

Quantitative NO and Temperature Measurements in a Pulsed Laminar Hydrogen–Air Flame

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Keywords: Combustion, NO-PLIF, NO pollutant.

ABSTRACT

The transition to hydrogen as a carbon-free fuel introduces new combustion challenges, primarily nitrogen oxide (NO_x) emissions driven by elevated flame temperatures. While nitric oxide laser-induced fluorescence (NO-LIF) is a well-established technique to study NO formation in steady flows, quantitative planar measurements in periodically pulsating environments remain limited. This work demonstrates phase-resolved, quantitative multi-line planar LIF (NO-PLIF) to map temperature fields and absolute NO concentrations in a pulsed, laminar premixed conical H₂-air flame ($\phi = 0.8$). Controlled 10 Hz velocity modulations were introduced using a proportional valve. Temperature fields were resolved using multi-line NO-PLIF thermometry, while absolute NO fields were quantified on the Q₂+Q₁₂(27) transition calibrated via controlled NO seeding. Phase-locked measurements revealed that while local peak temperatures (~ 2000 K) and peak NO levels (~ 18 ppm) remain nearly identical between steady and pulsed conditions, significant structural widening occurs during the near-peak velocity phase. Area-weighted spatial integration shows that this widening increases the high-temperature reaction volume, driving the cross-sectional average NO concentration to be higher at mid-flame heights compared to the steady flame. These high-fidelity data provide a baseline for validating unsteady hydrogen combustion models.

1. Introduction

In recent years, hydrogen has emerged as a promising carbon-free alternative to replace traditional hydrocarbon fuels. However, hydrogen combustion presents new challenges, most notably the formation of nitrogen oxides (NO_x) resulting from the high temperatures characteristic of hydrogen flames. To characterize these emissions without disturbing the flow field, non-intrusive optical diagnostics are essential. To this end, NO laser-induced fluorescence (NO-LIF) has become a widely adopted technique for studying NO emissions in a broad range of flows, including laminar hydrogen flames (Meulemans et al., 2024; Mitra et al., 2025; Durocher et al., 2021, 2023). Because

LIF imaging is sensitive to both local species concentration and thermodynamic properties like temperature, it provides spatially resolved, instantaneous measurements of these quantities.

While NO-LIF is well-established for steady-state environments, applying the technique to periodically pulsating flows is much less common. Foundational studies investigating flame dynamics on pulsed flames have relied on techniques such as CH* or OH* chemiluminescence, paired with Laser Doppler Velocimetry (LDV) (Ducruix et al., 2000; Schuller et al., 2003) and Particle Image Velocimetry (PIV) (Birbaud et al., 2007). Nonetheless, Ratner et al. (2002) implements planar LIF (PLIF) to measure fields of NO in an acoustically forced CH₄-N₂ jet flame, observing that NO levels increase for frequencies that exhibit stronger acoustic coupling to the flame. To expand upon these planar measurements toward absolute metrics, this work presents multi-line NO-PLIF phase-resolved temperature and quantitative NO measurements in a pulsed premixed conical hydrogen–air flame. By using a proportional valve to introduce controlled velocity modulations, we establish a repeatable, time-periodic fluctuation to yield a high-fidelity baseline dataset.

2. Experimental setup

2.1. Burner configuration

Experiments were conducted on pulsed laminar premixed hydrogen-air flames stabilized at atmospheric pressure on a coflow burner. The burner, shown in Figure 1, consists of two concentric sections, both ending in converging nozzles. The outlet diameters are 4 mm and 19 mm for the inner and outer sections, respectively, with a 1 mm wall thickness in the inner section. The premixed hydrogen-air mixture is injected into the inner section through inlets located at the bottom of the burner. Before reaching the nozzle, the central flow passes through a conditioning section, identified as (1) in Figure 1, consisting of small glass beads and a honeycomb structure to ensure a uniform laminar velocity profile.

The hydrogen and air flow rates were regulated using Bronkhorst mass flow controllers. Flame pulsation was generated by sinusoidal modulation of the inlet velocity of the premixed hydrogen-air mixture using a proportional valve (Moog). The imposed forcing signal operated at 10 Hz with an amplitude of 4.5 V. The investigated flame corresponds to an equivalence ratio of $\phi = 0.8$, a thermal power of 220 W, with a hydrogen flow rate of 6.6 g/h. The reactants were injected into the burner at room temperature. Based on mass flow meter accuracies, the uncertainty in the equivalence ratio is $\pm 7.8 \times 10^{-3}$ and $\pm 1.1 \times 10^{-2}$ for the non-seeded and seeded flame, respectively.

