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Laminar burning speeds and Markstein lengths of *p*-cymene possibly involved in accelerating forest fires

B. Coudour^{1,2*}, K. Chetehouna², L. Courty¹, J.P. Garo¹, C. Mounaïm-Rousselle³, F. Halter³

¹ Institut P', CNRS, ENSMA, Université de Poitiers, 1 Av. Clément Ader, Téléport 2, BP 40109, 86961 Futuroscope Chasseneuil, France.

² ENSI de Bourges, Laboratoire PRISME, 88 Boulevard Lahitolle, 18020 Bourges, France.

³ Laboratoire PRISME, Université d'Orléans, 8 Rue Léonard de Vinci, 45072 Orléans Cedex 2, France.

Abstract

A potential implication of Volatile Organic Compounds (VOCs) emitted by vegetal species has been introduced in the literature to explain accelerating forest fires. These fires are characterized by the sudden increase of the rate of spread and of the energy released by the fire front. The main purpose of this paper is to determine the combustion characteristics of a major VOC emitted by *Thymus vulgaris* needles, namely *p*-cymene. The emission of this compound is studied for the temperature range 343-453 K with an emission peak observed at 443 K. Laminar burning speeds, Markstein lengths and flame thicknesses are determined using outwardly propagating spherical flames in a combustion vessel at atmospheric pressure. The effects of equivalence ratio (0.8 to 1.4) and unburned gas temperature (358 to 453 K) are investigated. Results are compared to experimental data of α -pinene and to computed data of two fuels, JP-10 and n-decane obtained with the PREMIX code of the CHEMKIN package.

Introduction

Accelerating forest fires are defined as extreme fires that appear without any change in the external conditions (ambient wind, vegetation type, etc.). They cause serious damages in terms of ecological and economical issues and human lives every year being the cause of many fatalities during the last decades. The Storm King and Guadalajara are some examples of these fires listed in Courty et al., 2012 [1]. They occurred in USA and Spain, killing respectively 14 and 11 persons.

A thermochemical approach deals with the possible ignition of gases accumulated ahead of the fire like unburned gases or Volatile Organic Compounds (VOCs). Indeed, almost all plants when heated produce and emit volatile compounds [2], forming a flammable mixture. The VOCs ignition hypothesis is supported by the empirical knowledge of fire-fighters in the terrain that usually link the strong smell of these VOCs with high risk. A flammability enhanced by the Lower Flammability Limits (LFLs) of VOCs which are lower than unburned products [3].

Physical forest fires modelling do not consider the possible ignition of VOCs to predict accelerating forest fires. Modelling such phenomenon requires the knowledge of VOCs combustion characteristics that have never been determined. This paper presents the determination of some combustion characteristics of *p*-cymene emitted by *Thymus vulgaris* shrubs : the laminar burning speeds, Markstein lengths and flame thicknesses. The results are obtained using a spherical combustion chamber and a nonlinear extraction methodology. The nonlinear model used in this methodology is given by the following relation [1,4]:

$$\left(\frac{s_b}{s_b^0}\right)^2 \ln\left(\frac{s_b}{s_b^0}\right) = -2 \frac{L_b}{s_b^0} \kappa \quad (1)$$

where s_b and s_b^0 are respectively the stretched and unstretched premixed flame speeds, L_b is the Markstein length and κ is the flame stretch. An analytical solution of this equation, considering the flame radius R_f as a variable, can be given as:

$$\begin{cases} t = \frac{2L_b}{s_b^0} \left[E_1(\ln \eta^2) - \frac{1}{\eta^2 \ln \eta} \right] + C \\ R_f = -\frac{2L_b}{\eta \ln \eta} \end{cases} \quad (2)$$

with $\eta \in [e^{-1}, 1]$ for $L_b > 0$, $\eta \in [1, \infty[$ for $L_b < 0$ and C an integration constant. The flame radius is linked to the stretched premixed flame speed by $s_b = \frac{dR_f}{dt}$.

The unstretched premixed flame speed s_b^0 and Markstein length can be estimated from equation (1) using the temporal evolutions of the stretched premixed flame speed obtained with experimental flame radii derivation and with the analytical solution of equation (2). A detailed description of the determination process of these parameters can be found in Courty et al. (2012) and Halter et al. (2010) [1,4].

