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APPROXIMATE CALCULATIONS OF NO$_x$ FORMATION IN AN OSCILLATING FLOW FIELD

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Abstract	121	0	0
Introduction	801	0	0
Background: Form...	1121	49(7) = 343	0
Coupling of ...	427	3(7) = 21	0
Calculations of Nox...	364	8(7) = 56	0
Pressure Dependence ...	522	19(7) = 133	0
Temperature Dependence ...	468	6(7) = 42	2(200) = 400
Pressure and Temperature ...	296	3(7) = 21	0
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Abstract

Chiefly for improved efficiency, the trend to increasing use of gas turbine engines in stationary powerplants has been firmly established. The requirement for minimum NOx production has motivated operation as close as practically possible near the lean flammability limit, to reduce formation of nitrogen oxides by the Zeldovich thermal mechanism. However, experience has shown that under these conditions, stability of the chamber is reduced, often leading to the presence of sustained oscillations in the combustor. That possibility raises the problem of the influence of oscillatory motions on the production of nitrogen oxides. The work represented in this paper covers the initial steps in constructing an analysis suitable for application to that problem in design and development of operational gas turbine combustors.

Introduction

The rate of formation of unwanted chemical species depends on both detailed local chemical kinetics and on the various aspects of fluid mechanics comprising both local and global influences. Precise predictions of the production of pollutants are not possible for 'steady' operation of a combustor. It is nevertheless necessary to begin investigating the corresponding problem under 'unsteady' conditions. In fact, the flow field in a combustor is never steady, owing to the inevitable fluctuations associated with local instabilities, including regions of flow separation, and to the presence of substantial noise arising from a variety of sources. Hence, it is quite possibly more appropriate and practically useful to examine the formation of pollutants, in particular nitrogen oxides, under dynamic conditions. In this paper, we do not consider other pollutants such as soot, carbon monoxide and unburned hydrocarbons, but we believe that the methods we initiate here, to be developed further in subsequent work, will accommodate those contributions.

Because combustors necessarily contain high densities of energy production, and possess relatively low losses for unsteady motions in the flow, the general problem of instabilities - generically called 'combustion instabilities' - has been encountered in all types of propulsion systems and stationary powerplants. Analyses have been carried out at various levels of complexity and with varying degrees of success. The approach taken here amounts to extension of the methods used with considerable success in analyzing combustion instabilities in rockets and ramjets [1-4]. The eventual purpose is to develop methods for predicting the formation of pollutants in the presence of organized oscillations and random motions or noise. In the present work, we are concerned with some elementary calculations directed to constructing representations of local 'response functions' for unsteady formations of NOx, i.e. the ratio of fluctuations of NOx produced locally by fluctuations of the thermodynamic variables. We do not attempt to model the possible effects of velocity fluctuations.

Environmental concerns have lead to increased restrictions on pollutant emissions of gas turbine combustion systems. The strict standards have increased the efforts by researchers to understand the mechanisms behind pollutant formation (NOx, CO, UHC) in order to reduce emission levels. One of the efforts in the reduction of emissions has been the development of lean premixed combustion (LPC).

Current efforts to examine NOx formation in LPC have been reviewed by Correa [5]. The general strategy is to decrease the NOx level with a decrease in the equivalence ratio and a corresponding decrease in flame temperature which reduces the amount of NOx produced according to the Zeldovich thermal mechanism.

Current computational and experimental works in this field [6-8] examine steady state or time averaged pollutant values at a set operating condition. The goals are scaling laws relating pollutant levels to pressure and temperature design conditions. Those relations apply to steady conditions; however, the predicted values may show variations within an oscillating flow field. The report by J.O. Keller and I. Hongo [9] provides some results for pollutant emissions in a pulse combustor under pulsing and non-pulsing conditions. Significantly less NOx was produced when the combustor operated under pulsing conditions. The explanation was based on the idea that the shorter residence time at high combustion temperatures reduced the impact of the Zeldovich thermal mechanism. In the non-pulsing case, the temperature remained high downstream of the flame, allowing more time for the completion of NOx production. However, in the pulsing case, the temperature field dropped off significantly after the peak flame temperature, reducing the residence time at high temperature and hence lowering the emission level. The interpretations given in that paper are largely speculative; neither sufficiently detailed experimental results nor any analysis exists to support the conclusions.

In this paper we will be concerned with an elementary examination of the effects of a given oscillating flow field on the time averaged production of NOx levels. The calculations will be presented for a single oscillating mode, but possible effects of higher modes will be examined. The analysis will be based on formulas for the levels of NOx concentration derived from computational simulations and experimental measurements aimed at determining the dependence of pollutant formation on pressure and temperature. Additional computational work will be performed in this paper using a single PSR (perfectly stirred reactor) model to compare with previous results for the effects of pressure and temperature on NOx levels with special consideration of behavior in the lean limit.

