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Effects of Fuel-Air Unmixedness on NO_x Emissions

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Unmixedness effects of lean fuel-air mixtures on emissions of nitrogen oxides (NO_x) are experimentally studied. The objective is to study the relative influence of temporal fuel concentration fluctuations and spatial nonuniformities of mean concentration profiles on NO_x emissions from lean premixed combustion. The NO_2 laser-induced fluorescence (LIF) technique is used to quantify unmixedness levels at the flameholder. Unmixedness data are related to NO_x emissions measurements from downstream of the flames. The results show that temporal unmixedness contributes significantly to higher NO_x emissions. It is not sufficient to eliminate only spatial nonuniformities of mean concentration in order to minimize NO_x emissions from lean premixed combustion.

Nomenclature

c = volumetric fuel concentration
c' = rms fluctuation component of c
$\bar{c}$ = time-mean fuel concentration
D_c = combustor diameter
D_{cf} = coflow diameter
D_j = jet diameter
Re_{cf} = coflow Reynolds number
Re_j = jet Reynolds number
U = unmixedness parameter
X_j = distance from jet to flameholder
z = vertical coordinate; origin at combustor centerline
ϕ = fuel-air equivalence ratio

Introduction

LEAN premixed combustion has received extensive attention recently as a method to reduce NO_x emissions from gas turbine combustors. This has been the case for both land-based power generation gas turbines[1,2] and aircraft engine gas turbines.[3,4] However, for lean fuel-air mixtures, reaction rates responsible for NO_x production increase exponentially with temperature. This nonlinear dependence of NO_x on equivalence ratio is shown clearly in the results of Leonard and Correa.[5] It is most desirable to burn always and everywhere at the overall equivalence ratio. Fuel-air mixtures that burn closer to stoichiometric result in higher levels of NO_x. Those that burn leaner than the overall equivalence ratio reduce NO_x. However, because of the nonlinearity, the net effect is an increase in NO_x emissions. It follows that the success of lean premixed combustion depends on the proper mixing of fuel and air prior to combustion. Because of the difficulties in mixing fuel and air fully in real combustors, the word "premixed" in lean premixed combustion often refers more to the goal of the process rather than its realization in practice.

In one sense, the quantity of NO_x emitted from lean premixed combustion is an indicator of fuel-air unmixedness just prior to combustion. The purpose of this work was to go one step further—to measure unmixedness levels at the combustor flameholder and directly relate them to NO_x emissions. The effects of both spatial nonuniformities in time-mean fuel mixture concentration and temporal fluctuations of fuel mixture fraction about the mean on NO_x emissions are quantified. In particular, our interest here is to study the relative impact of temporal fluctuations on NO_x.

A few examples of past works that address effects of fuel-air nonuniformities on NO_x emissions are cited here. Pompei and Heywood[6] studied the influence of initial conditions and subsequent mixing in a kerosene-fueled combustor. Atomizing pressure was varied to affect mixing and subsequently NO_x emissions. Similarly, the analytical and experimental study of Lyons[7] showed that spatial fuel-air nonuniformity leads to increased NO_x emissions for lean mixtures. Most recently, Maughan et al.[8] considered effects of initial gaseous fuel distribution from the injection process and effects of (premixer) mixing distance on NO_x emissions. They showed that an optimum fuel injection distribution at the upstream end of the premixer exists for minimum NO_x. In addition, longer mixing distances, i.e., longer premixers, reduced NO_x emissions. The influence of unmixedness on NO_x emissions is clearly implied. However, the results of Maughan et al. do not quantify unmixedness directly, and no distinction between spatial and temporal unmixedness was made.

For the experiments reported in this article, methane was used as the fuel. Although we consider only a gaseous fuel, the results should also be relevant to liquid-fueled combustors which rely on some premixing to reduce NO_x emissions.