The laminar flame thickness is determined according to Zeldovich definition using the thermal diffusivity:

$$\delta = \left(\frac{\lambda_u}{\rho_u C_{p,u}} \right) \frac{1}{s_u^0} \quad (3)$$

with λ_u , ρ_u and $C_{p,u}$ respectively the thermal conductivity, the density and the specific heat of the unburned gas mixture.

The literature does not contain many works studying the combustion of VOCs [5] and none about

*Corresponding author: bruno.coudour@ensma.fr

the detailed kinetic mechanism of *p*-cymene. Besides, there are no results on the laminar burning speed of this volatile organic compound. To be able to make a comparison with the literature data, we have to choose molecules with carbon atoms number close to *p*-cymene carbon one. The experimental data are therefore compared to computed values of JP-10 and n-decane (obtained using the PREMIX code of the CHEMKIN package [6] with respectively the San Diego detailed kinetic mechanism (http://web.eng.ucsd.edu/mae/groups/combustion/mechanism.html) and the one proposed by Honnet et al. (2009) [7] as well as to experimental ones of ethylbenzene, iso-octane, α -pinene and n-decane available in the literature.

Experimental section

~ VOCs emission measurement

This section details the characterization of the VOCs emitted by needles of a typical Mediterranean region species, namely *Thymus vulgaris*. The needles emission measurements are performed using a flash pyrolysis apparatus (CDS Pyroprobe 5150). This device is widely used to characterize the thermal degradation of various materials [8-10]. Needle samples with an average mass of 2.3 mg are placed inside a 40 mm x 2 mm i.d. quartz tube and heated up to the desired temperature during a variable lapse of time. The temperature rise can also be varied up to a maximum heating rate of 5000 K.s⁻¹. The flash pyrolysis device consists of an inductively heated coil to heat the samples from room temperature to the selected temperature. Helium is used as carrier gas to transport the emitted VOCs to the gas chromatograph coupled with mass spectrometer (GC/MS) apparatus via a heated (553 K) transfer line. GC/MS analyses are realized with a Trace Ultra GC-Thermo DSQ II equipped with a DB5 capillary column (30 m long, 0.25 mm i.d., film thickness 0.25 μ m). The column temperature was programmed from 333 to 473 K at a rate of 5 K.min⁻¹ and held for 5 minutes at 473 K. Mass spectra are recorded in the electron impact mode with ionization energy of 70 eV. A schematic overview of the described experimental process is presented on Figure 1.

Identification of VOCs is based on a comparison of their mass spectra with the NIST mass spectral library, with data from literature and with mass spectra and retention times of reference compounds (α -pinene, limonene). These reference compounds can also be used to make calibration curves for quantification. The range of temperature selected is between 343 and 453 K in order to determine VOCs emissions before their pyrolysis phase. Three experiments are performed for each temperature in order to ensure the repeatability.

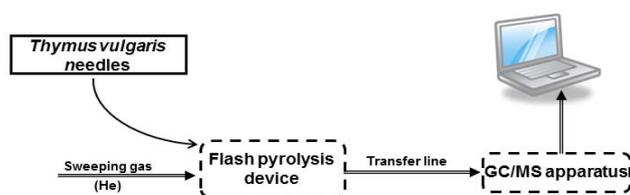


Figure 1: Schematic overview of the emission experimental setup.

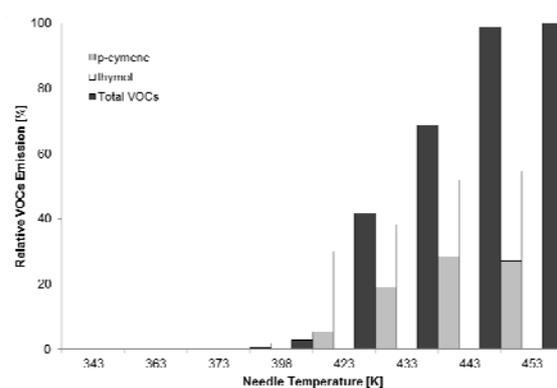


Figure 2: VOCs emissions of *Thymus vulgaris* needles for different temperatures.

Figure 2 exhibits the VOCs emissions of *Thymus vulgaris* needles for different temperatures: the two major compounds, thymol and *p*-cymene, and the total VOCs are presented. The Figure shows clearly that the emissions are important for temperature higher than 398 K and increase with temperature. The total amount emitted at 453 K is one thousand times higher than the total amount emitted at 343 K, whereas the amount of thymol emitted is ten thousand times higher between 343 and 453 K. The total amount of VOCs emitted is multiplied by 220 between 398 and 453 K and by 14 between 423 and 453 K. The emissions start to be really important from this last temperature.

