DÉTERMINATION IN SITU DE LA DISTRIBUTION DE TAILLE DES AGRÉGATS DE SUIE DANS UNE FLAMME DE DIFFUSION LAMINAIRE

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TITLE

IN SITU DETERMINATION OF THE SIZE DISTRIBUTION OF SOOT AGGREGATES IN A LAMINAR DIFFUSION FLAME

RESUME

Les particules de suie issues des processus de combustion nécessitent une caractérisation précise pour comprendre et contrôler leurs émissions. Cette étude présente une méthodologie permettant de déterminer de façon in situ et résolue spatialement les distributions de taille dans une flamme de diffusion laminaire d'éthylène en utilisant la diffusion angulaire et spectrale de la lumière. Les données résolues spatialement sont obtenues par inversion d'Abel avec correction de l'auto-absorption du signal. L'analyse montre que le modèle de distribution de taille auto-préservée capture une polydispersité plus élevée par rapport à la distribution log-normale. Le coefficient de diffusion avant est déterminé de manière robuste et sa dépendance spectrale est mise en évidence ouvrant de nouvelles perspectives d'application.

ABSTRACT

Soot particles from combustion processes require accurate characterization to understand and control their emissions. This study presents a methodology to determine spatially-resolved in-situ size distributions in a laminar ethylene diffusion flame using angular and spectral light scattering. Spatially resolved data is obtained through Abel inversion with correction for signal self-absorption. The analysis shows that the self-preserving size distribution model captures higher polydispersity compared to the log-normal distribution. The forward scattering coefficient is robustly determined and its spectral dependence is highlighted, opening new application perspectives.

MOTS-CLÉS: Suie, Distribution de taille, Flamme de diffusion, Diffusion angulaire de la lumière /**KEYWORDS**: Soot, Size distribution, Diffusion flame, Angular light scattering

1. INTRODUCTION

The emission of soot particles from fossil fuel combustion remains a significant environmental and health concern (Achakulwisut et al. 2023). Numerical simulations are used to study and design combustion systems, but these require accurate models for soot formation. To develop better control strategies, it is crucial to characterize particles in terms of their mass, concentration, and size using robust diagnostic tools (Santoro et al. 1983).

In-situ measurement techniques are preferred for their non-intrusive capabilities. While techniques like Small-angle X-ray Scattering (SAXS) and Laser-Induced Incandescence (LII) offer valuable insights (Di Stasio et al. 2006; Michelsen et al. 2022; J. B. A. Mitchell et al. 2013; Yon et al. 2018), multi-angle light scattering has emerged as a powerful method for determining aggregate size with fine spatial resolution. Various research groups have advanced the application of angular light scattering for soot characterization. Notable developments include the work of Oltmann et al. (2010, 2012) on turbulent jet-diffusion flames and Caumont-Prim et al. (2013) approach for full determination of soot size distribution. Recent advancements by Bouvier et al. (2019, 2023) and Yon et al. (2021a,b) have extended the technique to provide spatially-resolved data through Abel inversion.

This study combines measurements at different angles and wavelengths while correcting for signal trapping effects (Littin et al. 2024a,b). The focus is on determining soot aggregate size distribution using log-normal and self-preserving size distribution models. The research aims to demonstrate correlations between size and polydispersity of soot aggregate populations and provide two-dimensional maps of these parameters.

2. Methodology

Angular light scattering and line-of-sight attenuation measurements were performed in a previous work (Littin et al. 2024a,b; Yon et al. 2021a,b) using a co-flow laminar diffusion flame of ethylene. Measurements were conducted at multiple wavelengths and scattering angles using a Gülder-type burner operated at atmospheric pressure.

