Flame Response to Equivalence Ratio Fluctuations – Relationship Between Chemiluminescence and Heat Release

Shreekrishna\textsuperscript{i}, Tim Lieuwen\textsuperscript{ii}

School of Aerospace Engineering
Georgia Institute of Technology, Atlanta, GA, USA

This paper presents a comparative study of the global responses of heat release and chemiluminescence emissions of a laminar premixed flame excited by oscillations in fuel/air ratio. Chemiluminescence fluctuations are a commonly used marker of heat release oscillations in unsteady flames. While there is strong evidence of a good correlation between these two quantities in situations where the flame is excited by velocity oscillations, the situation is less well understood for cases where the excitation is due to fuel/air ratio oscillations. This is due to the fact that chemiluminescence is not only a function of the heat release rate, but also a strong function of the fuel/air ratio itself. This work attempts to assess this issue by theoretically evaluating linear transfer functions for the global chemiluminescence and heat release responses of the flame. It is shown that they are qualitatively similar in all cases, but that there are quantitative differences between the two that depend upon operating conditions, nominal flame geometry, and which radical species is being measured.

Nomenclature

\begin{align*}
h_R &= \text{Heat of reaction} \\
h_{R1} &= \text{(Linear) heat of reaction sensitivity to fuel/air ratio} \\
m &= \text{Local chemiluminescence sensitivity to fuel/air ratio} \\
q &= \text{Heat release of the flame per unit area} \\
s_L &= \text{Flame speed} \\
s_{L1} &= \text{(Linear) flame speed sensitivity to fuel/air ratio} \\
t &= \text{Time coordinate} \\
u_o &= \text{Mean flow velocity} \\
z &= \text{Axial coordinate} \\
A &= \text{Flame surface area} \\
CH_G &= \text{Global chemiluminescence intensity} \\
F &= \text{Transfer function} \\
F_C &= \text{Chemiluminescence response transfer function} \\
F_Q &= \text{Heat release response transfer function} \\
L_f &= \text{Quiescent flame length} \\
Q &= \text{Global heat release by the flame} \\
R &= \text{Burner radius} \\
St &= \text{Strouhal number} \\
\alpha &= \cos^2 \psi, \text{ where } \psi \text{ is the half angle of the flame} \\
\beta &= \cot \psi \\
\varepsilon &= \text{Amplitude of equivalence ratio perturbations} \\
\phi &= \text{Equivalence ratio}
\end{align*}

\textsuperscript{i} Corresponding Author : shreekrishna@gatech.edu, Graduate Research Assistant, AIAA Student member
\textsuperscript{ii} Associate Professor, AIAA Associate Fellow

Copyright © 2009 by Shreekrishna, Tim Lieuwen. Published by the American Institute of Aeronautics and Astronautics, Inc., with permission.
\( \phi_{eff} = \) “Effective” (area-weighted) equivalence ratio
\( \rho = \) Reactant mixture density (assumed constant)
\( \sigma^* = \) Chemiluminescence intensity per unit flame area
\( \omega = \) Angular frequency of equivalence ratio perturbations
\( \langle \cdot \rangle_o = \) Mean value of a variable
\( \langle \cdot \rangle' = \) Fluctuations in the value of a variable
\( \langle \cdot \rangle = \) Fourier transform of a time domain variable

I. Introduction

This paper considers the relationship between global heat release and chemiluminescence oscillations of flames excited by fuel/air ratio oscillations. This comparison is important because chemiluminescence response measurements may not directly reflect the heat release response of flames when the fuel composition is oscillating.

This study is motivated by the phenomenon of combustion instabilities. These instabilities are caused by unsteady heat-release processes in the combustor that couple with one or more acoustic modes of the system causing acoustic pressure and flow oscillations. These flow oscillations then feed-back on the unsteady heat release perturbations completing a feedback loop. This can potentially result in high amplitude pressure oscillations in the combustor causing degradation in system performance, hardware damage or both.

The focus of this paper is on the response of flames to fuel/air ratio oscillations. The manner in which these oscillations disturb the heat release can be understood by considering the instantaneous global heat release rate, given by the following integral over the flame surface area

\[
Q(t) = \int_{\text{flame}} \rho s_t h_x dA
\]

Equation (1) shows four fundamentally different mechanisms generating heat-release disturbances in a premixed flame, viz., fluctuations in reactant density, flame speed, heat of reaction, or flame surface area.

To understand how equivalence ratio oscillations lead to heat release oscillations, consider Figure 1, which provides a pictorial representation of the different possible “routes” by which the heat release oscillates. First, spatio-temporal fluctuations in equivalence ratio directly perturb the heat release by perturbing flame speed (route 2a in Figure 1) and the heat of reaction (route 1). Second, these perturbations in the flame speed induce fluctuations in flame position, hence causing fluctuations in burning area (route 2b in Figure 1). This in turn causes perturbations in the heat release. This is an indirect route.

