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(UV, VIS) laboratory evaluation of the **lidar** depolarization ratio of freshly emitted soot aggregates from pool fire in ambient air at exact backscattering angle

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Abstract

In this paper, a controlled-laboratory experiment is carried out to evaluate the **lidar** depolarization ratio of freshly emitted soot aggregates in the exact backward scattering direction at 180.0°. The experiment is performed at two wavelengths simultaneously, namely 355 and 532 nm, often used in polarimetric lidar remote sensing. The soot aggregates are generated from a kerosene JET A-1 pool fire in laboratory ambient air and microscopic images confirm the fractal morphology of generated soot aggregates. Then, the Superposition T-Matrix (STM) method is applied to numerically simulate the soot aggregates backscattering properties for different soot particles refractive indices, monomer radii and monomer numbers. The range of these parameters **which ensures** the lowest discrepancy between the laboratory measurement and the STM-computations is discussed within experimental and numerical error bars. *We find that the polydisperse monomers model gives an overall better evaluation of the ratio $F_{22}(\pi)/F_{11}(\pi)$. In the polydisperse case, our numerical and laboratory experimental findings agree at both wavelengths for a refractive index $m = 2.65 + i1.32$ and monomer number $N_m > 40$ at a mean monomer radius of $r_p = 30$ nm ($N_m > 160$ at $r_p = 27.5$ nm).* We believe this work may be useful for the light scattering and remote sensing communities and may also help future studies aimed at better understanding the impact of soot particle aggregates on the Earth's climate, which still needs to be precisely quantified.

Keywords

Backscattering; laboratory; soot particles; **lidar depolarization ratio**; polarimetry; STM.

1. Introduction

Soot aggregates are particles emitted during the incomplete combustion of organic matter (e.g. biomass, hydrocarbon fuel), through natural and anthropogenic processes. These particles are formed by clustered **nearly**-spherical primary particles which form irregularly-shaped aggregates. Different combustion processes can lead to variations in the aggregates size and structure [1], the latter being usually described with a fractal model [2], [3]. Moreover, when airborne, the soot particles can induce air-quality issues and **are part of complex** atmospheric chemistry **processes** [4]–[6]. Indeed, the molecular composition and sub-micronic size of soot aerosols can induce pulmonary and arterial diseases, as well as cancers [4], [7]. Soot particles also contribute to the Earth's climate warming by inducing a positive radiative forcing, through both direct and indirect effects. However, the uncertainty on these effects remains high [8], [9] as these impacts are partly dependent on the morphology of the particles [10]. Hence, the detection of soot particles and their in-situ characterization are required to better understand and predict their impact on air-quality and on the Earth's climate, by reducing the corresponding uncertainties.

55 In this context, the scattering of light in the backward direction (*i.e.* backscattering) is of utmost
56 importance for remote sensing applications such as Light Detection and Ranging (lidar). The
57 inversion of the received lidar signal and the analysis of the resulting lidar products (*i.e.*
58 backscattering and extinction coefficients) allows retrieving the particles concentration, the
59 particles size distribution or morphological properties [11], [12]. The lidar problem is however
60 under-determined as several microphysical parameters (*e.g.* size distribution, radius) can lead
61 to the same backscattered radiation [13], [14]. Hence, lidar inversion algorithms and analysis
62 require a-priori knowledge of several input parameters, such as the lidar ratio or the Lidar
63 Depolarization Ratio (LDR), as emphasized in [15, 19]. The LDR is defined as the ratio
64 between the cross and co-polarized components of the backscattered radiation. It can be
65 used to specify the type of particle present in the medium [16], [17]. To detect soot particles
66 with a lidar is however a challenging task, mainly due to the complex morphology of soot
67 particle aggregates. Recently, Miffre et al. [18] remotely measured the very low Planck
68 thermal radiation emitted by absorbing soot particles by incandescence, after their heating by
69 the lidar laser source. Though promising, as being a clear signature of the absorption process,
70 this methodology is rather difficult to apply during daytime. Hence, complementary
71 methodologies have been developed such as polarization lidars, by considering the low
72 depolarization of soot particles [19]. However, deriving the soot particles depolarization ratio
73 from atmospheric lidar measurements is difficult, at least because the measured
74 depolarization is nevertheless that of a particles mixture: as explained in [12], [15], [17], the
75 measured depolarization ratio may indeed differ from that specific to the soot particles under
76 study.

77
78 To address the scattering properties of soot particle aggregates is also complex from a
79 numerical point of view, mainly due to their complex shape. Numerical models are extensively
80 described in the literature and interested readers can refer to recent papers by Kahnert and
81 Kanngießer [20], Liu and Mishchenko [21], [22]. These numerical models can be used to
82 study the effect of different morphologies on the radiative properties in the lidar backward
83 scattering direction. Indeed, the Lorenz-Mie theory can be applied to calculate the radiative
84 properties of the equivalent sphere based on the particles morphological properties but has
85 shown poor accuracy when applied to soot aggregates [23], [24]. The Rayleigh-Debye-Gans
86 for Fractal Aggregates (RDGFA) theory is also commonly used to compute soot aggregates
87 scattering properties [25]. This method has shown accuracy in the computation of angle-
88 integrated radiative properties (*e.g.* extinction cross section) but its applicability is limited
89 when considering large size parameters and angle-dependent features such as the scattering
90 matrix elements [26]. The Lorenz-Mie theory and the RDGFA method also do not reproduce
91 the particles LDR in the lidar backward scattering direction. The Discrete Dipole
92 Approximation method allows the computation of the electromagnetic field scattered by
93 particles of arbitrary shape [10], [27], [28]. Although this method is often used as a reference
94 to assess the accuracy of other numerical methods, its highly demanding computational cost
95 makes it rather difficult to implement in numerical studies covering a large range of
96 morphological parameters and number of aggregate realizations. Finally, the Superposition T-
97 Matrix (STM) method can also be used to compute soot aggregates scattering properties [29],
98 providing that the particles structures are composed exclusively of non-overlapping spheres.
99 Despite these major recent advances, the underlying assumptions, inherent to numerical
100 simulations, should be discussed.

101
102 In this context, and to face the problem of backscattering by irregularly-shaped particles,
103 controlled-laboratory experiments are proposed to evaluate the lidar particles depolarization
104 ratio [30], [31]. Strictly speaking however, all these experimental set-ups do not operate in the
105 lidar backward scattering direction corresponding to the π -angle, which may lead to potentially
106 quite considerable errors in the evaluation of the LDR [32]. Indeed, at scattering angles
107 approaching the π -angle, the LDR is indeed determined from the ratios F_{22}/F_{11} and F_{21}/F_{11}
108 of the scattering matrix elements, which a priori depend on the scattering angle in a way that
109 still needs to be quantified. In particular, it is only at strict π - backscattering angle that F_{12} is

110 null. Hence and as discussed in [32], [33], precise evaluations of the lidar LDR of soot
111 particles can only be performed at exact backscattering angle, by precisely evaluating the
112 ratio F_{22}/F_{11} at specific backscattering angle of 180.0° . If precise enough, such laboratory-
113 measurements can then be used to discuss, within experimental error bars, on the ability of
114 existing light-scattering numerical models to faithfully reproduce the LDR of soot particles.

