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Deriving Cool Flame Propagation Speeds by Means of an Ozone-Seeded, Stagnation Plate Burner Configuration

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Full length article

Abstract

This study aims to investigate the feasibility of experimental determination of DME/O₂/O₃ cool flame propagation speeds using Particle Image Velocimetry (PIV) in a stagnation plate burner operated at atmospheric pressure. A specific PIV data analysis procedure was developed in order to improve the accuracy of the measurements in this particular configuration. Five flame conditions, with equivalence ratio varying from 0.3 to 0.5 and ozone mole fraction varying from 1.5 to 2% were investigated to compare experimental results with kinetic modeling. Three ozone-submechanisms, respectively from Jian et al. (Jian et al., 2022), Halter et al. (Halter et al., 2011) and Zhao et al. (Zhao et al., 2016), were coupled with our previously developed DME mechanism (Panaget et al., 2021) and used to compare experimental and simulated axial velocity profiles. Results show that a thoughtful choice of the ozone-submechanism is of particular importance in predicting an accurate cool flame velocity in these conditions. A numerically assisted non-linear extrapolation method is proposed for the determination of the unstrained cool flame speed $S_{u,0}$. Additionally, simulations for which the plate temperature reaches the maximal flame temperature (adiabatic conditions) were performed, demonstrating a negligible effect of the plate temperature on the determined $S_{u,0}$. A kinetic analysis is also presented to highlight the most sensitive chemical reactions influencing the reference cool flame speed $S_{u,ref}$, showing the preponderant role of the fuel low temperature chemistry.

Keywords: *Cool flame, Propagation speed, Low temperature combustion, Particle Image Velocimetry, Ozone assisted combustion*

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1. INTRODUCTION

From their discovery two centuries ago [1] to their recent observation under micro-gravity conditions [2], cool flames raised a number of issues throughout the years [3,4]. They are known to play a critical role in advanced combustion technologies [5] as they control both autoignition and assisted-ignition processes in the low-temperature combustion regime. Flame propagation speeds are fundamental parameters in combustion due to their influence on the rate of heat release within the reaction zone of flames. Cool flame speeds were numerically investigated by Ju et al. [6,7] and by Zhao et al. [8]. Using dimethyl ether (DME)/O₂ mixtures in both freely-propagating and counter-flow conditions, it was reported that cool flame speeds typically range from 6 to 20 cm.s⁻¹, depending on the experimental conditions. Interestingly, they are almost insensitive to the equivalence ratio within their stability range, while their hot flames counterparts are known to be highly dependent on this parameter. Numerical investigations of the effect of ozone on the cool flame speed have shown that it increases as the ozone concentration increases, as in the case of hot flames [9,10].

To the best of our knowledge, experimental cool flame propagation speeds were only reported under microgravity conditions [11] or at sub-atmospheric pressure by the Belmont group [12–16]. Foster and Pearlman [11] measured the speed of a propane/O₂ cool flame at low-pressure and microgravity conditions, using the spherically-propagating flame method. It was however observed that heat was released by pre-ignition of the mixture, prior to the cool flame formation. Thus, the measurement could not be used to determine the propagation speed of the propane cool flame. Recent work from Belmont group at the University of Wyoming [12–16] showed the feasibility of measuring cool flame speeds using a Hencken Burner operated at sub-atmospheric pressure. They reported low pressure data for ozone-seeded cool flames of dimethyl ether, propane and *n*-heptane. This method consists of varying the gas inlet flow rate while keeping the equivalence ratio fixed, and measuring the position of the flame

78 above the burner by excited formaldehyde chemiluminescence or formaldehyde planar laser
79 induced fluorescence (PLIF). Two distinct regimes are then identified: a/ a first regime where
80 the flame remains anchored to the burner by the heat losses, and where the variation of the inlet
81 flow rate causes only a minimal variation of the flame position; b/ a second regime, where the
82 flame detaches from the burner surface and is considered freely-propagating and nearly
83 adiabatic, the heat transfer being considered negligible. In this case, the flame position varies
84 linearly with the inlet flow rate. The transition between these two regimes was suggested to
85 provide a reasonable estimate of the cool flame speed. This method was validated for hot flames
86 by comparing flame speeds measured by Particle Image Velocimetry (PIV) [16,17] and data
87 from the literature and simulations. The cool flame speeds obtained experimentally by Hajilou
88 et al. were simulated with different kinetic models [13-15] using freely-propagating flame
89 simulations.

90 Ozone has been widely used in experimental cool flames studies [15,18] as its thermal
91 decomposition releases highly reactive oxygen atoms at temperatures close to 450 K at the
92 atmospheric pressure, facilitating the initiation of low-temperature reactivity [19]. Several
93 ozone specific submechanisms have been developed in the past years, and are commonly
94 coupled with hydrocarbon mechanisms to describe the decomposition of ozone and its impact
95 on combustion [20–22]. Knowing the important influence of ozone on the combustion process,
96 it is important to wisely choose the ozone submechanism when simulating the behaviour of
97 ozone-seeded cool flames.

98 In this work, we investigated the feasibility of measuring atmospheric pressure lean
99 DME/O₂/O₃ cool flames propagation speeds from the measurements of the axial velocity
100 profiles using PIV in a newly implemented stagnation plate burner [23]. A specific PIV data
101 analysis procedure, developed in order to improve the accuracy of the measurements, is
102 presented. Five different flames conditions have been studied with equivalence ratio varying

103 from 0.3 to 0.5 and O₃ mole fraction from 1.5 to 2%. Comparison with simulations is performed
104 using three different O₃-submechanisms from the literature [20–22] coupled to a detailed kinetic
105 mechanism recently developed in our group [23] for DME low temperature combustion. The
106 performance of the coupled mechanisms is first assessed in predicting experimental axial
107 velocity profiles in the strained cool flames, then a method to extract the unstrained propagation
108 speed is proposed. Finally, a kinetic analysis is performed to highlight the most sensitive
109 chemical reactions influencing the cool flame speed.

110

111 **2. EXPERIMENTAL AND NUMERICAL METHODS**

112

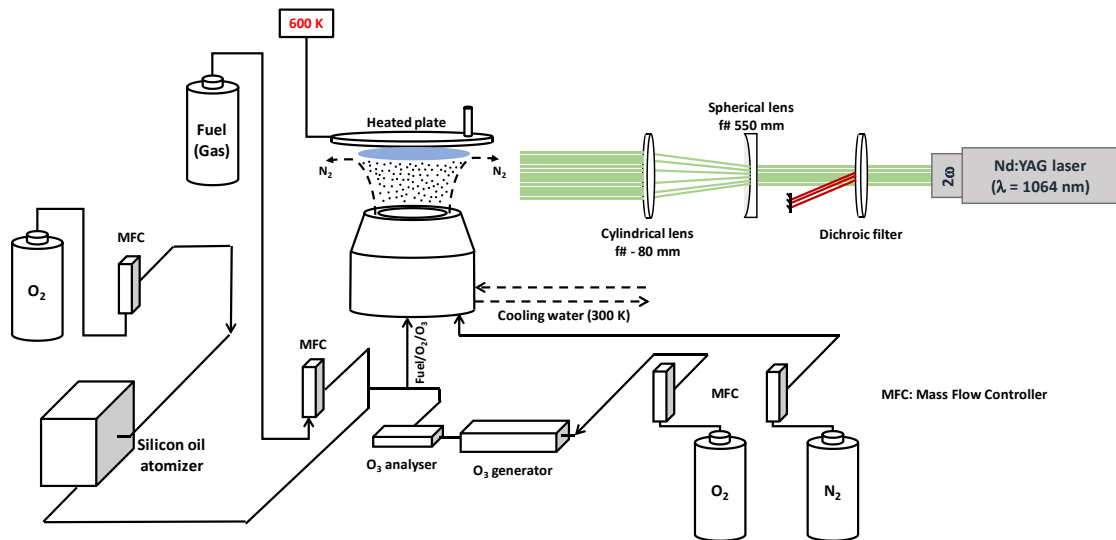
113 **2.1. Experimental setup for PIV measurements**

