to 4 times more costly in CPU time compared to a LES using global or tabulated chemistry, which is considered by the authors a small price considering the increase of accuracy and predictability.

The ARC-DTFLES approach appears therefore to have a very strong potential to increase the accuracy of turbulent combustion predictions in practical systems. With a new description of the involved chemistry, it allows to revisit many combustion problems such as ignition, pollutant emissions or thermo-acoustic instabilities. In particular it is of great interest to evaluate the impact of fuel composition on these phenomena in the current context of development of alternative fuels containing variable proportions of synthetic or organic oils.

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# Appendices

# A. Stoichiometry of intermediate species

#### CO equivalence ratio in ethylene flame

CO is considered as an example. The global ethylene oxidation reaction reads:

$$C_2H_4 + 3 O_2 \rightarrow 2 CO_2 + 2 H_2O$$
 (11)

with the mass stoichiometric factor  $s^{C2H4} = 3 W_{O2} / W_{C2H4} = 3.42$ . A two-step mechanism, including CO oxidation, reads:

$$C_2H_4 + 2 O_2 \rightarrow 2 CO + 2 H_2O$$
 (12)

$$CO + \frac{1}{2} O_2 \to CO_2 \tag{13}$$

where, in reaction 13, the CO mass stoichiometric factor is  $s^{CO} = 0.5 \text{ W}_{O2} / \text{ W}_{CO} = 0.57$ .

The local equivalence ratio  $\Phi$  of a mixture of pure ethylene ( $Y_{C2H4}^0 = 1$ ) and air ( $Y_{O2}^0 = 0.233$ ), can be expressed in function of a fuel based mixture fraction  $Y_z$  as:

$$\Phi = s^{C2H4} \frac{Y^m_{C2H4}}{Y^m_{O2}} = s^{C2H4} \frac{Y^0_{C2H4}}{Y^0_{O2}} \frac{Y_z}{1 - Y_z}$$
(14)

where  $Y_{C2H4}^m$  and  $Y_{O2}^m$  are the mass fractions of fuel and oxidizer if they would have mixed without burning, and therefore correspond to a *pure mixing solution* [69]. With this definition, the  $Y_z$  at stoichiometry (corresponding to  $\Phi = 1$ ) is  $Y_{z,st} = 0.0637$ .

Likewise, the equivalence ratio  $\Phi_{CO}$  can be expressed in function of a theoretical pure mixing solution, involving the available CO and  $O_2$ :

$$\Phi_{CO} = s^{CO} \frac{Y_{CO}^m}{Y_{O2}^m} \tag{15}$$

However, since the mixture fraction  $Y_z$  is based on ethylene, an expression for the mixing of CO with  $O_2$  depending on it is not straightforward. Indeed, according to the global reaction Eq. 12, the transformation of  $C_2H_4$  into CO requires  $O_2$ . Assuming an initial mixture composed of A moles of  $C_2H_4$ , B moles of  $O_2$  and C moles of  $N_2$ , by mass conservation, a mixture composed of 2A moles of CO, 2A moles of  $H_2O$ , (B-2A) moles of  $O_2$  and C moles of  $N_2$  is obtained. Since A and B are functions of  $Y_z$ , with  $A = Y_{C2H4}^0 Y_z$  and  $B = Y_{O2}^0(1-Y_z)$ , an expression for 2A and (B-2A) required to express the evolution of  $Y_{CO}^m$  and  $Y_{O2}^m$  in the  $Y_z$  diagram is now straightforward:

$$\Phi_{CO} = s^{CO} \frac{2 Y_z Y_{C2H4}^0 W_{CO} / W_{C2H4}}{(1 - Y_z) Y_{O2}^0 - 2 Y_z Y_{C2H4}^0 W_{O2} / W_{C2H4}}$$
(16)

or more conveniently:

$$\Phi_{CO} = s^{CO} \frac{Y_z Y_{CO}^0}{(1 - Y_z) Y_{O2,r}^0}$$
(17)

with  $Y_{CO}^0 = 2 Y_{C2H4}^0 W_{CO}/W_{C2H4}$  and  $Y_{O2,r}^0 = Y_{O2}^0 - 2 Y_z Y_{C2H4}^0 W_{O2}/(W_{C2H4} (1 - Y_z))$ . Replacing  $Y_z$  in Eq. 17 by

