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► **To cite this version:**

Jérôme Yon, José Morán, Florian Lespinasse, Felipe Escudero, Gilles Godard, et al.. Horizontal Planar Angular Light Scattering (HPALS) characterization of soot produced in a laminar axisymmetric coflow ethylene diffusion flame. *Combustion and Flame*, 2021, 232, pp.111539. 10.1016/j.combustflame.2021.111539 . hal-03265019

HAL Id: hal-03265019

<https://normandie-univ.hal.science/hal-03265019>

Submitted on 2 Aug 2023

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Horizontal Planar Angular Light Scattering (HPALS) characterization of soot produced in a Laminar Axisymmetric Coflow Ethylene Diffusion Flame

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Abstract

In-flame optical characterization of soot is of vital importance to understand soot formation mechanisms as well as to develop and validate accurate soot models. The present work introduces an unconventional methodology adapted to laminar axisymmetric flames that avoids the issue of variable measurement volume with varying scattering angle in the existing light scattering techniques and thus enables the determination of aggregate size with a higher spatial resolution. Coupled with multi-wavelength line-of-sight attenuation measurements, the proposed Horizontal Planar Angular Light Scattering at 532 nm was found able to provide radial profiles of aggregate size, number and diameter of primary spheres, soot volume fraction, and number density in a laminar axisymmetric coflow ethylene/air diffusion flame established over a Gülder burner. The spatial variation of soot optical properties associated with soot maturity was considered in data analysis.

Keywords: Planar angular light scattering, Axisymmetric flames, soot maturity, Line-of-sight attenuation, RDGFA

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1. Introduction

Soot formation and oxidation mechanisms are still not completely understood and very challenging to model. Yet, it is becoming imperative to design energy systems that are cleaner and have less impacts on human health and on the environment. To this end, numerical simulation plays a vital role to speed up the design and optimization processes. To gain improved fundamental understanding of soot formation mechanisms and to validate soot models, it is essential to obtain complete and robust databases of soot properties in well-controlled and highly stable laminar target flames. Such efforts have been continuously made in the last four decades through developing sophisticated *in-situ* optical techniques. Among these techniques, Laser Induced Incandescence (LII) has received widespread attention [1].

LII enables the determination of soot volume fraction in complex, transient environments [2] or in very low sooting flames [3]. The LII signal temporal decay rate can be interpreted in terms of soot primary sphere diameters [4, 5].

By conducting LII measurements using two different laser wavelengths, it is possible to determine the ratio of soot absorption functions at the two laser wavelengths, which in turn can be linked to the soot maturity [6–8]. The line-of-sight attenuation (LOSA) and spectral soot emission (SSE) methods are also often used in laminar axisymmetric flames to determine the soot volume fraction and flame temperature [9]. A recent study has shown that LOSA measurements at different wavelengths in the visible and near infrared can provide instructive information on the soot maturation process with a high spatial resolution [10]. Measurement of the volume fraction of incipient soot is extremely challenging due to the very low volume fraction associated with the very small incipient soot particles and their weak absorption in the visible. Although the soot aggregate size is important to validate the aggregation and oxidation mechanisms modeled by Population Balance Equation [11, 12] or discrete element methods [13, 14], the above optical techniques are not able to provide information of soot aggregate size, morphology, and aggregate number density, which are also important quantities to fully characterize soot. To complement the measurements of LII, LOSA, or SSE, the soot aggregate size information has often been obtained through an *ex-situ* method through soot sampling in the flame [15, 16]. However, sampling of nanoparticles is intrusive in nature and can alter the structure of the flame [17, 18]. Moreover, the particle aggregation process can occur

38 in the sampling line as well as smaller particles can be lost by deposition [3].
39 Finally, the aerosol sizing instruments provide the particle size distributions
40 in either mobility or aerodynamic diameter that have to be interpreted to
41 obtain aggregate size, such as gyration radius, since soot particles are not
42 spherical [12]. These considerations suggest that it is necessary to develop
43 *in-situ* techniques to measure soot aggregate size. It has been demonstrated
44 some 40 years ago by Santoro et al. [19] that scattering based techniques
45 are able to provide instructive information on soot size. Köylü and Faeth
46 [20] have shown that the interpretation of scattering measurements of soot
47 particles should be performed by a dedicated theory of light interaction with
48 fractal aggregates called RDGFA [21], but not by Mie theory.

49 The reader is invited to refer to Sorensen [22] for more details about that
50 the RDGFA theory. This approach has been extensively used to interpret
51 light scattering measurements of sooting flames [23–25]. Wang and Sorensen
52 [26] have shown the reliability of the RDGFA theory for TiO₂ and SiO₂ ag-
53 gregates and later Chakrabarty et al. [27] for carbonaceous chain aggregates.
54 It has also been shown in some studies that there is good agreement between
55 the size of both aggregates and primary spheres obtained by light scatter-
56 ing compared to Transmission Electron Microscopy image analysis [28–30].
57 However, light scattering techniques have been less favored in recent years
58 compared to LII. Indeed, the current angular light scattering method remains
59 challenging in the sense that the change of scattering angle is accompanied
60 by a change in the measurement volume and thus suffers the problem of a
61 significantly reduced spatial resolution for small and large scattering angles,
62 leading to large uncertainties in the inferred soot aggregate sizes in diffusion
63 flames where physical and chemical quantities display high spatial gradients.

64 Nevertheless, the light scattering techniques are still being improved. An
65 integrated system enabling the measurement of soot size distribution with
66 high temporal resolution has been developed by Oltmann et al. [31]. Bouvier
67 et al. [32] and Zhang et al. [33] have shown the potential of exploiting the
68 spectral dependency of the scattered light. The vertical planar configura-
69 tion in an ethylene diffusion flame [34, 35] and in premixed 1D flat flame
70 (McKenna-type burner) [36] has been developed in order to provide a planar
71 description of the soot size. Finally, the light scattering based techniques
72 have been applied in more and more challenging configurations [37, 38] and
73 some efforts have been made to improve the light scattering theories [39, 40]
74 and the inversion algorithms to accurately recover aggregate size distribution
75 [27, 41, 42]. These studies illustrate the potential of light scattering tech-

76 niques and coupled techniques based on light scattering and other optical
77 diagnostics [43, 44].

78 Up to now, 2D Multi Angle Light Scattering is the technique providing
79 the aggregate size with the finest spatial resolution [36]. However, the data
80 processing is not simple. Indeed, as explained by Ma and Long [34], the
81 observed image of the laser light sheet interaction with the flame "became
82 narrower in shape and the signal on each pixel was increased due to an
83 increased sample volume". In other terms, the image resolution and the
84 volume of measurement change with the scattering angle when rotating the
85 camera around the burner. Then, an intensity calibration and a correction
86 of distortion of the images are required [34, 36]. These corrections may have
87 an impact on the spatial resolution of such planar light scattering technique.

88 The present work intends to overcome the issue of scattering angle depen-
89 dent measurement volume and to improve the spatial resolution of laser-based
90 light scattering technique. This is achieved by the use of an unconventional
91 horizontal laser sheet configuration and the application of this technique is
92 demonstrated in a laminar axisymmetric diffusion flame. This technique
93 enables the determination of soot aggregate size distribution with a high
94 spatial resolution along the radial direction. The coupling of this technique
95 with multi-wavelength LOSA enables the determination of additional infor-
96 mation, such as volume fraction, number density, and primary particle size.
97 The combined technique also permits to analyze the data by taking into
98 account the maturity dependence of soot optical properties.

99 **2. Experimental setup, theoretical method, and data analysis**

100 *2.1. Experimental setup*

101 Figure 1 shows the experimental setup used in this study. The Gülder
102 laminar coflow diffusion flame burner can be accurately moved vertically by
103 means of an Electromechanical cylinder EMC Rexroth. This linear stage
104 facilitates the scanning of scattering measurements at different height above
105 the burner (HAB) with a precision $< 10 \mu\text{m}$. The laminar coflow diffusion
106 flame is generated by delivering pure ethylene through the central fuel tube
107 of the burner at a flowrate of $0.194 \text{ l}_n/\text{min}$. The inner and outer diameters of
108 the central fuel tube are 10.9 mm and 12.7 mm, respectively. The oxidizer is
109 ambient air introduced through the co-annular region of the burner between
110 the central fuel tube and the 90 mm inner diameter of the oxidizer port at

111 150 l_n /min. These conditions result in a stable laminar diffusion flame with
112 a visible flame height of 75 mm.

113 Unlike conventional light scattering techniques that utilize a laser beam
114 or a laser sheet orthogonal to the camera axis, the present setup employs
115 a horizontal laser sheet that is wider than the largest flame diameter. This
116 original arrangement can overcome the issue of scattering angle dependent
117 measurement volume at the expense of conducting Abel inversion as shown
118 below. To apply the horizontal planar angular light scattering (HPALS)
119 technique proposed in this study, a Quantel Brilliant Easy laser pumped at
120 1064 nm and frequency-doubled at 532 nm is employed. The laser is set for a
121 maximum energy peak per pulse of 185 mJ at 10 Hz with a pulse duration of
122 4 ns (FWHM). The laser output energy is modulated by an internal polarized
123 based attenuator. It is first spatially filtered by using a pin-hole (2.44 mm
124 diameter) in order to preserve the uniform region of the laser beam. Two
125 532 nm dichroic mirrors M1 (CVI Y2-1025-45-A) and M2 (CVI Y2-2037-
126 45) are employed to modify the direction of the laser beam and laser sheet,
127 respectively. A variable dielectric attenuator VA1 (Laseroptik L-01543) is
128 used to redirect a part of the laser beam toward a power meter (Coherent
129 PowerMax Model PM10) in order to measure the energy of laser beam during
130 each measurement.