Figure 1 - Physical mechanisms causing heat release oscillations due to fluctuations in reactant equivalence ratio

Hemchandra et al. showed that route 1, i.e., heat of reaction oscillations, is dominant for lean flames at very frequencies when \( St << 1 \). At higher values of the Strouhal number, all three routes are seen to be of comparable importance. 

2 American Institute of Aeronautics and Astronautics
Having overviewed the mechanisms by which fuel/air ratio oscillations lead to heat release oscillations, we next discuss how equivalence ratio oscillations are manifested through oscillations in flame chemiluminescence. Measurements of naturally occurring flame chemiluminescence emissions from premixed flames have been used in numerous studies as an indicator of the local and global heat release rates. Detailed chemical kinetic calculations of lean premixed laminar CH₄-air flames show that CH*, OH* and CO₂* radicals occur within the reaction zone, indicating that the reaction zone is the source of these chemiluminescence emissions 5-7. While certainly not a perfect approach, as discussed further below, chemiluminescence is really the only practical method for inferring heat release rates at present.

There is significant experimental data 8-11 showing that at a fixed equivalence ratio, the quasi-steady chemiluminescence emission intensity from the flame exhibits a linear dependence on the reactant flow rate. For these reasons, unsteady chemiluminescence appears to be a good marker of unsteady heat release for flames responding to low frequency flow velocity oscillations. For example, the data below illustrate gain curves for global CH* and OH* chemiluminescence showing similar frequency sensitivities. Given the different chemical pathways by which these species are formed, it appears unlikely that their gains would be so similar if they were not both primarily functions of the heat release rate.

![Figure 2: Chemiluminescence transfer function gain (left) and phase (right, with respect to velocity perturbations) for velocity-coupled flame response, for φₒ=0.75, uₒ=25 m/s, 5% perturbation amplitude](image)

However, chemiluminescence emissions are not only a function of the instantaneous heat release rate, but other parameters as well, including fuel/air ratio, fuel type, strain rate, and unsteady effects. We start first with unsteady effects. Due to finite rate kinetics, one can expect there to be a certain phase lag between heat release and chemiluminescence in unsteady flames, even if they track each other perfectly in the quasi-steady limit. While we are not aware of studies explicitly considering this effect, data suggests that it may be negligible for a variety of frequencies of interest. This may be seen from the difference in phases of CH* and OH* chemiluminescence signals with respect to those of velocity perturbations at the flame base, as shown above. Given the different chemical pathways (and presumably time scales) by which these species are formed, the fact that they give essentially the same phase strongly suggests that they are tracking the heat release in a quasi-steady manner. More data and analyses are needed, however, to further understand these effects.

We next consider other effects such as turbulence and strain rate and flame curvature. John and Summerfield 11, Hurle et al. 12 and, more recently, Lauer and Sattelmayer 13 have shown that turbulence reduces the global chemiluminescence emission intensity. Additionally, other studies 7, 12, 14 have systematically characterized the relationship between chemiluminescence and heat release fluctuations, showing that these are correlated as long as the strain rate and flame curvature are not “too large” (e.g., within a flame cusp). For highly strained flames, these studies indicate that the local chemiluminescence emission can go to zero, even without local extinction. The

---

§ B. Jones, J. G. Lee, D. A. Santavicca, Pennsylvania State University, University Park, PA - Personal communication
sensitivity to strain rate has also been discussed extensively by Nori\textsuperscript{15}. It appears that the strain rate sensitivity of chemiluminescence is much less than its sensitivity to equivalence ratio.

Fuel sensitivities have been studied by, e.g., Clark et al.\textsuperscript{9} and Nori\textsuperscript{15}, who reported measurements using methane, propane, ethylene, H\textsubscript{2}/CO blends, and Jet A. Given the caveats already noted in this section, these studies show that chemiluminescence emissions tracked the unsteady heat release, although the specific sensitivities varied with the fuel and radical species.

Finally, we consider the fuel/air ratio sensitivity of chemiluminescence emissions, which forms the main focus of this study. The sensitivity of chemiluminescence to fuel/air ratio is well known from a number of experiments, showing that the variation of global chemiluminescence intensity with fuel-flow rate itself is an exponential function of the equivalence ratio. This is demonstrated by global OH* and CH* measurements\textsuperscript{14-16} and occurs due to the exponential dependence of the reaction rate upon the temperature\textsuperscript{5}. Due to this exponential sensitivity of chemiluminescence to fuel/air ratio, there are problems associated with its interpretation if both the fuel/air ratio and heat release rate are oscillating. In particular, it can be anticipated that the relationship between the global chemiluminescence intensity and the global heat release is not one-to-one when there is a spatial variation in equivalence ratio along the flame surface area. This occurs because the chemiluminescence intensities of the local area elements are functions of the local equivalence ratio, which is now varying along the flame surface. Hence, the overall chemiluminescence emission intensity is a non-equally weighted sum of individual elements over the flame.