115
116 In recent papers [22], [34], lidar measurements of the LDR of stratospheric smoke have been
117 compared with numerical results using spheroid morphological models with varying size,
118 aspect ratio and refractive indices. In [35], the scattering matrix elements of soot particles
119 have been measured from 5° to 175° at 532 nm wavelength, and subsequently modelled
120 using the STM method with different set of morphological parameters and complex refractive
121 index. A different approach is used in this study, where the LDR of freshly emitted soot
122 aggregates is for the first time to our knowledge evaluated in a controlled-laboratory
123 experiment operating at the exact backscattering angle of 180.0° . Moreover, the experiment is
124 achieved at two wavelengths simultaneously, namely $\lambda_{UV} = 355$ nm and $\lambda_{VIS} = 532$ nm, often
125 used in polarimetric lidar remote sensing [16], [17], [19], [36]. The soot particles are emitted
126 from the combustion of a kerosene JET A-1 pool fire in laboratory ambient air. Because of the
127 vicinity of the emission source, we refer to these particles as freshly emitted soot aggregates.
128 Transmission Electron Microscopy (TEM) images of the freshly emitted soot aggregates are
129 also provided and from these TEM-observations, we find the fractal-like morphological model
130 to be the most relevant in our case, as it closely describes our observed structures. Then,
131 using the STM method, several ensembles of freshly emitted soot aggregates scattering
132 properties are computed with varying morphological parameters and refractive indices, before
133 identifying the soot parameters (refractive indices, radii and number of monomers) ensuring
134 the lowest discrepancy between the laboratory-measured and the numerically-computed
135 LDR-values. The paper is hence organized as follows. Section 2 is dedicated to our
136 laboratory experimental set-up at the exact backscattering angle of 180.0° , based on the
137 scattering matrix formalism. Section 3 presents our LDR-laboratory measurements of freshly
138 emitted soot aggregates at wavelengths λ_{UV} and λ_{VIS} . The numerical methods are then
139 presented in Section 4, where the LDR is numerically computed at both wavelengths by
140 applying the STM method. Finally, the soot parameters (refractive indices, radii and number
141 of monomers) ensuring the lowest discrepancy between the laboratory-measured and the
142 numerically-computed LDR-values are discussed in Section 5, followed by our conclusions
143 and outlooks.

144
145

146 2. Light backscattering by soot particles from pool fire

147

148 2.1 Scattering matrix formalism

149

150 Considering an incident electromagnetic wave defined by the Stokes vector I^{inc} , with a
151 direction vector defined as \mathbf{n}^{inc} on an arbitrary particle ensemble, the scattered wave, with
152 propagation direction defined by the vector \mathbf{n}^{sca} , is defined as :

153

$$I^{sca}(r\mathbf{n}^{sca}) = \frac{1}{r^2} \mathbf{Z}(\mathbf{n}^{sca}, \mathbf{n}^{inc}) I^{inc} \quad (1)$$

154

155 where r is the distance from the scattering event to the observer and \mathbf{Z} is the 4×4 scattering
156 phase matrix specific to the particles orientation, shape and refractive index. The orientation
157 averaging [37] of the normalized phase matrix allows to reduce the two propagation vectors
158 dependence to only one scalar property as [38]:

159

$$\langle \mathbf{Z}(\mathbf{n}^{sca}, \mathbf{n}^{inc}) \rangle_{\zeta} = \mathbf{f}(\theta) = \begin{pmatrix} F_{11}(\theta) & F_{12}(\theta) & 0 & 0 \\ F_{21}(\theta) & F_{22}(\theta) & 0 & 0 \\ 0 & 0 & F_{33}(\theta) & F_{34}(\theta) \\ 0 & 0 & F_{43}(\theta) & F_{44}(\theta) \end{pmatrix} \quad (2)$$

where θ is the scattering angle and the notation $\langle \rangle_{\zeta}$ refers to orientation averaging. In the strict backward scattering direction (i.e. $\theta = \pi$), the phase matrix further simplifies to [39] and exhibits only two non-vanishing elements, namely $F_{11}(\pi)$ and $F_{22}(\pi)$. The lidar depolarization ratio (LDR) of soot particles from pool fire is then determined by the ratio $F_{22}(\pi)/F_{11}(\pi)$ of the scattering matrix elements:

$$\text{LDR} = \delta(\pi) = \frac{1 - F_{22}(\pi)/F_{11}(\pi)}{1 + F_{22}(\pi)/F_{11}(\pi)} \quad (3)$$

In this study, the ratio $F_{22}(\pi)/F_{11}(\pi)$ has been experimentally evaluated in laboratory at two wavelengths. Numerical computations of this ratio are also conducted in order to assess the agreement between simplified numerical models and laboratory experimental findings. From Eq. (3), it is clear that increasing the ratio $F_{22}(\pi)/F_{11}(\pi)$ leads to a decrease in the corresponding LDR.

2.2 Laboratory experiment for evaluating the soot particles depolarization

As schemed in Fig 1, light backscattering by soot particles from pool fire is simultaneously measured at two wavelengths ($\lambda_{UV} = 355$ nm, $\lambda_{VIS} = 532$ nm) by two laboratory polarimeters, each measuring the intensity of the backscattered wave at a given wavelength. The backscattering angle is addressed with accuracy and $\theta = (180.0 \pm 0.2)^{\circ}$. The principle of this (UV, VIS) controlled-laboratory experiment at exact backscattering angle ($180.0 \pm 0.2^{\circ}$) has been extensively described in [33] where the same experiment has been used for mineral dust particles embedded in ambient air. As a detailed description is available in this former publication, we here only briefly recall the methodology in order to ease the reading. Following Fig 1, the scattered Stokes vector can be related to the incident Stokes vector by considering the successive Muller matrices corresponding to each optical component present between the light source and the light detector:

$$\mathbf{I}^{sca} = [\mathbf{PBC}][\mathbf{QWP}][\mathbf{F}(\pi)][\mathbf{QWP}][\mathbf{PBC}]\mathbf{I}^{inc} \quad (4)$$

Where $[\mathbf{F}(\pi)]$ is given by Eq. (2) for $\theta = \pi$ and $[\mathbf{QWP}]$ and $[\mathbf{PBC}]$ are the Mueller matrices of the quarter-wave plate and the reflecting polarizing cube respectively. The detected light intensity measured by each polarimeter corresponds to the first component of the scattered Stokes vector. Developing the expression of the different Muller matrices that appear in Eq. (4) for incident light polarized parallel to the scattering plane (e.g. $\mathbf{I}^{inc} = [1, 1, 0, 0]^T$), the intensity of the detected backscattered wave is given by [33]:

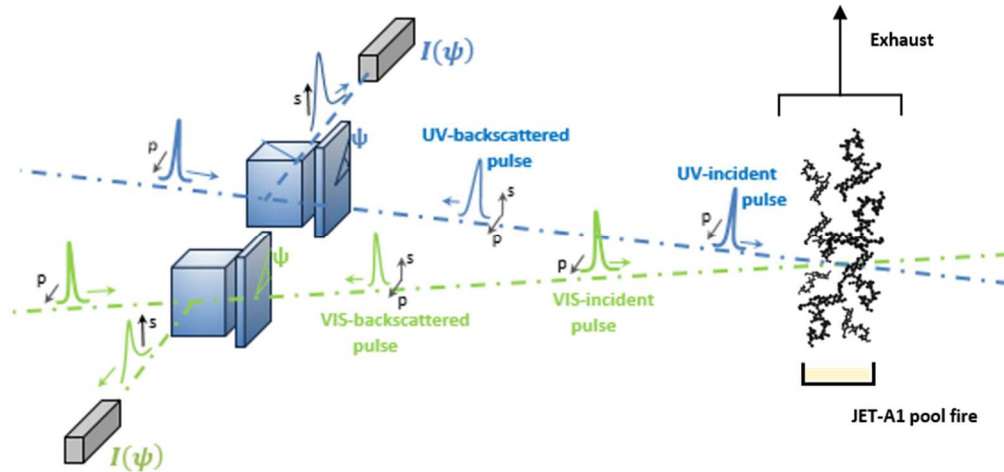
$$I(\psi) = I_0 \times [a - b \cos(4\psi)] \quad (5)$$

where the intensity I_0 mainly depends on the intensity of the laser source and on the distance from the detector to the soot particles while the angle ψ is the angle between the fast axis of the quarter-wave plate and the scattering plane. Interestingly, the coefficients a and b are wavelength-dependent as determined by the scattering matrix elements $F_{11}(\pi)$ and $F_{22}(\pi)$: $2a = F_{11}(\pi) + F_{22}(\pi)$ while $2b = 3F_{22}(\pi) - F_{11}(\pi)$. The $I(\psi)$ -curve hence has $a - b = F_{11}(\pi) - F_{22}(\pi)$ for minimum, which is null for spherical particles only. Likewise, the maxima of the $I(\psi)$ -curve are equal to $2F_{22}(\pi)$. Moreover, the a and b coefficients can be precisely evaluated by adjusting the laboratory-measured $I(\psi)$ -values with Eq. (5), as detailed below in