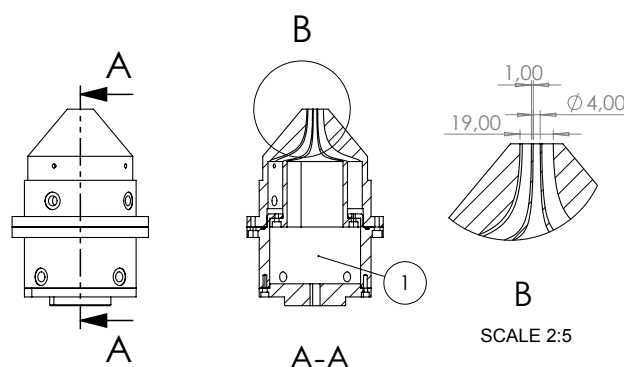


Figure 1. Burner schematic.

2.2. Laser and optical setup

Quantitative NO-PLIF measurements were performed using a tunable dye laser (Radiant Dyes NarrowScan HP), pumped by the second harmonic (532 nm) of an injection-seeded Nd:YAG laser (Continuum PL-PLUS, 10 ns, 10 Hz). The dye laser output at approximately 615 nm was frequency-mixed with the third harmonic (355 nm) of the Nd:YAG laser in a BBO crystal to generate an ultra-violet laser beam at around 225 nm for NO excitation. Four Pellin-Broca prisms were used to isolate the desired UV wavelength. The laser energy was attenuated to ensure linear LIF conditions, and the beam was shaped into a 40 mm-high vertical laser sheet using cylindrical and spherical lenses, enabling two-dimensional measurements.

The NO fluorescence signal was collected at 90° from the excitation axis using a two-dimensional imaging configuration. The fluorescence signal was directly recorded using an ICCD camera (Princeton Instruments PM4-1024-F) equipped with a 100 mm UV lens and a bandpass filter (Thorlabs FGUV5S), collecting the accessible NO vibrational bands while also permitting OH* chemiluminescence imaging. The spatial resolution of the imaging system was approximately $40 \mu\text{m}/\text{pixel}$. Image acquisition was synchronized with the imposed flame pulsation, allowing phase-resolved measurements. Laser pulse-to-pulse energy fluctuations were monitored using a photodiode connected to an oscilloscope, while wavelength variations were tracked using a wavemeter (HighFinesse WS/5) measuring the residual dye laser wavelength exiting the Pellin-Broca prism assembly.

3. Experimental methods

3.1. Temperature measurements

Temperature distributions were obtained using multi-line NO-PLIF thermometry in flames seeded with 1000 ppm NO, allowing for measurements in both the unburnt and burnt gas regions. The

laser excitation wavelength range was selected based on the study by Bessler & Schulz (2004), and has also been adopted in subsequent works (Kronemayer et al., 2005; Meulemans et al., 2024). This range was chosen as an optimal compromise, balancing the spectral sensitivity required for hydrogen flame temperatures with practical experimental constraints such as rapid scanning. LIF excitation spectra were obtained by scanning the laser from 225.06 to 225.12 nm (air), using 0.002 nm increments. At each wavelength step, a two-dimensional single-shot fluorescence image was recorded, yielding 270 images per scan over approximately one minute. Eighteen scans were recorded and averaged for each flame.

From the recorded planar fluorescence images, excitation spectra were independently extracted at each pixel, allowing for spatially resolved temperature fields. Temperature at each pixel was determined using a custom MATLAB routine, which compared the experimental NO-LIF excitation spectrum with a database of simulated spectra generated using LIFBASE (Luque & Crosley, 1999). The algorithm identified the temperature that minimized the squared Euclidean distance between the simulated and measured spectra. Before performing the fitting procedure, the experimental spectra were corrected for pulse-to-pulse laser energy fluctuations and baseline offsets, and were filtered to reduce noise. The custom-fitting routine was validated using selected experimental spectra by comparing results with those obtained from the Thermo NO-LIF software developed by Foo et al. (2020). Under the present experimental conditions, the temperature measurements exhibited a variability between 6 and 7.8 %.

