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Soot measurement in diluted methane diffusion flames by multi-pass extinction and laser-induced incandescence

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ABSTRACT

Multi-pass cavity line-of-sight extinction (MPC-LOSE) and laser-induced incandescence (LII) techniques are deployed to measure the soot volume fraction in a series of nitrogen-diluted flames, which produce only ppm volume mass fractions of soot. The separate suppression effects on soot formation of direct fuel dilution and indirect effects of temperature and residence time are interpreted by using a numerically calculated flow velocity and temperature field using a one-step fast chemistry model. The experimentally determined rate of soot formation is shown to obey approximately the same function of the local temperature for all dilution cases. The results show that a simple one-step reaction model using previously measured activation energies can account for the dilution effect with good accuracy. The results show that the direct effect of dilution on concentration is comparable to the effects of changing the temperature estimated local temperature and residence time.

1. Introduction

Soot from combustion sources is both a significant atmospheric pollutant, as well as a contributor to climate change [1,2]. Understanding the process of soot production, and creating predictive models is part of the research into the development of clean, soot-free combustion systems. Here, we investigate inert-diluted hydrocarbon flames, which typically produce significantly lower soot than in undiluted flames, thus presenting challenges to usual measurement methods [3–8].

Line-of-sight extinction (LOSE) methods [9–11] have been widely used for soot detection and measurement, as they can yield *absolute* values of soot volume fraction (f_v) in uniform or symmetric systems, and are relatively simple and inexpensive to implement using a low power continuous wave (CW) laser and photodiodes. Laser-induced incandescence (LII) imaging produces two-dimensional maps of *relative* soot volume fractions, but requires calibration. The techniques are therefore complementary, and have been used together in the past [12,13].

In our previous work, multi-pass cavity line-of-sight extinction (MPC-LOSE) was utilised [11] to measure the f_v in low-soot, diluted flames. The MPC-LOSE increases the level of absorption by a order of magnitude relatively to a single pass technique, while maintaining spatial resolution.

Previous studies conducted using the LOSE technique on diluted diffusion flames have used fuels that produce high levels of soot [4,7,8] and thus can be measured with single pass extinction techniques. In this paper, we extend the technique to measure low sooting methane flames, and investigate the sensitivity limits of the multi-pass technique. The resulting signal to noise ratio is sufficiently large to allow the extraction of spatially resolved f_v along both radial and axial directions via LII.

A series of nitrogen-diluted laminar diffusion flames produced on a burner similar to one used by Shaddix et al. [12,13], Santoro et al. [14,15] and Puri et al. [16] are investigated. LII is used for imaging the soot distribution, which is quantitatively calibrated with MPC-LOSE. The two measurements reveal the effect of dilution on the total soot volume fraction.

Dilution results both in a direct lowering of the soot volume fraction via a concentration effect, as well as an indirect effect by lowering the flame temperature and changing the residence times. In order to separate these effects, a simplified fast chemistry model of a jet co-flow diffusion flame is implemented numerically to estimate the local temperature. This allows the assessment of the direct (concentration) and indirect (temperature and residence time) effects of dilution on soot, via a one-step chemistry model, as detailed further on.

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Nomenclature

Y_F	mass fraction of fuel
Y_O	mass fraction of oxygen
X_F	mole fraction of fuel
X_O	mole fraction of oxygen
T	temperature
T_f	flame temperature
T_{ad}	adiabatic temperature as a function of mole fraction of fuel
$T_{ad,0}$	adiabatic temperature of undiluted flame
T_l	local temperature
$T_{O,0}$	temperature of oxygen in co-flow
$T_{F,0}$	temperature of methane in fuel flow
c_p	heat capacity of gas
Q	combustion enthalpy of methane
$Y_{O,0}$	mass fraction of oxygen in co-flow
$Y_{F,0}$	mass fraction of fuel in fuel flow
$X_{O,0}$	mole fraction of oxygen in co-flow
$X_{F,0}$	mole fraction of fuel in fuel flow
f_v	soot volume fraction
f_n	normalised soot volume fraction
f_v^*	maximum soot volume fraction
ξ_{st}	stoichiometric mixture fraction
u_z	axial velocity
u_r	radial velocity
s	stoichiometric ratio of air and fuel
λ	heat conductivity
ρ	density
ω	chemical reaction rate
T_{ref}	reference temperature of cold mixture
W	molecular weight
D_f	mass diffusivity
C_s	Sutherland's constant
r_p	inner diameter of fuel port
E_a	global activation energy for soot formation
A_p	pre-exponential coefficient for soot reaction rate
A	single pass laser extinction factor
B_p	pre-exponential coefficient for soot reaction rate
μ_v	viscosity
T_p	temperature of soot particle
K_e	local extinction coefficient
P_t	total laser extinction projection
P_0	single pass laser extinction projection
m	complex refractive index of soot particles
S_{LII}	integrated LII signal intensity with probe volume
K_{LII}	LII calibration coefficient
$E(m)$	absorption function of soot particle, $E(m) = -\mathrm{Im}\left(\frac{m^2-1}{m^2+2}\right)$, where Im is the imaginary part
R_m	product of the reflectivity of the two cavity mirrors
T_m	product of the transmittance of the two cavity mirrors
r	radial coordinate