131 The horizontal laser sheet is formed by the combination of three cylindri-
132 cal lenses, a plano-concave L1 ($f=-40$ mm, CVI SCC-20.0-20.3-C), a plano-
133 convex L2 ($f=300$ mm, Thorlabs LJ1996L1), and a second plano-convex L3
134 ($f=500$ mm, Thorlabs LJ1144L2). This horizontal laser sheet is then pro-
135 jected across the flame with a width of 15 mm (larger than the maximum
136 flame diameter ~ 10 mm). A reasonably uniform thickness is obtained with
137 $141 \mu m$ (corresponding to 2σ) measured with a Precision Beam Profiler (Pho-
138 ton Inc. Model 2323 USB 2.0 Camera). Note that the polarization state is
139 orthogonal to the laser sheet, i.e., in the vertical direction in Fig.1.

140 The detection system is mounted on a motorized rotating arm (Danaher
141 motion N33HRLG-LEK-M2-00) to enable the signal collection at different
142 scattering angles designated by θ (see Fig. 1). The optical collection is com-
143 posed of a vertical polarizer P1 (Linear Pola 49 mm, second white arrow
144 in Fig.1) and a long distance microscope system (Infinity CF2+basic unit)
145 mounted with an 1.5 cm extension ring on an ICCD camera (PI-MAX-4,
146 model PM4). The internal laser PTU is employed to trigger the camera to
147 capture the scattering signal for each laser pulse. In order to limit the col-
148 lection of the flame natural emission, the ICCD exposure time is reduced to

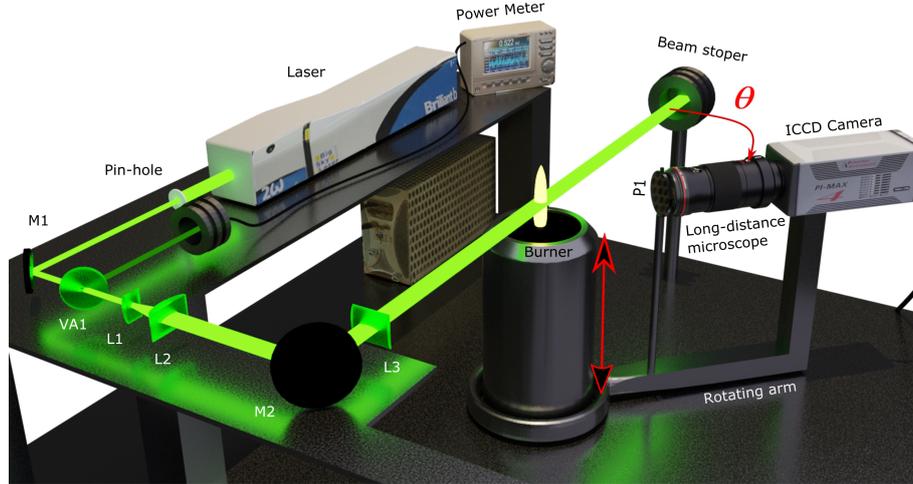


Figure 1: Experimental setup: The horizontal laser sheet is arranged to cross the flame. The Height Above the Burner can be modified (red arrow) as well as the scattering angle (θ) by rotating the camera around the flame axis. M1 and M2 are two dichroic mirrors, L1, L2 and L3 are respectively a cylindrical, plano-concave, and plano convex lenses. VA1 is a variable dielectric attenuator and P1 is a polarizer.

150 2.2. Raw data treatment

151 Up to 2000 images of 1024×1024 pix² are accumulated or averaged
 152 per condition depending on the signal intensity. The image resolution is
 153 ~ 51 pix/mm, and the field of an image can cover the width of the flame as
 154 depicted in Fig. 2-A. Also, in this figure the region of interest (ROI) is high-
 155 lighted by the white rectangle of dashed-line. This ROI corresponds to the
 156 image of scattering signal of the horizontal laser sheet captured by the ICCD
 157 at $HAB = 30$ mm and a scattering angle of 14.9° . The same ROI is also
 158 employed with the laser off in order to obtain the background. The profiles
 159 after subtracting the aforementioned background are divided by the corre-
 160 sponding laser energy, the number of accumulations, and the camera gain.
 161 The laser sheet uniformity was checked and it was found quite good. The
 162 lateral bumps observed in Fig. 2-A are likely caused by multiple scattering
 163 in the dense regions (high soot concentration) of the flame. This phenomenon,
 164 which is not considered in the present analysis, will be investigated in our fu-
 165 ture research. Nevertheless, its contribution is considered very small (please

166 note that the scattering intensity is displayed in log scale). Fig. 2-B presents
 167 two normalized profiles of two different scattering angles at a given HAB.
 168 To avoid a noisy deconvolution, the Python “Savgov filter” (polynomial of
 169 order 2 and 31 elements in the averaging window and mirror boundary) is
 170 used. The PyAbel’s “Hansenlaw” method [45] is used to deconvolute the
 171 experimental profiles (Abel inversion, see the dash curves in Fig. 2-B). The
 172 obtained radial profile is re-convoluted and compared to the acquired sig-
 173 nal to check the validity of the deconvolution process (the continuous curves
 174 in Fig. 2-B). The re-convoluted scattering signal profiles are in very good
 175 agreement with the measurements.

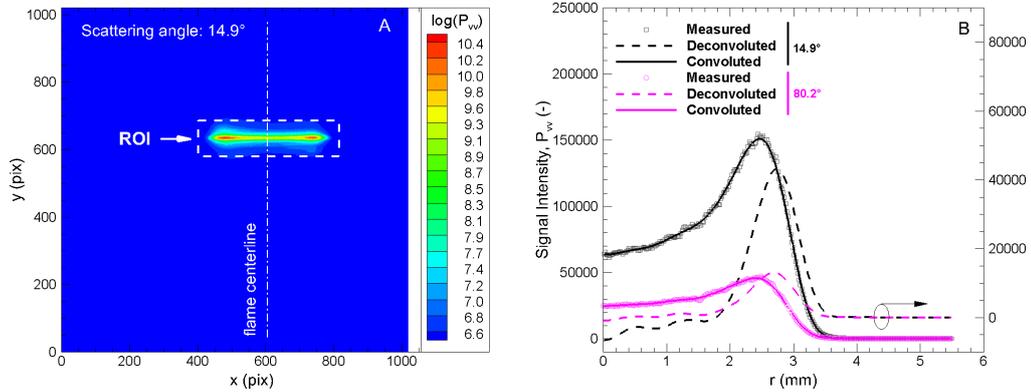


Figure 2: A typical light scattering image (left plot) and profiles captured at HAB = 30 mm and at two angles, 14.9° (or $q = 0.0030 \text{ nm}^{-1}$) and 80.2° (or $q = 0.0152 \text{ nm}^{-1}$) (right plot): (A) Raw scattering signal field and ROI employed (at a scattering angle of 14.9°) and (B) Measured scattering signal profile (symbols), deconvoluted profile (Abel inversion in dash curves), and re-convoluted profiles using the deconvoluted signal (continuous).

176 3. Light scattering theoretical background

177 3.1. Determination of the absolute scattering coefficient

178 As illustrated in Fig. 3, the radiative power of scattered light reaching the
 179 detector by molecules or particles from an elementary volume dV located at
 180 position M and exposed to a planar incident light source with power density
 181 I_0 (in W/m^2) can be expressed as,

$$dP_{vv} = I_0 \frac{\overline{dK_{vv}}}{d\Omega} \Omega dV, \quad (1)$$

182 where $\overline{\frac{dK_{vv}}{d\Omega}}$ is the population averaged differential scattering coefficient for
 183 vertical-vertical polarization and Ω the solid angle related to the detection
 184 aperture. The elementary volume can be expressed as $dV = dx dy dz$, where x
 185 and y are respectively the positions along the detector and orthogonal to the
 186 detector (see Fig. 3) and z is the position along the flame height. The term
 187 $dS = dx dz$ corresponds to the elementary surface of the pixel of the ICCD
 188 camera which collects the scattered light originated from dV at the scattering
 189 angle θ . This elementary volume can also be expressed as $dV = \frac{r dr dS}{\sqrt{r^2 - x^2}}$ with
 190 r being the distance of M from the flame center line.

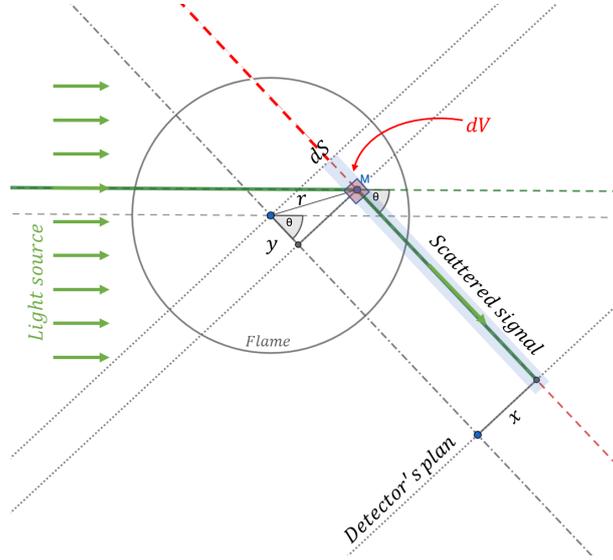


Figure 3: Schematic of the HPALS principle (top view).

191 By assuming negligible laser energy attenuation and self absorption of
 192 the scattered signal, the collected signal at position x on the detector can be
 193 expressed as an Abel transform of the averaged scattering coefficient, namely
 194 the integrated scattering signal along the cord corresponding to position x :

$$P_{vv} = I_0 \Omega dS \times 2 \underbrace{\int_x^\infty \frac{\overline{dK_{vv}}}{d\Omega} \frac{r}{\sqrt{r^2 - x^2}} dr}_{\text{Abel transform of } \frac{\overline{dK_{vv}}}{d\Omega}}. \quad (2)$$

195 It should be pointed out that laser energy attenuation and signal trapping at
 196 532 nm in the ethylene flame studied in this work may not be negligible at

197 flame heights where soot volume fractions are fairly high. Nevertheless, such
 198 effects are neglected in the first attempt to demonstrate the capabilities of
 199 the HPALS technique and will be evaluated and taken into account in future
 200 studies.