Thus, an important question must be addressed: How does the global chemiluminescence response of the flame compare with the global heat release response? The current work aims to address this question by analytically calculating and comparing transfer functions for the chemiluminescence and heat release responses, which are respectively defined as follows:

\begin{equation}
F_C = \frac{CH^G}{CH^G} \frac{\phi_{\text{base}}'}{\phi_o}
\end{equation}

\begin{equation}
F_Q = \frac{Q'}{Q_o} \frac{\phi_{\text{base}}'}{\phi_o}
\end{equation}

Here, $\phi_{\text{base}}'$ denotes the fluctuations in equivalence ratio at the flame base and the hats (^) denote Fourier-transformed time domain variables. We will show that the comparison between $F_C$ and $F_Q$ depends upon the chemiluminescing species considered, area-averaged fuel/air ratio, and flame geometry.

The remainder of the paper is organized as follows. Section II provides a brief outline of the analytical formulation used to model the problem. Section III presents analytical results for the transfer functions and presents a detailed comparative study of the heat release and chemiluminescence responses for representative conditions and fuel composition, by utilizing the transfer functions. Finally, Section IV concludes the paper with a summary of the key accomplishments of this work and suggestions for further research.

## II. Analytical Formulation

The analytical framework adopted to estimate the heat release transfer function closely follows that of Hemchandra et al.\textsuperscript{2} and is not detailed here. We relate the fluctuations in global chemiluminescence intensity to those in equivalence ratio as described below.

### A. Global Chemiluminescence Response Modeling

The global chemiluminescence intensity can be calculated as an integral of the local chemiluminescence intensity per unit area. We write this local chemiluminescence intensity as a function of the local instantaneous heat release rate per unit flamelet surface area and local equivalence ratio. Mathematically, this may be expressed as follows.

\begin{equation}
CH^G(t) = \int_{\text{flame}} \sigma^L(q(\phi),\phi(z,t))dA
\end{equation}

Here, $q$ is the heat release per unit area of the flame. By definition,

\begin{equation}
Q(t) = \int_{\text{flame}} q(t)dA = \int_{\text{flame}} \rho S_\phi(\phi) h_\phi(\phi)dA
\end{equation}

Further, note that the local heat release density may be estimated as the product of the local mass burning rate of the reactants and the heat of reaction of the reactants, both of which depend on the local equivalence ratio. Mathematically,
We shall assume here that the fuel/air ratio oscillations occur at constant reactant density. From Eq. (6), it follows that \( q = q(\phi) \), so that we may write

\[
\sigma^L(q, \phi) = \sigma^L(q(\phi), \phi) = \sigma^L(\phi(z,t))
\]

With these considerations, the global chemiluminescence intensity \( CH^G \), as expressed in Eq. (4), may be rewritten as follows.

\[
CH^G(t) = \int_{\text{flame}} \sigma^L(\phi(z,t)) dA
\]

For the sake of illustration, we will now assume that the mean equivalence ratio, \( \phi_o \), is spatially uniform, so that \( \phi(z, t) = \phi_o + \phi'(z, t) \). Expanding \( \sigma^L(\phi(z,t)) \) about \( \phi_o \) yields,

\[
CH^G(t) = \int_{\text{flame}} \left( \sigma^L(\phi_o) + \frac{\partial \sigma^L}{\partial \phi} \bigg|_{\phi_o} \phi'(z,t) \right) dA
\]

Note that \( \sigma^L(\phi_o) = CH^G_o / A_o \) is the chemiluminescence intensity per unit flame surface area of the undisturbed flame, which is a constant.

Further, define a flame surface area averaged “effective” equivalence ratio fluctuation as

\[
\phi_{\text{eff}}'(t) = \int_{\text{flame}} \phi'(z,t) dA
\]

The instantaneous global chemiluminescence intensity may be written in terms of \( \phi_{\text{eff}}'(t) \) as

\[
\frac{CH^G(t)}{CH^G_o} = \frac{A'(t)}{A_o} + m(\phi_o) \frac{\phi_{\text{eff}}'(t)}{\phi_o}
\]

On subtracting the mean quantities, this can be written as

\[
\frac{CH^{\text{eff}}(t)}{CH^G_o} = \frac{A'(t)}{A_o} + m(\phi_o) \frac{\phi_{\text{eff}}'(t)}{\phi_o}
\]

where

\[
m(\phi_o) = \frac{\partial \left( \sigma^L(\phi)/\sigma^L(\phi_o) \right)}{\partial \phi} \bigg|_{\phi_o}
\]

The constant \( m \) is the sensitivity of the chemiluminescence intensity of the flame per unit area to fluctuations in equivalence ratio. This will be obtained from experimental data relating chemiluminescence intensity/flow rate to equivalence ratio \(^{14}\).