2.1. Size distribution determination

The forward scattering coefficient for polydisperse aggregates, under the Rayleigh-Debye-Gans for Fractal Aggregates (RDGFA) theory, is expressed as:

$$\kappa_{vv}(\lambda,\theta) = \underbrace{\frac{9\pi}{\lambda^4}F(m)f_s\frac{\overline{V_{agg}^2}}{\overline{V_{agg}}}}_{\kappa_{vv}(0°,\lambda)}\underbrace{\frac{\overline{R_g^{2D_f}f(qR_g,D_f)}}{\overline{R_g^{2D_f}}}}_{\text{Size dependent}} \quad (1)$$

where f_s is the soot volume fraction, $F(m,\lambda)$ is the scattering function, D_f is the fractal dimension, f is the structure factor, and q is the scattering wave vector. Two probability density functions (PDFs) are considered: log-normal (Equation 2) and self-preserving (Equation 3).

$$\frac{dp(D_g)}{d\ln D_g} = \frac{1}{\sqrt{2\pi}\ln\sigma_g}\exp\left[-\frac{1}{2}\left(\frac{\ln(D_g/D_{g,geo})}{\ln\sigma_g}\right)^2\right] \quad (2) \qquad \frac{dp(D_g)}{d\ln D_g} = \frac{D_f}{a^d\Gamma(d/D_f)}\left(\frac{D_g}{\widetilde{D_g}}\right)^d\exp\left[-\left(\frac{D_g}{a\widetilde{D_g}}\right)^{D_f}\right] \quad (3)$$

where $a = (1-\lambda_{SP})^{-1/D_f}$, $d = D_f(1-\lambda_{SP})$. λ_{SP}, $\widetilde{D_g}$, $D_{g,geo}$ and $\sigma_{g,geo}$ are the governing parameters of the self-preserving and log-normal distributions. A conversion between the self-preserving and log-normal distributions is proposed (Equation 4) to facilitate comparison:

$$D_{g,geo} = \widetilde{D}_g\exp\left[\frac{1}{D_f}\psi^0(1-\lambda_{SP})\right] \qquad \sigma_{g,geo} = \exp\left[\frac{1}{D_f}\sqrt{\psi^1(1-\lambda_{SP})}\right] \quad (4)$$

The aggregate size distribution is determined using the ratio of scattering coefficients at a fixed reference angle (Equation 5). The size distribution parameters are obtained by fitting an equivalent gyration diameter (D_g^*) using the cost function in Equation 6, which employs root median squared error (RMedianSE) to reduce outlier influence.

$$\frac{\kappa_{vv}(\lambda,\theta)}{\kappa_{vv}(\lambda,\theta_{ref})} = \frac{\int_0^\infty D_g^{2D_f}f(qR_g,D_f)p(D_g)\,dD_g}{\int_0^\infty D_g^{2D_f}f(q_{ref}R_g,D_f)p(D_g)\,dD_g} \quad (5) \qquad \xi = \frac{1}{\overline{D_g}}\text{RMedianSE}\left(D^*_{g,\text{meas}}, D^*_{g,\text{ret}}\right) \quad (6)$$

Results for different radial positions are illustrated in Figure 2, showing consistent trends of increasing aggregate size and polydispersity for both distribution models. The methodology is subsequently extended to the entire flame map.

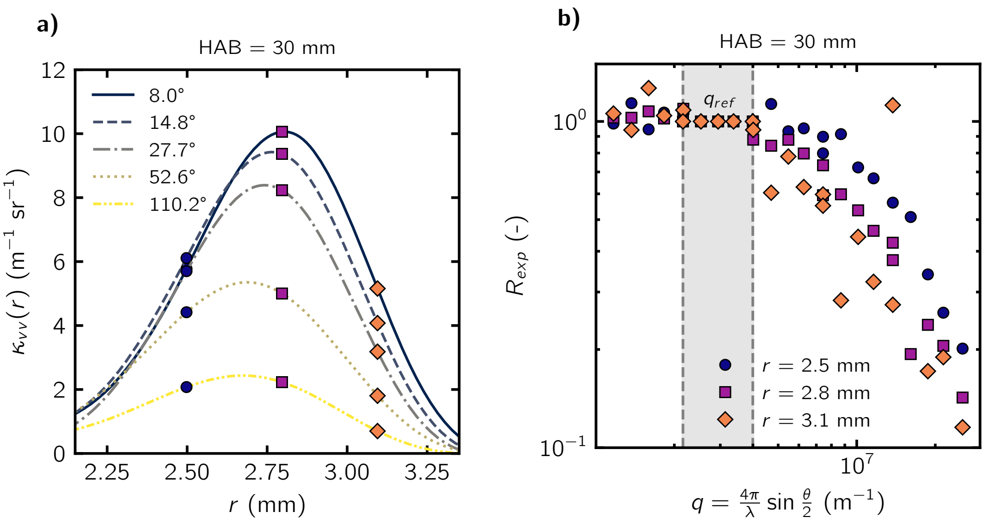

Figure 1. (a) Measured κ_{vv} values at 550 nm for various angles and radial positions. (b) Measured ratios of κ_{vv} for wavelengths between 405 nm and 750 nm across all angles.