The unmixedness parameter used to quantify temporal fluctuations in fuel concentration c is the same one used by Danckwertz[9] and, more recently, by Dimotakis and Miller.[10] Danckwertz sought quantitative expressions for "goodness of mixing" and introduced the quantity, here called the level of unmixedness

$$U = \frac{c'^2}{\bar{c}(1 - \bar{c})}$$

where c'^2 is the variance of fuel concentration fluctuations and $\bar{c}$ is the time-mean fuel concentration at a location. The elegance of using U to quantify unmixedness is that its value ranges from 0 for a perfectly mixed case to 1 for a case of maximum variance, where it can be said that the fuel and air are stirred but not mixed.[10]

Further quantitative information on unmixedness is given here by profiles of $\bar{c}$, c', and U for several different degrees of premixing. Probability density functions (pdfs) of fuel concentration, obtained experimentally, are also shown.

Experimental Details

In the design of this experiment there were two aspects that were considered essential. First, a diagnostic that could adequately resolve temporal concentration fluctuations and spatial nonuniformities at the flameholder location was nec-

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essary. The study of Mansour et al.,[11] which considered spatial-averaging effects of probe volume size on mean, rms, and pdf measurements, was used as a guide to help determine what probe volume size could be considered adequate. Second, upon varying the degree of premixing upstream of the flameholder, the flame needed to be held steadily at the same location in every case. Therefore, a flameholder was required; a lifted flame would have made it difficult to determine where to make unmixedness measurements. However, a sufficiently benign flameholder was desired so it would not mask the upstream differences in unmixedness by adding significant mixing itself. The first requirement led us to use the NO_2 laser-induced fluorescence (LIF) technique to quantify unmixedness. This technique was specifically developed by Gulati and Warren[12] for the purpose of measuring cold flow mixing. Briefly, this technique involves using NO_2 gas as a seed marker in a stream one would like to detect. For example, if the fuel supply is seeded with NO_2, subsequent dilution and mixing of that fuel can be measured in the premixer. NO_2 fluoresces under excitation of the 488.0-nm line of a continuous wave argon-ion laser. Here, a sampling rate of 20 kHz was used, resolving fluctuations to 10 kHz. The laser probe volume was approximately $0.5 \times 0.5 \times 2.0$ mm. This probe volume was primarily limited by obtaining an adequate signal to noise ratio to resolve fuel concentration fluctuations at the 5% level. We expect that this spatial resolution will produce somewhat lower rms values and smoother pdfs than are actually present.[11] A general discussion of NO_2 LIF and further particulars to the present setup are available in Gulati and Warren.[12] The second aspect led us to use a simple jet in a coflow arrangement as the fuel-air mixer. Such a setup was effective in altering the amount of mixing prior to the flameholder.

The experimental setup is shown in Fig. 1. The combustor is located inside a suction wind tunnel. The fuel and air supplies consist of a concentric jet in a coflow arrangement where the jet inside diameter D_j is either 3.2 or 4.2 mm, and the coflow diameter D_{cf} is 40 mm. Screens and honeycombs were used in the coflow tube to help make the coflow sufficiently uniform. In general, arbitrary amounts of fuel and air could be fed through the jet and coflow tubes. However, the jet typically injected most of the fuel while the coflow provided the air. The one exception was the premixed case, where both fuel and air were carried by the coflow, and the jet was plugged. Both the fuel and air supplies were metered with choked flow orifices.

The flameholder was located at the juncture of the coflow and combustor tubes. After significant trial and error, the flameholder settled on consisted of the combination of a rearward-facing step and a cross hair-like bluff body. (The flameholder is visible in the photographs of Fig. 2.) The flameholder was required to hold the flame at the same location (i.e., without liftoff) for a variety of unmixedness levels. In addition, the minimal possible disturbance to the mixture of unburned fuel and air was sought. The reason for this was that the unmixedness measurements were made at the location of the flameholder without the flameholder in place. (As explained below, the NO_2 LIF measurements were performed under cold flow conditions, whereas the NO_x emissions measurements were made in a separate set of combusting experiments with the flameholder in place.) In essence, we did not want the flameholder to mask the measured differences in unmixedness by being a significant mixer itself.