$$Y_z = \frac{\Phi}{(\Phi + s^{C2H4} Y^0_{C2H4} / Y^0_{O2})}$$
(18)

eventually reduces to:

$$\Phi_{CO} = \frac{\Phi}{(3-2\Phi)} \tag{19}$$

Now, obviously, if  $\Phi = 1$  in Eq. 19, then  $\Phi_{CO} = 1$ , and vice versa. It is thus demonstrated that CO diffusion fronts and C<sub>2</sub>H<sub>4</sub> diffusion fronts will gather around the same Y<sub>z</sub> value: Y<sub>z,st</sub> = 0.0637.

#### Generalization to any intermediates

Note that this derivation can be generalized to any intermediate species I (and virtually any fuel species but we will only consider ethylene): as long as the fuel global consumption Eq. 11 is decomposed as:

$$C_2H_4 + \alpha \ O_2 \to \beta \ I + y \ H_2O \tag{20}$$

$$I + \gamma O_2 \to x' C O_2 + y' H_2 O \tag{21}$$

The rescaled local equivalence ratio reads:

$$\Phi_I = s^I \frac{Y_z Y_I^0}{(1 - Y_z) Y_{O2,r}^0}$$
(22)

with:

$$s^{I} = \gamma \frac{W_{O2}}{W_{I}} \tag{23}$$

$$Y_I^0 = \beta \ Y_{C2H4}^0 \ \frac{W_I}{W_{C2H4}} \tag{24}$$

$$Y_{O2,0}^{0} = Y_{O2,0} - Y_z Y_{C2H4}^{0} \frac{\alpha}{(1 - Y_z)} \frac{W_{O2}}{W_{C2H4}}$$
(25)

As in the case of CO, replacing for  $Y_z$  in Eq. 22 and setting  $\Phi = 1$  leads to:

$$\Phi_I = \frac{\gamma\beta}{3-\alpha} = 1 \tag{26}$$

since  $\gamma\beta + \alpha = 3$ . As such, diffusion fronts of any intermediate species I will also gather around  $Y_{z,st} = 0.0637$ .

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# Supplementary Materials: Additional chemistry validations

# P = 3 bar

# Unstrained Premixed Flames (UPF)

Spatial profiles of major species through the stoichiometric ethylene/air UPF are presented in Fig. S1. Note the under-prediction of peak levels of  $C_2H_2$  observed with FPI.



Figure S1: Profiles of a) CO, b) OH, and c)  $C_2H_2$  across a UPF. Fresh gas temperature is 300 K, P = 3 bar and  $\phi = 1$ . Detailed mechanism - Mix (solid line), ARC 18 C2H4NARA (o), Detailed mechanism - Le=1 ( $\Delta$ ), FPI (x).

Strained Diffusion Flames (SDF)



Figure S2: Structure of a SDF at  $a = 50 \ s^{-1}$ . Fresh gas temperature is 300 K, P = 3 bar. Phase space evolutions of a) CO, b) OH, and c) C<sub>2</sub>H<sub>2</sub> mass fractions. Detailed mechanism - Mix (solid line), ARC\_18\_C2H4NARA (o), Detailed mechanism - Le=1 ( $\Delta$ ), FPI (+).

A SDF structure in phase space for a small strain rate (a = 50 s<sup>-1</sup>) is provided in Fig. S2, illustrating the discussed discrepancies between detailed chemistry, ARC and FPI in Section 2.3.2. The poor agreement observed between detailed chemistry and FPI, in particular, is consistent with previous observations of Fiorina *et al.* [15] in similar cases. These authors concluded that a UPF-based FPI table, when applied to a diffusion flame, was only valid in a small inner reaction zone around stoichiometry, where diffusive processes are dominated by chemistry. Outside of this zone, they reported large differences in CO and CO<sub>2</sub> mass fractions, consistent with the over-predictions observed here (Fig. S2 (a)). This also holds for C<sub>2</sub>H<sub>2</sub> (Fig. S2 (c)) which is under-predicted over the entire mixture fraction range. OH is consistently better recovered (Fig. S2 (b)), as it is present only inside the reaction zone near stoichiometry. Note that the linear interpolation outside of the flammability range  $(0.03 < Y_z < 0.17)$ , also visible from Fig. S2 (a) & (c), is another source of error. These FPI results could be improved by extending the table to include non-premixed canonical flames, as proposed by [17, 70] for example.