The paper has two purposes: (1) to outline the application of an approximate analysis to the problem of determining the effects of coherent oscillations and noise on the formation of NOx; and (2) to give some initial results for the influences of pressure and temperature oscillations on the level of NOx,

based on the simplest possible models. Although the approach taken here can be extended to actual combustors, no reportable progress has been made.

Background: Formulation of an Approximate Analysis

It is presently not reasonable to attempt a complete analysis of a flow in a combustor to determine the production of pollutants, whether or not organized oscillations are present. The numerical methods of CFD might be adequate but the basis for applying them has not been developed. A comprehensive theory encompassing large scale non-uniform motions, small-scale turbulence, detailed finite-rate chemistry and interactions between turbulence and chemical kinetics is not available [5-10]. In any event, analytical work is necessarily approximate. Since our attention here is directed to the possible influences of oscillations in the flow on the rate at which pollutants are formed, it seems a rational strategy to investigate the possibility of extending an existing approximate analysis. In this section, we first describe briefly a framework that has been used to predict and interpret the characteristics of linear and nonlinear oscillations in combustion chambers. We then indicate how that analysis may be applied to the problem of pollution. It is an intrinsic property of this approach that careful modeling and explicit calculations of contributing physical processes are prerequisite to obtaining quantitative global results. The discussion in this section suggests what processes must be accommodated and where they fit in the approximate analysis.

Formulation of the analysis begins with the conservation equations of mass, momentum and energy. To those we must eventually add equations governing the time and spatial evolution of chemical species, and a set of equations representing the kinetic mechanism. The analysis is concerned essentially with matters of coupling between unsteady fluid motions and the chemical processes. In the framework constructed here, the coupling or interactions are strongly biased in one direction. The unsteady motions (i.e. the pressure, temperature and velocity fluctuations) determine unsteady formation of chemical species, but are themselves weakly influenced by the detailed chemistry. Here we ignore the latter, to concentrate on a method for calculating the time-dependent formation of chemical species in a combustor. Of course, the comparable assumption cannot be made with respect to steady motions: the flow itself depends on the production of energy released in chemical reactions; even for steady flow much useful information is

gained even with quite severe approximations to the chemistry. In analysis of the unsteady flows we assume that the properties of the steady flow are known.

Much of the formulation may therefore be constructed without regard for the species equations and begins with the equations for the conservation of mass, momentum and energy written in the primitive form.

MASS
$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{u}) = \mathcal{W} \tag{1}$$

MOMENTUM
$$\frac{\partial}{\partial t}(\rho \vec{u}) + \nabla \cdot (\rho \vec{u}\vec{u}) + \nabla p = \nabla \cdot \bar{\bar{\tau}} + \vec{F} \tag{2}$$

ENERGY
$$\frac{\partial}{\partial t}(\rho e_0) + \nabla \cdot (\rho \vec{u} e_0) + \nabla \cdot (\rho \vec{u}) = \nabla \cdot (\bar{\bar{\tau}} \cdot \vec{u}) + \nabla \cdot \vec{q} + Q + \vec{u} \cdot \vec{F} \tag{3}$$

where ρ, $\vec{u}$ and p are the density, velocity and pressure; $e_0 = e + u^2/2$ is the stagnation internal energy (de = C_vdT where T is temperature); $\bar{\bar{\tau}}$ is the viscous stress tensor; $\vec{q}$ is internal heat transfer; and $\mathcal{W}$, $\vec{F}$ and Q represent sources of mass, momentum and energy, the last being essentially the heat released in chemical reactions. The equation of state is

$$p = \rho RT \tag{4}$$

Standard manipulations of these equations lead to the set

$$\frac{D\rho}{Dt} = -\rho \nabla \cdot \vec{u} + \mathcal{W} \tag{5}$$

$$\rho \frac{D\vec{u}}{Dt} = -\nabla p + \vec{\mathfrak{F}} \tag{6}$$

$$\rho C_v \frac{DT}{Dt} = -p \nabla \cdot \vec{u} + \mathfrak{Q} \tag{7}$$

$$\frac{Dp}{Dt} = -\gamma p \nabla \cdot \vec{u} + \mathcal{P} \tag{8}$$

$$T\frac{Ds}{Dt} = \mathfrak{Q} - RT\mathcal{W} \tag{9}$$

where,

$$\vec{\mathfrak{F}} = \nabla \cdot \bar{\bar{\tau}} + \vec{F} - \vec{u}\mathcal{W} \tag{10}$$

$$\mathfrak{Q} = (\bar{\bar{\tau}} \cdot \nabla) \cdot \vec{u} + \nabla \cdot \vec{q} + Q - (e - u^2/2)\mathcal{W} \tag{11}$$

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$$\mathcal{P} = \frac{R}{C_\upsilon}\left(\mathcal{Q} + C_\upsilon T \mathcal{W}\right) \qquad (12)$$