3.2. Concentration measurements

NO concentrations in flames were quantified using NO-PLIF. The NO molecule was excited on the $Q_2+Q_{12}(27)$ transition of the $A2\Sigma^+-X2\Pi(0,0)$ vibrational band at 225.509 nm (air). For each investigated flame condition, LIF images were recorded at on- and off-resonance wavelengths, each consisting of 50 images with 100 on-chip accumulations per image to improve signal-to-noise ratio. Before NO quantification, raw images were corrected for shot-to-shot laser energy fluctuations and spatial inhomogeneities in the laser sheet. These corrections were based on the Rayleigh-scattering signal observed outside the flame region in the raw fluorescence images, where the gas composition remains constant and at room-temperature. Due to the selected camera parameters, the Rayleigh signal was sufficiently strong to extract the laser sheet profile and the relative laser pulse energy per image, which were used to normalize the LIF images.

The NO fluorescence signal was extracted from the averaged corrected on- and off-resonance images by subtracting the off-resonance contribution. Absolute NO concentrations were determined through comparison with a calibrated reference flame ($\phi = 0.9$ H₂-O₂-N₂). Calibration was performed by seeding known NO concentrations (0-100 ppm) into the reference flame and measuring the corresponding PLIF response, a technique widely used in the literature (Meulemans et al., 2025; Brackmann et al., 2020; Mulla et al., 2019), allowing quantification of the native NO in the reference

flame. The NO concentration in the investigated flames was subsequently given by:

$$X_{\text{flame}} = X_{\text{ref}} \cdot \frac{S_{\text{flame}}}{S_{\text{ref}}} \cdot \frac{f_B(T_{\text{ref}})}{f_B(T_{\text{flame}})} \cdot \frac{T_{\text{flame}}}{T_{\text{ref}}}, \quad (1)$$

where subscripts *flame* and *ref* stand for the investigated and reference flames, respectively, X is the NO molar fraction (ppm), S is the intensity of the fluorescence signal (a.u.), f_B is the Boltzmann fraction, and T is the temperature. The temperature fields previously measured following the procedure described in Section 3.1 were used for both flames. For each flame condition, six independent measurements were acquired and subsequently averaged. The detection limit for the current NO-PLIF configuration was determined to be approximately 0.33 ppm, evaluated from the background noise floor (2σ) of the corrected signal images. Furthermore, the uncertainty for the absolute NO measurements is estimated at 20% across all investigated conditions.

4. Results

4.1. Phase-resolved OH* chemiluminescence measurements

To characterize the flame response to the imposed flow pulsation, OH* chemiluminescence images were recorded together with the forcing and camera trigger signals. The OH* emission was used as a marker of the heat release zone to track the flame evolution throughout the oscillation cycle. The images were recorded at different instants of the forcing cycle and were associated with their respective phases using the camera signal. This procedure allowed the reconstruction of the mean evolution of the flame emission over one complete oscillation period, as observed in Figure 2.

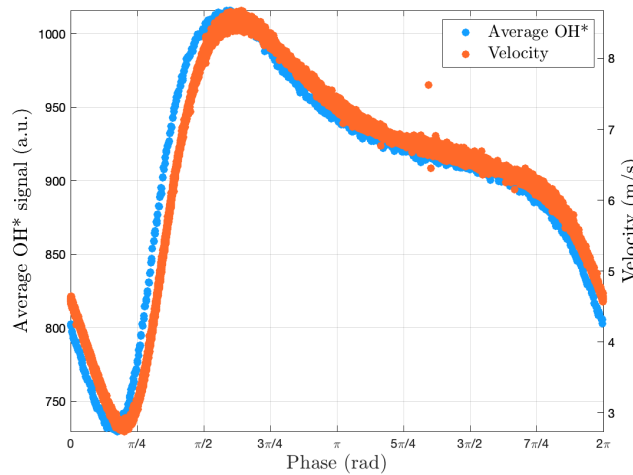


Figure 2. Evolution of the averaged OH* chemiluminescence intensity as a function of the phase of the forcing cycle over one complete oscillation period.