Figure 3 presents the composition of VOCs mixture emitted by *Thymus vulgaris* needles at different temperatures. It is interesting to note that thymol represents more than 50 % of the total emissions for the temperatures where high amount of VOCs are emitted (i.e. after 398 K). The highest percentage of thymol in the mixture is reached at 423 K, it represents then more than 72 % of the total VOCs. Thymol is the major component for every temperature studied, even if it represents only 24 % of the mixture at 363 K. The percentage of the second more important compound, *p*-cymene, is more constant, between 15 and 25 % for all the temperatures. Except thymol and *p*-cymene, the other compounds represent more than 40 % of the total mixture for temperatures lower than 398 K whereas above this temperature they never represent more than 18 % of all of the compounds.

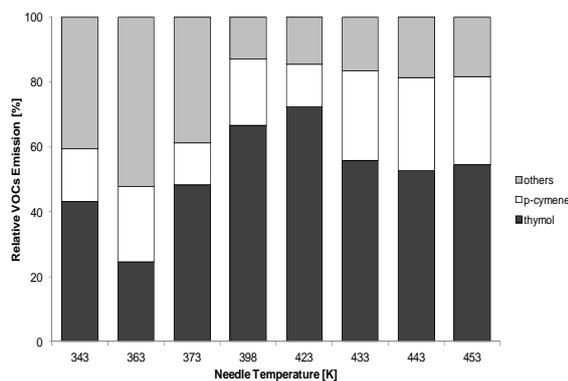


Figure 3: Composition of VOCs mixture emitted by *Thymus vulgaris* needles at different temperatures.

We can notice here the contributions of Parra et al. [11] and Nezhadali et al. [12] for the emissions of *Thymus vulgaris* under natural conditions. Parra et al. [11] estimated the magnitude of non-methane volatile organic compounds emitted by vegetation in Catalonia, Spain, along with their superficial and temporal distribution. They first defined mathematically an emission factor and then found that for *Thymus vulgaris* it was lower for monoterpenes than for other VOCs. This result is in agreement with our study. In their recent paper, Nezhadali et al. [12] compared the composition of *Thymus vulgaris* emissions using two different extraction methods: hydrodistillation and headspace solid phase microextraction. They found that for both of the methods, the major compounds were thymol, *p*-cymene, γ -terpinene, myrcene, α -pinene and caryophyllene. They found similar results for the two studied methods and concluded that hydrodistillation needs more time and much more amount of plant. They studied the emissions at 298 and 323 K and we can say that they obtained results very similar to those of our study, even if they only work at low temperatures.

Owen et al. [13] also studied the natural emissions of *Thymus vulgaris*. These authors studied the VOCs emitted from forty Mediterranean plant species. They found that the two major compounds for *Thymus vulgaris* were *p*-cymene and thymol; the following ones were α -pinene and β -myrcene. We observe slight differences between their results and ours, especially for the major compound. These differences can be explained by the differences in the plant characteristics and in the experimental protocol. Indeed, they used plants in the field and ours were grown in a greenhouse and they used a sampling technique called Teflon branch enclosure. This technique is very efficient for natural conditions but is impossible to use while simulating the conditions of an ongoing forest fire.

The emissions behaviour of *Thymus vulgaris* heated needles is similar to the one of other heated vegetal species that can be found in the literature, with slight differences. The emissions of *Rosmarinus officinalis* also increase with temperature and we can see two

peaks: one around 393 K and one around 450 K [1,14]. The first one is due to the water evaporation process that increases the transport of VOCs and the second one corresponds to the boiling point of these VOCs. A similar evolution has been observed for *Pinus laricio* and *Pinus pinaster* by Barboni et al. [15] with an important decrease of the emissions after 448 K [15]. On the contrary, the emissions of *Cistus monspeliensis* species do not reach a maximum and start to be important at 473 K [15]. According to these studies, it appears that the behaviour of *Thymus vulgaris* is closer to the evolution of *Cistus monspeliensis* than to the one of *Rosmarinus officinalis*, *Pinus laricio* or *Pinus pinaster*. This behaviour probably depends on the boiling point value of the major compounds. Indeed, above the boiling point, thermal cracking phenomena can be observed. Oxygenated compounds, which are the major compounds of *Cistus monspeliensis* and *Thymus vulgaris*, have boiling point values much higher than monoterpenes. For instance, the boiling points of monoterpenes like α -pinene or β -myrcene are respectively 428 and 440 K, whereas the boiling point of thymol is 506 K [616]. For this study we chose *p*-cymene for our experiments because of the high temperature of fusion of thymol which makes the measurements impossible.