201 Thus, for each scattering angle imposed by the angular position of the
 202 camera, the radially-resolved scattering coefficient can be retrieved by a sim-
 203 ple inverse Abel transform (Ab^{-1}) of the convoluted scattering signal profile
 204 acquired over the entire range of x ,

$$\frac{d\overline{K_{vv}}}{d\Omega} = Ab^{-1} \left(\frac{P_{vv}}{I_0 \Omega dS} \right). \quad (3)$$

205 In order to determine the absolute value of the averaged scattering co-
 206 efficient, a calibration of the detection system is necessary. In this study
 207 the calibration is performed by measuring the scattered signal produced by
 208 a free jet of ethylene P_{vv, C_2H_4} . At ambient conditions, the molecule density
 209 and the corresponding scattering cross section [22] are known. The resultant
 210 scattering coefficient is found to be $K_{vv, C_2H_4} = 8.92 \times 10^{-6} m^{-1}$.

211 It is noticed that the collected scattering signals from the flame soot as
 212 well as from the ethylene jet are systematically normalized by the corre-
 213 sponding laser intensity, number of accumulated images and camera gain,
 214 the absolute scattering coefficient of soot is then given by:

$$\frac{d\overline{K_{vv, soot}}}{d\Omega} = K_{vv, C_2H_4} \times \frac{Ab^{-1}(P_{vv, soot})}{Ab^{-1}(P_{vv, C_2H_4})}. \quad (4)$$

215 The measured profile (P_{vv, C_2H_4}) and its deconvolution $Ab^{-1}(P_{vv, C_2H_4})$ are
 216 given in [Appendix A](#).

217 3.2. Interpretation of scattering coefficient based on the RDGFA theory

218 For an ensemble of monodisperse aggregates and under conditions where
 219 the RDGFA theory is considered valid [22], the aggregate differential scat-
 220 tering coefficient can be expressed as,

$$\frac{dK_{vv, soot}}{d\Omega} = N_{agg} \frac{9\pi^2}{\lambda^4} F(m) V_{agg}^2 f(qR_g, D_f). \quad (5)$$

221 where N_{agg} and V_{agg} are respectively the aggregate number density and the
 222 individual aggregate volume, m is the soot complex refractive index, $F(m) =$

223 $\left|\frac{m^2-1}{m^2+2}\right|$ is the scattering function of m , λ is the incident laser wavelength, f
 224 is the structure factor, R_g is the gyration radius (the radius of an equivalent
 225 sphere having the same inertia moment), and $q = \frac{4\pi}{\lambda} \sin\left(\frac{\theta}{2}\right)$ is the magnitude
 226 of scattering wave vector. Symbol D_f is the aggregate fractal dimension.
 227 The scaling law correlating the aggregate volume to its gyration radius is
 228 expressed as

$$N_p = \frac{V_{agg}}{V_p} = k_f \left(\frac{D_g}{D_p}\right)^{D_f}, \quad (6)$$

229 where N_p is the number of primary spheres, V_p is the volume of primary
 230 sphere, $D_g = 2R_g$ is the aggregate gyration diameter, D_p is the diameter
 231 of the primary spheres, k_f is the fractal prefactor (assumed equal to 2.2 in
 232 the present study, consistent with TEM measurements, e.g. [46]). We will
 233 also assume a typical value of $D_f = 1.77$ for flame-generated soot, which is
 234 also the value of aggregates generated by diffusion-limited cluster aggregation
 235 (DLCA).

236 The structure factor formulated by Dobbins and Megaridis [47] is used in
 237 the present study because it has been shown to be accurate for the determi-
 238 nation of the aggregate gyration radius based on light scattering at 532 nm
 239 even when considering the aggregate internal multiple scattering [40].

240 For polydisperse aggregates, by introducing the soot volume fraction as
 241 $f_v = N_{agg} \times \overline{V_{agg}}$ and by using the fractal scaling law (Eq. 6), the population
 242 averaged scattering coefficient can be expressed as,

$$\frac{\overline{dK_{vv,soot}}}{d\Omega} = \underbrace{\frac{9\pi^2}{\lambda^4} F(m) f_v \overline{V_{agg}^2}}_{\text{Amplitude}} \underbrace{\frac{\overline{R_g^{2D_f} f(qR_g, D_f)}}{\overline{R_g^{2D_f}}}}_{\text{Size dependent}}. \quad (7)$$

243 In this expression, we can distinguish the amplitude of the signal, which
 244 corresponds to the magnitude of the forward scattering signal ($\theta = 0$), and
 245 the ratio $\frac{\overline{R_g^{2D_f} f(qR_g, D_f)}}{\overline{R_g^{2D_f}}}$ that contains the angular dependence through the q
 246 parameter. In this study, the angular dependence term is interpreted in terms
 247 of aggregates size distribution. Moreover, the scattering amplitude is coupled
 248 with LOSA measurements (see [10]) and interpreted in order to determine
 249 the aggregates volume, density and primary sphere diameter.

250 *3.3. Soot aggregate size distributions*

251 Interpretation of Eq. 7 requires some *a priori* knowledge of the size distri-
 252 bution density probability function. In the present study, three assumed
 253 distributions are considered, namely Monodisperse, Lognormal and Self-
 254 Preserving.

255 Due to the large particle number density and fairly high temperatures
 256 in flames, it is highly unlikely that the aggregate sizes are monodisperse.
 257 Nevertheless, this case is considered in order to test the sensitivity of the
 258 methodology proposed to the assumed shape of aggregate size distribution
 259 and to the magnitude of polydispersity. The Lognormal size distribution of
 260 soot aggregates is considered because it is usually observed when expressed in
 261 mobility diameter. Nevertheless, there is no theoretical basis to this empirical
 262 observation in particular when the aggregate size is expressed by gyration
 263 diameter. On the contrary, aggregation mechanisms for fractal aggregates
 264 suggest a self-preserving size distribution [48]. Sorensen [22] pointed out that
 265 this distribution can shorten the tail of the distribution for larger aggregates
 266 compared to the Lognormal distribution, which is suspected to overestimate
 267 the moments used for the evaluation of $\overline{V_{agg}}$, $\overline{V_{agg}^2}$, $\overline{R_g^{2D_f} f}$ and $\overline{R_g^{2D_f}}$ in Eq. 7.

268 The lognormal distribution is governed by the geometric diameter D_{geo}
 269 and the geometric standard deviation σ_{geo} . The former is not so different from
 270 the modal diameter of the distribution (most probable size). Caumont-Prim
 271 et al. [30] reported $\sigma_{geo} = 1.7$ based on angular light scattering measurement
 272 in an ethylene flame for mature soot. The same author found 1.6 when
 273 applying the technique on soot at the exhaust of a miniCAST generator.
 274 The latter value (1.6) will be considered here since the exhaust of miniCAST
 275 provides a uniform soot source avoiding issues related to the spatial resolution
 276 of the conventional light scattering technique. This is also in agreement with
 277 the values reported by Bouvier et al. [32]. However, it is noticed that some
 278 authors considered larger values of the geometric standard deviation [29, 48].

279 The self-preserving distribution is governed by a \widetilde{D}_g and by the self-
 280 preserving homogeneity coefficient λ_{SP} . The latter parameter controls the
 281 shape of the distribution. It has been reported to vary over a range of phys-
 282 ical values between -0.5 and 0.5 depending on the aggregation mechanisms,
 283 which are principally driven by the primary sphere diameter (positive when
 284 ballistic and negative when diffusive). In the present study, we hypothesize
 285 that this parameter takes the value of null ($\lambda_{SP} = 0$) as a result of aggregation
 286 occurring in the transition regime with a typical primary particle diameter

287 about 20 nm (see [48]). The size driving parameter \widetilde{D}_g is related to a moment
 288 of the size distribution at power D_f , indeed $\widetilde{D}_g = \left(\overline{D_g^{D_f}}\right)^{1/D_f}$ [48]. In con-
 289 sequence, this parameter becomes larger as the aggregates are increasingly
 290 more polydisperse.

291 A theoretical analysis of the self-preserving distribution, which has been
 292 shown to be a generalized gamma function [48], has shown that $D_{g,modal} =$
 293 $\widetilde{D}_g \left(1 - \frac{1}{D_f(1-\lambda_{SP})}\right)^{1/D_f}$ with $D_{g,modal}$ being the mode of the distribution. In
 294 the present case where D_f and λ_{SP} are fixed respectively at 1.77 and 0, one
 295 can find $\widetilde{D}_g = 1.57 \times D_{g,modal}$. Thus, it is important to keep in mind that
 296 \widetilde{D}_g is consistently larger than the most probable gyration diameter (mode of
 297 the distribution).

298 The analytical expressions for $\overline{R_g^{2D_f} f}$ and $\overline{R_g^{2D_f}}$ under the hypothesis
 299 of Monodisperse, Lognormal and Self-Preserving distributions are presented
 300 in [Appendix B](#).

301 4. Results and Discussion

302 4.1. Results at $HAB = 30$ mm

303 Figure 4 presents in black dots the radially-resolved scattering signals ex-
 304 pressed in K_{vv} measured at 22.4°, 33.9°, 51.1°, 80.2°, 90°, 119.8° and 148.1°,
 305 for $HAB = 30$ mm through Abel inversion. The scattering wave vector q is
 306 preferred over the direct use of scattering angle for the representation of an-
 307 gular dependence of scattering because this parameter is used in the RDGFA
 308 theory. The scattered intensity decreases with increasing q , in particular for
 309 large aggregates. Thus, at a given radial position r , the aggregate size dis-
 310 tribution can be obtained by fitting the curve in the q direction (e.g. the red
 311 arrow in the graph). It should be pointed out that the aggregate size distri-
 312 bution determined does not depend on the amplitude of K_{vv} (green arrow in
 313 graph). This point will be explained after.