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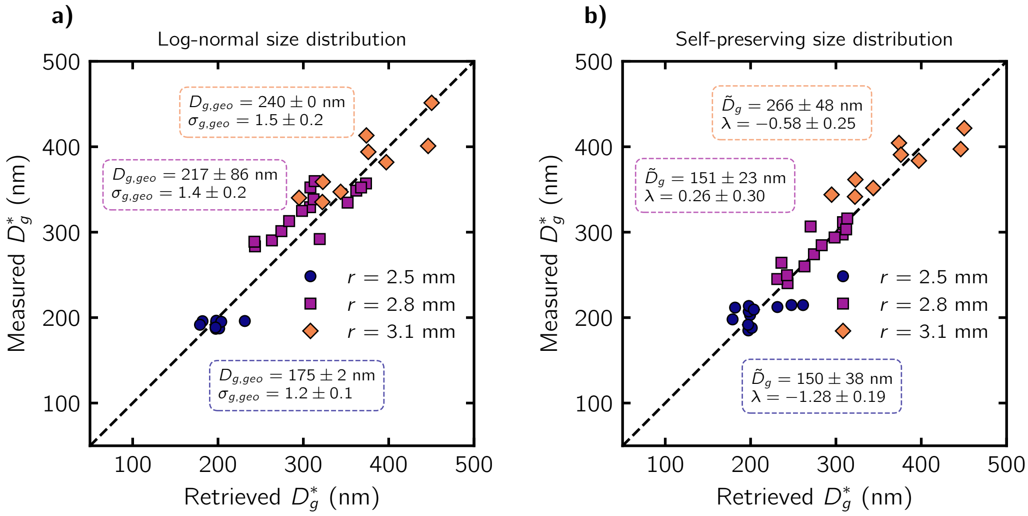

Figure 2. Comparison of optically measured and retrieved gyration diameters using log-normal (a) and self-preserving (b) size distribution models.

3. Results and perspectives

Two-dimensional probability density functions (PDFs) of the self-preserving and log-normal parameters across the entire flame are shown in Figure 3. The PDFs use a logarithmic color scale, with symbols indicating the most probable values. Higher polydispersity is captured by the self-preserving PDF, which exhibits asymptotic behavior at large gyration diameters, compared to the log-normal distribution.