So far as combustion instabilities are concerned, the contributions from $\vec{\mathcal{F}}, \mathcal{W}, \vec{\tau}$ and $\vec{q}$ are normally negligible, giving the simple result for $\mathcal{P}$,

$$\mathcal{P} = R\mathcal{Q}/C_v = RQ/C_v \qquad (13)$$

Nonlinear gasdynamic processes are important in the kinds of problems envisioned here, acting to couple the turbulent and acoustic fields [11,12]. Experience with analysis of nonlinear combustion instabilities has demonstrated that it is sufficient to carry nonlinear contributions to second order: third order terms seems to generate only quantitative, not qualitative, corrections. The flow field is represented as a sum of a steady mean field and an unsteady fluctuation, $p = \overline{p} + p'$, $\vec{u} = \overline{\vec{u}} + \vec{u}'$, etc. For simplicity here we will assume $\overline{p}, \overline{\rho}$ and $\overline{T}$ uniform but $\overline{\vec{u}}$ must be non-uniform.

The next step in the formulation is construction of a nonlinear wave equation for the pressure fluctuation and its associated boundary condition, i.e.

$$\nabla^2 p' - \frac{1}{a^2}\frac{\partial^2 p'}{\partial t^2} = h$$
$$\hat{n} \cdot \nabla p' = -f \qquad (14)a,b$$

with $a^2 = \gamma\, \overline{p}/\,\overline{\rho}$. Details of the derivation and the formulas for h and f may be found in the references cited above; they are unimportant in the present discussion.

Application of a form of Galerkin's method, a convenient way of spatially averaging the governing equations (14) a,b, produces the set

$$\ddot{\eta}_n + \omega_n^2 \eta_n = F_n \qquad (15)$$

where

$$F_n = -\frac{a^2}{\overline{p}E_n^2}\left\{\int h\psi_n dV + \oiint f\psi_n dS\right\}$$
$$E_n^2 = \int \psi_n^2 dV \qquad (16)$$

The $\psi_n(\vec{r})$ are the eigenfunctions for the unperturbed classical acoustic problem set in the same geometry as the combustion chamber in question; $\omega_n = ak_n$ is the frequency of the nth normal mode and k_n is the

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wavenumber. Derivation of (15) for the amplitudes η_n rests on spatially averaging (14)a,b after substituting the series representation of the pressure field,

$$p' = \bar{p} \sum_{j=1}^{\infty} \eta_j(t) \psi_j(\vec{r})$$

(17)

With the approximation to second order noted above, the forcing function F_n, defined by (16), can eventually be written

$$-\frac{\bar{p} E_n^2}{a^2} F_n = \bar{p} I_1 + \frac{1}{a^2} I_2 + \bar{p} I_3 + \frac{1}{a^2} I_4 + \iint \bar{p} \frac{\partial \vec{u}'}{\partial t} \cdot \hat{n} dS - \int \frac{1}{a^2} \frac{\partial p'}{\partial t} \psi_n dV$$

(18)

where,

$$I_1 = \int \left(\bar{\bar{u}} \cdot \nabla \vec{u}' + \vec{u}' \cdot \nabla \bar{\bar{u}} \right) \cdot \nabla \psi_n dV$$

$$I_2 = \frac{\partial}{\partial t} \int \left(\gamma p' \nabla \cdot \bar{\bar{u}} + \bar{\bar{u}} \nabla \cdot p' \right) \psi_n dV$$

$$I_3 = \int \left(\vec{u}' \cdot \nabla \vec{u}' + \frac{\rho'}{\bar{\rho}} \frac{\partial \vec{u}'}{\partial t} \right) \cdot \nabla \psi_n dV$$

(19)a,b,c,d

$$I_4 = \frac{\partial}{\partial t} \int \left(\gamma p' \nabla \cdot \vec{u}' + \vec{u}' \nabla \cdot p' \right) \psi_n dV$$

In deriving these formulas for the integrals I_1-I_4, some use has been made of the classical acoustics relations, $\bar{\rho} \vec{u}_t' = -\nabla p'$ and $p_t' = -\gamma \bar{p} \nabla \cdot \vec{u}'$, a step consistent with the order to which the equations have been written. The approximation can be justified by carrying out a two-parameter expansion of the original conservation equations, the two small parameters being Mach numbers characterizing the steady and unsteady fields.

A significant feature of the general problem defined by (15), (16), (18) and (19) is that the influences of a turbulent reacting flow on acoustic waves can be determined simultaneously with the noise field. The analysis supporting that statement is in progress and will be described but briefly here. The fundamental idea is that developed most completely by Chu and Kovasznay [13], that any disturbance in a compressible flow can be treated as a synthesis of three 'modes' of propagation, or classes of waves: acoustic, vortical, and entropic (or temperature) waves. In the linear limit, these three modes of propagation possess the following fundamental properties not proved here: (i) pressure fluctuations are carried only by acoustic waves which propagate with the speed of sound a; (ii) vortical and entropic disturbances are

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carried by the mean flow and hence propagate with the local mean flow velocity; and (iii) vorticity and entropy fluctuations are associated respectively with vortical and entropic waves only.