**B. Global Heat Release Response Modeling**

Let us next consider the expression for the instantaneous heat release by the flame. Assuming constant density we may linearly perturb Eq.(5) to obtain

\[
\frac{Q'(t)}{Q_o} = \frac{A'(t)}{A_o} + \left( \int_{\text{flame}} \frac{s'(\phi)}{s_{\text{Lo}}} dA + \int_{\text{flame}} \frac{h'_{\text{He}}(\phi)}{h_{\text{He}}} dA \right)
\]

The first term on the RHS of the above isolates the contribution of flame surface wrinkling and consequent changes in the flame surface area, to the heat release (indirect route, 2b, Figure 1). The terms in the parentheses denote the contribution to the heat release because of the dependence of flame speed and heat of reaction on equivalence ratio (direct route, 1 and 2a, Figure 1). The variation of equivalence ratio leads to different parts of the flame responding to different local equivalence ratios.

Next, define flame speed and heat of reaction sensitivities to equivalence ratio respectively, as
\[ s_{L1} = \frac{\partial (s_{L1}/s_{L0})}{\partial (\phi/\phi_s)} \]  \hspace{1cm} (15) \\
\[ h_{R1} = \frac{\partial (h_{R1}/h_{R0})}{\partial (\phi/\phi_s)} \]  \hspace{1cm} (16)

Note that, for fixed operating conditions, these sensitivities are functions of the mean equivalence ratio alone. Eq.(14) may be written in terms of these sensitivities as:

\[ \frac{Q'(t)}{Q_o} = \frac{A'(t)}{A_o} + \left[ s_{L1} (\phi_s) + h_{R1} (\phi_s) \right] \frac{\phi_{\text{eff}}' (t)}{\phi_s} \]  \hspace{1cm} (17)

Note from Eq.(12) and Eq.(17) that evaluation of the instantaneous global chemiluminescence intensity and global heat release of the flame requires us to estimate the fluctuations in burning surface area and the effective equivalence ratio.

Expressions for the flame surface area may be estimated using the knowledge of the instantaneous flame surface location. For this purpose, a two-dimensional front tracking formulation is employed (Markstein\textsuperscript{17}, Marble and Candel\textsuperscript{18}, Yang and Culick\textsuperscript{19}, Fleifil et al.\textsuperscript{20}, Kerstein et al.\textsuperscript{21}, Preetham and Lieuwen\textsuperscript{22}). The flame is assumed to consist of a thin sheet whose surface can be represented implicitly by the zero contour of a two dimensional function \( G(r,z) \). For complete details regarding flame surface location calculation and evaluation of fluctuations in instantaneous area and area-averaged equivalence ratio, see Hemchandra et al.\textsuperscript{2} and Shreekrishna and Lieuwen\textsuperscript{4}.

For purposes of illustration, a spatio-temporally harmonically varying equivalence ratio fluctuation advecting at the mean velocity of the reactants is assumed in this work. Mathematically,

\[ \phi = \phi_s \left( 1 + \epsilon \cos \left( \omega \left( t - \frac{z}{u_o} \right) \right) \right) \]  \hspace{1cm} (18)

We non-dimensionalize the axial coordinate \( z \) by the length of the quiescent flame, \( L_f \) and \( t \) by the characteristic convection time for the disturbances from the flame base to the flame tip, \( L_f/u_o \). Retaining the same notations for the non-dimensional variables as their original dimensional counterparts for the sake of notational convenience, we may rewrite Eq.(18) in non-dimensional form as

\[ \phi = \phi_s \left( 1 + \epsilon \cos \left( St (z - t) \right) \right) \]  \hspace{1cm} (19)

Here, the Strouhal number is a non-dimensional frequency defined as

\[ St = \frac{\omega L_f}{u_o} \]  \hspace{1cm} (20)

C. Global Transfer Functions

With the framework presented in the previous subsections, we may evaluate transfer functions for the global chemiluminescence response, \( F_C \) and global heat release response, \( F_Q \), by taking the Fourier transforms of Eq.(12) and Eq.(17) at the forcing Strouhal number, to yield:

\[ F_C = F_A + m F_{\phi_s} \]  \hspace{1cm} (21)
\[ F_Q = F_A + (s_{L1} + h_{R1}) F_{\phi_s} \]  \hspace{1cm} (22)

Here, \( F_A \) and \( F_{\phi_s} \) denote the transfer functions for the burning area and effective equivalence ratio fluctuation responses whose definitions are similar to those of \( F_C \) and \( F_Q \), see Eq.(2) and Eq.(3)

\[ F_A = \frac{\bar{A}'}{A_o} \]  \hspace{1cm} \left[ \frac{\phi_{\text{base}}'}{\phi_o} \right] \]  \hspace{1cm} (23)
\[ F_{\phi_s} = \frac{\phi_{\text{eff}}'}{\phi_o} \]  \hspace{1cm} \left[ \frac{\phi_{\text{base}}'}{\phi_o} \right] \]  \hspace{1cm} (24)