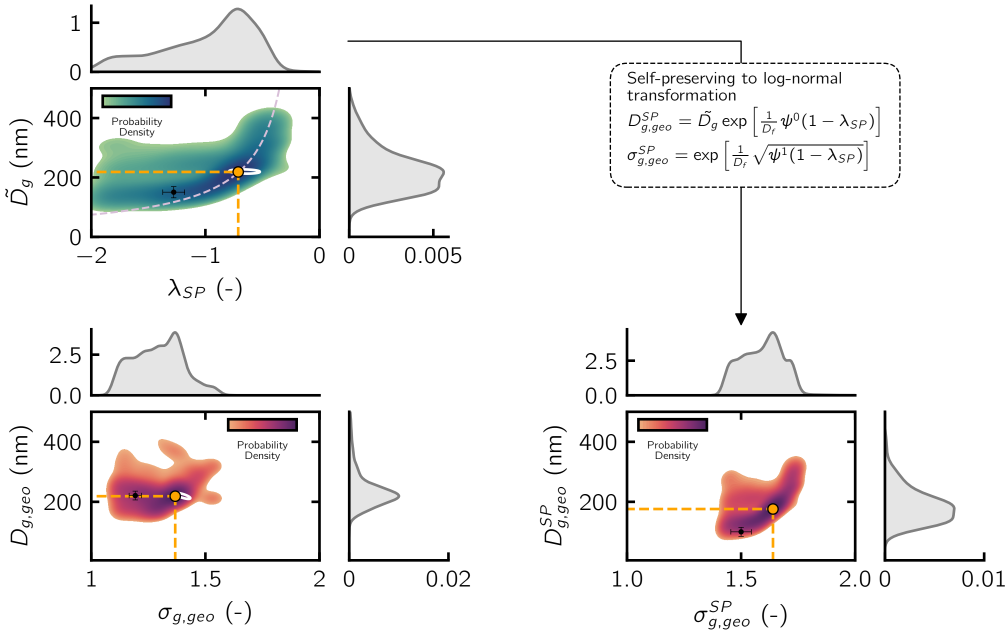

Figure 3. Two-dimensional PDFs of self-preserving (a) and log-normal (b) parameters, with geometric conversion (c). Log-scale probability density shown with most probable values (orange circles) and uncertainties (Monte Carlo in white, covariance in error bars).

An empirical relationship between size and polydispersity parameters of SP is established using a 4-parameter function:

$$\lambda_{SP}(\widetilde{D_g}) = D + \frac{(A-D)}{(1+(\widetilde{D_g}/C)^B)}. \tag{7}$$

This shows polydispersity (λ_{SP}) increases with modal diameter ($\widetilde{D_g}$). The relationship is used to determine parameters which are converted to log-normal form, as shown in Figure 4 a) and b).

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As a perspective for future work, the determination of the forward scattering coefficient ($\kappa_{vv}(0°)$) is extended across multiple wavelengths. Figure 4 illustrates $\kappa_{vv}(0°)$ at 550 nm and the spectral variation of $\kappa_{vv}(0°)\lambda^4$ for two distinct regions of the flame. A linear fitting approach is employed to quantify this spectral variation, providing insights into the behavior of the scattering function across different wavelengths.

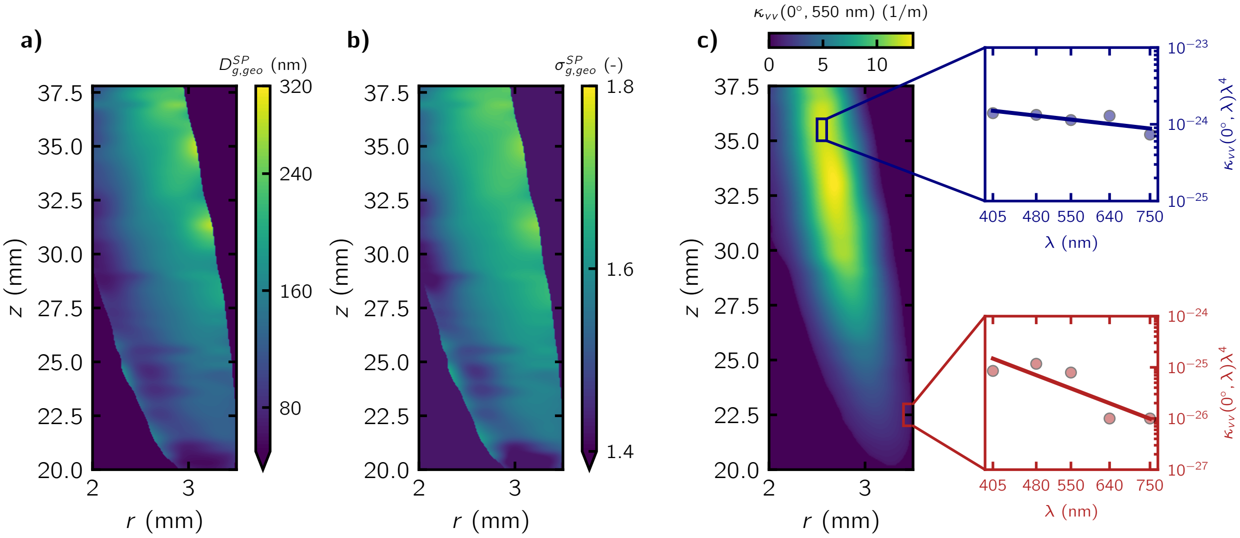

Figure 4. a) Geometric gyration diameter ($D^{SP}_{g,geo}$) and standard deviation ($\sigma^{SP}_{g,geo}$) for the self-preserving size distribution. c) Forward scattering coefficient $\kappa_{vv}(0°)$ at 550 nm and its spectral variation $\kappa_{vv}(0°)\lambda^4$ for two flame regions, with linear fit analysis.

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