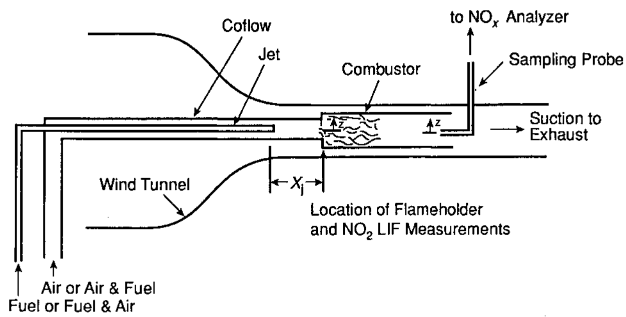

Fig. 1 Experimental setup showing coflow, jet, and combustor (not necessarily drawn to scale).

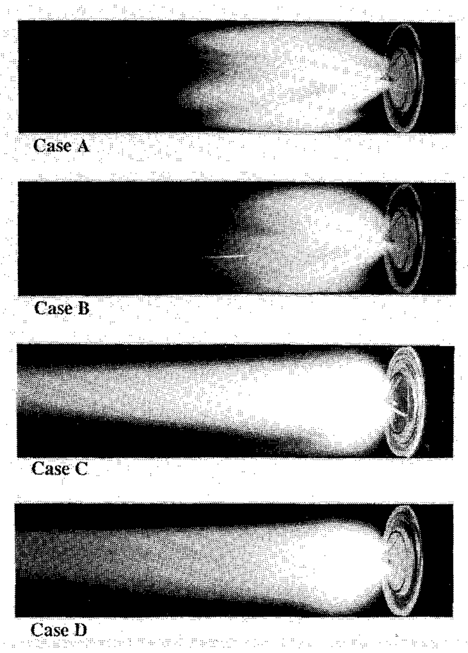

Fig. 2 Photographs of flames from the four cases. Flow is from right to left.

Table 1 Flow conditions

Case	X_j/D_{cf}	D_j, mm	Re_j	Re_{cf}	ϕ
A	—	—	—	8900	0.5
B	10	3.2	5100	8500	0.5
C	4	4.2	7800	8100	0.5
D	4	3.2	5100	8500	0.5

The amount of mixing, or the level of unmixedness of the methane-air mixture was varied by changing the distance from the jet to the flameholder and/or by diverting some air from the coflow to the fuel jet. The practical extremes in unmixedness and choices of flow rates were constrained by requiring the flame to be stabilized at the same location, maintaining equal equivalence ratios, for all cases. For example, if the flow rate through the coflow tube was too low and the mixture was sufficiently well-mixed the flame would flash back upstream of the flameholder. If the flow rate was too high or if the mixture was too unmixed the flame would lift off from the stabilizer. Since the flame stabilization location and the location of the LIF measurements were considered essential constants in the experiments, this constraint was acceptable.

The combustor portion of the setup had a diameter D_c of 81 mm. NO_x emissions data was sampled with a quartz probe about 7.5 combustor diameters downstream of the flameholder. A Beckman 951 NO_x analyzer was used to detect NO_x levels to ppm levels. The probe was traversed vertically to obtain profiles of NO_x near the exit of the combustor. The supply tubes and combustor were installed inside the wind tunnel. The tunnel flow provided cooling flow and, more importantly, a NO_2 flow path under suction relative to ambient conditions. Calibrated source bottles of 5090 ppm NO_2 in N_2 were used, and since NO_2 is extremely toxic at levels much lower,[13] the wind tunnel provided a safety factor if any leaks in the system would have occurred.

The premixer portions of the coflow tube and combustor were made of quartz, for visual access to the flame. The wind-tunnel test section allowed for optical access, which was necessary for the NO_2 LIF measurements. The focused laser beam passed vertically through the test section. The scattered light fluorescence signal was collected onto a photomultiplier tube located near a side wall of the tunnel.

Results of four cases are reported on here, and their flow conditions are summarized in Table 1. Re_j is based on the jet velocity, D_j, and the kinematic viscosity of methane or a methane-air mixture, as the case may be. Likewise, Re_{cf} is based on the coflow velocity, D_{cf}, and the kinematic viscosity of air or a methane-air mixture. All experiments were performed at an overall equivalence ratio ϕ equal to 0.5. ϕ is based on the net flow of fuel and air injected either through the jet or coflow. All tests were done at atmospheric pressure with no preheating of combustion air.