The fluctuations of pressure, vorticity and entropy (or nonisentropic temperature) are nonzero only for their corresponding modes of propagation. An immediate consequence is that coherent acoustic waves and the noise field can be represented, at least in principle, by the synthesis (17) of classical normal modes. Development of this idea requires lengthy formal calculations [11,12] leading finally to the more explicit form for the set of coupled oscillator equations,

$$\ddot{\eta}_n + \omega_n^2 \eta_n = \alpha_n \eta_n + \theta_n \dot{\eta}_n + F_n^{NL} + \xi_n \eta_n + \xi_n^v \dot{\eta}_n + \sum_{i=1}^{\infty} \left[\xi_{ni} \eta_i + \xi_{ni}^v \dot{\eta}_i \right] + \Xi_n \qquad (20)$$

Here F_n^{NL} stands for the well-known terms arising from second-order nonlinear gasdynamics (see ref. [2], for example) and the $\xi_n(t)$, $\xi_n^v(t)$, ..., $\Xi_n(t)$ represent stochastic processes responsible for the background noise evident in experimental data. They are defined in terms of the integrals (19) a,b,c,d with the velocity fluctuation replaced by its values for one of the three modes of propagation. Thus, the first term in I_3 gives rise to nine distinct integrals which contribute to the stochastic functions in (20).

Coupling of Unsteady Motions with Chemical Kinetics

The formulation described in the preceding section leads to the representation (20) of the unsteady pressure field as a collection of nonlinear oscillators driven by energy released by chemical reactions, coupling with the mean flow field, and interactions with the turbulence field. Nearly all published works on combustion instabilities neglect the influences of turbulence and are concerned with stability of small disturbances and some aspects of nonlinear behavior. The details of the chemistry are buried in representations of the unsteady heat release and normally receive little attention, although that omission is being corrected in recent works on unsteady burning of solid propellants.

As we remarked earlier, we assume here that the unsteady motions can be computed (i.e. by solving the set (20)) without specifying details of the chemistry. That assumption implies assigning the fluctuation Q' of heat release values that depend only on the velocity and thermodynamic variables. Having, in principle, determined the unsteady field, we then turn attention to its influence on the chemical

kinetics required to describe the formation of pollutants - here nitrogen oxides. Each of the species, having concentration $Y_i = \rho_i/\rho$ is described by the equation

$$\frac{\partial Y_i}{\partial t} + \rho \vec{u} \cdot \nabla Y_i = \nabla \cdot \left(\rho \vartheta \nabla Y_i \right) + w_i \tag{21}$$

where w_i is the production rate, having dimension s^{-1}.

Given the unsteady field (p', ρ', T' and $\vec{u}'$) and a formula for the source w_i' in terms of the field variables, the formation of species is to be computed by solving the set (21) written for the fluctuations. Presently there is no formal approximate method for doing so and solutions must be obtained numerically. That procedure would then give the global distributions of the Y_i' and the rates at which the species depart the chamber. A complete treatment of the problem must also include computation of nonisentropic temperature fluctuations which are lightly coupled (in both directions) to the species concentrations; we ignore that problem here.

The remainder of this paper is concerned with only a small but important part of the general problem just described, namely fluctuations w_i' of the local production rate of constituent i when the unsteady field is specified. We examine several simplified elementary modes of the kinetic mechanism as a means of investigating the response of the chemical kinetics to imposed oscillations. The results amount to an initial determination of models of the local "response function" for the reacting system. Such a response function could then be used as a representation of w_i'. We are far from carrying out the program suggested by the preceding discussion which here serves chiefly to provide the context for the calculations given in the following sections.

Calculations of NOx Formation

A PSR model with a single module was used in our simulations to obtain NOx levels as a function of temperature and pressure for a range of results. The model uses the Miller-Bowman mechanism [14] with residence time equal to 5 ms. The PSR model predicts the steady state temperature and species composition in a perfectly stirred reactor. The reactor is defined by a given volume, residence time, heat

loss parameter, inlet temperature and mixture composition. The contents of a PSR are uniform throughout due to high-level mixing.

For one set of simulations the pressure was varied between 5 atm and 20 atm for three different equivalence ratios. The relationships derived from these simulations have NOx levels proportional to p^n, where n is 0.155 for the stoichiometric case and changes to 0.198 and -0.173 for equivalence ratios 0.7 and 0.5 respectively.