314 The results of the aggregate size distribution parameters are reported in
 315 Fig. 5. In this figure, the points of red-circle are the gyration diameters of
 316 Monodisperse size distribution (D_g), while the points of green-star and blue-
 317 square are respectively the \widetilde{D}_g and D_{geo} for the self-preserving and lognormal
 318 distributions.

319 It is interesting to see that \widetilde{D}_g at power D_f corresponds to the popu-
 320 lation average gyration diameter at power D_f ($\widetilde{D}_g^{D_f} = \overline{D_g^{D_f}}$). Due to the

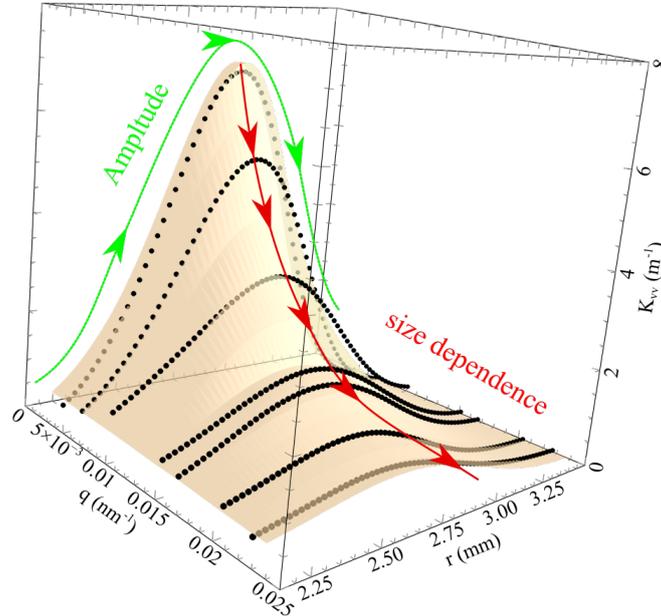


Figure 4: Radial and angular distribution of the scattered signal at HAB = 30 mm after Abel inversion and expressed in terms of K_{vv} . The angular dependence of scattering is expressed through $q = \frac{4\pi}{\lambda} \sin\left(\frac{\theta}{2}\right)$, which is the magnitude of the scattering wave vector. The measured scattering signal is presented in black dots after the fitting process generating a continuous surface.

321 polydispersity, those parameters are lower than the equivalent gyration di-
 322 ameter under the assumption of monodisperse aggregates. Nevertheless, the
 323 assumption of monodisperse soot aggregates is clearly unrealistic in flames.

324 Therefore, we consider the lognormal or self-preserving hypothesis more
 325 representative of soot aggregates. \widetilde{D}_g is found to be somewhat larger than
 326 D_{geo} even though the self-preserving size distribution is lower than the log-
 327 normal one for small aggregates (see the inset of Fig. 5). This behavior is
 328 attributed to the definition of \widetilde{D}_g which represents a moment of power D_f ,
 329 whereas D_{geo} is the median diameter.

330 It is interesting to notice that larger aggregates are found at a radial
 331 position between $r = 2.9$ and 3.0 mm whereas the signal amplitude starts to
 332 decrease at this radial distance at HAB = 30 mm (see Fig. 4).

333 The band that bounds each curve by dotted lines in Fig. 5 indicates the
 334 fitting quality. The band height is evaluated by considering the variance of

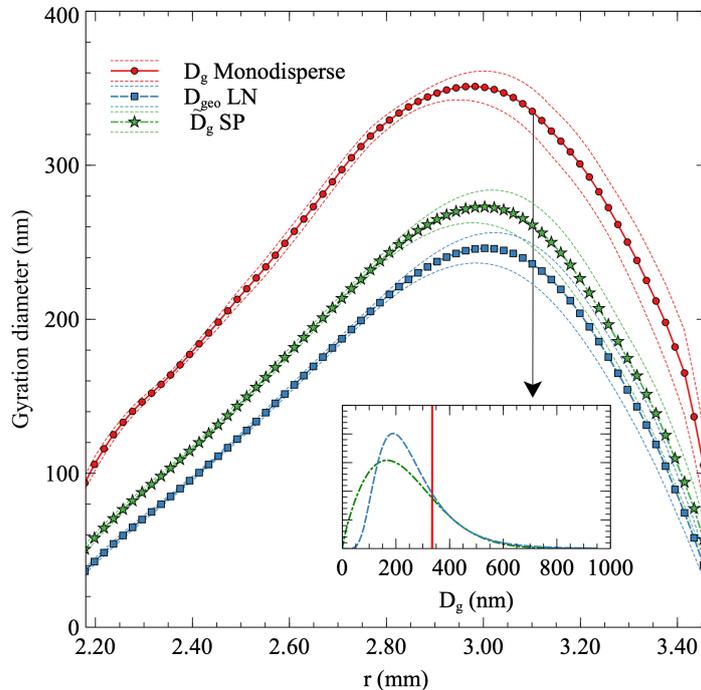


Figure 5: Radial profiles of soot gyration diameter distribution parameters at HAB=30 mm. The three profiles correspond to monodisperse, logNormal ($\sigma_{geo} = 1.6$), and self-preserving ($\lambda_{SP} = 0$) density probability functions. The bands between the dotted lines around each curve are an indication of the reliability level of the results. The inset illustrates the three aggregate size distributions at $r = 3.1$ mm.

335 experimental measurements around the fitted model. It should be noticed,
 336 however, that the band height is not the measurement uncertainty since it is
 337 very difficult to evaluate the latter. Nevertheless, we can consider the band
 338 height as a proxy to the measurement uncertainty. In consequence, we can
 339 see the uncertainty relative to the soot aggregate size is larger on the outer
 340 edge of the flame wing. This is in part caused by the lower signal intensity
 341 in this region. For $r \leq 2.8$ mm, the fitting quality is slightly better for
 342 the Lognormal and the Self-Preserving distributions than the Monodisperse
 343 one. For the two polydisperse distributions (LN and SP) the fitting quality is
 344 essentially the same over the entire radial range at HAB = 30 mm, suggesting
 345 that it is impossible to say which distribution is a better representation of
 346 soot aggregate size based on the fitting quality alone.

347 In addition to the determination of aggregate size distribution parame-
 348 ter, the fitting process of the recorded signals (Fig. 4) also enables an ex-
 349 trapolation of the forward scattering intensity ($K_{vv}(0^\circ)$) corresponding to
 350 the amplitude term in Eq. 7. The results are reported in Fig. 6-A for the
 351 three assumed aggregate size distributions. $K_{vv}(0^\circ)$ is clearly affected by the
 352 hypothesis of the aggregate size distribution primarily in the amplitude but
 353 also slightly in the radial position of the peak.

354 The forward scattering intensity is determined by the soot volume fraction
 355 f_v , scattering function of the refractive index $F(m)$, and ratio of aggregate
 356 volume moments $\overline{V_{agg}^2}/\overline{V_{agg}}$. In order to extract the moment ratio, we di-
 357 vide the amplitude term by f_v and $F(m)$, which are reported in Fig. 6-B
 358 at HAB = 30 mm. The volume fraction f_v has been determined by LOSA
 359 measurements at multiple wavelengths (500, 532, 660 and 810 nm) in order
 360 to take into account the influence of maturity dependence of soot optical
 361 properties [10]. The value of $F(m)$ is derived from the spectral dependence
 362 of absorption function $E(m)$, which in turn provides the soot maturity in-
 363 dex and finally the absolute absorption function $E(m)$ (see [10]). The same
 364 methodology can be used to infer the scattering function $F(m)$ in the present
 365 study (see Appendix C by utilizing the relationship between maturity in-
 366 dex and $F(m)$). Nevertheless, it should be pointed out that according to
 367 the model used the change of scattering function at 532 nm is fairly weak
 368 compared to the change of absorption function (see [10]). Thus, the impact
 369 of soot maturity on the scattering function is less pronounced.

370 Figs. 6-B and 6-A clearly show a shift among the maximum volume frac-
 371 tion (at $r \approx 2.6$ mm), scattered intensity ($r \approx 2.8$ mm) and sharp increase
 372 of maturity ($r \approx 2.95$ mm). The corresponding ratio $\overline{V_{agg}^2}/\overline{V_{agg}}$ for the three
 373 assumed distributions is reported in Fig. 6-C. This ratio remains almost the
 374 same as the average aggregate volume for $r \leq 2.3$ mm; however, it shifts
 375 slightly towards larger values at larger radial positions for polydisperse ag-
 376 gregate distributions. In addition, the ratios for the lognormal distribution
 377 are seen to be slightly larger than those of the self-preserving distribution.

378 The increase in $\overline{V_{agg}^2}/\overline{V_{agg}}$ with increasing r suggests an increase in the av-
 379 erage aggregate volume and in the aggregate polydispersity up to the outer
 380 edge of the flame. The even higher values of this ratio beyond $r = 3.25$ mm,
 381 where soot volume fraction drops to zero, are unphysical and can be consid-
 382 ered a result of noise.