Note, finally, that the unity Lewis assumption impact on the results is mitigated in these SDF cases.

# P = 5 bar

#### Unstrained Premixed Flames (UPF)

Results are here presented only for the detailed mechanism computed with complex transport, and the ARC\_18\_C2H4NARA mechanism, as the same conclusion as in the 3 bar case holds for unity Lewis numbers. Burnt gas temperature as well as laminar flame speed are plotted versus equivalence ratio, respectively, in Fig. S3 (a)-(b). The laminar flame speed is consistently lower than in the P = 3 bar case (Fig. 1). The agreement is excellent between both mechanisms, for the entire range considered. With the ARC mechanism, the relative error on  $s_l$  never exceeds 5%, while the maximum relative error on the burnt gas temperature is below 1%.



Figure S3: a) Burnt gas temperature b) laminar flame speed c) total CO production and maximum of d) CO e) OH and f) C2H2 reached in UPF. Fresh gas temperature is 300 K, P = 5 bar. Detailed mechanism - Mix (solid line), ARC\_18\_C2H4NARA (o).

CO total production is plotted on Fig. S3 (c). The evolution with the mixture fraction is comparable to that of the case P = 3 bar, but levels are about 35% higher. The accuracy of the ARC mechanism is similar to what was discussed in the case P = 3 bar.

Maximums of species of interest are plotted against mixture fraction, in Fig. S3 (d)-(f) for the two chemical schemes. The curves are almost indistinguishable, except for the maximum of  $C_2H_2$  for which the ARC mechanism exhibit about 10% of relative error over the entire equivalence ratio range. Figure S4 displays the evolution of the species of interest across a stoichiometric flame front. As expected, it can be seen that the flame is thinner than in the P = 3 bar case. Despite the small offset on the predicted peak of  $C_2H_2$  that is observed on Fig. S4 (c), results with the ARC scheme are considered excellent. It is noted, here also, that both detailed and ARC mechanisms eventually converge towards the same burnt gas state.



Figure S4: Profiles of a) CO b) OH and c) C2H2 across a UPF. Initial temperature is 300 K, P = 5 bar and  $\phi = 1$ . Detailed mechanism - Mix (solid line), ARC\_18\_C2H4NARA (o).

Strained Diffusion Flames (SDF)



Figure S5: a) Maximum temperature b) total  $C_2H_4$  and c) CO production and maximum of d) CO e) OH and f) C2H2 reached in a series of SDF. Fresh gas temperature is 300 K, P = 5 bar. Detailed mechanism - Mix (solid line), ARC\_18\_C2H4NARA (o).

Similar to the P = 3 bar case, the response to strain of the ARC mechanism, presented in Fig. S5, is very good. The predicted extinction strain rate is  $a_{ext} \approx 3260 \ s^{-1}$  and  $a_{ext} \approx 3190 \ s^{-1}$  with the detailed and ARC mechanisms, respectively. Those values are significantly higher than for the P = 3 bar case. Here also, the level of C<sub>2</sub>H<sub>2</sub> presents the highest offset, with very similar levels of error than what was found in the P = 3 bar case. This error in peak values of C<sub>2</sub>H<sub>2</sub> is also seen on the profiles of Fig. S6 (c), (d) & (f), featuring the structure of a diffusion flame for a very high strain rate ( $a = 2600 \ s^{-1}$ ), in both physical and phase space. CO and OH evolutions are remarkably well predicted.

Those results emphasize once more the main asset of ARC, which is that the derived mechanisms remain accurate outside of their strictly specified targeted operating range.



Figure S6: Structure of a SDF with  $a = 2600 \ s^{-1}$ . Fresh gas temperature is 300 K, P = 5 bar. Spatial evolutions of a) CO b) OH and c) C<sub>2</sub>H<sub>2</sub> as well as evolutions in the phase space of c) CO d) OH and e) C<sub>2</sub>H<sub>2</sub> mass fraction. Detailed mechanism - Mix (solid line), ARC\_18\_C2H4NARA (o).