205 Section 3.2. Precise evaluations of $F_{22}(\pi)/F_{11}(\pi)$ for soot particles embedded in laboratory
 206 ambient air can then be retrieved from a and b since:
 207

$$[F_{22}(\pi)/F_{11}(\pi)]_{lab} = (a + b)/(3a - b) \quad (6)$$

208
 209 As expected, $F_{22}(\pi)/F_{11}(\pi)$ equals unity for spherical particles for which coefficients a and b
 210 are equal. Moreover, the LDR of soot particles can then be retrieved from $[F_{22}(\pi)/$
 211 $F_{11}(\pi)]_{lab}$ by applying Eq. (3).



212
 213
 214 *Figure 1 : Principle of the laboratory backscattering experimental set-up to evaluate the particles depolarization*
 215 *ratio LDR in laboratory ambient air, identical to that presented in [33], but here applied to the particles released*
 216 *from a JET A-1 pool fire. In a few words, the experiment relies on two laboratory polarimeters (one at $\lambda_{UV} = 355$*
 217 *nm in blue, one at $\lambda_{VIS} = 532$ nm in green) operating simultaneously in the exact backward scattering direction $\theta =$*
 218 *$(180.0 \pm 0.2)^\circ$. Based on the Mueller matrix formalism, each laboratory polarimeter allows precise evaluations of*
 219 *the soot particles LDR by adjusting the variations of the detected scattered intensity $I(\psi)$ with Eq. (5) to retrieve*
 220 *$[F_{22}(\pi)/F_{11}(\pi)]_{lab}$ then the corresponding LDR $\delta_{lab}(\pi)$, by applying Eqs. (3,6), as detailed in Section 3.2 below. A*
 221 *distance of 5 meters separates the pool fire from the detectors.*
 222

223 3. Laboratory light backscattering by freshly emitted soot 224 particles from pool fire

225 3.1 Soot emitted from small-scale kerosene pool fire

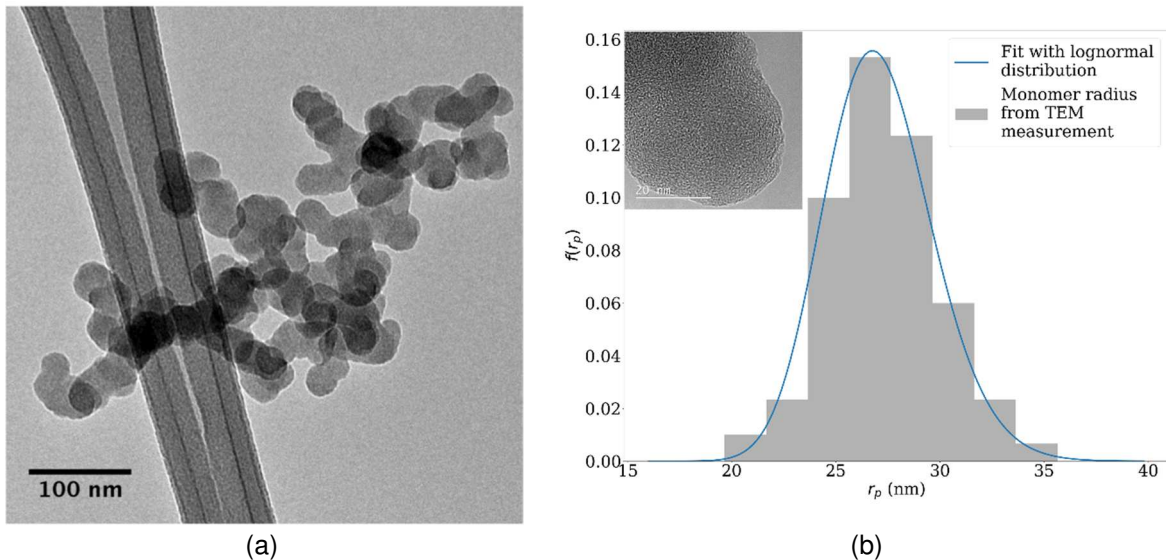
226
 227
 228 Pool fires are defined as self-sustaining turbulent diffusion flames generated over a plain
 229 liquid fuel surface. Such fires are of particular interest for phenomenological investigations of
 230 combustion [40], [41]. For the purpose of this study, we have designed a small-scale
 231 kerosene pool fire which consists in a round container of kerosene with a diameter of 50 mm
 232 and 20 mm height. Such an experimental set-up enables the production of a constant soot
 233 emission from a defined volume of fuel. Following theoretical knowledge about liquid pool
 234 fire, the pool fire heat release rate and burning duration are evaluated for a kerosene burning
 235 rate $m'' = 0.039 \text{ kg}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$. In this study, JET A-1 (NATO code F-35) kerosene was used.
 236 Thus, the heat release rate is estimated at 5 kW for a burning duration of 30 seconds. The
 237 flame height is comprised between 35 cm and 55 cm estimated from the Heskestad and
 238 Thomas method [42], [43]. The light backscattering measurements were carried out for a few
 239 seconds after ignition and during the steady burning time of the pool fire combustion.

240
 241 Soot particles emitted from the small-scale kerosene pool fire have been analyzed by
 242 Transmission Electron Microscopy (TEM), as shown in Fig. 2(a). More precisely, particles
 243 have been captured and transferred on TEM grids covered by a holey carbon film in order to
 244 observe suspended aggregates. TEM grid were placed at the laser height (i.e. 30 cm above

245 the recipient base), and approximately 3 cm off the axis formed by the container center and
 246 the laser beam. The Zeiss LIBRA 200 microscope is equipped with an in-column filter to
 247 achieve filtered images and operates at 200 kV. No degradation of the soot structure has
 248 been observed during the light backscattering experiment. Typical aggregates, as illustrated
 249 in Fig. 2(a), are composed of several tens of monomers with narrow size dispersion. These
 250 monomers exhibit a degree of non-sphericity and overlapping. High resolution images have
 251 shown that the monomers exhibit an onion-like structure fully graphitized. Due to restraints
 252 imposed by the process of particles sampling, the monomer radius reported here represents
 253 radii averaged over several TEM grids at different sampling location in the pool fire. Analysis
 254 of the TEM image allows the retrieval of the monomer radius size distribution, as displayed in
 255 Fig. 2(b). To characterize this size distribution, we use the lognormal size distribution defined
 256 as:

$$f(r_p) = \frac{1}{r_p \ln(\sigma_p) \sqrt{2\pi}} e^{-\frac{1}{2} \left(\frac{\ln(r_p) - \ln(R_p)}{\ln(\sigma_p)} \right)^2} \quad (7)$$