383 On the other hand, it can be seen from Fig. 6-C that the uncertainty in
 384 $\overline{V_{agg}^2}/\overline{V_{agg}}$, which is based on the propagation of error of the amplitude and

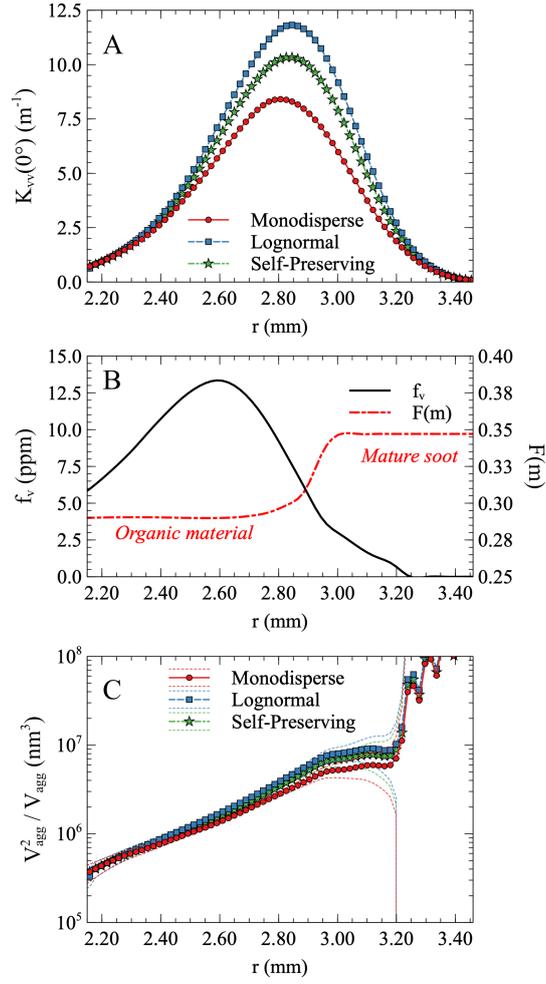


Figure 6: Radial profiles of selected properties of soot particles at HAB = 30 mm. A) Amplitude of the signal extrapolated at forward scattering $K_{vv}(0)$. B) Radial distribution of soot volume fraction f_v and scattering function $F(m)$ according to [10]. C) Ratio of aggregates volumes momentum's $\overline{V_{agg}^2} / \overline{V_{agg}}$. The three profiles in the top and bottom plots correspond to data interpretation for the assumed three aggregate gyration diameter distributions: monodisperse, lognormal ($\sigma_{geo} = 1.6$), and self-preserving ($\lambda_{SP} = 0$).

385 volume fraction terms, also increases with increasing r due to the decrease
 386 of f_v . Up to $r \approx 2.9$ mm, it is not surprising to observe an increase of the
 387 aggregate volume since the aggregate size increases (see Fig. 5). However, it
 388 is less obvious for the increase in the ratio at larger r where the aggregate
 389 size decreases, Fig. 5. This may be interpreted by an increase of the primary
 390 sphere radius. Indeed, as shown in Table 1, the representative aggregate
 391 volume term (the ratio of aggregate volume moments) depends on not only
 392 the aggregate gyration diameter but also the primary sphere diameter D_p .
 393 At this point, the aggregate size distribution is already determined thus the
 394 radial behaviour of the primary sphere diameter can be established (see Fig 7-
 395 A). Because this quantity depends not only on the amplitude and angular
 396 variation of the scattered signal but also on the soot volume fraction and the
 397 morphological parameters (D_f and k_f), its uncertainty is relatively large. For
 398 example, a lower fractal prefactor will cause an increase in the determined
 399 primary sphere diameter. Again, the band bounded by two dotted lines
 400 around each curve indicates the uncertainty based on error analysis. These
 401 curves clearly show large uncertainties in the inner and outer regions of the
 402 flame along the radial direction.

403 It is noticed that the obtained primary sphere diameters are in reasonably
 404 good agreement with results reported in the literature. At the same HAB,
 405 Botero et al. [49] found by TEM image analysis in a laminar coflow diffusion
 406 flame over the Yale burner and fueled with nitrogen diluted ethylene that
 407 the primary soot particles have a modal diameter of 21 nm with a high
 408 polydispersity. Slightly larger values have been reported by Kempema and
 409 Long [35]. In the same burner as in the present study and also for pure
 410 ethylene, Cortés et al. [46] also performed TEM image analysis and found at
 411 a similar location (HAB = 27 mm) and oxygen index (21%) that the primary
 412 soot particles have a geometric mean diameter of 16 nm and a geometric
 413 standard deviation of 1.6. Considering that Rayleigh scattering is a process
 414 proportional to D_p^6 , the results of Cortés et al. [46] can be reanalysed in terms
 415 of the optical equivalent primary sphere diameter by using the Hatch-Choate
 416 equation (average primary diameter at power 6) to yield 31 nm, in very close
 417 agreement with the present results. Indeed, we observe $D_p \approx 32$ nm in the
 418 region where the uncertainties are low (see Fig 7-A)).

419 The region of low uncertainties ($2.6 \leq r \leq 2.85$) displays a locally in-
 420 creasing trend of the primary sphere diameter with increasing r that can be
 421 related to the surface growth process. This region also corresponds to the
 422 rapid increase in soot maturity as shown by the red dashed curve in Fig 6-B.

Table 1: Analytical expressions for the ratio of aggregate volume moments for monodisperse, lognormal, and self-preserving size distributions.

Distribution	$\frac{V_{agg}^2}{V_{agg}}$
Mono	$\frac{\pi}{6} D_p^{3-D_f} k_f D_g^{D_f}$
Lognormal	$\frac{\pi}{6} D_p^{3-D_f} k_f D_{geo}^{D_f} e^{(2-\frac{1}{2})D_f^2 \ln^2(\sigma_{geo})}$
Self-Preserving	$\frac{\pi}{6} D_p^{3-D_f} k_f \widetilde{D}_g^{D_f} \frac{2-\lambda_{SP}}{1-\lambda_{SP}}$

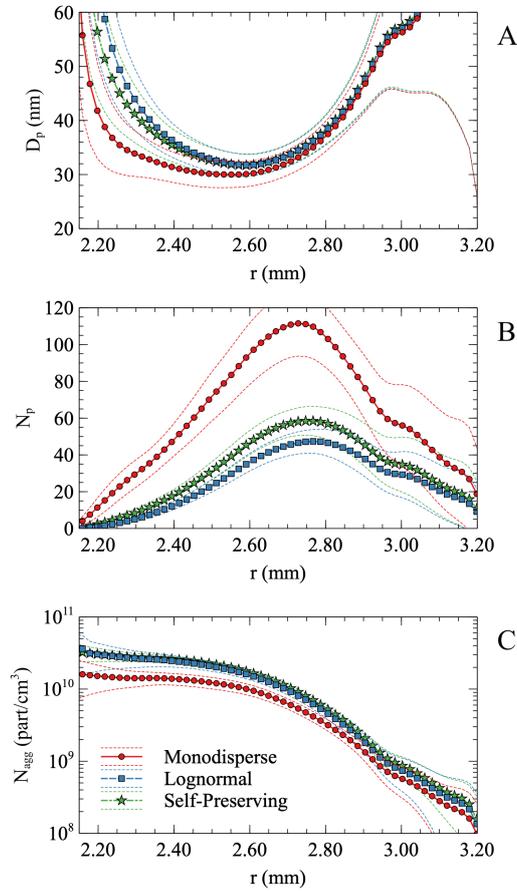


Figure 7: Radial profiles of some soot properties inferred from light scattering measurements at HAB = 30 mm. A) Primary sphere diameters D_p , B) Number of primary spheres per aggregate N_p related to the representative diameter and C) Aggregates number density N_{agg} . The three curves correspond to results for Monodisperse, Lognormal ($\sigma_{geo} = 1.6$) and Self-Preserving ($\lambda_{SP} = 0$) density probability function hypotheses.

423 In addition, since gyration diameter and primary sphere diameter are
 424 determined, we can evaluate the corresponding representative number of pri-
 425 mary spheres (N_p) per aggregate employing the fractal law (Eq. 6). The
 426 result is reported in Fig 7-B. As already discussed for the gyration diameter,
 427 N_p is found to be larger if the aggregates are assume to be monodisperse.

428 The maximum average number of primary spheres per aggregate (at about
 429 $r = 2.74 \text{ mm}$) is found slightly shifted toward the flame centerline compared
 430 to the maximum representative gyration diameter (at about $r = 3.0 \text{ mm}$,
 431 Fig 5). This is explained by the increase of the primary sphere diameter due
 432 to the surface growth. Assuming lognormal and self-preserving aggregate
 433 gyration diameter distributions, the determined maximum average number
 434 of primary spheres per aggregate is found to be 46 and 58 at HAB = 30 mm,
 435 respectively.

436 Finally, the aggregate number density can be determined. Indeed, it is
 437 evaluated as $N_{agg} = f_v/\overline{V_{agg}}$, with $\overline{V_{agg}} = \pi D_p^{3-D_f} k_f \overline{D_g^{D_f}}/6$ and $\overline{D_g^{D_f}}$ is given
 438 in Appendix B according to the type of assumed aggregate size distribution.
 439 The results are reported in Fig 7-C. The aggregate number density decreases
 440 with increasing r due to aggregation. At radial positions larger than about r
 441 = 2.75 mm, we observe a decreasing aggregate number density (Fig 7-C) of
 442 smaller aggregates (Fig 7-B), which seems composed of large primary spheres
 443 (Fig 7-A).

444 4.2. Results at different HAB

445 The methodology described above has also been applied to different heights
 446 above the burner. To highlight the capability of the proposed light scat-
 447 tering technique and focus on the measured soot properties in the laminar
 448 diffusion flame, we only report below the results based on the hypothesis of
 449 self-preserving aggregate size distribution, which is supported by theoretical
 450 considerations as mentioned in Section 3.3. It is also useful to recall that
 451 the corresponding governing parameter \widetilde{D}_g is larger than the modal diam-
 452 eter of the distribution by a factor 1.57, which results from its moment at
 453 a power D_f (see discussion in Section 3.3). Therefore, the corresponding
 454 average number of primary spheres per aggregate will be also slightly larger
 455 as seen in Fig 7-B.