114 The stagnation plate burner used in the present study has already been extensively
115 described in a previous study [23], and will therefore only briefly be presented in the present
116 work (Figure 1). It consists of a stainless-steel body, with a coaxial converging nozzle of an
117 inner diameter of 10 mm for the DME/O₂/O₃ mixture. This mixture is injected through four
118 orthogonal inlets and flows through a stainless-steel porous disc before entering the convergent
119 section of the burner. This geometry is chosen as it provides a laminar and uniform flow at the
120 burner exit. A N₂ co-flow exits through a 20 mm internal diameter co-axial nozzle also equipped
121 with a stainless-steel porous section to protect the cool flames from external perturbations. The
122 burner body is kept at a constant temperature of 300 K by a temperature-regulated water
123 circulation system, and is operated at atmospheric pressure. A cylindrical heated plate, with a
124 diameter of 65 mm and a thickness of 8 mm, is placed at 13 mm from the burner nozzle, with
125 a strictly parallel geometry, and is heated at 600 ± 0.1 K. Gas flow rates are controlled by
126 Bronkhorst mass flow controllers, whose calibration was performed using DryCal DC-Lite
127 Primary Flow Meters. Part of the O₂ flows through two ozone generators (BMT Messtechnik
128 GMBH) arranged in parallel. The ozone mole fraction is measured at the entrance of the burner

129 with a Teledyne API 452 ozone analyser, the relative uncertainty on the reported ozone
130 concentrations being $\pm 0.02\%$ for every studied condition. The ozone mole fraction at the burner
131 outlet has been measured both in reactive and non-reactive conditions using an Omnistar GSD
132 301 O₂ Pfeiffer Vacuum mass spectrometer, showing no ozone reactivity or decomposition
133 before the burner outlet.

134 Measurements of the axial velocity profiles between the burner and the plate are
135 performed using the PIV technique which is based on the diffusion of illuminated particles
136 seeded in the flow. In our case, part of the oxygen flows through a liquid atomizer, allowing
137 the seeding of small particles in the flow. Silicon oil, provided by Chem-Lab, is chosen for its
138 well-defined properties (viscosity, surface tension, etc.) and its high vaporization temperature,
139 around 600 K. In the studied conditions, the temperature does not exceed 900 K downstream
140 from the flame, and the temperature in the flame front is around 600 K [23]. The particle
141 diameter is estimated to be in the range 1 – 5 μm , as usually met for this kind of atomizer [24].
142 The flame position did not change with the addition of particles, demonstrating that the seeding
143 by silicon oil droplets does not affect the cool flame speed. The use of silicon oil is therefore
144 well adapted for such PIV measurements, as will be demonstrated further later in this
145 manuscript.

146



147

148 **Figure 1.** Schematic diagram of the experimental setup.

149

150 Particles are illuminated by a dual cavity Nd:YAG Splitlight Compact laser from
 151 Innolas, delivering a 50 mJ pulse at 532 nm, at a frequency of 10 Hz. The laser sheet, centred
 152 on the burner, is generated using spherical (f# 550 mm) and cylindrical (f# - 80 mm) lenses.
 153 The laser sheet dimensions are ~ 12 mm height x 600 μm thick. The particles displacement
 154 between two laser pulses is captured using a LaVision Scmos camera of 2560 by 2160 pixels,
 155 equipped with a 105 mm Nikkor lens at an aperture of f#8, the resulting magnification being
 156 equal to 20 μm per pixel. The time between two pulses (Δt) was fixed to 220 μs, in order to
 157 record a 10-pixel displacement between the pulses at the lowest flow rate. A total of 1000
 158 images were recorded for each condition.

159 In the vicinity of the flame, the particle concentration is estimated to 0.005 particle per
 160 pixel. Such a low concentration is caused by the difficulties of effectively seeding the flow, as
 161 the oxygen flow rate had to be split between the ozone generators and the atomizer. For
 162 conventional PIV, this very low concentration requires a minimum interrogation window size
 163 (IWS) of 32 x 32 pixels (0.64 x 0.64 mm²), which leads in our conditions to an insufficient
 164 spatial resolution for accurate measurements. To tackle this difficulty, a more advanced and

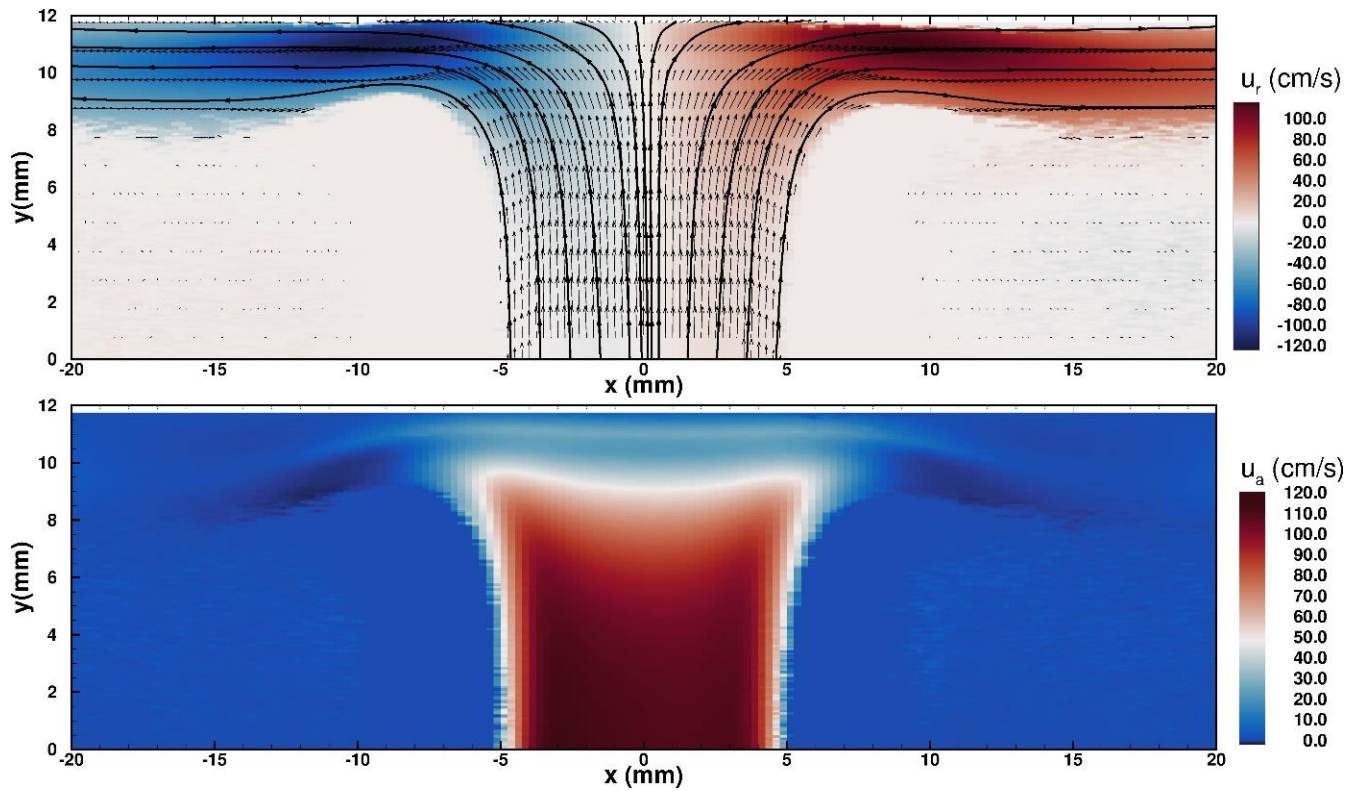
165 adapted post-processing approach has been specifically developed at the CORIA laboratory,
166 and is detailed below.