258 where $f(r_p)dr_p$ is the size distribution giving the number of monomers with radius between r_p
 259 and $r_p + dr_p$, R_p is the mean radius and σ_p is the monomer radius deviation. This distribution
 260 is often used in order to characterize soot aggregates monomers polydispersion [44]. Using
 261 this method, we find that the monomer radius size distribution is characterized by a mean
 262 radius of $R_p = 27 \text{ nm}$ and a standard deviation of $\sigma_p = 1.1$. This value for primary soot particle
 263 size is relatively large compared with soot produced by laminar diffusion flames but remains
 264 consistent with previous results obtained from soot emitted by pool fires [45].
 265
 266

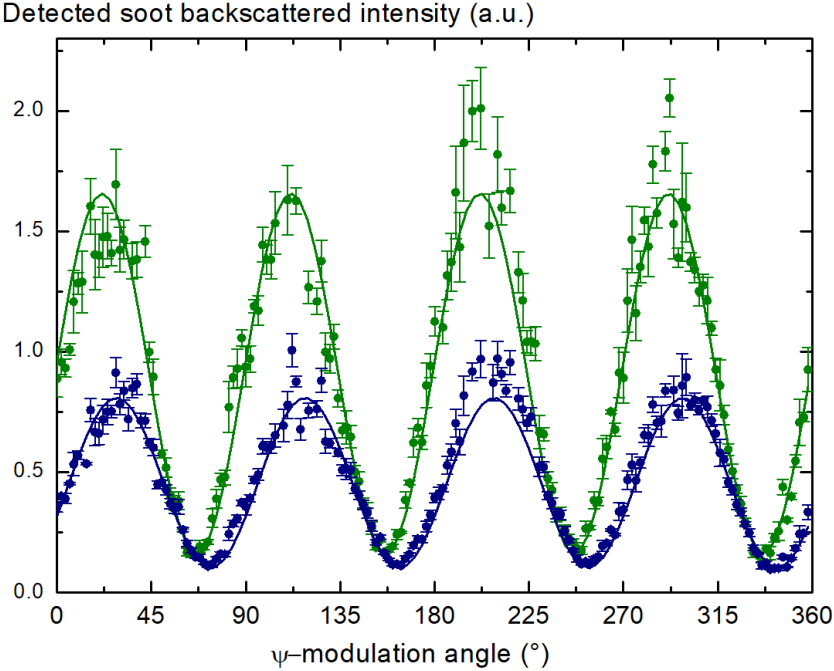


267 Figure 2: (a) HR TEM image of a soot aggregate generated from our JET A-1 pool fire, (b) Corresponding
 268 monomer radius size distribution. In order to represent this distribution, 151 monomers have been characterized.
 269 The top-left insert is a zoomed in view of a monomer, showing the onion-like structure.
 270

271 3.2 Light backscattering by freshly emitted soot particles from pool fire

272
 273 By applying the experimental set-up presented in Fig. 1, we display in Fig. 3 the variations
 274 of the detected backscattered intensity $I(\psi)$ as a function of the modulation angle ψ at
 275 wavelength λ_{UV} and λ_{VIS} . Light backscattering from the soot particles generated from the
 276 pool fire is distinguished from that due to particles in laboratory ambient air by using pulsed
 277 laser light. Special care has been taken to minimize the potential polarization and wavelength
 278 cross-talks affecting the detected backscattered light intensity (see [33] for more details). At
 279 each wavelength, within our experimental error bars, the detected backscattered intensity

280 exhibits constant minima, which mean that the shape distribution of the generated soot
 281 particles did not vary during the acquisition. Indeed, these minima, which are equal to
 282 $F_{11}(\pi) - F_{22}(\pi)$ (see Section 2.2), are a clear optical signature of the soot particles non-
 283 sphericity and interestingly, the precision of our experimental set-up allows to unambiguously
 284 measure these minima at two wavelengths to evaluate the soot particles LDR at two
 285 wavelengths. A precise evaluation of the soot LDR can then be achieved from Fig. 3 where
 286 the detected scattered light intensity is recorded for each position of the QWP, to gain in
 287 accuracy. Our experimental data points are then adjusted with Eq. (5) to get precise
 288 evaluations of the ratio $F_{22}(\pi)/F_{11}(\pi)$ from Eq. (6), as given in Table 1. As a result, the LDR of
 289 soot particles released from the pool fire in laboratory ambient air is retrieved from Eq. (3) and
 290 is equal to $(11.7 \pm 2.3) \%$ at wavelength λ_{UV} and to $(8.7 \pm 2.1) \%$ at wavelength λ_{VIS} . Table 1
 291 summarizes our findings from this laboratory backscattering experiment.



292
 293 *Figure 3 : Detected backscattered light intensity $I(\psi)$ from soot particles (JET A-1) generated in a pool fire at*
 294 *exact backscattering angle of π and wavelengths $\lambda_{UV} = 355$ nm (in blue) and $\lambda_{VIS} = 532$ nm (in green). The*
 295 *experimental data points are adjusted with Eq. (5), represented in full lines, to retrieve $(F_{22}/F_{11})_{lab}$ then $\delta_{lab}(\pi)$ by*
 296 *applying Eqs. (3,6).*

297
 298 *Table 1 : Evaluation of the ratio $[F_{22}(\pi)/F_{11}(\pi)]_{lab}$ and LDR $\delta_{lab}(\pi)$ of soot particles emitted from at JET A-1*
 299 *pool fire. These ratios are specific to the freshly emitted soot aggregates from JET-A1 pool fire. LDR measurement*
 300 *derived from airborne or spaceborne lidar instruments may differ as explained in [12], [15], [17].*

Wavelength (nm)	$[F_{22}(\pi)/F_{11}(\pi)]_{lab}$	LDR $\delta_{lab}(\pi)$ (%)
355	0.79 ± 0.03	11.7 ± 2.3
532	0.84 ± 0.03	8.7 ± 2.1

301
 302
 303 **4. Numerical backscattering simulations**

304
 305 **4.1 Soot fractal aggregate models and scattering matrix computation**

306
 307 In order to model the soot morphology, two shape models have been considered. As a first
 308 shape model, the aggregates morphology is described by monodisperse spheres in point
 309 contact. The morphology of soot aggregates is described using a fractal model [2], [3], and
 310 this monodisperse model is described using the following equation:
 311

$$N_m = k_f \left(\frac{R_g}{r_m} \right)^{D_f} \quad (8)$$

312 where N_m is the number of monomers (i.e. spherical primary particles), k_f is the fractal
 313 prefactor, R_g is the radius of gyration, r_m is the monomer radius and D_f is the fractal
 314 dimension.
 315

316 For **aggregates formed by polydisperse monomers**, a similar expression is used as [46] :

317

$$N_m = k_f \left(\frac{R_g}{r_p} \right)^{D_f} \quad (9)$$

318 where r_p is the monomers geometric mean radius.

319

320 Monodisperse aggregates have been generated using a tunable cluster-cluster aggregation
 321 algorithm [47], [48], allowing to predetermine the morphological parameters as defined in Eq.
 322 (8). In order to be consistent with aggregates generated through classical Diffusion-Limited
 323 Cluster Aggregation (DLCA) algorithms, we generated aggregates with a fractal dimension of
 324 $D_f = 1.8$ and a fractal prefactor of $k_f = 1.3$ [49], with varying numbers of monomers and
 325 monomer radii. Using this algorithm, the generated aggregates present a monodisperse
 326 radius size distribution. In this method, the monomer radius has been varied from $r_m = 10$ nm
 327 to $r_m = 30$ nm with steps of $\Delta R_m = 2.5$ nm. The monomer numbers have been varied from
 328 $N_m = 20$ to $N_m = 200$ with steps of $\Delta N_m = 20$. For each set of morphological parameters, 50
 329 aggregates have been generated.