Equations (21) and (22) are very revealing for understanding the comparison between \( F_C \) and \( F_Q \). First, both transfer functions have similar structures and the same two principal contributing terms –fluctuating flame surface area and fluctuating effective equivalence ratio. Furthermore, Eq.(21) and Eq.(22) differ only in the coefficient which precedes the latter contribution, \( F_{\phi_s} \). The former contribution, \( F_A \) affects both \( F_C \) and \( F_Q \) in an identical
manner. This has important implications. Specifically, it means that $F_C$ and $F_Q$ are the same only if either of the following two conditions is satisfied –

\begin{align}
(1) \quad & \quad m \sim s_{L1} + h_{ke} \\
(2) \quad & \quad |F_A| \gg |F_{pe,ef}| \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad (25)
\end{align}

The first condition reflects the fact that $F_C$ and $F_Q$ are identical if the sum of the heat of reaction and flame speed sensitivities of the heat release identically equal the fuel/air ratio sensitivity of the local chemiluminescence intensity per unit area. The second condition states that even if there is a disparity between the two coefficients, their net effect on the total transfer function would be swamped out because of the heavy domination of the area response.

Equation (25) also isolates the two ways in which either response can change – viz., (a) by altering the burning area and effective equivalence ratio responses, or (b) by altering the chemiluminescence intensity sensitivity, flame speed sensitivity and heat of reaction sensitivity. The former can be achieved if the flame geometry and/or the mean flame shape changes. The latter occurs if the mean equivalence ratio, reactant pressure or reactant temperature change. Hence, for fixed operating and geometric conditions, a change in fuel composition would affect flame response through affecting the sensitivities alone.

To understand these effects, we present results from illustrative calculations for these transfer functions in the next section that account for each of these effects. For these calculations we estimate the constant $m$ from the slope of global chemiluminescence intensity/fuel flow rate versus equivalence ratio data from Lee and Santavicca \textsuperscript{14} and Nori \textsuperscript{15}. For example, a typical curve using CO\textsubscript{2}* as the chemiluminescing species is presented below. Similarly, the flame speed and heat of reaction sensitivities were calculated from the Premix module of Chemkin (using GRIMech 3.1) and GasEq, respectively.

![Figure 3: Variation of CO\textsubscript{2}* chemiluminescence intensity/fuel flow rate with mean equivalence ratio \textsuperscript{14}](image)

**III. Illustrative Results**

This section presents illustrative results comparing $F_C$ and $F_Q$. We intend to understand how factors such as different flame geometries, mean equivalence ratios, and thermodynamic conditions of the reactants affect $F_C$ and $F_Q$. Additionally, different chemiluminescing species are considered to estimate $F_C$, in order to determine which of those compare best with $F_Q$.

Two different flame geometries are considered, axisymmetric conical and V-shaped flame, and are shown in Figure 4. We assume that in either case, the flame base remains attached to the burner lip at all times.
As noted in the earlier section, the relationship between global responses is influenced by the flame geometry or in the three sensitivity parameters. These two possibilities are discussed under subsections ‘A’ and ‘B’ below.

A. Flame Geometric Effects and Burning Area Response

We start by presenting expressions for the V-flame and conical flame transfer functions. For an axisymmetric V-flame, expressions for $F_A$ and $F_{\phi_{\text{eff}}}$ are

$$F_A = -s_{t_{1}} \left\{ \frac{2\alpha}{1-\alpha} \left[ \frac{1-\alpha + (iSt-1)\exp(iSt) - \alpha(iSt/\alpha - 1)\exp(iSt/\alpha)}{St^2} \right] \right\}$$

$$F_{\phi_{\text{eff}}} = \frac{2(1+iSt e^{\text{St}})}{St^2}$$

Here $\alpha = \cos^2 \psi$ is a flame geometric parameter.

For a premixed conical flame, transfer function expressions may be obtained as follows.

$$F_A = s_{t_{1}} \left\{ \frac{2\alpha}{1-\alpha} \left[ \frac{1-\alpha - \exp(iSt) + \alpha\exp(iSt/\alpha)}{St^2} \right] \right\}$$

$$F_{\phi_{\text{eff}}} = \frac{2(1+iSt e^{\text{St}})}{St^2}$$

Using the above, $F_C$ and $F_Q$ may be evaluated using Eq.(21) and Eq.(22). Further, the low Strouhal number limits of the above may be evaluated for either geometry as :

$$\lim_{St \to 0} F_A = -s_{t_{1}}$$

$$\lim_{St \to 0} F_{\phi_{\text{eff}}} = 1$$

For the total responses, these limits hence become, 

$$\lim_{St \to 0} F_C = m - s_{L_{1}}$$

$$\lim_{St \to 0} F_Q = h_{R_{1}}$$

Consider next the contribution of the area fluctuations and effective equivalence ratio fluctuations to the total transfer functions.
Figure 5: Contributions to $F_Q$ by area and effective equivalence ratio fluctuations for (a) V-flame (b) conical flame at 1 atm, 300 K, $\phi_o=0.6$, $\beta=4$.