In Figure 2, the flame response is presented alongside the gas velocity at the burner outlet, measured using a hot-wire probe. It can be observed that the flame response follows the same phase dynamics as the velocity variations imposed by the proportional valve. The averaged velocity of the pulsed flow was found to be 6.38 m/s, representing a minor deviation ($<1.5\%$) from the steady-state flame velocity of 6.48 m/s. This small discrepancy can be attributed to the fact that the hot-wire measurement was taken 3 mm above the burner outlet. The phase-resolved response was subsequently analyzed to identify characteristic instants in the forcing cycle. Based on this analysis, the phases at 0.2 rad and 1.56 rad were selected for further investigation, as they represent two contrasting states: the initial acceleration phase and the near-peak velocity regime, respectively. The OH^* chemiluminescence images corresponding to the non-pulsed flame and the two representative phases are shown in Figure 3, where it can be observed that at 0.2 rad the flame is shorter than the non-pulsed case due to the lower local velocity, whereas at 1.56 rad, the near-peak velocity results in a significantly taller flame structure.

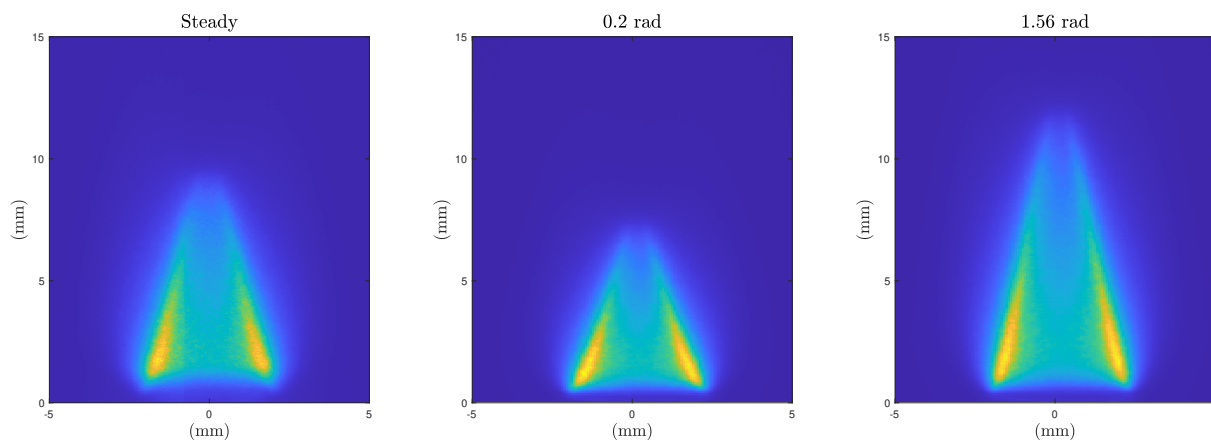


Figure 3. Average OH^* chemiluminescence images of the steady flame and of the pulsed flame at the selected forcing phases of 0.2 rad and 1.56 rad.

4.2. Temperature and NO distributions

Figure 4 presents the two-dimensional spatial distributions of temperature and NO molar fraction for the steady flame alongside the two characteristic phases (0.2 rad and 1.56 rad) of the pulsed flame. To provide a quantitative comparison, localized radial profiles extracted at heights of $H = 10, 20$, and 27 mm above the burner outlet are plotted below the maps.

In all three investigated cases, a maximum temperature of approximately 2000 K is consistently reached. Consequently, these identical temperature peaks yield similar peak NO values close to 18 ppm across all conditions. However, significant differences in the shape of the flame are observed, particularly for the flame at 1.56 rad, which exhibits a noticeably wider structure in both temperature and NO distributions, resulting in broader radial profiles at all investigated heights,