330

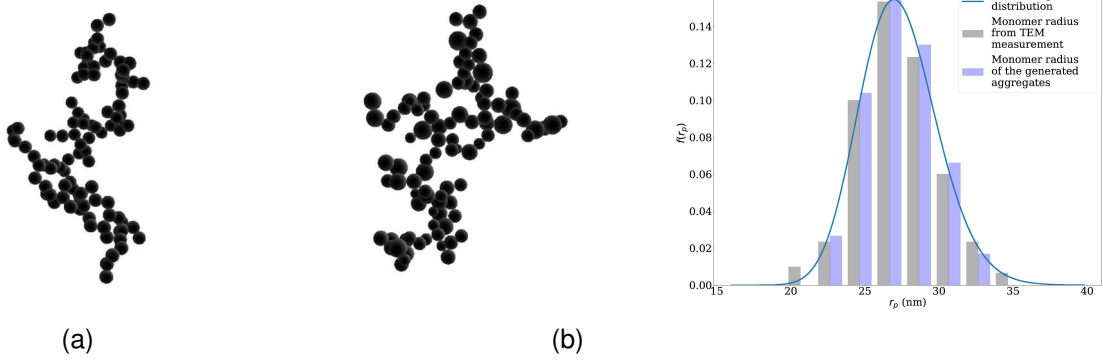
331 The second shape model is also described using a fractal model. However, the size
 332 distribution of the primary particles is accounted for, based on our TEM-laboratory
 333 observations. This second method replicates the monomer size distribution observed using
 334 the TEM measurements, in which case the aggregates have been generated using the
 335 FracVAL code [50]. This numerical program allows generating aggregates with a fixed set of
 336 morphological parameters (e.g. number of monomers, fractal dimension) **and with either a**
 337 **monodisperse or a polydisperse monomer size distribution**. The monomer radius size
 338 distribution is described by the lognormal function as defined in Eq. (7). Each aggregate has
 339 been generated with a fractal dimension of $D_f = 1.8$ and fractal prefactor $k_f = 1.3$ and a
 340 standard deviation of $\sigma = 1.1$. Moreover, the number of monomers has been varied within
 341 $N_m \in [20,200]$ with a step of $\Delta N_m = 20$, and the mean monomer radius within $r_p \in [10,30]$
 342 with steps of $\Delta r_p = 2.5$ nm. 50 aggregates **are generated for each set of morphological**
 343 **parameters and are then gathered into ensembles**.

344

345 To summarize, Figure 4 presents two examples of aggregates generated with a
 346 monodisperse (Figure 4(a)) and a polydisperse (Figure 4(b)) monomer radius size distribution,
 347 both with a number of monomers of $N_m = 100$. In Figure 4(b) is also represented the
 348 monomer radius size distribution of the aggregates generated with the FracVAL code, using
 349 the previously defined parameters.

350

351



352
 353 *Figure 4 : Examples of generated aggregate with a monodisperse size distribution (a) and a polydisperse size*
 354 *distribution (b). For the polydisperse case, the TEM-observed size distribution (in grey) is compared with that from*
 355 *numerically-generated aggregates (in blue).*
 356

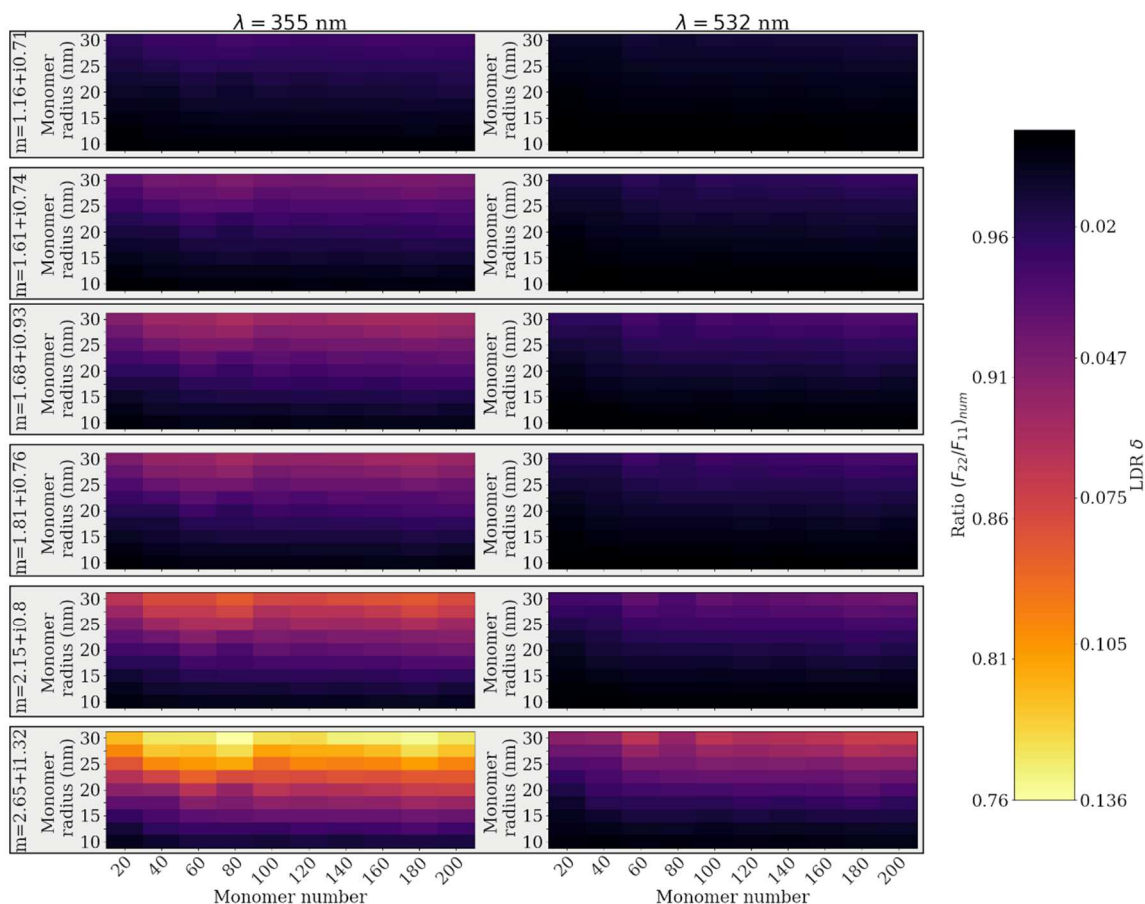
357 The Superposition T-Matrix (STM) method is here used for the computation of soot fractal
 358 aggregate scattering properties [29], [51], [52]. This method consists in expressing the
 359 scattered field by an assembly of spheres as the superposition of the scattered field by each
 360 sphere. Both incoming and outgoing field of each sphere is expressed using Vector
 361 Spherical Wave Function (VSWF). The Transition Matrix (T-Matrix), links the outgoing
 362 VSWF expansion coefficient to those of the incoming VSWF, therefore considering the
 363 coupling between the different spheres. The Multiple Sphere T-Matrix (MSTM) code [53] has
 364 been used in order to compute the scattering matrix of soot fractal aggregates. This code
 365 takes as inputs the position of each primary particles composing the aggregates, as well as
 366 their refractive index and size parameter.
 367

368 For both shape models, the **orientation-averaged** ratio $(F_{22}/F_{11})_{num}$ of each individual
 369 aggregate is computed using the MSTM code. Results are then averaged according to the set
 370 of morphological parameters used in the generation of the aggregates. Hence, in the
 371 numerical study, we report results of ensembles of aggregates with same morphological
 372 parameters and the associated numerical standard deviation. In order to compute the
 373 scattering matrix of the generated aggregates, the complex refractive index is needed. A wide
 374 range of complex refractive indices of soot particles are reported in the literature, and the
 375 refractive index of the particles generated under the conditions of the experiment described in
 376 Section 3 is unknown. In order to cover the range of possible indices, several values of the
 377 soot complex refractive index are used during the computations: $m = 1.16 + i0.71$, $m =$
 378 $1.61 + i0.74$, $m = 1.68 + i0.93$, $m = 1.81 + i0.76$, $m = 2.15 + i0.8$ and $m = 2.65 + i1.32$.
 379 These indices are used for both the UV and the VIS computation. The choice of these values
 380 is motivated by an already existing study [54] aiming, among other things, at studying the
 381 impact of the optical index on the depolarization ratio. **All computations are operated within**
 382 **the random orientation settings. Indeed, as the particles are measured in ambient air and in**
 383 **the close vicinity of the pool-fire, the airflow resulting from the combustion process may**
 384 **prevent the particles from keeping a preferred orientation during the backscattering**
 385 **experiment, especially at this size range.**
 386