167 **2.2. Particle Image Velocimetry (PIV) images post-processing**

168 Before applying the PIV image processing, an advanced background correction is
169 performed from conditioned averaged images, one at t and one at $t + \Delta t$. A standard background
170 correction is not adapted in the case of low-densities due to the difficulty to remove the
171 individual particle signal on averaged images, even when 1000 images are considered. In our
172 approach, the conditioned background images are then obtained by averaging all the
173 instantaneous images but without integrating the signal of particles in the process using a
174 Kuwahara filter. This specific image processing improves the estimation of background images
175 in particular in low density regions. Following the background correction, only the particle
176 signal remains on the corrected images. Then, the spatial calibration is done with a well-defined
177 target (0.977 x 0.977 mm), and is used to estimate a polynomial camera model of 3rd order used
178 to dewarp each image, ensuring a perfect alignment of both wall and burner axes within the (x,
179 y) frame. This step also corrects any image distortion induced by the camera lens. From this
180 step, PIV processing starts by an extraction of the particles position in the images using a 2D
181 correlation pattern recognizing technique. Between 1500 and 3500 particles are detected on
182 each image, depending on the experimental conditions. The vector calculation is then initiated
183 at the particle location, and not on a regular mesh as it is usually performed when the particles
184 density is higher. This calculation is based on an iterative continuous window shift technique
185 [25,26], starting with an interrogation window size (IWS) of 64 x 64, and reaching an IWS of
186 8 x 8 pixels (0.16 x 0.16 mm²) in the final pass. This results in a series of non-regular velocity
187 fields which are validated with a rate higher than 85%. The scaling in the real coordinated
188 system is realized with the burner plate as reference, with an accuracy of $\pm 20 \mu\text{m}$. The final
189 step consists in computing a mean velocity field on a regular mesh by averaging every

190 instantaneous vector in small cells at each mesh node, the cell size being compatible with the
191 PIV resolution of 8 x 8 pixels. This processing method allows a significant improvement in the
192 particle detection in the area close to the flame front. A comparison of the axial velocity profiles
193 determined using a conventional image processing method, i.e., with an IWS of 32 x 32 pixels,
194 and the present one is presented in Figure S1 of the Supplementary Material.

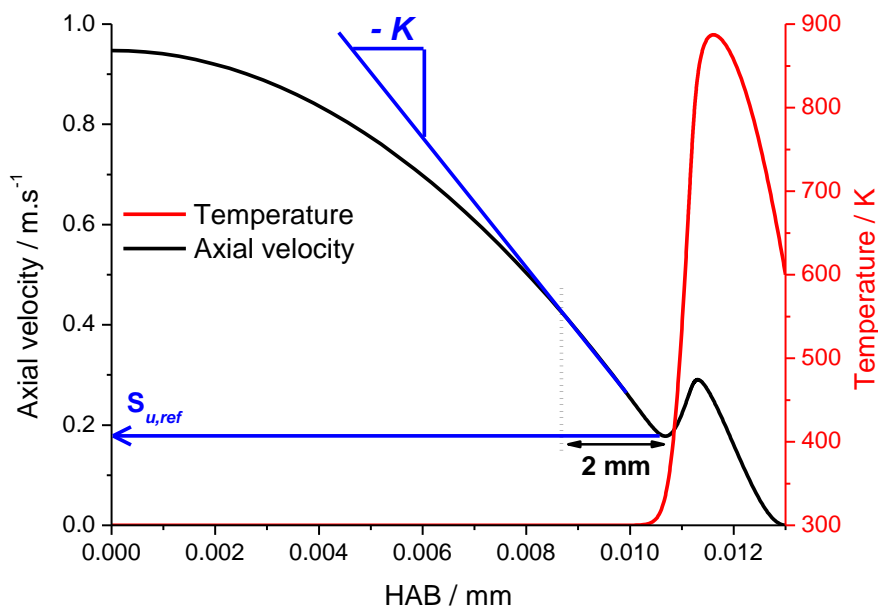
195 Axial velocity profiles are finally extracted from the mean velocity field by averaging
196 on a radial width of ± 1 mm around the centre of the burner. A two-dimensional velocity field
197 in reactive conditions is shown in Figure 2. It demonstrates that the radial (u_r) distribution of
198 the velocities is axisymmetric, and that the axial velocity (u_a) profile at the burner exit is almost
199 uniform over the entire diameter of the burner (10 mm). The flat cool flame front, stabilized
200 two millimetres under the heated plate, can be distinguished on the axial velocity profile. This
201 confirms the efficiency of the converging nozzle geometry, and constitutes further justification
202 of the averaging process over a region of ± 1 mm used in the image post-processing, as the axial
203 velocity can be considered constant over a significant portion of the burner diameter. From
204 Root Mean Square (RMS) analysis, the uncertainty on the determination of the axial velocity
205 with the PIV technique is estimated to be $\pm 5\%$. This lead to an uncertainty in the flame speed
206 determination lying between 0.83 and 1.05 $\text{cm}\cdot\text{s}^{-1}$, depending on the value of the reference axial
207 velocity $S_{u,\text{ref}}$ (minimum velocity upstream of the flame front, as shown in Figure 3), which has
208 been averaged at $\pm 1 \text{ cm}\cdot\text{s}^{-1}$.