~ Spherical combustion chamber experiments

The characteristics of *p*-cymene/air laminar premixed flames are measured using a spherical combustion vessel. High purity *p*-cymene ($\geq 99\%$ pure from Sigma-Aldrich) is injected through a Coriolis mass flow meter. The effects of equivalence ratio (0.8 to 1.4) and unburned gas temperature (358 to 453 K) are investigated. This range of temperatures is similar to the one of the emission measurements. An electric fan, located inside the chamber, mixes all the gases to ensure homogenous mixtures *p*-cymene/air. The visualisation method is based on shadowgraph technique using an optical system with a high-speed digital camera. Then, we obtain temporal evolutions of *p*-cymene/air spherical flame expansion. Each experiment is performed in triplicate. A detailed description of the experimental protocol is given by Zhou et al. (2011) [17].

Results and discussion

~ Laminar burning speeds of *p*-cymene/air mixtures

The determination of laminar burning speeds of VOCs leads to a better understanding of their combustion and of their possible implication in accelerating forest fires. This section focuses on the determination of the laminar burning speeds s_u^0 of *p*-cymene/air mixtures from the unstretched premixed flame speeds s_b^0 and the expansion factor σ by means of the relation $s_u^0 = \sigma \cdot s_b^0$. As indicated above, there are few works in the literature dealing with VOCs combustion [5] and none about the detailed kinetic mechanism of *p*-cymene. Therefore, the expansion factor is evaluated using the adiabatic flame

calculation via the combustion reaction of *p*-cymene with air.

Analysis of experimental results for different equivalence ratios and preheat temperatures can lead to the derivation of a correlation that have the following form [18]:

$$s_u^0 [\text{cm/s}] = s_u^{0\text{ref}} \left(1 + \alpha_1 (1 - \Phi) + \alpha_2 (1 - \Phi)^2 \right) \left(\frac{T_u}{T_u^{\text{ref}}} \right)^{\alpha_3} \quad (4)$$

with $s_u^{0\text{ref}}$ the laminar burning speed at a reference point ($\Phi = 1$ and $T_u^{\text{ref}} = 358 \text{ K}$) given in cm/s, T_u (in K) the unburned gas temperature and α_1 , α_2 and α_3 constants. These coefficients are obtained by the least squares method applied to the experimental data of laminar burning speeds. Figure 4 presents laminar burning speeds of *p*-cymene/air flames as well as the results of this fitting based on a minimisation genetic algorithm (Goldberg, 1989). The coefficient values of the empirical correlation (4) are $\alpha_1 = -0.4749$, $\alpha_2 = -2.3632$ and $\alpha_3 = 1.9076$. This Figure exhibits clearly that the empirical correlation (4) is a good approximation of *p*-cymene experimental results for different temperatures with a coefficient of determination $R^2 = 0.9929$. As expected, measured laminar burning speeds increase with preheat temperature.

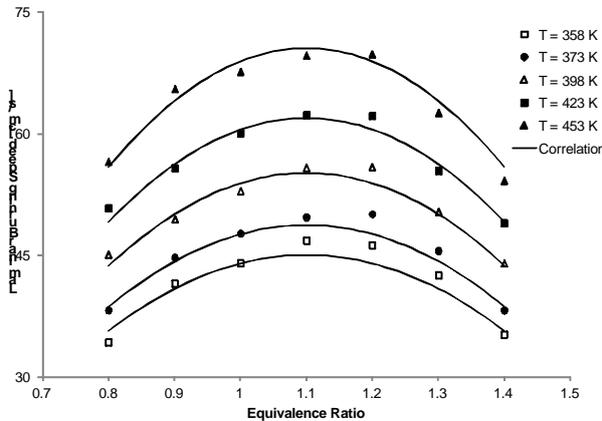


Figure 4. Experimental laminar burning speeds of *p*-cymene/air mixtures at different temperatures and their best-fit curves.