387 4.2 Numerical results

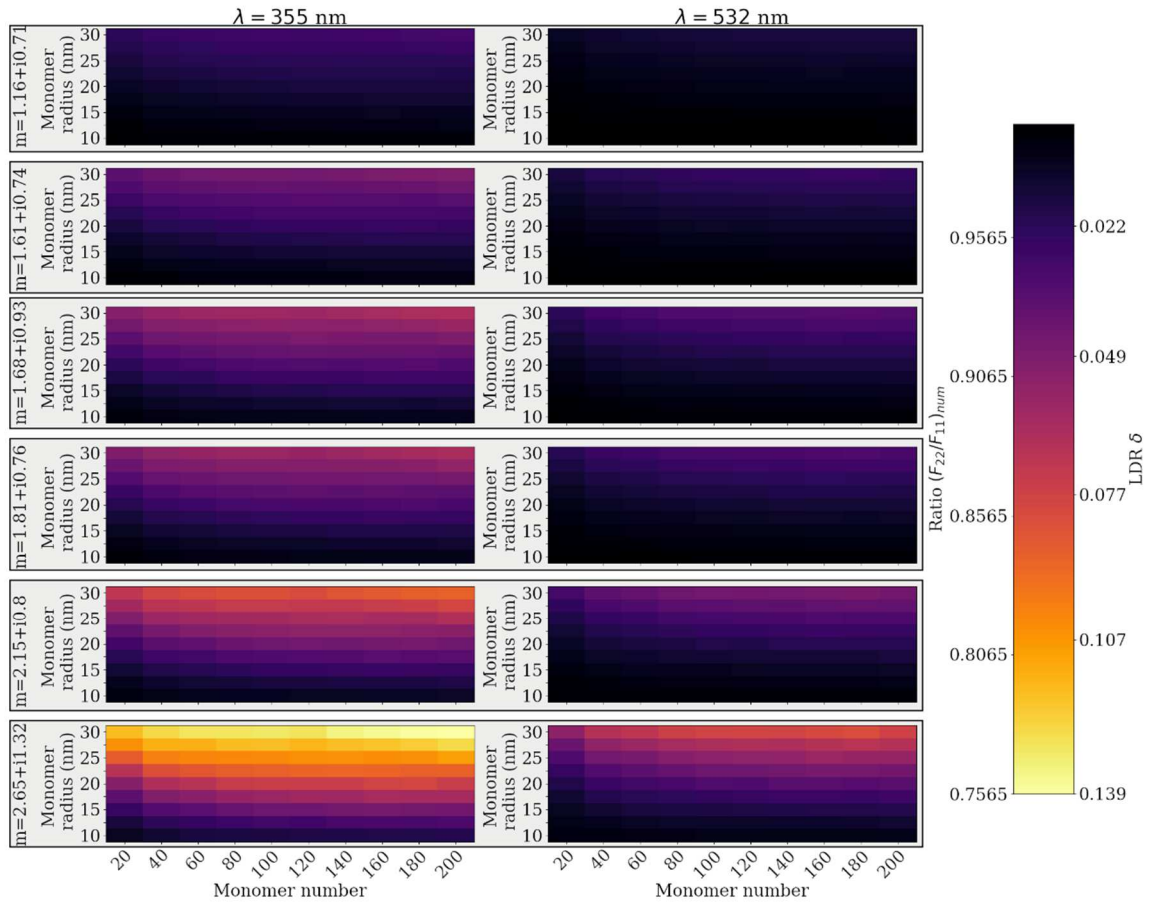
388
 389 Figure 5 presents the results of the numerical modelling of the ratio $(F_{22}/F_{11})_{num}$, together
 390 with the corresponding LDR values derived from Eq. (3) These numerical results show several
 391 trends across all subfigures. First, we can observe smaller ratios as the monomer radius is
 392 increased, which implies that soot fractal aggregates with large monomers produces larger
 393 lidar depolarization ratio at both wavelengths. A wavelength dependence of the ratio can also
 394 be observed. Indeed, at constant morphological parameters and refractive index, the LDR at

395 532 nm wavelength is lower than at 355 nm. The chosen complex refractive index also
 396 induces large variation of the $(F_{22}/F_{11})_{num}$ ratio and consequently of the LDR. Both real part
 397 and imaginary part increase induce larger LDR. Finally, in some cases, a dependence on the
 398 number of monomers can be observed. Aggregates with a number of monomers under 80
 399 show a decrease in the ratio $(F_{22}/F_{11})_{num}$ with increasing monomer number. In the other
 400 cases, the variation is less significant. These numerical results are consistent with results
 401 present in the literature as in [21], [51], [54].
 402



403
 404 *Figure 5 : Numerical results of the ratio $(F_{22}/F_{11})_{num}$ and LDR for ensembles of aggregates formed by*
 405 *monodisperse monomers. The column of subplots refers two different wavelengths, while the rows refer to different*
 406 *refractive index. Inside the figure themselves, the x-axis represents variable monomer number, and the y-axis*
 407 *represents variable monomer radius. The colour scale indicates variation in the numerically computed ratio*
 408 *$(F_{22}/F_{11})_{num}$ and LDR.*
 409

410 The numerical results for the **ensembles of aggregates formed by polydisperse**
 411 **monomers** are presented in Figure 6. As in the monodisperse case, the F_{22}/F_{11} ratio
 412 decreases as the refractive index increases and also as the monomer radius or number
 413 increases. Compared with the monodisperse results presented in Figure 5, results for the
 414 polydisperse **case** present slightly lower F_{22}/F_{11} values (*i.e.* higher LDR). In [54], a slight
 415 decrease of the LDR with polydispersity is reported. However, the author used a normal
 416 monomer radius size distribution function, in comparison with the lognormal function used in
 417 this study, which could explain the discrepancy between our results. This is further supported
 418 by the results presented in [55] using a lognormal distribution. Polydisperse results also
 419 present an almost monotonic decrease of ratio F_{22}/F_{11} with monomer number, in contrary to
 420 the monodisperse results where more variability is present. **We attribute this smoothing effect**
 421 **to the polydispersity of the monomer radii.**
 422