A preliminary step in the experiments required a survey of combinations of jet velocities, coflow velocities, and jet-to-flameholder distances that produced a stable flame, one which would not flash back or lift off. Following this, the experiments were performed in two steps. First, the NO_2 LIF technique was used to measure unmixedness profiles at the exit of the coflow tube; the flameholder was removed as was the combustor tube. For the reasons of increased signal strength and linearity of NO_2 concentration vs signal, fuel was represented by nitrogen seeded with NO_2 (5090 ppm) instead of methane seeded with NO_2.[12] The effects from fuel density differences which this introduces are considered to be small. The second step involved combusting runs, where the fuel was methane, and all pieces of the combustor were in place. This is when NO_x emissions profiles near the combustor exit were taken. Volume flow rates, or velocities, were matched in going from the cold flow mixing experiments to the combusting runs. To prevent confusion, it is noted again that the NO_2 used to seed the fuel flow was only used in the cold-flow portions of the experiments; the NO_2 in NO_x emissions was due solely to the combustion processes between pure methane and air.

Results

The flames produced by cases A, B, C, and D are shown in Fig. 2. Flow is from right to left in these photographs, with the flameholder just visible at the right side of each picture. In each of the four cases the flame is anchored at the exit of the coflow tube. Case A is considered to be fully and uniformly premixed combustion. Methane and air were injected into the coflow tube approximately 50 coflow diameters upstream of the flameholder. The following results indicate that this length is sufficient for complete mixing. Case D is the most poorly mixed since the jet is pure methane, the coflow is pure air, and the jet is only four coflow diameters from the flameholder. Its mixing length is therefore relatively short. Cases B and C are in between cases A and D in terms of unmixedness (see Table 1). Two observations can be made from the photographs: 1) flame length increases with increased unmixedness; and 2) the flames, particularly the longer ones, show that flow rises in the combustor. The asymmetry caused by flow rising will also be evident from the NO_x profiles at the combustor exit. These observations suggest that buoyancy effects are present in the combustor. In fact, Froude number based on combustor velocity and diameter is estimated to be about 5, and so the presence of buoyancy effects is not surprising. Flow velocities could not be increased here since then the flame would lift off. We assume that the main results presented here, relating unmixedness to NO_x, are not affected by this flow asymmetry.

The main objective of this work was to consider the influence of temporal mixture fluctuations on NO_x. The most direct way to investigate this influence appeared to be to create two cases with equivalent mean fuel concentration profiles at the flameholder, but with different mixture fluctuation levels—the best example obtained with this setup is the comparison of cases A and B.

Mean fuel concentration $\bar{c}$ and rms fuel concentration fluctuations c' profiles are shown in Fig. 3 for cases A and B. As

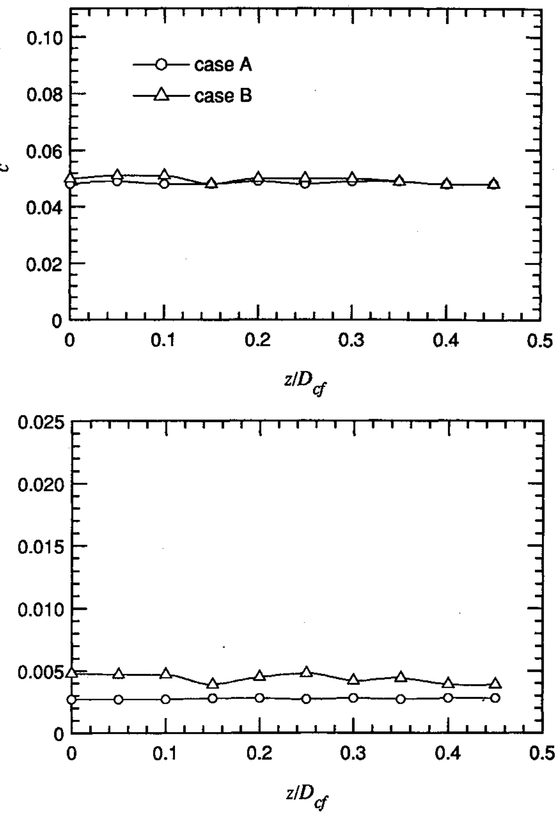

Fig. 3 Profiles of $\bar{c}$ and c' for cases A and B.