Figure 5 compares the laminar burning speeds of *p*-cymene obtained using the above correlation with the computed values of JP-10 and n-decane at the emission peak temperature. As indicated in the introduction, these last values are computed by means of the PREMIX code of the CHEMKIN package using respectively the San Diego detailed kinetic mechanism and the one of Honnet et al. (2009) [7]. This Figure shows that our measurements are close to n-decane computational values for equivalence ratios around the stoichiometry and very close to JP-10 computational values for rich mixtures. For lean mixtures, our measured values are in

better agreement with computed results of n-decane than with JP-10 ones.

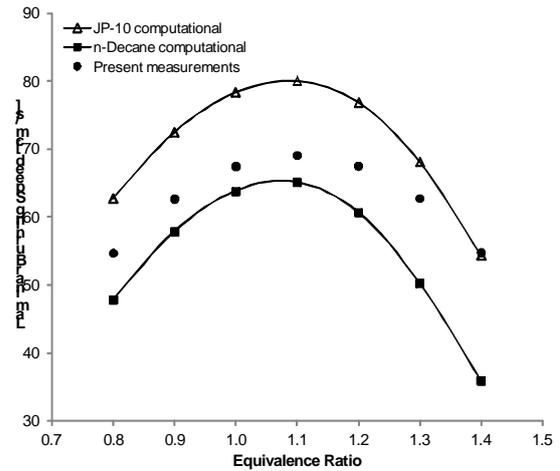


Figure 5. Measured laminar burning speeds of *p*-cymene/air mixtures compared with computed ones of JP-10 and n-decane at 448 K.

Figure 6 compares measured laminar burning speeds of *p*-cymene/air mixtures at 398 K to experimental data of α -pinene [1], to those of iso-octane and ethylbenzene [19] obtained using their empirical correlation as well as to the values of n-decane [20] at 400 K. This figure shows that laminar burning speeds of α -pinene are close to our values from lean mixtures up to $\Phi = 1.1$. Concerning ethylbenzene laminar premixed flame speeds, we can say that they are in good agreement with our measured data for equivalence ratios 1, 1.1 and 1.2. Before these equivalence ratios, they underestimate *p*-cymene values and overestimate them after $\Phi = 1.2$. Iso-octane laminar premixed flame speeds are very close to our values for an equivalence ratio of 1.4 and underestimate them for other equivalence ratios. Singh et al. (2011) [20] experimental values of n-decane overestimate *p*-cymene values up to $\Phi = 1.1$ and underestimate them after this equivalence ratio with slighter differences at equivalence ratios 1.1, 1.2 and 1.3.

~ Markstein lengths of *p*-cymene/air mixtures

Markstein length is an important parameter to determine stretch influence on the flame and to characterize flame stability. Our results show that Markstein length decreases with the increase of equivalence ratio and there is a low effect of the preheat temperature. The same little variation for different preheat temperatures has already been observed by Gu et al. (2011) [21] for tert-butanol/air mixtures. We can also notice that there is a sign change between the equivalence ratios 1.3 and 1.4 for all temperatures except 358 K, which corresponds to the stable to unstable flames transition. We can also notice that the hydrocarbon studied here is a heavy molecule and presents a different behavior compared to lighter fuels [22,23]. There are few Markstein lengths data reported in the literature for heavy molecules. Therefore, measured values of *p*-

cymene at 398 K are compared to those of α -pinene [1] at the same temperature and to the ones of iso-octane (Halter et al., 2010) and n-decane [20] at 400 K. Figure 7 illustrates this comparison.

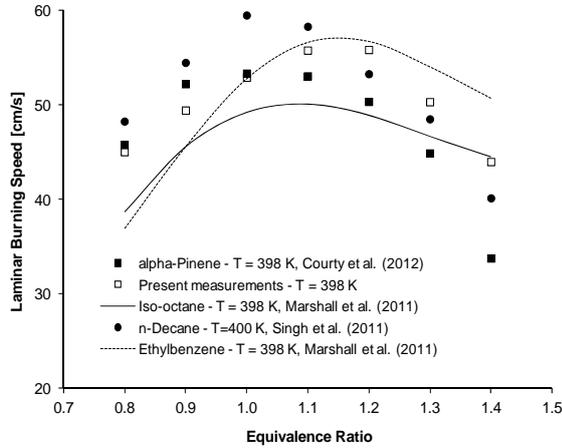


Figure 6. Laminar burning speeds of p -cymene/air mixtures compared with experimental ones of α -pinene, iso-octane and ethylbenzene at 398 K and n-decane at 400 K.