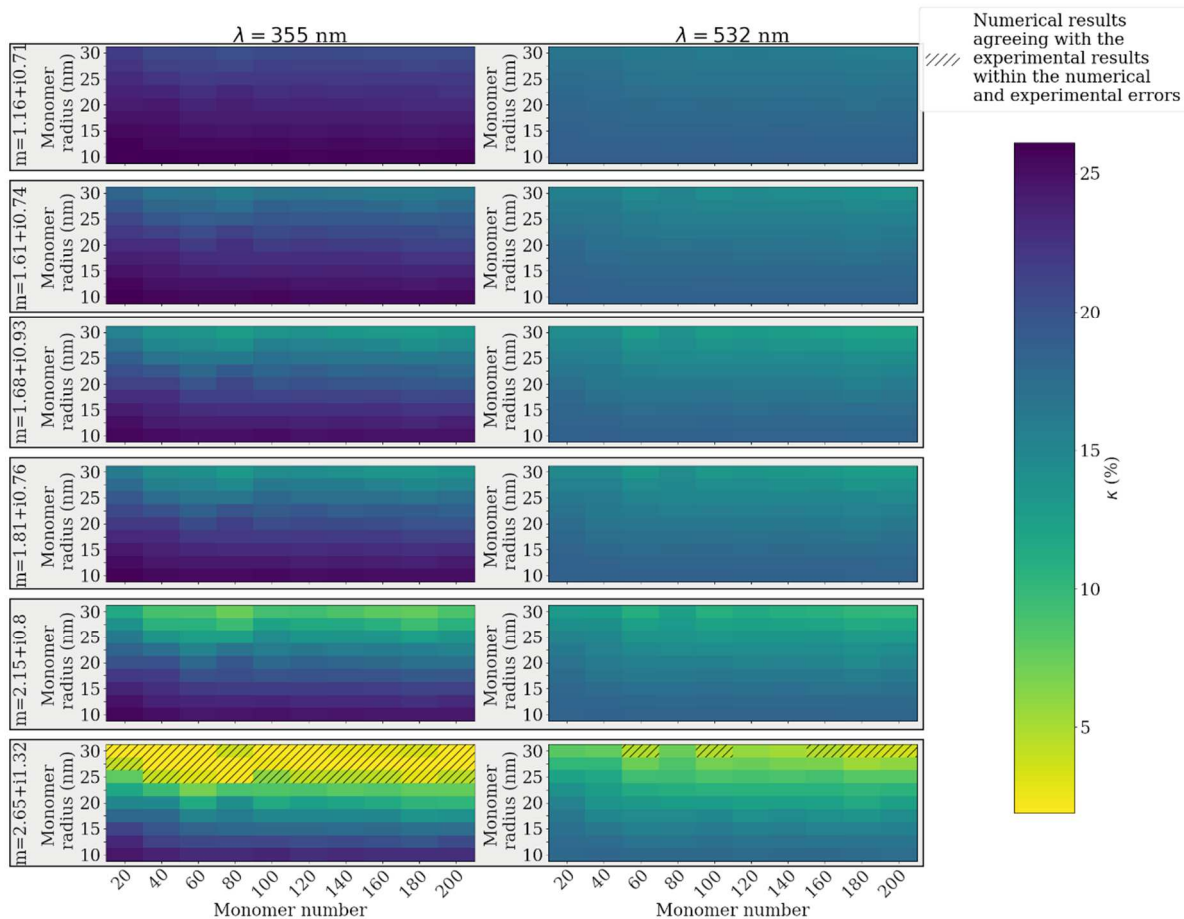
423
 424 *Figure 6 : Numerical evaluation of the ratio $(F_{22}/F_{11})_{num}$ and LDR for ensembles of aggregates formed by*
 425 *polydisperse monomers. The column of subplots refers two different wavelengths, while the rows refer to different*
 426 *refractive index. Inside the figure themselves, the x-axis represents variable monomer number, and the y-axis*
 427 *represents variable monomer radius. The colour scale indicates variation in the numerically computed ratio*
 428 *$(F_{22}/F_{11})_{num}$ and LDR.*

431 5. Discussion

432
 433 In order to assess the discrepancies between the laboratory-measured $(F_{22}/F_{11})_{lab}$ and the
 434 numerically-computed $(F_{22}/F_{11})_{num}$ ratios, we introduce the percent disagreement, defined as
 435 :

$$436 \quad \kappa = 100 * \frac{|(F_{22}/F_{11})_{num} - (F_{22}/F_{11})_{lab}|}{(F_{22}/F_{11})_{lab}} \quad (10)$$

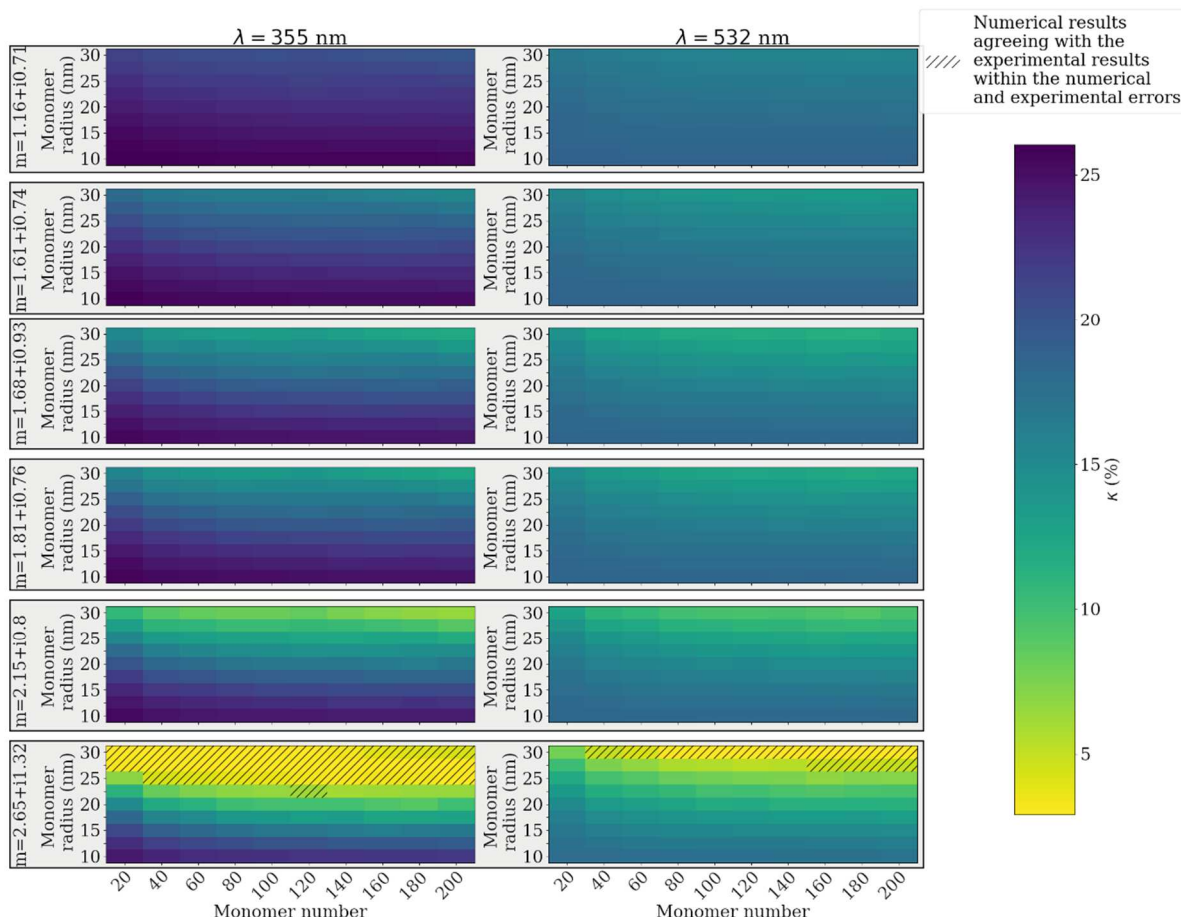
437
 438



439
 440 *Figure 7 : Discrepancy κ between the laboratory and numerical experiments for monodisperse aggregates*
 441 *(colour scale, with better agreement as the value closes to 0, in yellow). The column of subplots refers two*
 442 *different wavelengths, while the rows refer to different refractive index. Inside the figure themselves, the x-axis*
 443 *represents variable monomer number, and the y-axis represents variable monomer radius.*
 444