456 Figures 8 and 9 report respectively the radial profiles of the self-preserving
 457 governing parameter \widetilde{D}_g and the corresponding average number of primary
 458 spheres per aggregate N_p at different HAB. The corresponding uncertainties

459 are represented by the error bars. Clearly, an intense particle aggregation
 460 takes place along the flame wing region up to about $HAB = 45$ mm, followed
 461 by a significant decrease of aggregate size at higher HAB. Indeed, simulations
 462 performed with the CoFlame code [50] (reported in [Appendix D](#)) indicate
 463 that particle nucleation, coagulation, and surface growth via surface reactions
 464 (HACA) and PAH adsorption processes govern soot production at the lower
 465 part of the flame, leading to increased number and diameter of primary
 466 particles. These particles are convected downstream where the oxidation
 467 processes by OH radicals and O₂ molecules are dominant, causing a decrease
 468 in their size and eventually complete oxidation.

469 To our knowledge, there have been no studies of *in situ* determination
 470 of the aggregates size in the flame wing region of a pure ethylene diffusion
 471 flame established over the Gülder burner. Therefore, it is challenging to di-
 472 rectly compare our present results with those of previous studies. Kempema
 473 and Long [35] applied 2D-MALS (multi-angle light scattering) in a nitrogen
 474 diluted ethylene flame (20% C₂H₄ and 80% N₂) over the Yale coflow burner
 475 and found similar profiles. The monodisperse equivalent gyration diameters
 476 they reported was about 440 nm, which is much smaller than the maximum
 477 values reported in the present work. However, it should be noted that for
 478 larger aggregates ($\widetilde{D}_g \geq 200$ nm), the fractal dimension can affect the size
 479 measurement since part of the collected signal emanates from the power-law
 480 regime. Thus, the uncertainty associated with the largest diameters, and
 481 thus in particular for $HAB = 45$ mm, can become significant. Moreover,
 482 the different fuels and burners between our work and previous studies may
 483 largely explain the difference. Finally, we believe that the present technique
 484 offers a greater spatial resolution, enabling to properly resolve the aggregate
 485 sizes in the flame region of high gradients. Indeed, we show in [Appendix E](#)
 486 that the classical 2D-MALS can underestimate the peak gyration diameter
 487 detected at $HAB = 40$ mm by about 100 nm due to the change of mea-
 488 surement volume with the scattering angle. Nevertheless, this strong effect
 489 seems to appear only in the presence of large spatial gradients of aggregate
 490 size distribution parameter.

491 The largest aggregates in terms of \widetilde{D}_g are found at $HAB = 40$ mm,
 492 whereas larger values of N_p seem to be at $HAB = 55$ mm. This difference
 493 may be explained by the smaller primary spheres at $HAB = 55$ mm (see
 494 [Figure 10](#)). Kempema and Long [35] reported median numbers of monomers
 495 per aggregate between 47 and 71 along the centerline, and between 14 to 16
 496 along the wing of a flame established over the Yale coflow burner. Although

497 the presently reported values are larger than those of Kempema and Long
 498 [35], it should be noticed that in addition to the different flame conditions
 499 the present results correspond to the number of primary sphere of the rep-
 500 resentative size parameter \widetilde{D}_g , which is not the median parameter reported
 501 by Kempema and Long [35].

502 Due to the large uncertainties related to the primary sphere diameter
 503 determination, we only report in Figure 10 the representative values along
 504 HAB. These values are the primary sphere diameter whose uncertainty is
 505 the lowest in the radial profile at the given HAB. The overall decreasing
 506 trend in the representative primary sphere diameter above HAB = 50 mm
 507 shown in this figure confirms the strong oxidation process in the upper part
 508 of the flame, which is also qualitatively observed from the predictions of the
 509 simulated flame (see Appendix D). The results are also in reasonably good
 510 agreement with the TEM image analysis reported in [35].

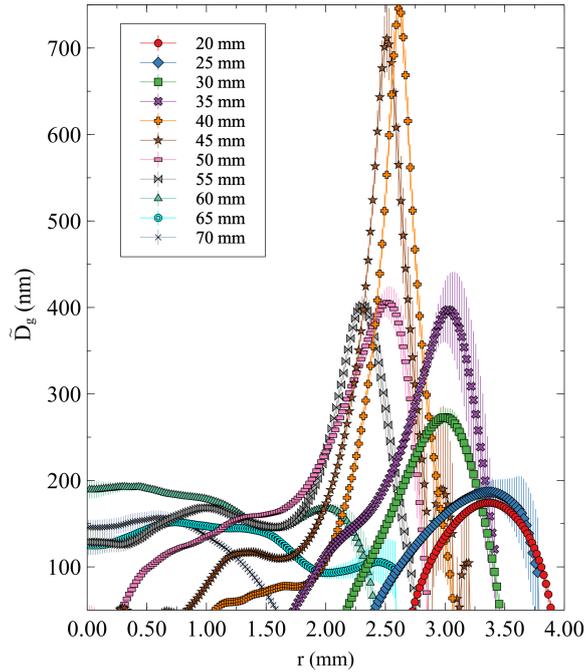


Figure 8: Radial profiles of the aggregate size distribution parameter in the self-preserving distribution assuming ($\lambda_{SP} = 0$) at different HAB.

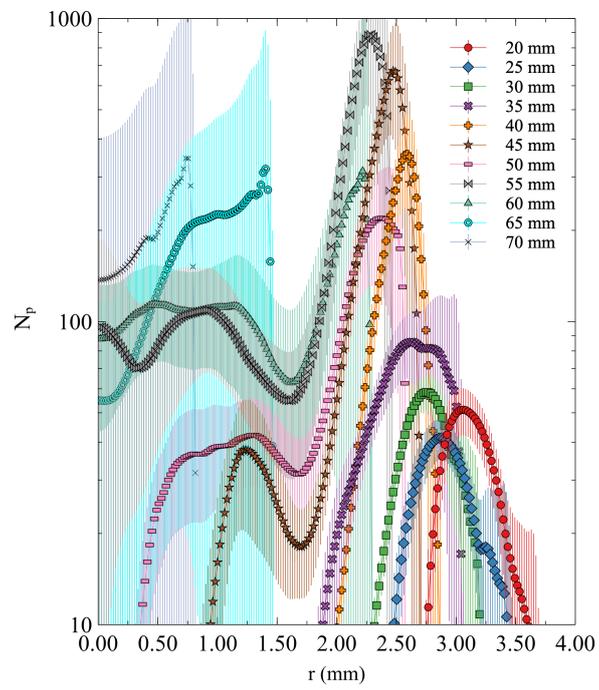


Figure 9: Radial profiles of the average number of primary spheres per aggregate at different HAB.

511 At low HAB, soot is present only in the flame wing region. With increas-
 512 ing HAB, soot starts to appear in the flame centreline region. At upper part
 513 of the flame ($55 \leq HAB \leq 70$ mm), the aggregate size seems relatively uni-
 514 form in the centerline region within about $r \leq 1.5$ mm. Aggregation seems
 515 less intense in this region compared to the outer flame edge. Nevertheless, at
 516 the flame centerline large aggregates can be found at $HAB = 60$ mm (up to
 517 about 200 nm) but then decrease to about 150 nm at $HAB = 70$ mm. This
 518 decrease is mainly caused by the decrease of primary sphere diameters while
 519 the number of primary spheres still increases. Because this region is marked
 520 by a strong reduction of soot material (decrease of volume fraction shown in
 521 [Appendix C](#)), we can speculate that the soot oxidation in this region of the
 522 flame is likely a surface process which mainly reduces the diameter of the
 523 primary spheres. Moreover, the aggregation process seems to compensate
 524 the possible fragmentation process to some extent.

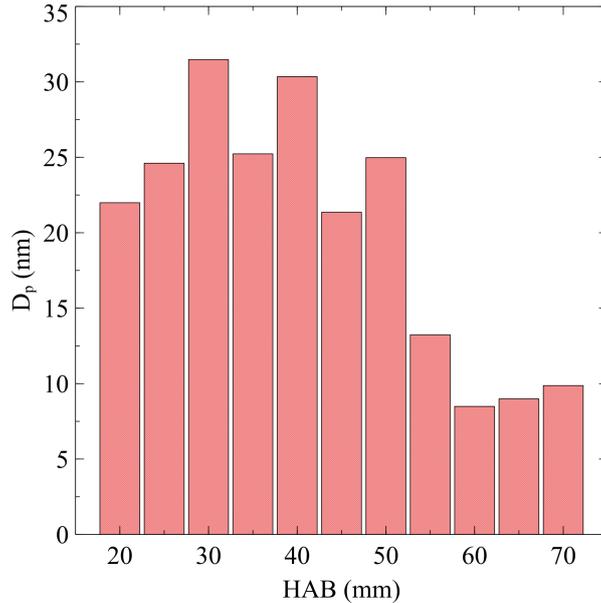


Figure 10: Variation of the representative primary sphere diameter along HAB. The representative primary sphere diameter at each HAB is the value whose uncertainty is the lowest.

525 Figure 11 presents the radial profiles of aggregate number density at dif-
 526 ferent HAB. The vertical bars represent the uncertainty for each value of
 527 N_{agg} . The gradient of this parameter remains very large on the flame wings

528 up to about $HAB = 50$ mm, above which soot particles in the centerline
 529 region can be detected.

530 Although some soot particles in the centerline region may be convected
 531 from the outer region, soot particles in this region are believed to be mainly
 532 composed of nucleation soot with smaller aggregates and immature (lower
 533 $E(m)$ and $F(m)$). In this region, the number density is seen to be between
 534 3×10^{10} and 8×10^{10} particles/ cm^3 , in good agreement with [23, 51] which
 535 reported $N_{agg} \sim 10^{10}$ cm^{-3} in similar ethylene diffusion flames.