discussed earlier, case A is considered to be a fully premixed case. As is expected, both profiles for A are uniform from the centerline to the edge of the coflow tube. However, c' is about 5% of $\bar{c}$. Ideally, the fully premixed condition would have $c' = 0$. This discrepancy could be accounted for by a mixture that is not fully premixed. However, it appears that this c' level is primarily signal noise. The NO_2 LIF noise level for a diluted NO_2 concentration of 270 ppm is close to 5% of signal strength. Recall that fuel, for the unmixedness data, is nitrogen seeded with 5090 ppm NO_2. Since approximately 19 parts air mixes with 1 part fuel to achieve $\phi = 0.5$, the LIF signal strength expected for case A is that corresponding to approximately 270 ppm NO_2. Therefore we deduce that nearly all of the c' level in Fig. 3 for case A is signal noise and not mixture fluctuation. Figure 3 shows that $\bar{c}$ profiles for both cases A and B are spatially uniform. In this sense, both are mixed well; spatial unmixedness is very low. However, temporal concentration fluctuations, as indicated by c', are found to be larger for case B than case A.

Degrees of spatial and temporal unmixedness are also shown by concentration pdfs. pdfs for cases A and B are shown in Figs. 4a and 4b, respectively. In both cases the pdfs are centered near $\bar{c} = 0.5$ for all z/D_{cf}. However, the pdfs for case B are lower and broader, as is expected from the c' data. Finally, Fig. 5 compares U. For both cases U is very low, in fact, much less than 1. As is again expected, the more poorly (temporally) mixed case B has higher levels of U than case A.

Figures 3–5 show that both cases A and B are spatially well-mixed, i.e., all quantities (and in particular $\bar{c}$) are uniform in z at the flameholder location. However, temporal unmixedness, as indicated by the rms, pdf, and U data, are greater for case B. The significant effect of temporal unmixedness on NO_x emissions is seen in Fig. 6. Figure 6 shows NO_x profiles downstream of the flame. NO_x is about twice as large for case B than for case A. This result shows that temporal unmixedness can significantly increase NO_x emissions. Since $\bar{c}$ data are spatially uniform for both cases, the effects of nonuniformities in mean profiles are removed and only

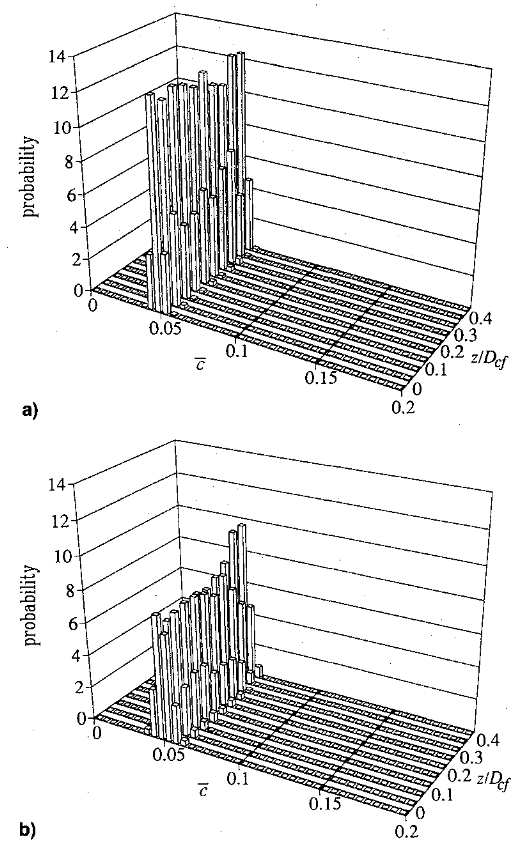

Fig. 4 Probability density functions of fuel concentration: a) case A and b) case B.