445 In Fig. 7 is represented the percent disagreement κ between the numerical and
 446 experimental results when monodisperse aggregates are considered. The dashed area
 447 represents the range of numerical input parameters (*i.e.* complex refractive index, monomer
 448 radius, monomer number) which reproduce the experimental results within the numerical and
 449 experimental errors. At 355 nm wavelength, we can observe one main range of agreement.
 450 Using a complex refractive index of $m = 2.65 + i1.32$, the soot aggregates with monomer
 451 radius $r_m \geq 25$ nm also show good agreement, with the number of monomers within $N_m \in$
 452 $[20,200]$. At 532 nm wavelength, the results noticeably differ. Indeed, the range of parameters
 453 that reproduces the laboratory experimental results is more reduced; only the computation
 454 using a complex refractive index of $m = 2.65 + i1.32$, with monomer radius of $r_m = 30$ nm and
 455 monomer number $N_m = 60,100$ and within $N_m \in [160,200]$ is able to reproduce the
 456 experimental results. As stated in Section 2.2, $(F_{22}/F_{11})_{lab}$ has been experimentally evaluated
 457 simultaneously at 355 and 532 nm wavelengths. This indicates that a unique ensemble of
 458 soot aggregates is responsible for the observed F_{22}/F_{11} ratio and LDR. We may then expect
 459 the agreeing numerical results to have the same morphological parameters at both
 460 wavelengths. We can then reduce the range of agreement of the morphological parameters
 461 (*i.e.* N_m and r_m) by considering only those which coincide at both 355 nm and 532 nm
 462 wavelengths. Hence, the ensemble of monomer radius $r_m = 30$ nm and monomer number
 463 $N_m = 60,100$ and within $N_m \in [160,200]$ are the only ensembles reproducing the
 464 experimental results.
 465

466 Likewise, the κ -values for the polydisperse aggregates results are presented in Fig. 8.
 467 Results are similar to those observed in the monodisperse case at 355 nm wavelength, but
 468 present differences at 532 nm wavelength. At 355 nm wavelength, results are in agreement
 469 for ensembles of monomer radius $r_p \geq 25$ nm and with monomer number within $N_m \in$
 470 $[20,200]$ with a refractive index of $m = 2.65 + i1.32$. At 532 nm wavelength, the lesser
 471 variability in the results induced by the polydispersity causes a clearer range of agreement.
 472 For a refractive index of $m = 2.65 + i1.32$, ensembles with monomer radius $r_m = 30$ nm and
 473 monomer number $N_m \in [40,200]$ are in agreement, as for ensembles with $r_p = 27.5$ and $N_m \in$
 474 $[160,200]$.
 475



476
 477 *Figure 8 : Discrepancy between the laboratory and numerical experiments for aggregates formed by*
 478 *polydisperse monomers (colour scale, with better agreement as the value closes to 0, in yellow). The column of*
 479 *subplots refers two different wavelengths, while the rows refer to different refractive index. Inside the figure*
 480 *themselves, the x-axis represents variable monomer number, and the y-axis represents variable monomer radius.*
 481

482 During the numerical study, we identified several ensembles which backscattering properties
 483 are in agreement with the laboratory results. The monomer radius of these ensembles is
 484 consistent with results from the TEM-measurement. The number of monomers is also in a
 485 realistic range. Although the numerical results coincide with the experimental results in these
 486 cases, the refractive index used remains very large. Indeed, the complex indices correspond
 487 to spark-generated soot [56], which microstructure is composed almost exclusively of
 488 amorphous carbon [57]. In comparison, the soot aggregates emitted during our experiment
 489 clearly exhibits an onion-like structure. Considering that the inclusion of monomer radius
 490 polydispersity did not significantly improve the agreement between the numerical and
 491 experimental results, this strongly suggests that the morphological models used in this study
 492 are in need of further refinement in order to better interpret the laboratory experimental

493 measurements. Several morphological effects can be considered in order to do so. Particles
494 overlapping and necking effect (*i.e.* supplementary material at the monomers surface), could
495 produce significant changes of the lidar depolarization ratio. As described in [54], particle
496 overlapping has an effect similar to a decrease in monomer number, while necking produces
497 a scaling effect on the LDR in the forward direction. Still, interrogation remains whether these
498 morphological parameters also produce similar results in the backward scattering direction,
499 and in which proportion. To account for these specific morphological effects, the STM method
500 can't be used during the scattering properties computation. Hence, methods with higher
501 computational cost are required, such as DDA methods, which would strongly reduce the
502 considered number of aggregates realization per ensembles and range of parameters.
503 Spheroidal monomers can also be considered in order to better replicate soot morphology.
504 Wu et al. [58] investigated the effects of both prolate and oblate spheroid monomers on the
505 scattering properties of soot aggregates, and showed a decrease in the F_{22}/F_{11} ratio in the
506 backward scattering direction. This morphology type also requires DDA calculation, as the
507 STM method cannot be used for spheroidal monomers currently. Another possible
508 explanation might involve more complex fractal structures such as soot super-aggregates,
509 which have been recently reported in both pool fires [41] and laminar diffusion flames [59],
510 [60] with large depolarization properties [61]. The multiple scattering effect might also be
511 responsible for an increase in the measured LDR. In dense medium composed of spherical
512 droplets, this effect can be responsible for non-zero values of the LDR [62], whereas in the
513 single scattering framework, spherical particles do not depolarize light in the backward
514 scattering direction [39].

515

516 6. Conclusion

517

518 In this paper, the LDR of freshly emitted soot particles is evaluated at both 355 and 532 nm
519 wavelengths in a controlled-laboratory experiment operating in the exact backward scattering
520 angle of 180.0° . This laboratory experimental set-up allows accurate evaluations of the ratio
521 $F_{22}(\pi)/F_{11}(\pi)$, which in turn leads to the evaluation of the corresponding LDR. We report
522 values of $\delta_{UV} = (11.7 \pm 2.3) \%$ at wavelength $\lambda_{UV} = 355$ nm and to $\delta_{VIS} = (8.7 \pm 2.1) \%$ at
523 wavelength $\lambda_{VIS} = 532$ nm, for soot particles generated from a pool fire of JET A-1 in
524 laboratory ambient air. In complement, a numerical study is associated to this laboratory
525 measurement. **Considering the TEM measurements, the structure of the soot aggregates is**
526 **described by a fractal-like model.** Two morphological models for freshly emitted soot
527 aggregates are used to simulate the soot aggregates structure. Using these morphologies,
528 the scattering properties are computed with the MSTM code. *We find that the morphological*
529 *model of aggregates formed by polydisperse monomers provides an overall better*
530 *performance than the monodisperse monomers model. Moreover, in the polydisperse case,*
531 *the laboratory findings are reproduced at both wavelengths only with the highest considered*
532 *refractive index ($m = 2.65 + i1.32$) and for monomer number $N_m \geq 40$ with mean monomer*
533 *radius of $r_p = 30$ nm ($N_m > 160$ at $r_p = 27.5$ nm).* This supports the use of more realistic
534 morphological models when computing soot aggregates backscattering properties **and the**
535 **independent measurement of the complex refractive index of soot particles, at the aggregate**
536 **and monomer scale. As we do not have direct evidence of the random orientation of the**
537 **particles, complementary experiments should be performed to tackle this specific issue,**
538 **which is however beyond the scope of this paper.** Likewise, the laboratory experiment may
539 be further improved by analyzing other soot particle aggregates, **and by evaluating other**
540 **radiative properties such as the asymmetry parameter or the single-scattering albedo.** This
541 work is however beyond the scope of this contribution. Still as is, the lidar remote sensing
542 community may use our contribution to better interpret polarimetric lidar signals of freshly
543 emitted soot aggregates since, for the first time to our knowledge, accurate laboratory-
544 measured LDR-values of freshly emitted soot aggregates are provided at exact
545 backscattering angle of 180.0° , supported by STM-light backscattering computations.

546

547
548

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550

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553

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555

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