



Assessment of chemical scalars for heat release rate measurement in highly turbulent premixed combustion including experimental factors

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ABSTRACT

This study uses direct numerical simulation (DNS) data from two turbulent premixed flames to explore the variables involved in experimental measurement of the heat release rate, \dot{h} . Several commonly used markers of \dot{h} are compared, namely the CH, OH, and HCO radicals, as well as the spatial overlap of OH and CH₂O and of H-atom and CH₂O. The DNS data were also used to generate synthetic Planar Laser-Induced Fluorescence (PLIF) images that account for confounding experimental factors such as laser sheet thickness, optical blur, and shot noise. The overlap of OH and CH₂O and the HCO radical appear to be the most promising markers of \dot{h} , as they show excellent correlation with the high-intensity heat release. The synthetic-PLIF images show that increasing the laser sheet thickness and optical blur progressively de-correlates the measurement and the underlying DNS structures. However, for a sufficiently thin laser sheet ($\delta_{sheet}/\delta_L \lesssim 1$), the correlation between the synthetic-PLIF and DNS remains strong even with significant optical blur. Similar results are observed in the measured layer thickness and the peak-to-peak distance between the PLIF structure and the heat release layers. The resolution of \dot{h} features was evaluated by measuring the average turbulent flame length, the spatial power spectral density, and the flame surface curvature. As the laser sheet thickness or optical blur is increased, the measured flame surface area progressively decreases, less fine-scale features are resolved, and regions of high curvature are eroded. Shot noise strongly affects spatial correlation with the DNS data and the layer thickness, but has relatively small impact on the turbulent flame length. Significantly greater deviations are observed for the overlap techniques requiring simultaneous measurement from two cameras, as optical blur and/or shot noise present in the two camera systems will both influence the resulting overlapped PLIF signal.

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1. Introduction

The rate of sensible enthalpy release (\dot{h}) is one of the most fundamental quantities in combustion, but remains difficult to assess experimentally [1]. In general, attempts are made to infer \dot{h} through measurable chemical markers that are linked in some way to exothermic chemical reactions [2]. A myriad of methods (reviewed below) have been proposed that range in measurement complexity, accuracy, and the features of the \dot{h} field that they attempt to capture. Assessments of several of these markers have been performed using direct numerical simulations (DNS)

of flame/vortex interactions and turbulent flames in the flamelet regime, assuming a perfect measurement system [3–5]; the focus typically has been on the local correspondence between the proposed chemical markers and \dot{h} . However, such assessments do not address many confounding experimental factors (i.e. realistic signal generation and detection), how data typically are used by experimentalists, or deviations in chemical structure induced by high turbulence levels. This paper uses two recent DNS data sets to address these issues for the case of highly-turbulent atmospheric-pressure methane/air flames.

The particular focus is on signals that would be obtained through planar laser-induced fluorescence (PLIF) of naturally occurring chemical species. Such species, individually or in combination, may reflect \dot{h} both directly through a local instantaneous correlation, or by having some aspect that mimics the topology of the \dot{h} field [2,5]. The latter, while not providing direct quantification of \dot{h} , can yield important information regarding surface area,

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curvature, etc. Specifically, we consider fields of formyl (HCO), the overlap between formaldehyde (CH₂O) and hydroxyl (OH), the overlap between formaldehyde and atomic hydrogen (H), and the methylidyne (CH) radical. The gradient of OH, which is typically very large near the flame front [1], also is evaluated as a topological marker of the heat release. We also directly assess the influence of experimental factors on measurements of an ideal marker, viz. the \dot{h} field itself.

The formyl radical (HCO) has a number of properties that make it an attractive marker of the heat release field in combustion processes. It is produced through the consumption of formaldehyde (CH₂O) that occurs at the start of the high temperature inner layer, it rapidly reacts with a large number of flame-zone intermediates, and it plays an important role in the CO-production pathway. In a study comparing numerous chemical markers of heat release in nitrogen diluted premixed V-flames, Najm et al. [2] demonstrated the acquisition of temporally-averaged HCO-PLIF, and verified through numerical simulations that formyl has high correlation with the heat release field for a variety of curvatures and strain rates. However, it was noted that single-shot measurement of HCO-PLIF in turbulent flames was not feasible at the time due to low HCO concentrations. More recent work using an Alexandrite laser with a broad spectral bandwidth and long pulse duration has demonstrated that single-shot measurement of HCO-PLIF is possible, and makes this a promising tool for describing heat release fields [6]. In a follow up study, strategies for obtaining HCO-PLIF in turbulent premixed flames were discussed, including optimal transitions, spectral interference, and a realistic range of conditions for measurement [7]. In particular, it was observed that large hydrocarbon species in rich flames produce considerable interference that cannot be eliminated, and therefore limits the application of HCO-PLIF to lean premixed flames.

Recognizing the strong correlation of the formyl radical with exothermic regions, but noting the above problems inherent to HCO-PLIF detection, Paul and Najm [8] searched for alternative reaction pathways that might correlate with HCO-production. They note that, unfortunately, the attributes that are likely to make a particular species a strong tracer of the heat release regions (formation at a relatively moderate rate followed by rapid consumption through reaction or quenching) also act to limit concentrations, often below detectable levels. Therefore, measurements of the heat release field using PLIF of a single species are inherently difficult to obtain. An alternative approach is to measure the reaction rate of some pathway that leads to HCO production. The reaction $\text{OH} + \text{CH}_2\text{O} \rightarrow \text{H}_2\text{O} + \text{HCO}$ is one such pathway, and was first investigated in nitrogen-diluted premixed laminar V-flames [8].