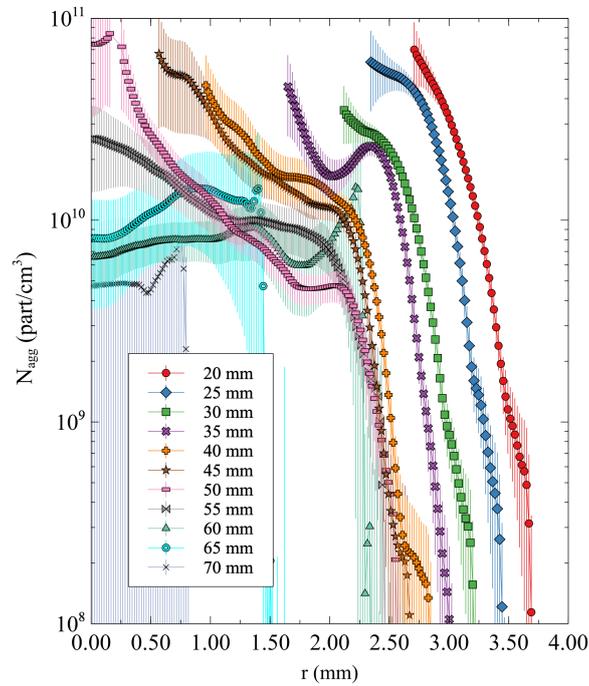


Figure 11: Radial profiles of the aggregates number density at different HAB.

536 5. Conclusion and perspectives

537 The present experimental investigation complements our recent study us-
 538 ing a multiple-wavelength LOSA analysis for the determination of soot vol-
 539 ume fraction and maturity in a laminar coflow ethylene/air diffusion flame
 540 produced using a Gülder burner. In the present work, the spatial distri-
 541 bution of aggregate size is determined by elastic angular light scattering.

542 The originality of the proposed technique lies in optical setup using a laser
543 sheet arranged horizontally through the flame and we named this method
544 the Horizontal Planar Angular Light Scattering (HPALS). This setup avoids
545 the issues related to a significant change of the measurement volume with
546 the scattering angle making the conventional light scattering techniques chal-
547 lenging to apply to flames where soot properties display strong gradients.

548 The Horizontal Planar Angular Light Scattering technique introduced in
549 this work enables a description of soot characteristics with an extremely high
550 spatial resolution, though it is necessary to conduct Abel inversion.

551 The coupling of HPALS with LOSA enables a comprehensive description
552 of soot volume fraction, size (aggregate and primary sphere), and number
553 density. The limitation of this technique is that it can only be applied to
554 axisymmetric laminar flames. The coupling of HPALS and multi-wavelength
555 LOSA paves the way to provide a more complete description of soot proper-
556 ties and hence helps improve our understanding of soot formation and oxida-
557 tion mechanisms. The present work also enables a complete database of soot
558 properties that is useful for validation of soot models able to consider the
559 aggregation mechanisms. Additionally, the high spatial resolution of HPALS
560 allows to improve and validate the coagulation models in regions of steep
561 gradients where the conventional light scattering techniques cannot properly
562 resolve soot properties, such as N_{agg} and N_p . In summary, the combination
563 of HPALS and multi-wavelength LOSA offers a powerful diagnostic tool to
564 provide improved description of soot properties related to aggregate size.

565 **Acknowledgments**

566 The authors gratefully acknowledge the support provided by France's
567 ANR ASTORIA under the research grant ANR-18-CE05-0015 and the Re-
568 gion of Normandy RIN Gasprores project and Chile's National Agency
569 for Research and Development (ANID) through research program FON-
570 decyt/Regular 1191758. Also, this work was performed by the mobility France-
571 Chile joint program receiving a research grant provided by Ecos/ANID C19E01.

572 **Appendix A. Calibration of the scattered signal**

573 Figure A.12 shows the radial profile of the acquired scattering signal be-
 574 fore and after Abel deconvolution. The average level determined after de-
 575 convolution $Ab^{-1}(P_{vv,C_2H_4}) = 0.0653$ is used to calibrate the scattering mea-
 surements.

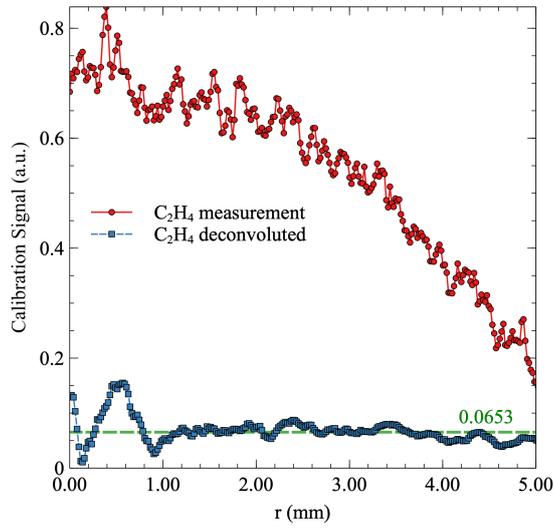


Figure A.12: The radial profiles of scattered signal obtained from a free jet of ethylene.

576

577 **Appendix B. Analytical expression of gyration radius related mo-**
 578 **ments**

579 Equations B.1, B.2 and B.3 provide the analytical expressions of $\overline{R_g^{2D_f}}$
 580 for monodisperse, lognormal, and self-preserving aggregate size distributions,
 581 respectively

$$\overline{R_g^{2D_f}} f = R_g^{2D_f} f(qR_g) \quad (\text{B.1})$$

$$\overline{R_g^{2D_f}} f = \int_0^\infty \frac{D_g^{2D_f-1} f(qR_g) e^{-\frac{1}{2} \left(\frac{\ln(D_g/D_{g_{eo}})}{\ln(\sigma_{g_{eo}})} \right)^2}}{2^{2D_f} \sqrt{2\pi} \ln(\sigma_{g_{eo}})} dD_g \quad (\text{B.2})$$

$$\overline{R_g^{2D_f} f} = \int_0^\infty \frac{D_f}{2^{2D_f}} \frac{(1 - \lambda_{SP})^{(1-\lambda_{SP})}}{\Gamma(1 - \lambda_{SP})} \frac{D_g^{D_f(3-\lambda_{SP})-2}}{\widetilde{D}_g^{D_f(1-\lambda_{SP})}} f(qR_g) e^{-(1-\lambda_{SP})\left(\frac{D_g}{\widetilde{D}_g}\right)^{D_f}} dD_g \quad (\text{B.3})$$

582 Equations B.4, B.5 and B.6 are the analytical expressions of $\overline{R_g^{2D_f}}$ for
 583 monodisperse, Lognormal, and self-preserving aggregate size distributions,
 584 respectively

$$\overline{R_g^{2D_f}} = R_g^{2D_f} \quad (\text{B.4})$$

$$\overline{R_g^{2D_f}} = \left(\frac{D_{geo}}{2}\right)^{2D_f} e^{2D_f^2 \ln^2(\sigma_{geo})} \quad (\text{B.5})$$

$$\overline{R_g^{2D_f}} = \left(\frac{\widetilde{D}_g}{2}\right)^{2D_f} \frac{2 - \lambda_{SP}}{1 - \lambda_{SP}} \quad (\text{B.6})$$

585 Equations B.7, B.8 and B.9 provide the analytical expressions of $\overline{R_g^{D_f}}$ for
 586 monodisperse, lognormal, and self-preserving size distributions, respectively

$$\overline{R_g^{D_f}} = R_g^{D_f} \quad (\text{B.7})$$

$$\overline{R_g^{D_f}} = \left(\frac{D_{geo}}{2}\right)^{D_f} e^{\frac{D_f^2 \ln^2(\sigma_{geo})}{2}} \quad (\text{B.8})$$

$$\overline{R_g^{D_f}} = \left(\frac{\widetilde{D}_g}{2}\right)^{D_f} \quad (\text{B.9})$$

587 Appendix C. $F(m)$ and f_v based on multi-wavelength LOSA mea- 588 surements

589 Figure C.13 shows the radial profiles of soot volume fraction at different
 590 HABs. These profiles were computed by considering the dependence of the
 591 scattering function $F(m)$ on the maturity coefficient β , as shown in Fig. C.14
 592 (see [10] for details).

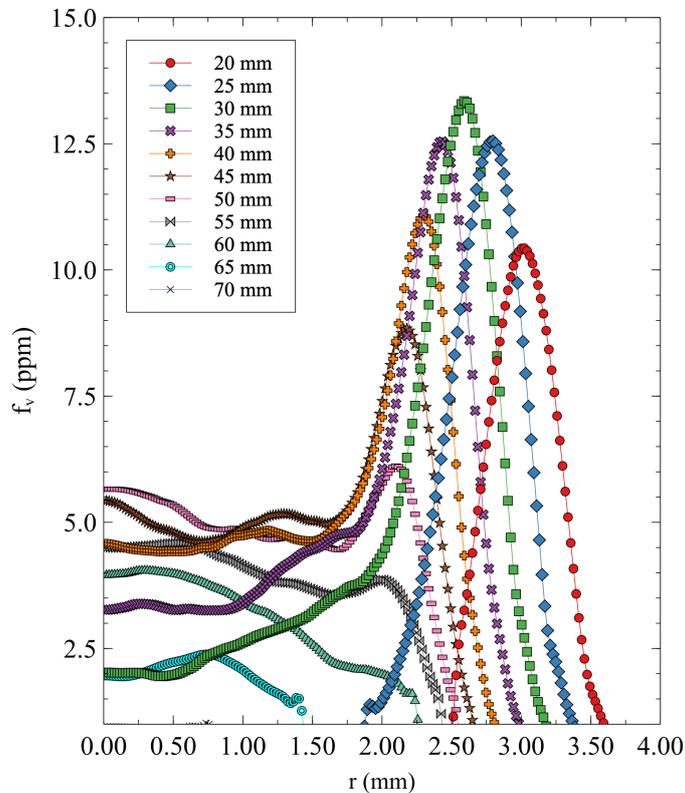


Figure C.13: Soot volume fraction as a function of the height above the burner (Data from [10])