Figure 5 plots the contributions of response of flame surface area fluctuations and effective equivalence ratio fluctuations to $F_C$ for a V-flame and conical flame. It can be clearly seen that the two processes contribute very differently to conical flames as compared to V-flames. In the case of V-flames, $F_A$ dominates over $F_{\phi_{\text{eff}}}$ over most of the Strouhal number range. Hence, the heat release response of the flame is primarily due to $F_A$. However, at lower Strouhal numbers, these effects are comparable. This is seen in the phase too, where the heat release response phase and burning area response phase are noticeably different in the quasi-steady limit and are identical at larger Strouhal numbers.

However, in the case of conical flames, these two effects contribute comparably over the entire Strouhal number range. This noticeable difference in the response characteristics is merely because of the fact that a V-flame has maximum surface area at the flame tip, while the conical flame has maximum surface area at the flame base. Hence, the maximum area in the V-flame is free to move and leads to higher response, while it is constrained at the flame attachment point for conical flames.

Figure 6: Contributions to $F_C$ by area and effective equivalence ratio fluctuations for (a) V-flame (b) conical flame at 1 atm, 300 K, $\phi_o=0.6$, $\beta=4$.
The points raised in the above discussion can be directly applied to anticipate the relationship between $F_C$ and $F_Q$. The V-flame is largely dominated by the area response at higher Strouhal numbers, while there is comparable contribution from the effective equivalence ratio response at lower Strouhal numbers. For the conical flame, these contributions are comparable over the entire range of Strouhal numbers considered. Based on the two conditions stated in Eq.(25), it can be seen that the V-flame $F_C$ and $F_Q$ reasonably agree over the range of Strouhal numbers where area response dominates. However, in the case of conical flames, there may be some differences to the extent that the sensitivity coefficients differ.

To investigate further into this issue, we next consider the comparison between $F_C$ and $F_Q$ for reactants at 1 atm, 300 K, which is plotted in Figure 7.

![Figure 7: Comparison between $F_C$ and $F_Q$ for (a) V-flame (b) Conical flame at 1 atm, 300 K. $\phi_o=0.6$, $\beta=4$](image)

The following observations may be made from Figure 7. The $F_C$ gain and phase for each of the three chemiluminescing species are qualitatively similar to $F_Q$ at all but very low Strouhal numbers. At very low Strouhal numbers, there is a disparity in gain as well as in phase. At these low Strouhal numbers, since $F_A$ and $F_{eff}$ are comparable; condition (1) of Eq.(25) needs to be satisfied, which is not the case. For conical flames, there is a noticeable disparity in phase if $CO_2^*$ is used, while there is a disparity in gain if $OH^*$ is used as the chemiluminescing species. These disparities are directly related to the degree that the different sensitivities satisfy condition (1) of Eq.(25).

### B. Effect of Variation of Sensitivities

We next consider the effect of chemiluminescence and local heat release sensitivity to fuel/air ratio. We shall start by considering the variation of the three sensitivities for reactants at 1 atm, 300 K, for different chemiluminescing species. This is plotted in Figure 8a. At STP, $CO_2^*$ and $OH^*$ sensitivities, $m_{CO2}$ and $m_{OH}$, are reasonably close to the sum of the flame speed and heat of reaction sensitivities, $s_{L1} + h_{R1}$, for mean equivalence ratios between 0.7 and 0.9. Hence, these can be expected to yield $F_C$’s that are close to $F_Q$. $CH^*$ might be preferable at lower equivalence ratios, while $OH^*$ seems to be preferable at near-stoichiometric fuel composition. Recall also that the value of $m$ relative to $s_{L1}$ is important in controlling the response at low Strouhal numbers; specifically, there is a 180 degrees phase difference between the quasi-steady $F_C$ phase and $F_Q$ phase if $m > s_{L1}$, see Eq.(32) and Eq.(33).