209
 210 **Figure 2.** Radial velocity streamlines (top) and axial velocity profile (bottom) of a DME/O₂/O₃ cool
 211 flame at $\phi = 0.4$ and $x_{O_3} = 1.9\%$. u_r represents the radial velocity, u_a the axial velocity, y the Height
 212 Above the Burner (HAB) and x the burner radius.

213

214



215
 216 **Figure 3.** Simulated temperature (red) and axial velocity (black) profiles of a cool flame. Determination
 217 of the couple ($S_{u,ref}$, K) is also shown.

218

219

2.3. Flame conditions

Five stable cool flame conditions were selected for the measurement of cool flame speeds, and are summarized in Table 1. Note that the inlet velocity (u_{in}) range is limited as it is constrained by the stability domain of the cool flames, which is known to be narrower than for hot flames. Furthermore, at high inlet velocity (and strain rate, usually calculated as the ratio between the flow velocity and the burner/plate distance), the cool flames tend to stabilize closer to the stagnation plate, leading to difficulties in resolving the entire velocity profile. On the contrary, at low inlet velocity, the particle seeding is not sufficient to provide accurate measurement of the velocity field in the burner.

The detailed flames conditions are provided in the Supplementary Material (Table T1).

ϕ	$x_{O_3}^* / \%$	$u_{in} / \text{cm.s}^{-1}$
0.5	1.5	65 – 80
0.45	1.7	75 – 110
0.4	1.7	65 – 85
0.4	1.9	75 – 110
0.3	2.0	65 – 95

*The indicated values correspond to the ozone mole fraction in the DME/O₂/O₃ mixture

Table 1. Experimental conditions for the investigated DME/O₂/O₃ cool flames.

2.4. Flame simulations

The axial velocity profiles for all flames conditions were simulated with multicomponent transport using the Pre-Mixed Burner Stagnation Flame module of Chemkin-Pro 2021 [27]. The size of the simulation domain (see Table T1 in Supplementary Material) was varied depending on the studied conditions, as detailed later. The GRAD and CURV parameters were fixed at 0.03 each, resulting in ~ 450 points per simulation, which is sufficient

239 to ensure accurate simulations of the reference axial velocity $S_{u,ref}$ within a 1% uncertainty, as
240 demonstrated in the Supplementary Material (Figure S2). The Soret effect option was disabled
241 for the axial velocity profiles simulations as it did not significantly modify the reference axial
242 velocity and allowed a drastic reduction of the computational time. The energy equation was
243 solved for simulation; hence the experimental temperature profile was not used as an input. The
244 experimental and simulated temperature profiles of lean DME/O₂/O₃ cool flames were
245 compared in our previous study [23] and showed an excellent agreement.

246 A detailed kinetic mechanism recently developed in our group [23] has been used, it is
247 based on the AramcoMech1.3 mechanism [28] with reassessed reaction pathways and modified
248 Arrhenius parameters. It was validated on the prediction of the mole fraction profiles of
249 intermediates species above the burner for DME/O₂/O₃ cool flames. Three different O₃-
250 submechanisms from the literature, respectively from Jian et al. [20], Zhao et al. [21] and Halter
251 et al. [22], were coupled to the aforementioned mechanism [23]. A summary of the different
252 reactions and their associated modified Arrhenius reaction rate coefficients is given in the Table
253 T2 of the Supplementary Material. Note that these three O₃-submechanisms have been very
254 recently [20] compared to experimental data on O₃ decomposition, O₃/O₂ flame speeds and
255 H₂/O₂/O₃ reaction in a flow reactor, showing satisfactory predictions in these experimental
256 conditions.

257

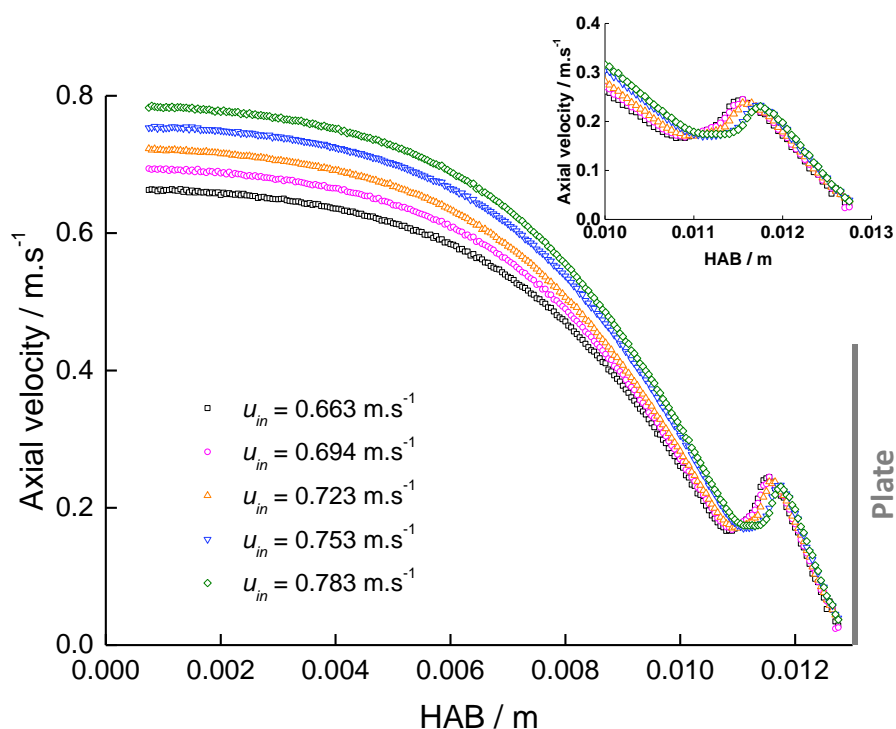
258 **3. RESULTS AND DISCUSSION**

259

260 **3.1. Axial velocity profiles**

261 The axial velocity profile measured for the strained cool flame at $\phi = 0.5$, $x_{O_3} = 1.5\%$ is
262 pictured for five different inlet velocities u_{in} (and strain rates) as an example of the obtained
263 results in Figure 4. The increase of velocity upstream of the flame front due to thermal
264 expansion is less pronounced than in hot flame cases since the heat release of cool flames is

265 significantly lower. The entire velocity profile is well defined in each condition supporting the
 266 use of silicon oil droplets and the applied PIV image processing. As the inlet velocity (and strain
 267 rate) increases, the flame moves towards the stagnation plate and the acceleration observed in
 268 the flame front is reduced in accordance with a decrease of the maximum flame temperature.