593 **Appendix D. CoFlame simulation of the ethylene diffusion flame**

594 To complement the experiments and to provide useful details of the
 595 soot formation processes, the laminar coflow flame was simulated with the
 596 CoFlame code [50]. Details of the simulation are provided in [10]. The
 597 gas-phase kinetics is modeled with the DLR chemical mechanism [52]. The
 598 transport equations for the flow, species, energy, and soot are solved in
 599 a fully-coupled fashion. The population balance equations of soot aggre-
 600 gates and primary particles were solved using a sectional method consid-
 601 ering soot nucleation due to collision of three five-ring polycyclic aromatic
 602 hydrocarbons (PAHs), surface growth via PAH adsorption (collision theory
 603 and sticking probability) of the same PAHs onto soot surface and by the

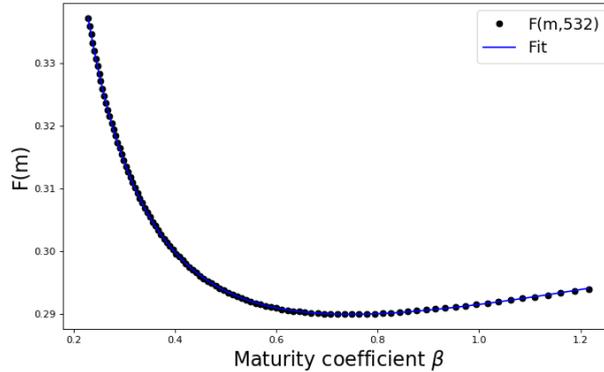


Figure C.14: Relationship between the maturity coefficient and the scattering function using the same methodology described in [10].

604 hydrogen-abstraction carbon-addition (HACA) mechanism [53], which in-
 605 cludes surface oxidation by O_2 (HACA) and OH (collision theory), particle
 606 aggregation and oxidation-driven aggregate fragmentation. The rates of pro-
 607 duction/consumption by these processes are computed from the soot particle
 608 characteristics and related species concentrations (see [50] for details). Fig-
 609 ure D.15 presents a qualitative description, from the converged solution of
 610 the flame simulation, of the different regions of the flame where nucleation,
 611 coagulation, PAH adsorption, HACA, OH and O_2 oxidation prevail.

612 Appendix E. Superiority of HPALS over conventional approaches

613 The main advantage of HPALS over conventional scattering techniques,
 614 such as 2D-MALS, is its ability to provide high spatial resolution over a wide
 615 range of scattering angle (except very small scattering angles in the forward
 616 direction or very large scattering angles in the backward direction), i.e., the
 617 spatial resolution of HPALS is independent of the scattering angle.

618 Indeed, in classical configuration of light scattering experiments (vertical
 619 laser light sheet or horizontal laser beam crossing the flame centerline, see
 620 Fig.E.16), the volume of measurement, which corresponds to the intersection
 621 between the laser and the signal collection optics, increases when scattering
 622 angle deviates from $\theta = \pi/2$. When the scattering signal is detected by a
 623 CCD camera, the spatial resolution of measurement, Δr , is related to a pixel
 624 on the camera with a width Δx , through $\Delta r = \Delta x / \sin(\theta)$.

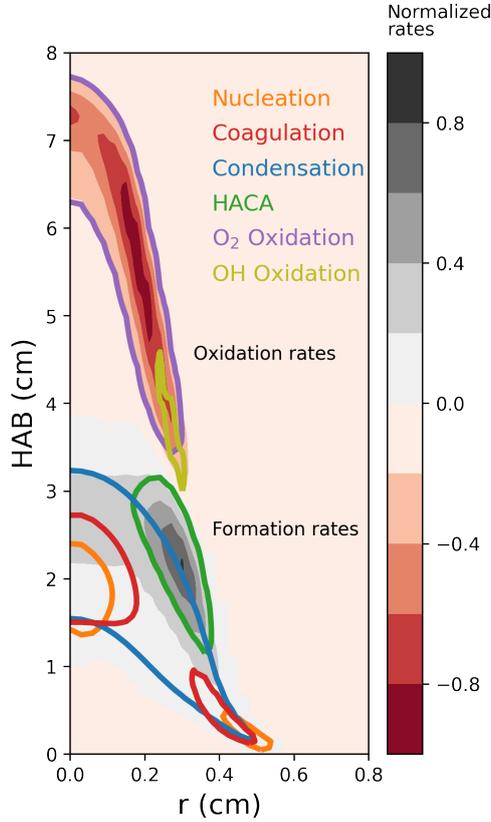


Figure D.15: Distributions of the different soot processes in the laminar ethylene diffusion flame based on the CoFlame simulation.

625 Therefore, even if the scattering intensity is corrected for the scattering-
 626 angle dependent measurement volume as in [34, 36], the detected scattering
 627 intensity is likely contributed from soot particles of different properties when
 628 the spatial resolution is poor at small and large scattering angles.

629 This tends to smooth the angle-resolved light scattering intensity, espe-
 630 cially in the forward and backward directions. In consequence, the fitting
 631 process and the resulting radial distribution of aggregate size distribution
 632 parameters can be affected. This impact is evaluated in the present section.
 633 For this purpose, we consider the radial profile determined by HPALS since
 634 its spatial resolution is independent of scattering angle. To simulate the
 635 collected signals that would be measured by 2D-MALS after correction of

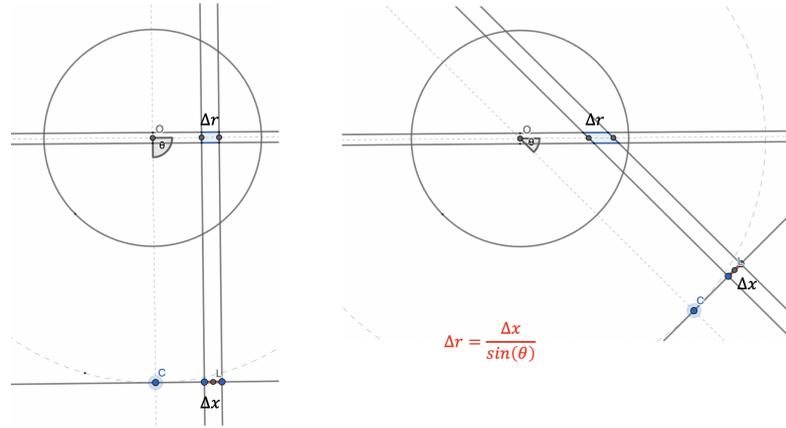


Figure E.16: Illustration of the change of measurement volume with changing the scattering angle in conventional vertical planar configuration (top view).

636 the intensity caused by angle-dependent measurement volume, the HPALS
 637 radial intensity profile is convoluted by a Gaussian function whose width is
 638 inversely proportional to $\sin(\theta)$.

639 The convoluted signals are then processed using the same method as in
 640 the present experiments. The results are compared at two HABs of 30 mm
 641 and 40 mm, the later being the height where larger aggregates and stronger
 642 gradients have been detected (see Fig.E.17). It is clear from this figure that
 643 the reduced spatial resolution of 2D-MALS with varying scattering angle
 644 leads to lower aggregate size distribution parameters in the radial region of
 645 of strong gradients and of large aggregates around $r = 2.6$ mm at HAB =
 646 40 mm. The effect is much less pronounced in regions where the gyration
 647 diameter profile is relatively smooth at both HAB = 40 mm (outside the
 648 peak region between about $r = 2.5$ mm and 2.7 mm) and 30 mm. It is also
 649 interesting to notice that the simulated 2DMALS experiments artificially
 650 enlarges the sooting region slightly at both HAB = 30 and 40 mm, which is
 651 again caused by the reduced spatial resolution with varying scattering angles.

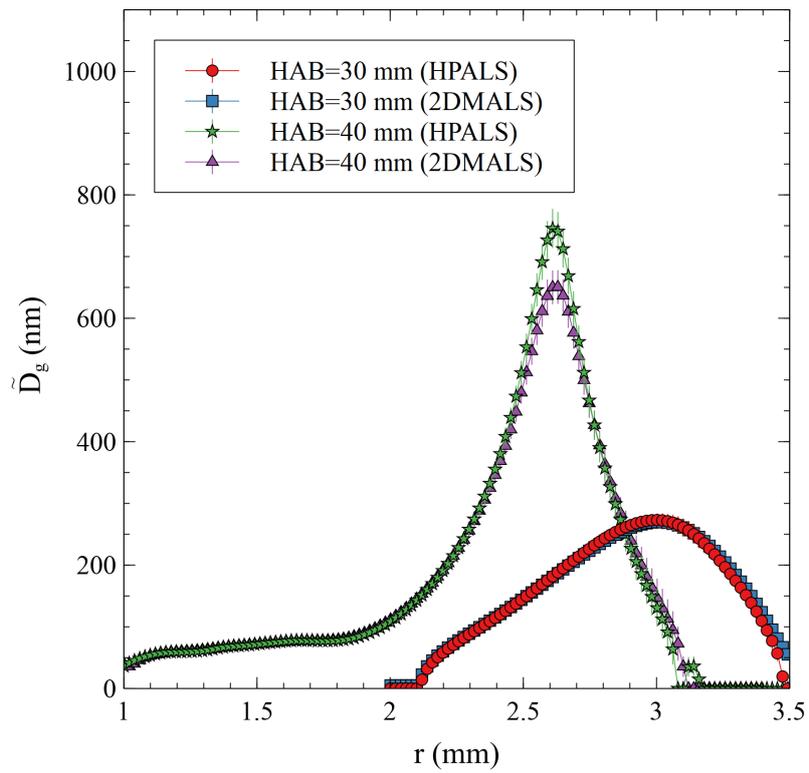


Figure E.17: Comparison of the gyration diameter governing parameter of the Self-Preserving size distribution obtained in the present study (HPALS) and as it should be measured with conventional vertical planar techniques as 2D-MALS. Comparison is performed at 2 HAB.

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