The PSR model was also run at a constant pressure, varying the temperature in the lean limit. The results show a temperature dependence of the form NOx ppmv(15% O_2) = $AT^k exp(-\theta_a/T)$ with A = 49.28, n = 2.00, θ_a = 3.03e4 for a pressure of 10 atm and A = 56.024, n = 2.03, θ_a = 3.099e4 for pressure equal to 15 atm. The numbers from the simulation vary slightly with pressure; those values can be used to approximate the levels of NOx when both the temperature and pressure change. These relationships will be used in the following analysis to approximate the changes in NOx production in the presence of organized oscillations of the thermodynamic variables. It is expected that this analysis will provide a local value for w'_i in the real flow. This analysis will be valid when the acoustic time is greater than the chemical time of the system and oscillation ampltitudes much less than one.

The analysis involves dividing the flow variables into mean and fluctuation values:

$$p = \overline{p} + p'$$
$$T = \overline{T} + T'$$
$$NOx = \overline{NO}x + NOx'$$

(22)a, b, c

Time averaged values over one cycle of the oscillation are denoted $(\ )_{avg}$,

$$(F)_{avg} = \frac{\omega}{2\pi} \int_{t}^{t+2\pi/\omega} (F)dt$$

(23)

We assume that $(p)_{avg} = \overline{p}$, $(T)_{avg} = \overline{T}$, but as will be shown, due to the nonlinear behavior of the kinetics, $(NOx)_{avg}$ is not necessarily equal to $\overline{NOx}$, the NOx level at the operating conditions $\overline{p}$ and $\overline{T}$ with no oscillations present.

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Pressure Dependence of NOx Levels

The analysis proceeds by examining the effects of an oscillating pressure field on the average NOx levels. Experimental and computational results have shown a power dependence on the pressure of order n. A value of n = 1/2 has been used for design considerations for non-premixed combustors. Leonard and Correa [8] show that this design condition does not match their results for a lean premixed combustor. Studies using PSR models have shown a p^n dependence with n varying for different pressure ranges. Correa [5] observes that n approaches zero as the combustion approaches the lean limit. Newburry and Mellor [7] also list a range of values for n, ranging from -0.44 for an inlet temperature of 600 K, an inlet pressure of 30 atm, and an equivalence ratio of 0.5, using a PSR model and the Michaud, Westmoreland and Feitelberg mechanism [15], to 1.13 for the same conditions using the Drake-Blint mechanism [16]. The analysis used here will assume a general n; the impact of the value of n will be studied when the analysis is complete. Expanding this dependence into mean and perturbation terms leads to:

$$NOx \propto p^n = (\bar{p} + p')^n \tag{24}$$

with $\overline{NOx} \propto \bar{p}^n$ and assuming $p'/\bar{p} \ll 1$ leads to:

$$\frac{NOx}{\overline{NOx}} = \left(1+\frac{p'}{\bar{p}}\right)^n \approx 1 + n\frac{p'}{\bar{p}} + \frac{n(n-1)}{2!}\left(\frac{p'}{\bar{p}}\right)^2 + \frac{n(n-1)(n-2)}{3!}\left(\frac{p'}{\bar{p}}\right)^3 + \dots$$

$$\tag{25}$$

$$\frac{NOx'}{\overline{NOx}} \approx n\frac{p'}{\bar{p}} + \frac{n(n-1)}{2!}\left(\frac{p'}{\bar{p}}\right)^2 + \frac{n(n-1)(n-2)}{3!}\left(\frac{p'}{\bar{p}}\right)^3 + \dots$$

If we let $p'/\bar{p} = \sum_i \eta_i \cos(\omega_i t + \phi_i)$ where η_i is the oscillation amplitude for mode i, and ignoring spatial dependence, then we can determine the time dependence of the NOx fluctuation. To simplify this, the analysis will proceed using a single pressure mode. Applying (25) for a single mode $p'/\bar{p} = \eta\cos(\omega t)$ leads to:

$$\frac{NOx'}{\overline{NOx}} \approx n\eta \cos(\omega t) + \frac{n(n-1)}{2!}\eta^2 \cos^2(\omega t) + \frac{n(n-1)(n-2)}{3!}\eta^3 \cos^3(\omega t) + \dots \tag{26}$$

The average of this perturbation over one period $0 < \omega t \le 2\pi$, is:

$$\left(\frac{NOx'}{NOx}\right)_{AVG} = \sum_{k=1}^{\infty} \frac{n(n-1)(n-2)\cdots(n-2k+1)}{(2k)!} \left[\frac{1(3)(5)\cdots(2k-1)}{2(4)(6)\cdots(2k)}\right]\eta^{2k} \qquad (27)$$

The sign of this series is given by the number n(n-1)...(n-2k+1) since the other components of the series are positive. For $0 < n < 1$, all of the coefficients of the series are less than zero. Therefore, setting,

$$\left(\frac{NOx'}{NOx}\right)_{AVG} = \sum_{k=1}^{\infty} A_{2k}\eta^{2k} \;, \qquad (28)$$

$A_{2k} < 0$ for all k if $0 < n < 1$, which indicates that the time average of the perturbed NOx concentrations is less than zero. Hence, the average NOx emissions are reduced slightly in the presence of pressure oscillations if $0 < n < 1$. However, for $n < 0$ and $n > 1$, this trend is reversed, with $A_{2k} > 0$ for all k and an increase in the NOx emission levels. The strength of these variations from the steady-state value is also strongly dependent on the amplitude of the pressure oscillations which were assumed small compared to one for the analysis. As can be seen from (28), increasing the amplitude of the perturbation causes a greater reduction of emissions for $0 < n < 1$ or a corresponding increase in emissions outside this range.