Through Arrhenius kinetics, the forward reaction rate will be proportional to the product of the respective number densities of OH and CH₂O, as well as a term which exhibits a functional dependence on temperature. The particular temperature dependence of the term will depend on the selected transitions of the excited species; however, it is possible to select transitions such that the temperature dependence mimics that of the rate constant $k(T)$. Over the narrow range of temperatures in which OH and CH₂O simultaneously exist, it was shown that the product of OH and CH₂O number densities, viz. $[\text{OH}] \times [\text{CH}_2\text{O}]$, is approximately equal to the forward reaction rate of the two species. Simulations of 1D laminar flames by Paul and Najm [8] verified that the reaction of OH and CH₂O has a high spatial correspondence with the \dot{h} field in premixed flames. Simultaneous OH and CH₂O PLIF was then applied in premixed laminar flames subjected to an isolated line-vortex pair, and the pixel-by-pixel product of the two PLIF images was obtained to produce a qualitative measure of the heat release field. Ayoola et al. [9] explored the effects of strain rate and curvature on the $[\text{OH}] \times [\text{CH}_2\text{O}]$ overlap in lean premixed ethylene flames stabilized between opposed jets and with a bluff body. They

showed that the metric tends to correlate better with \dot{h} when the strain rate was increased to near global extinction.

An alternative pathway for HCO production is the reaction $\text{H} + \text{CH}_2\text{O} \rightarrow \text{H}_2 + \text{HCO}$. In a numerical study comparing the correlation between several reaction pathways and the heat release rate, Najm et al. [5] demonstrated that $[\text{HCO}]$, $[\text{OH}] \times [\text{CH}_2\text{O}]$, and $[\text{H}] \times [\text{CH}_2\text{O}]$ all performed similarly. However, it was observed that whereas the former two proxies exhibited large variations with different heat release pathways, $[\text{H}] \times [\text{CH}_2\text{O}]$ was less sensitive. It therefore was suggested that if H-atom could be easily detected, it would be a more accurate marker of the heat release than the OH-based metric. Unfortunately, atomic hydrogen is very difficult to attain experimentally, and makes the technique somewhat impractical.

In a work comparing several potential heat release markers, Mulla et al. [10] performed one-dimensional H-atom-LIF (utilizing a two-photon excitation scheme) simultaneous with formaldehyde PLIF. Experimental measurements of $[\text{H}] \times [\text{CH}_2\text{O}]$ were compared with the $[\text{OH}] \times [\text{CH}_2\text{O}]$ signal, as well as with one-dimensional simulations. The results confirmed that the $[\text{H}] \times [\text{CH}_2\text{O}]$ method is feasible as an alternative marker of the heat release rate. Although the $[\text{OH}] \times [\text{CH}_2\text{O}]$ signal was found to have slightly less temperature sensitivity, the H-atom-based marker was less sensitive to variations in the equivalence ratio. However, it was noted that, while the two metrics performed equally well for lean mixtures, the OH-based metric was superior for flames in the range of $\phi = 1.0$ – 1.1 . Recent studies have demonstrated quantitative two-photon H-atom PLIF, increasing the potential attraction of this technique [11]. However, although planar H-atom fluorescence has recently been achieved using femto-second excitation [12], it appears unlikely that such techniques will be commonly available in the near future. This makes 2-D imaging of $[\text{H}] \times [\text{CH}_2\text{O}]$ a challenge.

More generally, despite having strong correlations with the heat release rate through the HCO-production pathway, $[\text{OH}] \times [\text{CH}_2\text{O}]$ and $[\text{H}] \times [\text{CH}_2\text{O}]$ both require dual excitation strategies. This essentially doubles the required experimental hardware, greatly complicating the data acquisition and image post-processing. Great care must be exercised when recording background images, white-field corrections, and the camera transform registration otherwise the resulting overlapped images will not properly reflect the desired field [13,14]. Experimentalists therefore often settle for other, easier to acquire measurements. Laminar flame calculations demonstrate that the CH radical has reasonable spatial correspondence with the heat release rate in terms of both location and spatial extent in the flame-normal direction. CH-PLIF therefore has been used as a topological marker of the heat release rate in numerous studies, and also has been proposed as an indicator of whether the structure of the heat release region is broadened relative to a laminar flame [15–19]. However, several measurements and simulations have indicated that it is not as accurate a measure of the heat release as other metrics [2].

CH presents measurement challenges due to its low concentration and somewhat difficult excitation. To evaluate the utility of CH-PLIF measurements for flames producing low CH concentrations, Chen and Mansour [20] applied saturated LIF of CH simultaneous with Rayleigh thermometry and reported sufficient signal-to-noise for CH concentrations as low as 1 ppm. In a study quantifying the range of premixed flame conditions for which CH PLIF is applicable, Sutton and Driscoll [21] found that CH signal is at a maximum at rich equivalence ratios around $\phi = 1.25$ for methane-air flames, and that CH-PLIF signal is detectable for equivalence ratios as small as $\phi = 0.85$, although the replacement of some of the hydrocarbon fuel with hydrogen permits even leaner equivalence ratios. Vagelopoulos and Frank [19] found that in nitrogen-diluted premixed counter-flow flames, the peak CH sig-

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