Next consider the effect of pressurizing the reactants to 5 atm. It merely suffices to consider the variation of sensitivities with equivalence ratio at 5 atm; conclusions regarding the comparison between $F_C$ and $F_Q$ can be drawn based upon the previous discussion and results. This is plotted in Figure 8b, which suggests that at lower equivalence ratios, $OH^*$ seems to be preferable; also, $m < s_{L1}$ for all chemiluminescing species and hence, the quasi-steady phase difference between $F_C$ and $F_Q$ would be 180 degrees. However, at higher equivalence ratios, say 0.9,
CO$_2^*$ seems to yield closer agreement between $F_C$ and $F_Q$, owing to its sensitivity, $m_{CO2}$ being closest to $s_{L1}+h_{R1}$. Also, it may be observed that at these equivalence ratios, because $m>s_{L1}$, irrespective of the chemiluminescing species, the phase predictions would be more accurate, even in the quasi-steady limit. Finally, it may be noted that, in general, one can expect very good agreement between $F_C$ and $F_Q$ if the mean equivalence ratio lies approximately between 0.7 and 0.85.

![Figure 8](image1.png)

**Figure 8**: Variation of various sensitivities with mean equivalence ratio at (a) 1 atm, 300 K (b) 5 atm, 300 K

Finally, we consider the effect of preheating the reactants to a higher temperature. The variation of various sensitivities versus equivalence ratio at 5 atm, 600 K is plotted in Figure 9.

![Figure 9](image2.png)

**Figure 9**: Variation of various sensitivities with mean equivalence ratio at 5 atm, 600 K.
Using similar arguments as before, it may be seen from Figure 9 that at all equivalence ratios it may be expected that CH* and OH* F_c’s would compare better with F_Q, while the low frequency phases would be off by 180 degrees for all of the chemiluminescing species, since \( m < S_L \) for all of them. At near-stoichiometric equivalence ratios (larger than 0.9, say), CH* appears to be capable of also yielding good comparison between the quasi-steady phase difference between \( F_C \) and \( F_Q \).

IV. Summary and Concluding Remarks

This paper studied the global chemiluminescence intensity response and global heat release response of premixed flame submitted to a spatio-temporally oscillating equivalence ratio field. The two responses were compared by comparing the respective transfer functions. A theoretical derivation of these linear transfer functions showed that both \( F_C \) and \( F_Q \) had similar forms, with the two constituent contributions coming from flame surface area response and response arising due to a spatio-temporally varying equivalence ratio oscillation. While the area response was seen to contribute identically to either transfer function, the contribution due to response of the fluctuating effective equivalence ratio differed in magnitude by a factor equal to the respective sensitivities, i.e., chemiluminescence intensity sensitivity for \( F_C \) and the sum of flame speed and heat of reaction sensitivities of \( F_Q \). This similarity of form yielded two criteria, at least one of which had to be satisfied for \( F_C \) and \( F_Q \) to compare reasonably well with each other (See Eq.(1.35)).

To develop an actual feel for how these two transfer functions compare, calculations were performed for CH_4-air mixtures at different conditions. In spite of quantitative differences, the qualitative relationship between \( F_C \) and \( F_Q \) seemed quite similar. The major difference seemed to be due to mean equivalence ratio and flame geometry.

For the V-flame geometry, \( F_C \) and \( F_Q \) almost always compared very well with each other owing to the dominating contribution of the area response over the effective equivalence ratio response. At lower equivalence ratios, the quasi-steady phases differed by a factor of 180 degrees, owing to the difference in signs of the values of the respective quasi-steady transfer functions. This disparity was eliminated at higher equivalence ratios, where the agreement between \( F_C \) and \( F_Q \) was excellent. For the conical flame geometry, both the area response and the effective equivalence ratio response contributed comparably over the entire frequency range. While the actual quantitative values between the \( F_C \) and \( F_Q \) were not as close as that of the V-flame, they did follow similar trends over most of the frequency range, especially in magnitude.

The overall comparison between \( F_C \) and \( F_Q \) were quite encouraging for both V-flames and conical flames, hinting that, to a reasonable degree of accuracy, chemiluminescence response can be used to understand heat release response even when the equivalence ratio is oscillating spatio-temporally. However, some caution seems to be required wherever direct quantitative comparisons are sought, as well as in studying the low Strouhal number responses.

Finally, it should be noted that this analysis is strictly valid for laminar flames. Clearly, the situation of most practical interest for performing these comparisons is in turbulent flames. The authors are not aware of any factors which would lead to fundamentally different qualitative conclusions than drawn here, but further work on this specific problem is needed.