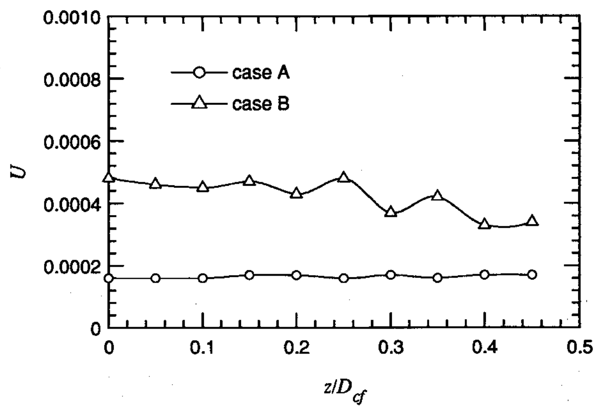

Fig. 5 Profiles of unmixedness parameter U for cases A and B.

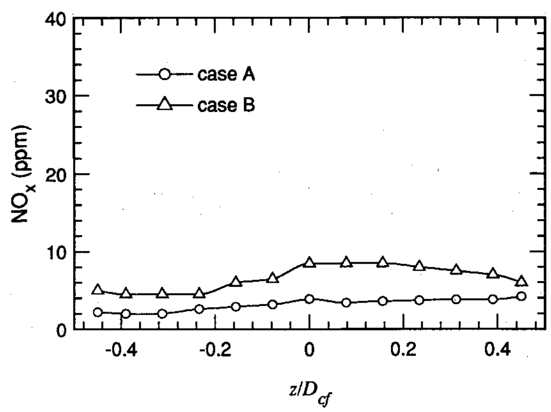

Fig. 6 Profiles of NO_x for cases A and B show increased emissions when temporal fluctuations are greater.

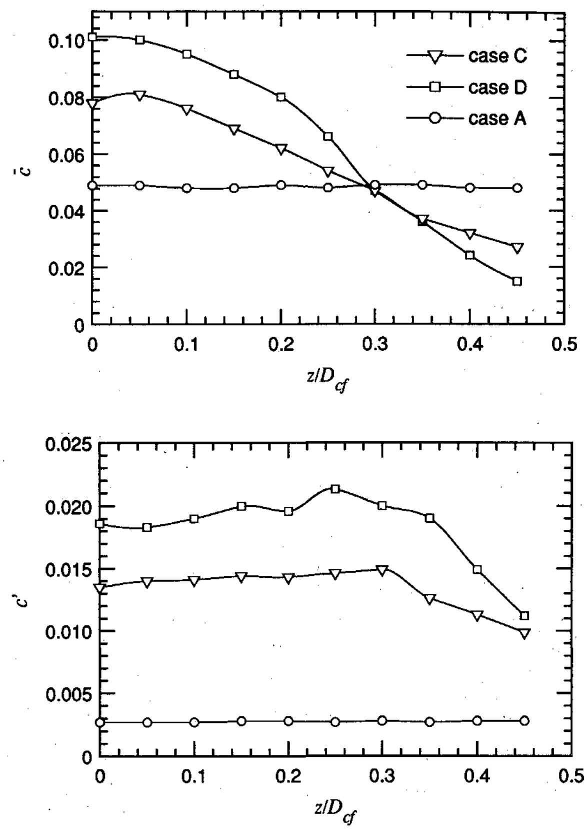

Fig. 7 Profiles of $\bar{c}$ and c' for cases C and D. Case A is shown for comparison.

those contributions of temporal fluctuations affect the increased NO_x of case B.

Since the amount of NO_x produced generally increases with either flame temperature or residence time, a couple of comments regarding the setup and NO_x production are pertinent. With radiation losses and convective cooling from the wind-tunnel flow, this combustor is clearly not adiabatic, and therefore, NO_x levels measured here are likely to be low relative to those in adiabatic flames. However, the relative amount of heat loss among each of these cases is likely similar; the degree to which this combustor is nonadiabatic should not affect comparisons made among them. The combustor residence times are about 0.2 s in these cases. Such residence times are relatively long compared to real gas turbine combustors, and it follows that NO_x levels are expected to be inflated. Again, since we are comparing cases of equal residence times, the comparisons made here should be valid.

Cases C and D data are shown in Fig. 7. The baseline data from case A is included for comparison case. With the jet closer to the flameholder, $\bar{c}$ profiles are nonuniform, and rms levels are much larger now. Likewise, the concentration pdfs are broader for both cases (Fig. 8). Figure 9 shows large values of U relative to the premixed case (case A) and case B. However, U is still much less than its maximum attainable value of 1, despite largely nonuniform $\bar{c}$ profiles and c' levels greater than 20% of $\bar{c}$.