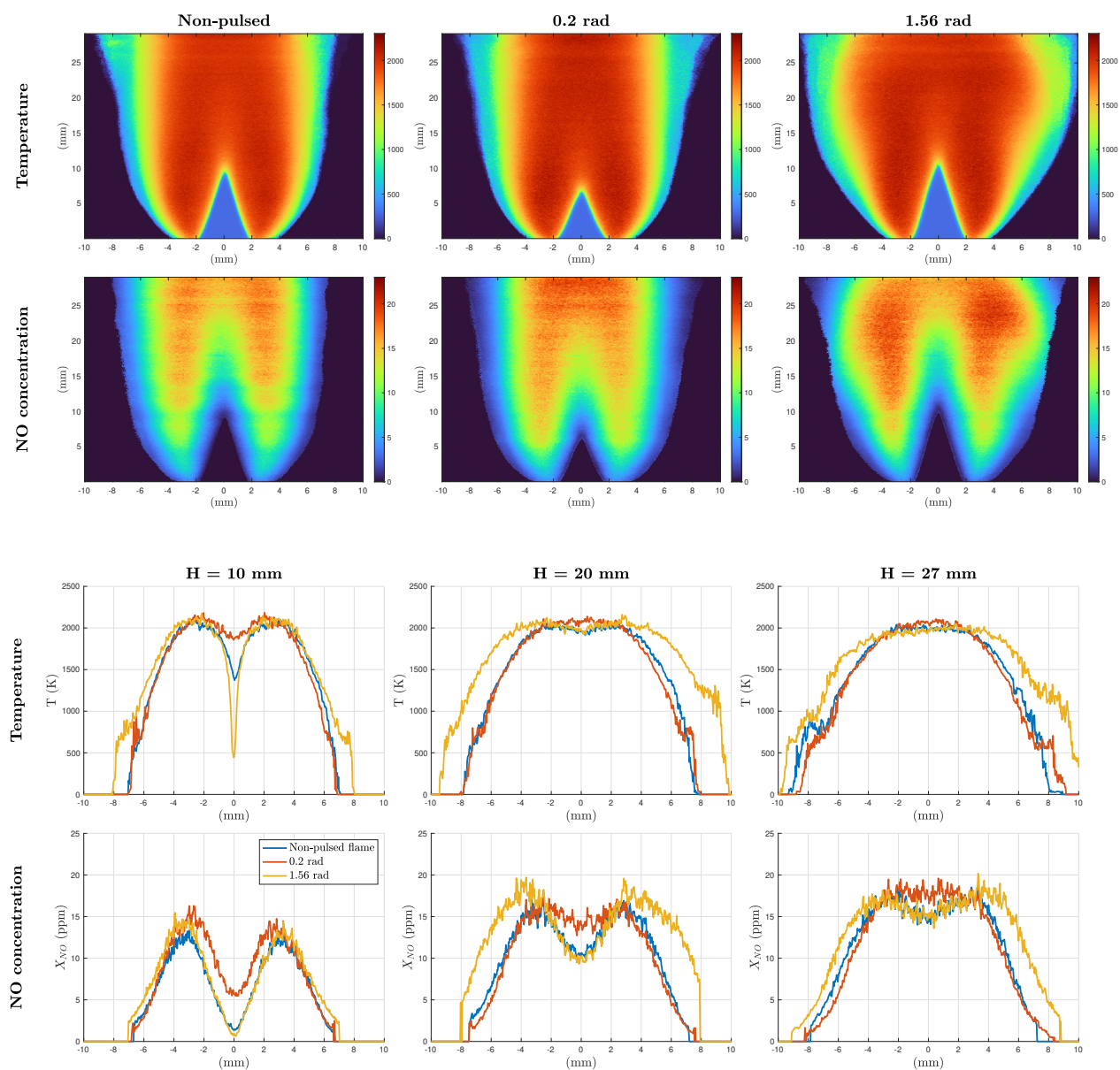


Figure 4. Temperature and NO concentration fields for the non-pulsed flame and for the pulsed flame at phases 0.2 rad and 1.56 rad. Radial profiles extracted at heights $H = 10$, 20, and 27 mm are shown below.

especially pronounced at $H = 20$ mm. This indicates that pulsation primarily affects the spatial distribution of the flame rather than the maximum values attained.

To further investigate the impact of the flow pulsation on total NO production, the two-dimensional concentration fields were integrated at each height H . A cylindrical integral was performed on the radial profiles to calculate the area-weighted average NO concentration across the flame cross-section. Furthermore, the radial length used for integrating is changing with the conditions, as the flame has a larger radial component at 1.56 rad compared to 0.2 rad. Assuming flame axisymmetry, this process was performed independently for the left and right sides of the profiles, and the resulting values were averaged.