2. Experiment

2.1. Multi-pass extinction

The methods have been described in our previous work [11], and only the key points are detailed here. The cavity extinction method is based on the fact that light passing through a medium is scattered and absorbed by particles, which results in an attenuation of the beam intensity, according to the Lambert-Beer law [17,18]:

$$\ln A = \ln\frac{I_t}{I_i} = -\int_{-\infty}^{+\infty} K_e(x)\,\mathrm{d}x = -P_0 \tag{1}$$

where K_e is the local extinction coefficient of the medium, determined by the local soot volume fraction and its optical properties, and P_0 represents the logarithmic loss of intensity across one pass. The total logarithmic loss of intensity for a cavity, P_t, can be related to the single pass extinction $A = I_t/I_i = \exp(-P_0)$ via:

$$P_t = \frac{T_m}{1 - A^2R_m}A \tag{2}$$

where R_m and T_m are the products of the reflectances and transmittances of the two cavity mirrors, respectively. The value P_0 can be obtained from the measurement of P_t and the calibrated mirror characteristics from Eqs. (1) and (2):

$$P_0 = -P_t - \ln\left(\frac{T_m}{2R_m}\right) - \ln\left[\sqrt{1 + \frac{4R_m\exp^2(-P_t)}{T_m^2}} - 1\right] \tag{3}$$

For a symmetric situation such as the current flame, the value of P_0 for each radial distance from the origin y can be deconvoluted to determine the local extinction coefficient $K_e(r)$ using the Abel Transform [19].

$$K_e(r) = -\frac{1}{\pi}\int_y^{\infty} \frac{P'_0(y)}{\sqrt{y^2 - r^2}}\,\mathrm{d}y \tag{4}$$

Assuming that the primary particles are sufficiently small relatively to the wavelength, the Rayleigh approximation for the emissivity is valid, so that the contribution of scattering to extinction [14,20] is negligible, and the local soot volume fraction f_v, can be obtained from

$$f_v = \frac{\lambda}{6\pi E(m)}K_e \tag{5}$$

where λ is the wavelength of the laser light and $E(m)$ is the soot absorption function, which is related to from the soot refractive index m via:

$$E(m) = \mathrm{Im}\left(\frac{m^2-1}{m^2+2}\right) \tag{6}$$

In the present study, we assume the value of m to be 1.57–0.56i, the value suggested by D'Alessio et al. for the visible range [21]. However, we note that the value of m is often considered wavelength dependent [22–24], and values used in studies around the present wavelength range suggest $E(m)$ values over a range of ±37% [11]. In addition, recent work has shown that the $E(m)$ also depends on the aging of particles [25–27], as their structure and composition changes with residence time through the high temperature field. Thus, $E(m)$ is expected to vary with HABs [26], especially in the central regions of diffusion flames in the present study. Given the continuing uncertainty in the optical properties, the values of for the extinction coefficient K_e are made available as supplemental material. These are independent of any assumptions about the value of $E(m)$, and can thus be used for comparison with any absorption/extinction model as they evolve in the future.

The discretisation of the Abel Transform generates uncertainty, which has been quantitatively analysed using the method in Ref. [11]. The estimated errors for the present case are around 10% for the cases where soot peaks away from the centre (typically short flames) and 18% for cases in which the soot peaks near the flame centre.

2.2. Laser cavity

The configuration of the laser cavity measurement system and measuring procedure for the total extent of extinction P_t can be