Clearly, cases C and D represent poorly premixed cases. Both spatial and temporal unmixedness are quite large. The effects of such high levels of unmixedness on NO_x are seen in Fig. 10. Here the influences of both nonuniform mean profiles and large fluctuations show in NO_x emissions. The shapes of the NO_x profiles across the combustors for cases C and D suggest that the structure of the flame is such that more NO_x is produced near the combustor axis. The peak in NO_x profiles is shifted toward the top of the tube under the effects of buoyancy. As noted previously when discussing the photographs of Fig. 2, the Froude number for these flows is about 5, indicating that the effects of buoyancy are not insignificant.

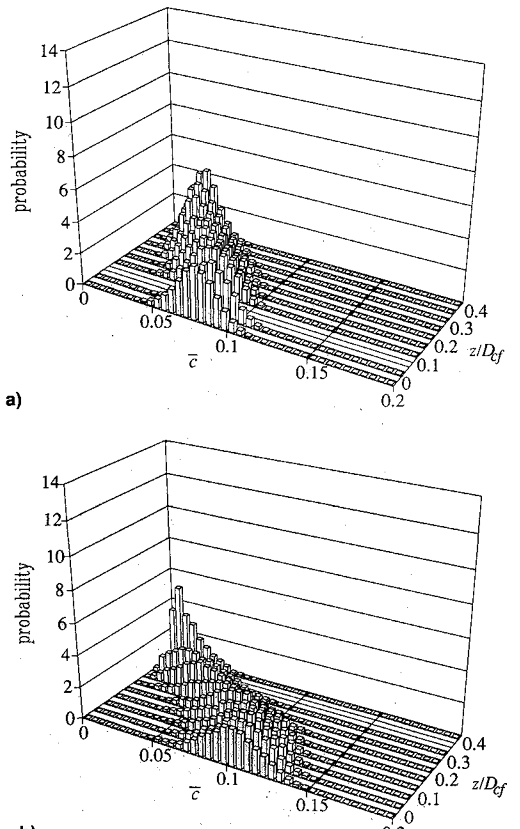

Fig. 8 Probability density functions of fuel concentration: a) case C and b) case D.

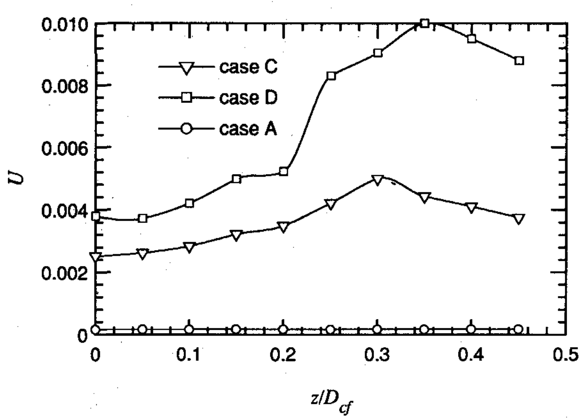

Fig. 9 Profiles of unmixedness parameter U for cases C and D. Case A is shown for comparison.

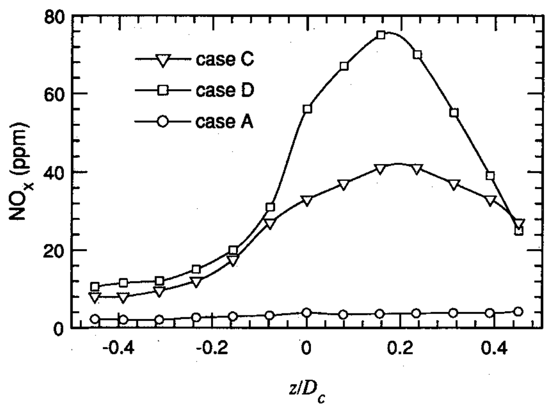

Fig. 10 Profiles of NO_x for cases C and D show effects of poorly mixed cases on emissions.