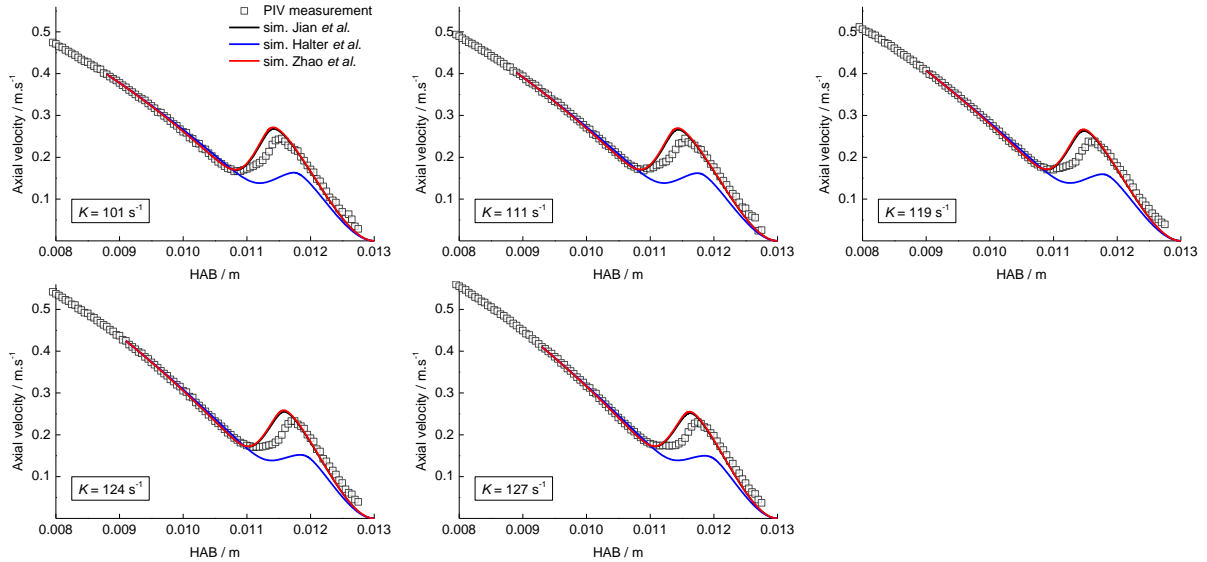
269
 270 **Figure 4.** Axial velocity profiles as a function of the Height Above the Burner (HAB) of a DME/O₂/O₃
 271 cool flame at $\phi = 0.5$, $x_{O_3} = 1.5\%$, for five different inlet velocities u_{in} .

272
 273 The measured axial velocity profiles in the strained cool flames were compared with
 274 simulations performed with the Pre-Mixed Burner Stagnation Flame module of Chemkin-Pro,
 275 following the methodology proposed in [29–31]. It consists in imposing the exact boundary
 276 conditions inferred from the PIV experiments, as it is recognized that the experimental flow
 277 field in stagnation plate burner configuration is usually neither a plug flow nor a potential flow
 278 [32] and that 1D approaches typically fail in providing accurate prediction of the corresponding
 279 velocity profile [33]. In this work, the inlet position of the simulation domain was fixed two
 280 millimetres before the minimum velocity upstream of the flame front position (defined as $S_{u,ref}$).

281 Experimental strain rates K and inlet velocities were inferred from the axial velocity profile 2
282 mm upstream from $S_{u,ref}$, as pictured in Figure 3. The effect of the domain size on the accuracy
283 of the velocity profile, targeting both the reference velocity $S_{u,ref}$ and its position above the
284 burner, was studied and the results are pictured in Figures S3 and S4 of the Supplementary
285 Material. Results showed that the simulated $S_{u,ref}$ were similar within 1% for domain sizes
286 below 8 mm, but that selecting a too small domain size, here less than 3.2 mm, led to a greater
287 deviation. The computational domain length therefore lies between 3.05 and 4.45 mm,
288 depending on the flame conditions. To ensure reproducible determination of the experimental
289 K , the experimental axial velocity profile was fitted using a 2nd-order polynomial function
290 beforehand.

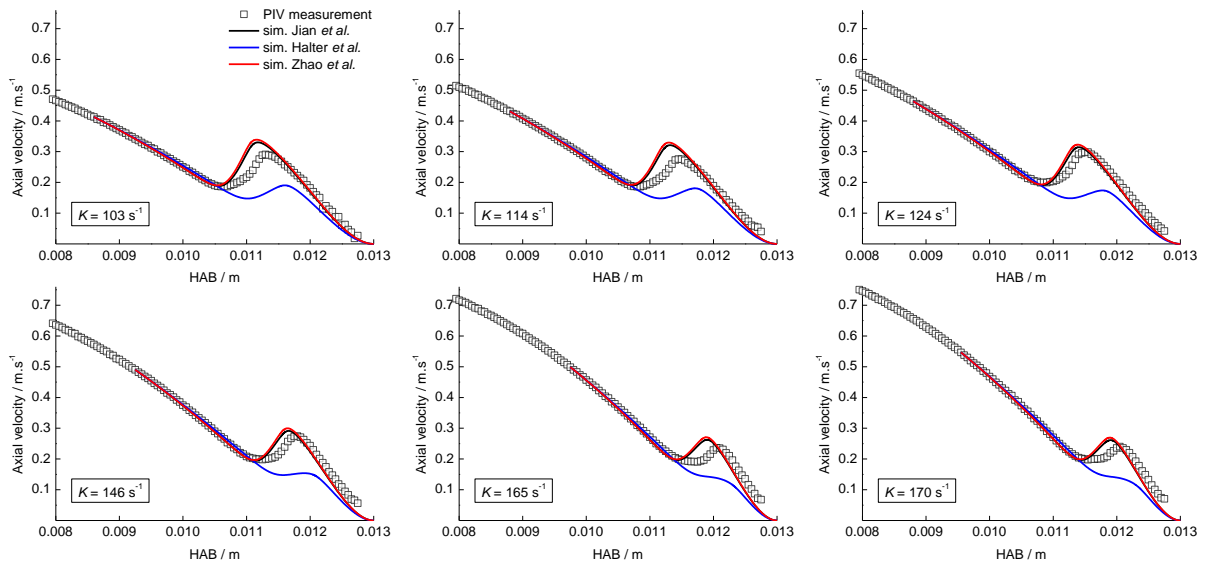
291 The axial velocity profiles simulated using the three aforementioned ozone-
292 submechanisms [20–22] were compared to the experimental ones. Two flame conditions at
293 $\phi = 0.5$, $x_{O_3} = 1.5\%$ and $\phi = 0.3$, $x_{O_3} = 2.0\%$ are presented in Figure 5 and Figure 6 respectively.
294 For the sake of brevity, the same comparisons for other flame conditions are presented in
295 Figures S5 to S7 in the Supplementary Material, and the exact boundary conditions used for the
296 simulation are given in the Table T1 of the Supplementary Material. Comparing the three
297 different O_3 -submechanisms, large variations in the prediction of the velocity profile can be
298 observed between the Halter et al. model and the two other models, with an important
299 underprediction of the minimum velocity $S_{u,ref}$ for the Halter et al. model. It should be noted
300 that in the case $\phi = 0.3$, $x_{O_3} = 2\%$ (Figure 6), the Halter et al. model predicts almost no minimum
301 velocity for higher strain rates, but only an inflection of the axial velocity curve upstream of the
302 flame front. The predictions from the Jian et al. and Zhao et al. models are comparable, as both
303 models accurately describe the overall velocity profile.

304



305 **Figure 5.** Comparison between experimental axial velocity profiles measured by PIV (□) and simulated
 306 ones with different O_3 -submechanisms, respectively from Jian et al., Halter et al. and Zhao et al., for the
 307 cool flame at $\phi = 0.5$, $x_{O_3} = 1.5\%$, at different strain rates.