Applying this analysis to higher modes in the system leads to similar behavior to that of a single mode, with slight variations due to phase differences between the modes. Examining this behavior for longitudinal modes in which $\omega_j = j\omega_l$, we find:

$$\left(\frac{NOx'}{NOx}\right)_{AVG} = \sum_{k=1}^{\infty} A_{2k} \sum_{j=1}^{\infty} \eta_j^{2k} + O(\eta_j^4) \qquad (29)$$

where the higher order terms are dependent on the phase difference between the modes. Results for higher modes show that a similar behavior depending on the value of n occurs, with slight variations to fourth order and higher depending on the amount of phase difference present.

Temperature Dependence of NOx Levels

Similar analysis can be performed using the effects of temperature oscillations alone on NOx levels. Previous work on temperature dependence has shown an exponential dependence similar to the Arrhenius form of the rate equations:

$$NOx \sim T^k \exp(-\theta_a/T) \qquad (30)$$

where θ_a has units of temperature. Due to this exponential dependence on temperature, any perturbation will have an adverse effect on pollutant levels by accelerating the reaction [17]. An increase in temperature will lead to a larger reaction rate and an increase in NOx levels.

Our PSR results used the form $NOx = AT^k exp(-\theta_a/T)$, which leads to:

$$\frac{NOx}{\overline{NOx}} = \left(1+\frac{T'}{\overline{T}}\right)^k exp\left[\frac{\theta_a}{\overline{T}}\left(\frac{T'/\overline{T}}{1+T'/\overline{T}}\right)\right]$$ (31)

Similar to the pressure effects, a single mode for the temperature oscillation $T' = \overline{T}\eta cos(\omega t)$ can be substituted into (31) and then averaged over one period to show how the NOx levels vary with temperature. Even for a single mode oscillation, the expansion analysis is lengthy, and a graphical representation will better demonstrate the fluctuations. Using the constants for the Miller-Bowman mechanism with a 5 ms residence time at a mean temperature of 1900 K as an example, a 5% oscillation in temperature leads to a 19.4% increase in the mean NOx level against the case with no oscillations.

This behavior is directly observable from the exponential dependence. The increase in NOx levels during one half of the cycle are much higher than the corresponding decrease during the other half of the cycle. Averaging this trend over one cycle leads to a distinct increase in emission levels. Figure 1 shows how $(NOx'/\overline{NOx})_{avg}$ varies with the amplitude of the temperature oscillation. As indicated in Fig. 1, a small oscillation, on the order of 5% can lead to a 20% increase in the average amount of NOx produced in the system. This is a substantial elevation in pollutant emissions levels in the presence of an oscillating field.

Fig. 1

Higher longitudinal modes in the oscillation were also examined for the temperature oscillations. Additional modes have a large impact on the increase of NOx levels, and the effect of phase difference is much more noticeable than for the case examining pressure dependence. Figure 2 shows results of the increase for two and three modes versus the phase offset of the highest mode of the oscillation. The amplitude of all the modes was taken as 0.05. The behavior of the temperature dependence is always towards an increase in the NOx levels, but the fractional amount of the increase depends strongly on not only the number of modes, but also the phase difference between the modes. As Fig. 2 demonstrates, the lowest fractional increase occurs: when the modes are 180° out of phase for a two mode system; and when

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the first and third modes are in phase and the second mode is 180° out of phase for a three mode system. This behavior for the lowest fractional increase occurs for even higher mode cases.

Pressure and Temperature Dependence of NOx Levels

Finally, the analysis examines combined pressure and temperature effects, which may not be applicable in lean systems where $n \approx 0$ and NOx is only temperature dependent. Of course, the p and T fields will still be dynamically coupled which will affect NOx production, but the levels will not be directly dependent on the pressure level. Combining the two effects leads to:

$$NOx \sim AT^k p^n exp(-\theta_a/T) \tag{32}$$

With $p'/\bar{p} = \eta_p cos(\omega t)$ and $T'/\bar{T} = \eta_T cos(\omega t + \phi)$, the effects of a single mode in the oscillation and a phase factor ϕ between the temperature and pressure oscillations can be examined.