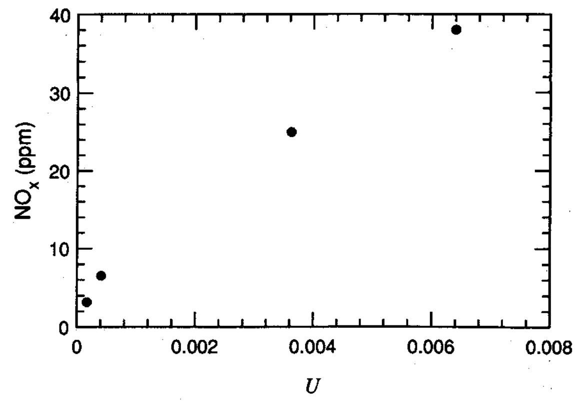

Fig. 11 Qualitative relationship between NO_x and U.

Finally, the qualitative dependence of NO_x emissions on U is suggested in Fig. 11. Here, profile-averaged NO_x data are plotted against profile-averaged U. It is assumed that velocity profiles are uniform at both measurement locations. No velocity profiles were measured at either the flameholder location or the position at which emissions data were taken. Figure 11, however, does not distinguish between the effects of temporal and spatial unmixedness on NO_x emissions. The first two data points (from cases A and B) show the effects of temporal fluctuations on NO_x. The higher two data points (from cases C and D) contain effects of both nonuniform mean profiles and temporal unmixedness.

Concluding Remarks

The primary conclusion from this work is that it is not sufficient to consider only spatial nonuniformities in fuel mixture when quantifying the effectiveness of a premixer to minimize NO_x emissions. Temporal unmixedness, or concentration fluctuations, contribute significantly to higher NO_x emissions. The results show that seemingly small levels of temporal unmixedness, $c'/\bar{c}$ on the order of 10% or $U \simeq 0.0004$, can double NO_x emissions relative to a perfectly premixed flame, given spatially uniform mean concentration profiles in both cases.

The results suggest that if both spatial and temporal unmixedness are measured prior to combustion of the mixture, then the NO_x performance caused by combustion can be anticipated. In these experiments the NO_2 LIF technique proved to be very effective in measuring mean and fluctuating fuel concentrations. We note that unmixedness at the flameholder is likely to be most influential when subsequent mixing caused by the flameholder and in-flame mixing effects are minimal. This study did not address the role of mixing downstream of the flameholder. Nonetheless, for lean premixed combustion, the goal of optimum mixing in the premixer section appears to remain critical.

The influence of mixture fluctuations may even be greater at higher, more realistic, pressures at which gas turbine combustors run. The experiments were performed in a lab-scale combustor at atmospheric pressure. We can anticipate that as pressure increases, increases in fluid mechanical mixing rates within the combustor may not keep up with increases in combustion reaction rates, and therefore, the effects of initial unmixedness would be even greater at higher pressures. Implied in this statement is that subsequent mixing of unburned fuel and air occurs in the flame region. Such additional mixing would likely help reduce NO_x. At higher pressures the opportunity for additional mixing prior to burning may lessen if reaction rates increase sufficiently with pressure.

Even though these experiments were carried out with gaseous fuel only, the significance of the results are believed to be relevant also to liquid-fueled combustors. With liquid fuel, the goal of achieving complete mixing is likely to be more difficult, however, since atomization and vaporization are required. The methods of atomization and vaporization are likely to be influential in determining the level of unmixedness in the mixture that burns.

An initial hope of these experiments was to specifically address whether the pattern of concentration fluctuations, as indicated by characteristic pdfs and spectra, is influential in NO_x emissions. In other words, we sought to create mixtures of equal rms or U levels and with uniquely shaped pdfs or unique spectra. However, within the constraints of this experimental setup, we were unable to produce such fluctuations.

Finally, what comprises a well-mixed mixture is essentially relative to the situation. Recall that U can range from 0 at the perfectly mixed state to 1 at the totally unmixed (but stirred) state. The largest value of U measured here was 0.01—far from the totally unmixed state—but still poorly mixed in terms of NO_x production. Order of magnitude reductions in NO_x emissions are possible by reducing U beyond 0.01. The results suggest that values of U much less than 1 are not sufficient for minimal NO_x production.

Acknowledgments

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