308



309 **Figure 6.** Comparison between experimental axial velocity profiles measured by PIV (□) and simulated
 310 ones with different O_3 -submechanisms, respectively from Jian et al., Halter et al. and Zhao et al., for the
 311 cool flame at $\phi = 0.3$, $x_{O_3} = 2.0\%$, at different strain rates.

312

313 In order to evaluate the capabilities of these models to predict cool flame speeds, the
 314 Root-Mean-Square Error (RMSE) between experimental and simulated $S_{u,ref}$ was calculated
 315 over the investigated strain rate range, for the five flames conditions.

316 The RMSE data are summed up in the Table 2, for each O₃-submechanism. One can see that
 317 the Halter et al. submechanism constantly underestimates the value of S_{u,ref}, while the two other
 318 models yield similar performance.

Condition	Root-Mean-Square Error (RMSE)		
	Jian et al.	Halter et al.	Zhao et al.
$\phi = 0.3, x_{O_3} = 2.0\%$	0.5	4.7	0.5
$\phi = 0.4, x_{O_3} = 1.9\%$	1.2	5.9	0.7
$\phi = 0.4, x_{O_3} = 1.7\%$	0.2	3.8	0.3
$\phi = 0.45, x_{O_3} = 1.7\%$	0.2	4.5	0.4
$\phi = 0.5, x_{O_3} = 1.5\%$	0.2	3.1	0.2

319 **Table 2.** Root Mean-Square Error (RMSE) calculated for each flame condition with the different O₃-
 320 submechanisms

321

322 A possible explanation for the constant underestimation of the flame speed by the model of
 323 Halter et al. is the definition of the ozone decomposition within the model, where only N₂, O₂
 324 and O₃ are declared as collision partners in the O₃ + M ⇌ O₂ + \ddot{O} + M reaction, in contrast with
 325 the other two mechanisms (for which all species are considered with a collision efficiency of 1,
 326 except if specified). In our experimental conditions, the fuel mole fraction lies between 10 and
 327 15% of the mixture. The absence of DME as a third-body in the ozone decomposition reaction
 328 can therefore lead to a lowered reactivity of the Halter et al. model (see Table T2 in the
 329 Supplementary Material). This aspect has been tested by replacing the O₃+M decomposition
 330 reactions of the model of Jian et al. by the O₃+M reactions of Halter et al., showing a
 331 deterioration in the prediction of the axial velocity profile, as seen in Figure S8 in the
 332 Supplementary Material.

333 For the following sections, the Jian et al. submechanism will be used solely, because of
 334 its more recent rate constants data and its good performance in predicting S_{u,ref}.

3.2. Determination of the unstrained cool flame speed $S_{u,0}$

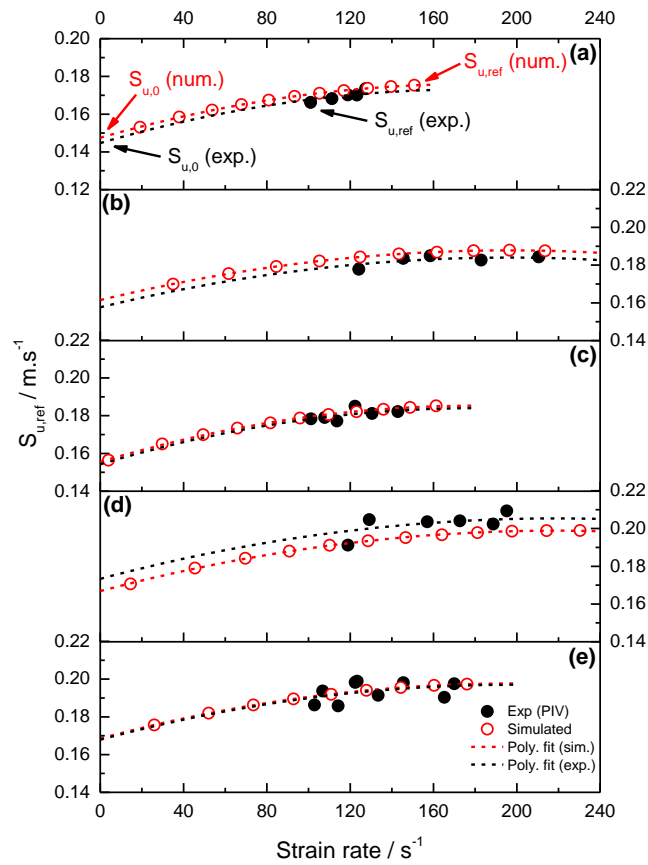
A well-established method [34] has been widely used to derive the unstrained flame speed $S_{u,0}$ of hot flames. It consists of measuring the axial velocity profiles for different strain rates (K) and determining the reference velocity ($S_{u,ref}$) as represented in Figure 3. From the plot of $S_{u,ref} = f(K)$, the extrapolation to strain rate $K = 0$ returns the unstrained laminar flame speed $S_{u,0}$.

Both linear [35] and non-linear [36] extrapolation methods have been used in the past. Vagelopoulos et al. [34] and Chong and Hochgreb [37] have demonstrated that for low-strain rate hot flames ($50 - 250 \text{ s}^{-1}$), the linear extrapolation method yields accurate results within a reported uncertainty of $\sim 1-2 \text{ cm.s}^{-1}$. In the case of cool flames, for which the speed is significantly smaller, this added uncertainty might however be problematic. The issue of non-linear extrapolation was revisited by Egolfopoulos et al. [38–41] with a computational approach, where the simulated $S_{u,ref}$ at various K (using a counter-flow flame code) and the simulated $S_{u,0}$ at $K = 0$ (using the freely propagating flame module of the PREMIX code) are fitted using a polynomial function. This computed curve is vertically translated to best fit the experimental data and the experimental flame speeds are derived from this non-linear extrapolation [40]. This method however requires using a kinetic mechanism that yields an accurate value of $S_{u,0}$.

In the present work, a similar approach has been applied but without using the $S_{u,0}$ calculated from a freely-propagating flame module. Axial velocity profiles were simulated using the Pre-Mixed Burner Stagnation Flame module of Chemkin-Pro by progressively decreasing the strain rate, from which a numerical curve $S_{u,ref} = f(K)$ was extracted, as shown in Figure 7. The simulated curve $S_{u,ref} = f(K)$ is then fitted using a 2nd order polynomial and vertically translated to best fit the experimental results [37-40]. The experimental unstrained cool flame speed $S_{u,0}$ is then considered at $K = 0$. Figure 7 shows the application of this method

360 for the five DME/O₂/O₃ cool flames conditions. As seen in Figure 7, the numerical and
361 experimental $S_{u,0}$ lie very close to each other (those values are given in Table 3 for the five
362 flames studied, columns 2 and 3).