Figure 5 presents the evolution of the area-weighted average NO concentration (left) and the corresponding flame width (right) as a function of height. In the near-burner region ($H < 5$ mm), all cases exhibit a similar initial increase in NO. However, between $H = 5$ mm and $H = 10$ mm, the 0.2 rad phase shows a more pronounced rise in average NO compared to the other conditions. This is attributed to the earlier closure of the flame cone at this phase, where the average is calculated across a cross-section fully occupied by hot combustion products. Figure 5 (right) shows that the flame width for the 0.2 rad phase remains close to that of the steady-state flame across the entire height. Despite this structural similarity, the 0.2 rad flame results in lower average NO concentration beyond $H = 15$ mm compared to the steady case. This occurs due to the combination of a slight increase in the flame width relative to the steady-state flame and slightly lower local NO values at the flame periphery that heavily weight the average, notwithstanding the higher NO concentrations measured near the centerline.

Further downstream, the flame at 1.56 rad reaches a higher peak concentration of approximately 10 ppm near $H = 23$ mm. This higher average is driven by the significant radial expansion of the flame at this phase, as observed in Figure 5 (right), which increases the total volume of the reaction zone and leads to a greater absolute production of NO. Beyond this peak, the average

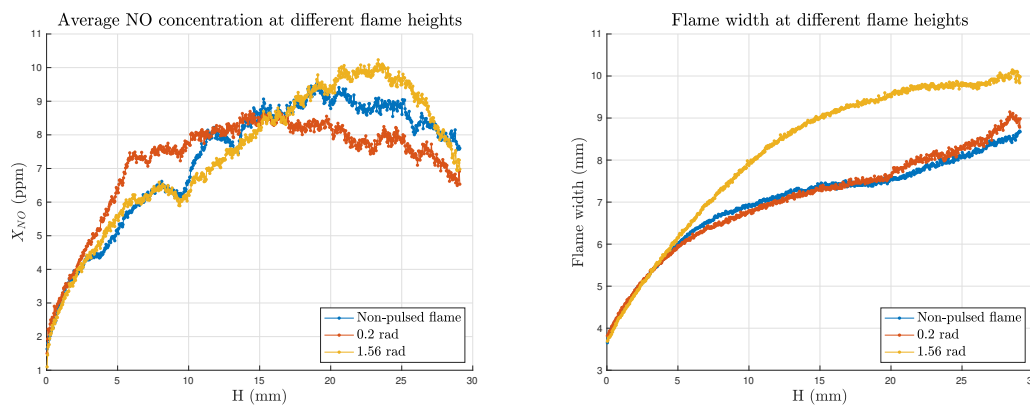


Figure 5. Area-weighted average NO concentration (X_{NO}) as a function of height H (left) and corresponding flame width (right) for the non-pulsed flame and the two investigated pulsed phases (0.2 rad and 1.56 rad).

NO concentration drops. As shown by the radial profiles in Figure 4, this drop occurs because the locations of the NO peaks shift inward toward the centerline, while the flame maintains a wider structure than the other cases.

These results demonstrate that the evolution of the average NO concentration is primarily governed by the phase-dependent flame structure rather than changes in local NO peak values. The variations observed throughout the cycle result from the interaction between the flame's radial expansion and the shifting distribution of the NO profiles. Ultimately, the total pollutant emission trend is dictated by how these structural changes interact with the area-weighted averaging across the flame cross-section.

5. Conclusions

This work reports the implementation of phase-resolved multi-line NO-PLIF thermometry and quantitative NO concentration measurements to investigate a pulsed premixed conical hydrogen–air flame. By utilizing a proportional valve to introduce controlled 10 Hz velocity modulations, a repeatable, time-periodic fluctuation was established and monitored via phase-locked OH* chemiluminescence tracking to select two contrasting states to study further.

The phase-resolved two-dimensional distributions and localized radial profiles revealed that the maximum temperature consistently reaches approximately 2000 K, yielding nearly identical local peak NO concentrations of approximately 18 ppm across the steady flame and both investigated forcing phases (0.2 rad and 1.56 rad). This suggests that the flow pulsation primarily affects the spatial distribution and shape of the flame rather than the maximum values attained. At the near-peak velocity phase of 1.56 rad, the flame exhibits a noticeably wider structure that increases the effective volume of the NO-producing region, leading to higher area-weighted average NO concentrations. Conversely, the initial acceleration phase at 0.2 rad results in a lower average NO concentration than the steady flame, driven by lower NO levels at the flame periphery. The resulting phase-resolved dataset provides a high-fidelity baseline suitable for the validation of unsteady combustion models.