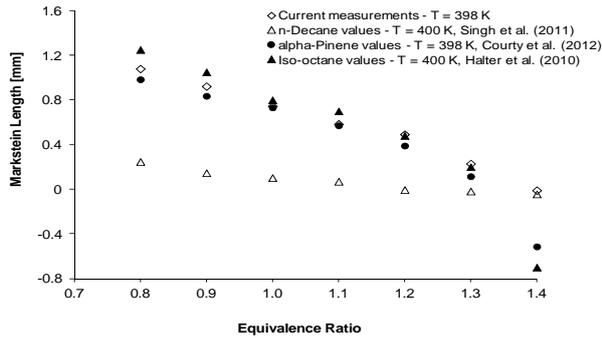


Figure 7. Markstein lengths of p -cymene/air mixtures compared to the ones of α -pinene at 398 K, iso-octane and n-decane at 400 K.

We can observe from Figure 7 that these four fuels present the same tendency with a decreasing steeper for equivalence ratios higher or equal to 1.3 for α -pinene and iso-octane. Unlike these two fuels, p -cymene and n-decane have a linear decreasing with a higher slope for the studied hydrocarbon. At 398 K, this linear relation can be expressed as:

$$L_b [\text{mm}] = 0.75 - 1.75(\Phi - 1) \quad (5)$$

with a coefficient of determination $R^2 = 0.9857$. The transition between stable and unstable flames occurs after $\Phi = 1.3$ for p -cymene, iso-octane and α -pinene but after $\Phi = 1.1$ for n-decane. We can also say that p -cymene, iso-octane and α -pinene have close Markstein length values in the stable flame domain (i.e. from lean mixtures to $\Phi = 1.3$) and they are higher than the n-decane ones.

~ Flame thicknesses of p -cymene/air mixtures

The laminar flame thickness is an important parameter to determine as it is useful to characterize hydrodynamic instabilities of the flames and its calculation is based on equation (3).

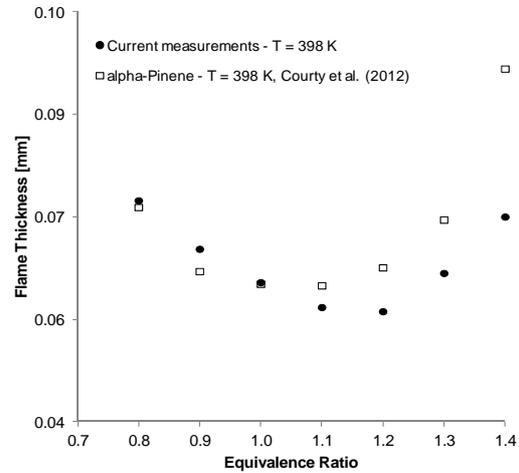


Figure 8. Flame thicknesses of p -cymene/air mixtures compared to those of α -pinene at 398 K.

Our results show that flame thicknesses of p -cymene are insensitive to the variation of unburned gas temperature and have minimum values at $\Phi = 1.2$. Figure 8 compares our measurements to the flame thicknesses of α -pinene [1] at 398 K. Our measured values are close to α -pinene data from lean mixtures up to $\Phi = 1.1$ and equal at the stoichiometry. For equivalence ratios higher than 1.1, flame thicknesses of p -cymene are lower than those of α -pinene. Let us notice that the flame thickness of p -cymene is minimal for an equivalence ratio of 1.2 whereas it is minimal at $\Phi = 1.1$ for α -pinene.

Conclusion

Some works in the forest fires literature have indicated the possible implication of VOCs emitted by fire heated vegetation in accelerating forest fires. Flames of such phenomenon can be seen as premixed flames and not as diffusion flames like usual wildland fires. The present study deals with the experimental determination of the combustion characteristics of p -cymene: laminar burning speeds, Markstein lengths and flame thicknesses. This compound is emitted by *Thymus vulgaris* shrubs and its emission peak is identified at 443 K. Laminar combustion characteristics are determined using the spherical expanding flame methodology based on a nonlinear model. The obtained results are compared to literature experimental data of fuels with large molecular weights and to the computed values of JP-10 and n-decane obtained by means of the PREMIX code of the CHEMKIN package. Measured values of combustion characteristics of p -cymene will be useful to integrate the potential VOCs ignition hypothesis in physical modelling of forest fires.

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