363 Limitations of this method should however be considered. First, unlike hot flames, the
364 fuel conversion is only partial in a cool flame. Therefore, thermochemical equilibrium is not
365 reached in the post flame region. This causes non-zero temperature and velocity gradients in
366 the downstream area of the flame. Such results can therefore not be considered as laminar
367 burning velocities, nor modelled using a freely propagating flame module. Secondly, since the
368 stagnation plate temperature (600 K) is below the measured flame peak temperatures of about
369 900 K [23], such flames are not adiabatic. To assess the effect of this non-adiabaticity on the
370 determined unstrained cool flame speeds $S_{u,0}$, additional simulations were carried out. The
371 stagnation plate module of Cantera 2.6 [42] was used in the same domain width and input
372 compositions as in the simulations of Figures 5-6, and the stagnation plate temperature was
373 progressively increased until it reached a temperature identical to the maximal flame
374 temperature within a 0.01% tolerance. The strain rate was varied, and the $S_{u,ref} = f(K)$ results
375 were fitted to a 2nd order polynomial leading to $S_{u,0}$ values at $K=0$ reported in Figure 8 and Table
376 3 (column 4). Because these cool flames are below 1000 K and sufficiently lifted to neglect
377 heat losses to the exit burner nozzle, they can be considered adiabatic, as for example in the
378 well-known heat flux method [43]. The adiabatic $S_{u,0}$ results (Table 3) are identical within the
379 uncertainty of the experiments to the simulated values at a plate temperature of 600 K,
380 demonstrating a negligible effect of the plate temperature on the measured unstrained cool
381 flame velocities.

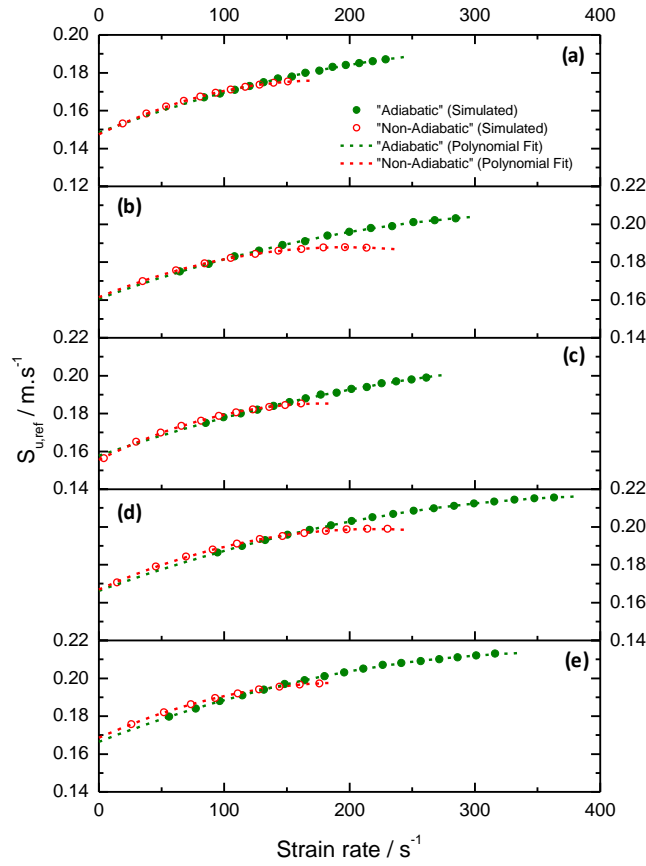


382

383

384 **Figure 7.** Variations of $S_{u,ref}$ with K and determination of $S_{u,0}$ using a numerically assisted non-linear
 385 extrapolation. Flame conditions: $\phi = 0.5$, $x_{O_3} = 1.5\%$ (a), $\phi = 0.45$, $x_{O_3} = 1.7\%$ (b), $\phi = 0.4$, $x_{O_3} = 1.7\%$
 386 (c), $\phi = 0.4$, $x_{O_3} = 1.9\%$ (d) and $\phi = 0.3$, $x_{O_3} = 2.0\%$ (e).

387



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389

390 **Figure 8.** Simulated variations of $S_{u,ref}$ with K using a non-linear extrapolation in adiabatic and non-
 391 adiabatic conditions. Flames conditions: $\phi = 0.5$, $x_{O_3} = 1.5\%$ (a), $\phi = 0.45$, $x_{O_3} = 1.7\%$ (b), $\phi = 0.4$, x_{O_3}
 392 $= 1.7\%$ (c), $\phi = 0.4$, $x_{O_3} = 1.9\%$ (d) and $\phi = 0.3$, $x_{O_3} = 2.0\%$ (e).

393

394

Flame conditions	$S_{u,0} / \text{cm}\cdot\text{s}^{-1}$		
	Experimental	Simulated	Simulated Adiabatic
$\phi = 0.5$, $x_{O_3} = 1.5\%$	$14.5 \pm 1 \text{ cm}\cdot\text{s}^{-1}$	14.7	14.8
$\phi = 0.45$, $x_{O_3} = 1.7\%$	15.8	16.1	16.1
$\phi = 0.4$, $x_{O_3} = 1.7\%$	15.4	15.6	15.8
$\phi = 0.4$, $x_{O_3} = 1.9\%$	17.3	16.7	16.6
$\phi = 0.3$, $x_{O_3} = 2.0\%$	16.8	16.9	16.6

395 **Table 3.** Experimental and simulated unstrained cool flame speeds $S_{u,0}$ for all the cool flames conditions.

396

397 By comparing both flames at $x_{O_3} = 1.7\%$, one can see that the cool flame speed increases
 398 moderately as the equivalence ratio increases from 0.4 to 0.45. This effect is captured by the
 399 simulation. A comparison of both flames at $\phi = 0.4$ ($x_{O_3} = 1.7\%$ and 1.9%) demonstrates the

400 important effect of ozone addition on the cool flame speed, which increases by about 2 cm.s^{-1}
401 (experimentally) when the ozone mole fraction increases only from 1.7% to 1.9%. This effect
402 is also captured in the simulation, yet underestimated.