Acknowledgments

This work has been partially supported by the US Department of Energy under contracts DE-FG26-07NT43069 and DE-NT0005054, contract monitors Mark Freeman and Richard Wenglarz, respectively. The authors would also like to acknowledge Prof. Dom Santavicca and Jongguen Lee for their inputs and comments, and Lynn Coldiron III and Jagannathan Pranarththikaran for their assistance in estimating the sensitivity coefficients used for the illustrative calculations.
References

1. Lieuwen, T. and Yang, V., (ed). *Combustion Instabilities in Gas Turbine Engines: Operational Experience, Fundamental Mechanisms, and Modeling*, Progress in Aeronautics and Astronautics, Vol. 210, AIAA.
2. Hemchandra, S., Shreekrishna, and Lieuwen, T., "Premixed Flame Response to Equivalence Ratio Perturbations". in *Joint Propulsion Conference*, 2007, AIAA#2007-5656.
3. Shreekrishna and Lieuwen, T., "Premixed Flame Response to Equivalence Ratio Oscillations : Non-quasisteady effects". in Fall Technical meeting of the Eastern States Section of the Combustion Institute, 2007,
4. Shreekrishna and Lieuwen, T., *High Frequency Premixed Flame Response to Acoustic Perturbations*, in 15th AIAA/CEAS International Conference in Aeroacoustics. 2009: Miami, FL.
5. Samaniego, J.M., Egolfopoulos, F.N., and Bowman, C.T., "CO$_2$ Chemiluminescence in Premixed Flames", *Combustion Science and Technology*, Vol. 109 (1), 1995, pp. 183-203.
6. Dandy, D. and Vosen, S., "Numerical and Experimental Studies of Hydroxyl Radical Chemiluminescence in Methane-Air Flames", *Combustion Science and Technology*, Vol. 82 (1), 1992, pp. 131-150.
7. Najm, H.N., Paul, P.H., Mueller, C.J., and Wyckoff, P.S., "On the Adequacy of Certain Observables as Measurements of Flame Burning Rate", *Combustion and Flame*, Vol. 113 (3), 1998, pp. 312-332.
8. Price, R., Hurle, I., and Sugden, T., "Optical Studies of the Generation of Noise in Turbulent Flames", *Proceedings of the Combustion Institute*, Vol. 12, 1968, pp. 1093-1102.
9. Clark, T., *Studies of OH, CO, CH and C$_2$ Radiations from Laminar and Turbulent Propane-Air and Ethylene-Air Flames*. 1958, NACA.
10. Diederichsen, J. and Gould, R.D., "Combustion Instability: Radiation from Premixed Flames of Variable Burning Velocity", *Combustion and Flame*, Vol. 9, 1965, pp. 22-31.
11. John, R. and Summerfield, M., "Effect of Turbulence on Radiation Intensity from Propane-Air Flames", *Jet Propulsion*, Vol. 27, 1957, pp. 169-178.
12. Hurle, I.R., Price, R.B., Sugden, T.M., Thomas, R.S., and Thomas, A., "Sound Emission from Open Turbulent Premixed Flames", *Proceedings of the Royal Society of London, Series A : Mathematical and Physical Sciences*, Vol. 303, 1968, pp. 409-427.
13. Lauer, M. and Sattelmayer, T., *On the Adequacy of Chemiluminescence as a Measure for Heat Release in Turbulent Flames with Mixture Gradients*, in ASME Turbo Expo. 2009, ASME: Orlando, FL.
14. Lee, J.G. and Santavicca, D.A., "Experimental Diagnostics for the Study of Combustion Instabilities in Lead Premixed Combustors", *Journal of Propulsion and Power*, Vol. 19 (5), 2003, pp. 735-750.
15. Nori, V.N., *Modeling and Analysis of Chemiluminescence Sensing for Syngas, Methane and Jet-A Combustion*, in Aerospace Engineering. 2008, Georgia Institute of Technology: Atlanta.
16. Miller, S.A., *Development of Flame Chemiluminescence Probe for Determination of Primary Zone Equivalence Ratio in Gas Turbine Combustors*, in Mechanical Engineering. 1999, Pennsylvania State University: University Park.
17. Markstein, G.H., *Non-steady flame propagation*, 1964, New York: Pergamon.
18. Marble, F. and Candel, S., "Acoustic Disturbance from Gas Nonuniformity convected through a nozzle", *Journal of Sound and Vibrations*, Vol. 55, 1977, pp. 225-243.
19. Yang, V. and Culick, F.E.C., "Analysis of Low Frequency Combustion Instabilities in a laboratory Ramjet Combustor", *Combustion Science and Technology*, Vol. 45, 1984, pp. 1-25.
20. Fleifil, M., Annaswamy, A.M., Ghoneim, Z.A., and Ghoneim, A.F., "Response of a laminar premixed flame to flow oscillations: A kinematic model and thermoacoustic instability results", *Combustion and Flame*, Vol. 106, 1996, pp. 487-510.
21. Kerstein, A.R., Ashurst, W.T., and Williams, F.A., "Field equation for interface propagation in an unsteady homogeneous flow field", *Physical Review*, Vol. A27, 1988, pp. 2728-2731.
22. Preetham and Lieuwen, T., "Nonlinear Flame-flow transfer function calculations: Flow disturbance celerity effects". in *AIAA Joint Propulsion Conference*, 2004, AIAA#2004-4035.