The time averaging was applied to the results for Miller-Bowman mechanism set to a 5 ms residence time. The amplitudes η_p and η_T were set equal to 0.05 and ϕ was varied. The NOx levels showed a dependence on the phase difference ϕ approximated by the formula $(NOx'/\overline{NOx})_{avg} = a + b \, cos\phi$, where $a = 0.2146$ and $b = 0.0131$ for this example. The fractional changes are higher than the mean NOx values without oscillations and show that the temperature oscillations play a major role in the variation of pollutant emissions. The pressure effects play some role in defining the exact levels, but are not strong enough to dominate the behavior. The effect of higher modes in the system displays similar behavior, with the higher modes of the temperture oscillation playing the largest role in the changes. The phase differences between the modes also behaves like it does when the pressure and temperature effects are considered separately. With similar amplitudes between the pressure and temperature fluctuations, the average NOx levels will always increase due to the strong dependence on the temperature field, with slight changes due to the phase differences between the modes and the pressure and temperature fields.

Conclusions

This preliminary analysis provides an elementary basis for determining how the production rate of NOx varies in the presence of an oscillating flow field. This effect will be important in future gas turbine

research due to the increase liklihood of combustion instabilities when combustors are operated near the lean blowout limit to reduce NOx production. Large scale oscillations within the gas turbine combustor may have negative consequences for pollutant emissions, leading to levels above the specified limits. The present analysis has shown that the exponential dependence on the temperature field causes a noticeable increase in the average amount of NOx produced, while the dependence p^n on pressure produces either an increase or a decrease in the levels depending on the value of n. These preliminary results will be extended and incorporated in more extensive analysis of the interactions between the kinetics and the acoustic oscillations in a turbulent reacting combustor.

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Captions for Figures

Figure 1 - Variation of Mean NOx Concentration in the Presence of a Single Mode Temperature Oscillation - $\overline{T}$ = 1900 K