403

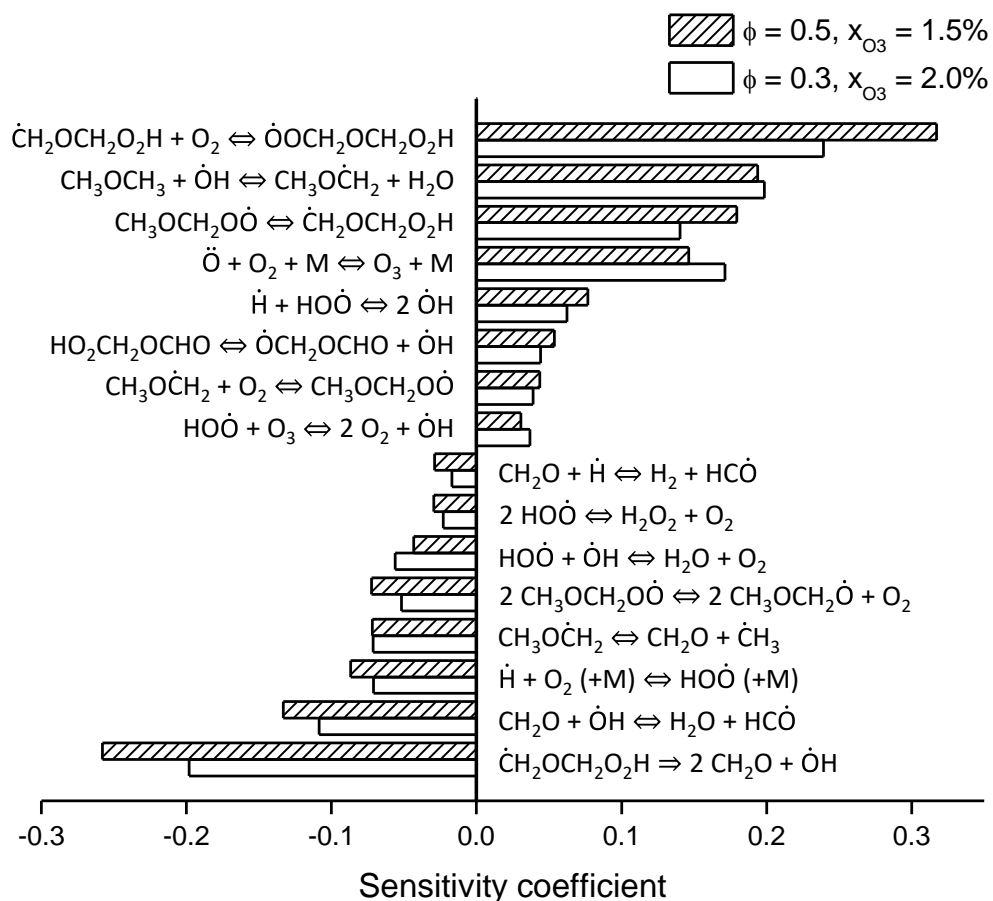
404 **3.3. Kinetic analysis**

405 A brute-force sensitivity analysis was performed on the experimental reference axial
406 velocity $S_{u,\text{ref}}$, using the Jian et al. O_3 -submechanism [20] in conjunction with our previously
407 published DME sub-mechanism [23], with the stagnation flame module of the Cantera 2.6
408 solver [42]. To do so, the domain size and input conditions were chosen as in the experiments
409 and simulations of Figures 5-6, and the sensitivity coefficients of the reactions on the minimum
410 of axial velocity associated with the cool flame were evaluated from a positive and negative
411 variation of the frequency factor of each reaction by 5%. A positive sensitivity coefficient
412 therefore means that increasing the reaction rate constant for the associated reaction increases
413 the reference cool flame velocity $S_{u,\text{ref}}$, and vice versa. The two most extreme conditions were
414 selected for the sensitivity analyses, respectively the flame at $\phi = 0.3$, $x_{\text{O}_3} = 2.0\%$, which
415 contains the highest ozone mole fraction in this study, and the flame at $\phi = 0.5$, $x_{\text{O}_3} = 1.5\%$,
416 which contains the lowest. The results are displayed for the 16 reactions with the highest
417 absolute sensitivity coefficients in Figure 9.

418 In both conditions, it is observed that the simulated $S_{u,\text{ref}}$ is strongly dependent on the
419 branching between the decomposition of the $\dot{\text{Q}}\text{OOH}$ into two molecules of formaldehyde and
420 an $\dot{\text{O}}\text{H}$ radical, which tends to decrease the cool flame velocity, and the addition of $\dot{\text{Q}}\text{OOH}$ to
421 O_2 , which will lead to indirect chain-branching. Among the other important features is the
422 competition between the $\text{RO}\dot{\text{O}} \Leftrightarrow \dot{\text{Q}}\text{OOH}$ pathway, and the $2 \text{RO}\dot{\text{O}} \Leftrightarrow 2 \text{R}\dot{\text{O}} + \text{O}_2$ route. The
423 latter is traditionally associated with atmospheric conditions, but was previously observed to be

424 significant in our conditions [23]. To a more reduced extent, the branching of \dot{R} radical between
 425 addition to O_2 or direct scission into formaldehyde and a methyl radical is also of importance.
 426 One can however note the large sensitivity coefficient of the H-atom abstraction reaction on
 427 DME by $\dot{O}H$. A striking feature of these results is that, in contrast with hot flames burning
 428 velocities, the most sensitive reactions are all directly part of the fuel submechanism, the $O_3 +$
 429 $M \rightleftharpoons O_2 + \ddot{O} + M$ reaction only ranking as the fourth most sensitive reaction. This is a strong
 430 argument in favour of these experimental data as potential targets for the validation of kinetic
 431 models in the low-temperature domain.

432



437 4. CONCLUSIONS

438 The measurement of ozone-seeded dimethyl ether cool flame speeds was successfully
439 realized in a stagnation plate burner for the first time. The PIV technique was used to measure
440 the 2-D velocity profiles, from which axial velocity profiles were inferred to measure the
441 unstrained cool flame speeds. For this purpose, an in-house PIV image processing method has
442 been developed and used in order to improve the accuracy of the measurement. Five flame
443 conditions, with equivalence ratios varying from 0.3 to 0.5 and ozone mole fraction varying
444 from 1.5 to 2% were used to compare experimental results with kinetic modeling. Three ozone-
445 submechanisms, namely from Jian et al., Zhao et al. and Halter et al., were coupled with our
446 previous mechanism validated in DME/O₂/O₃ cool flames and used to compare experimental
447 and simulated axial velocity profiles, emphasizing the importance of the choice of the O₃-
448 submechanism. The Jian et al. submechanism was selected because of its good predictive ability
449 for axial velocity profiles and was used to determine the unstrained cool flame speed $S_{u,0}$ for
450 each tested condition. A numerical assisted non-linear extrapolation method is proposed for the
451 determination of $S_{u,0}$. Additionally, simulations for which the plate temperature reaches the
452 maximal flame temperature (i.e., adiabatic conditions) were performed, demonstrating a
453 negligible effect of the plate temperature on the unstrained cool flame velocity. However, for
454 the purpose of mechanism validation, the direct comparison with the experimental axial
455 velocity profiles should be preferred as it requires no numerical assisted method. Values of $S_{u,0}$
456 were found to lie between ~ 14 and $17 \text{ cm}\cdot\text{s}^{-1}$ within the range of flame conditions studied here,
457 showing a limited effect of the equivalence ratio variations, and a more striking effect of the
458 ozone concentration. Finally, sensitivity analyses performed on the reference axial velocity $S_{u,\text{ref}}$
459 in two flame conditions emphasize the importance of the fuel low-temperature chain-branching
460 pathways, over the ozone submechanism.

461

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469

470 **Supplementary material**

471 Supplementary Material can be found in the attached file.

472

473 **Authors contributions**

474 Conceptualization: **L. Pillier, G. Vanhove**; Data curation: **All authors**; Formal analysis: **T.**
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477 **Cuvier**; Methodology: **All authors**; Project administration: **L. Pillier, G. Vanhove**; Resources:
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480 **Fenard**; Validation: **All authors**; Visualization: **All authors**; Roles/Writing - original draft: **T.**
481 **Panaget, L. Pillier, G. Vanhove**; and Writing - review & editing: **All authors**

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