Acknowledgements

This project has received funding from the European Climate and Environment Executive Agency under the European Union's Horizon 2021 research and innovation program (Grant Agreement 101056865, HESTIA).

References

- Bessler, W. G., & Schulz, C. (2004). Quantitative multi-line NO-LIF temperature imaging. *Applied Physics B*, 78(5), 519–533.
- Birbaud, A.-L., Durox, D., Ducruix, S., & Candel, S. (2007). Dynamics of confined premixed flames submitted to upstream acoustic modulations. *Proceedings of the Combustion Institute*, 31(1), 1257–1265.
- Brackmann, C., Methling, T., Lavadera, M. L., Capriolo, G., & Konnov, A. A. (2020). Experimental and modeling study of nitric oxide formation in premixed methanol+ air flames. *Combustion and Flame*, 213, 322–330.
- Ducruix, S., Durox, D., & Candel, S. (2000). Theoretical and experimental determinations of the transfer function of a laminar premixed flame. *Proceedings of the combustion institute*, 28(1), 765–773.
- Durocher, A., Meulemans, M., Bourque, G., & Bergthorson, J. M. (2023). Nitric oxide concentration measurements in low-temperature, premixed hydrogen-air stagnation flames at elevated pressures. *Proceedings of the Combustion Institute*, 39(1), 541–550.
- Durocher, A., Meulemans, M., Versailles, P., Bourque, G., & Bergthorson, J. M. (2021). Back to basics—no concentration measurements in atmospheric lean-to-rich, low-temperature, premixed hydrogen–air flames diluted with argon. *Proceedings of the Combustion Institute*, 38(2), 2093–2100.
- Foo, K. K., Lamoureux, N., Cessou, A., Lacour, C., & Desgroux, P. (2020). The accuracy and precision of multi-line NO-LIF thermometry in a wide range of pressures and temperatures. *Journal of Quantitative Spectroscopy and Radiative Transfer*, 255, 107257.
- Kronmayer, H., Bessler, W., & Schulz, C. (2005). Gas-phase temperature imaging in spray systems using multi-line no-lif thermometry. *Applied Physics B*, 81(8), 1071–1074.
- Luque, J., & Crosley, D. R. (1999). *LIFBASE: Database and Spectral Simulation Program (Version 1.5)* (Tech. Rep. No. MP 99-009). SRI International. (SRI International Report)
- Meulemans, M., Durocher, A., Bourque, G., & Bergthorson, J. M. (2024). NO measurements in high temperature hydrogen flames: The crucial role of the hydrogen oxidation chemistry for accurate NO predictions. *Combustion and Flame*, 261, 113279.
- Meulemans, M., Durocher, A., Versailles, P., Bourque, G., & Bergthorson, J. M. (2025). Calibration techniques for quantitative NO measurement using Laser-Induced Fluorescence. *Journal of Quantitative Spectroscopy and Radiative Transfer*, 330, 109221.

- Mitra, T., Lamoureux, N., & Desgroux, P. (2025). Understanding the chemical pathways of NO formation in low-pressure burner stabilized premixed lean-to-rich hydrogen flames. *Combustion and Flame*, 277, 114192.
- Mulla, I. A., Godard, G., Cabot, G., Grisch, F., & Renou, B. (2019). Quantitative imaging of nitric oxide concentration in a turbulent n-heptane spray flame. *Combustion and Flame*, 203, 217–229.
- Ratner, A., Pun, W., Palm, S. L., & Culick, F. E. (2002). Phase-resolved NO planar laser-induced fluorescence of a jet flame in an acoustic chamber with excitation at frequencies < 60 Hz. *Proceedings of the Combustion Institute*, 29(1), 85–90.
- Schuller, T., Durox, D., & Candel, S. (2003). A unified model for the prediction of laminar flame transfer functions: comparisons between conical and V-flame dynamics. *Combustion and flame*, 134(1-2), 21–34.