Figure 2 - Variation of Mean NOx Concentration in the Presence of Higher Mode Temperature Oscillations - Varying Phase Offsets Between Modes - $\overline{T}$ = 1900 K
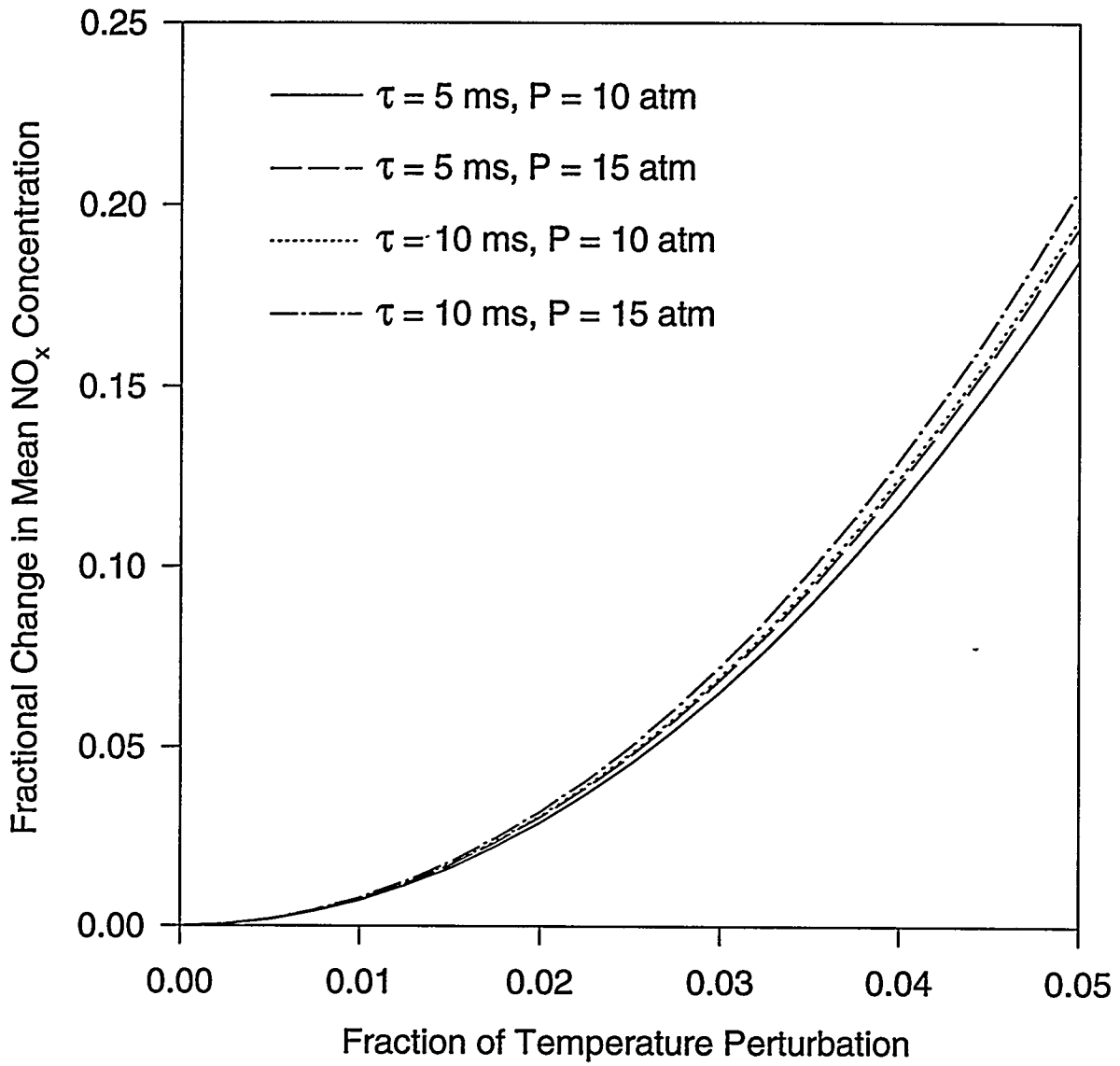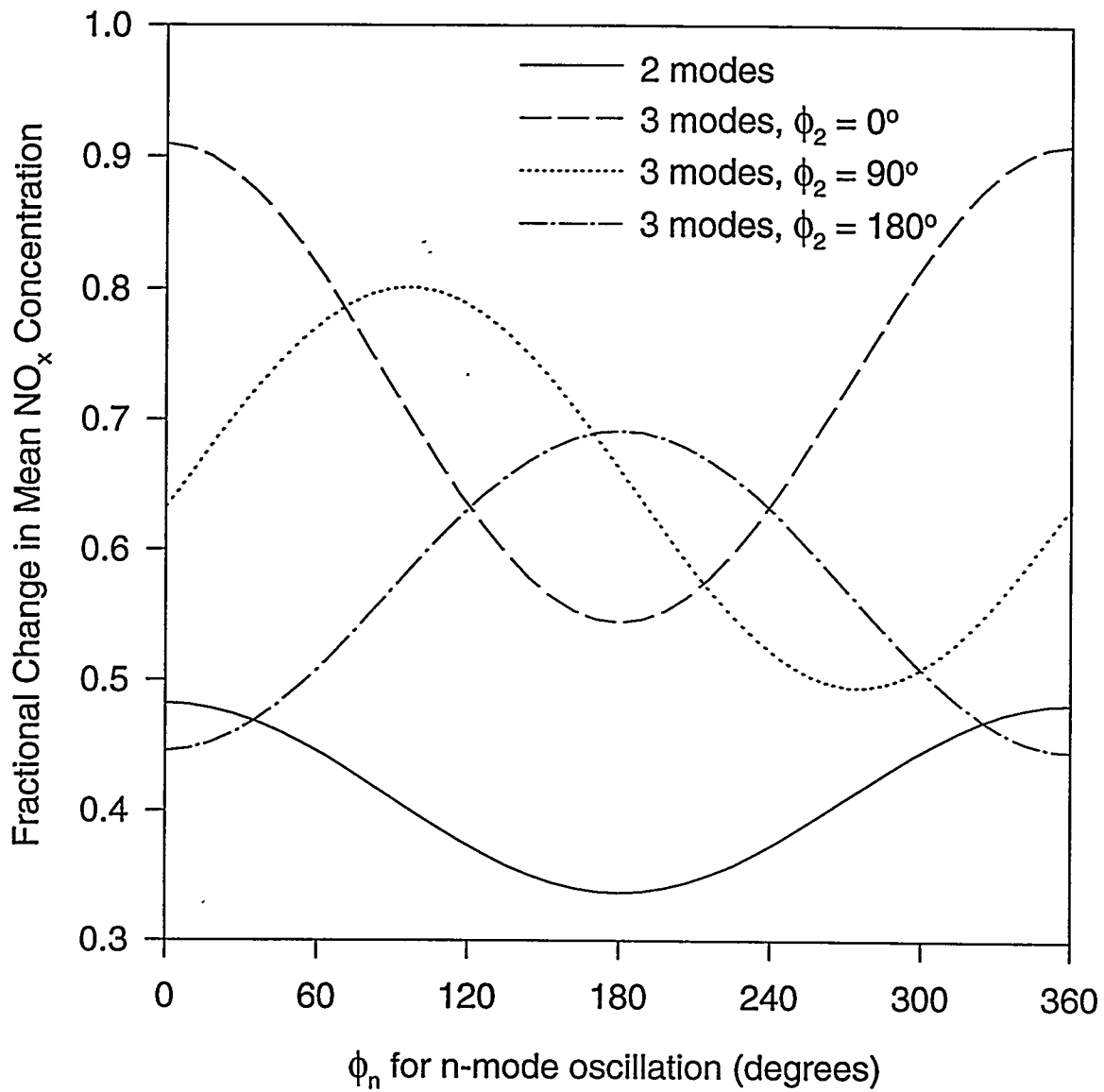
FIGURE 1

FIGURE 2

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