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Number of independent measurements required to obtain reliable mean scattering properties of irregular particles having a small size parameter, using microwave analogy measurements

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Abstract. Laboratory measurements of light scattered by a cloud of randomly oriented levitating particles are often used to interpret remote sensing measurements of dust in space and in Earth's atmosphere. It is necessary to know how many particles or how many different orientations of the same particles must be considered to retrieve the mean scattering function of brightness and polarization. New laboratory measurements were conducted using the microwave analogy method between frequencies of 3 to 18 GHz, where an "analog" particle with a small size parameter in a range of 0.5-12 will have a size of several cm. Twelve such "analog" particles from compact shapes to aggregates with small fractal dimensions were fabricated by additive manufacturing (3D printing) and were studied. The number of necessary measurements to reach the mean scattering properties of a particle with an accuracy of about 5% is obtained for less than 20 different orientations. To reach a 1.5% ($1-\sigma$) error in brightness and a 0.5% ($1-\sigma$) error in polarization, the number of necessary measurements is in a range of 20 to 70, depending on the shape, fluffiness, deviation from a perfect sphere, and surface irregularities of the particle. These results show that several tens of randomly oriented particles of the same size are sufficient to retrieve the mean light scattering properties. Also, several tens of orientations of the same particles provide mean scattering properties, compared to modelling calculations using the Finite Element Method, for an aggregate composed of identical monomers.

1. Introduction

Laboratory measurements of light scattered by a cloud of irregular particles are necessary to interpret the brightness and polarization remote sensing measurements of comets, interplanetary dust cloud, and circumstellar material orbiting stars, e.g., protoplanetary and debris disks. Such comparisons are conducted to retrieve bulk physical properties of particles such as composition, size distribution, albedo, and porosity [1-6]. The analyses often assume that the observed particles are randomly oriented inside the cloud, as well as during the reference laboratory measurements. On the opposite, the possible orientation of grains, like in a gas flow or in case of strong magnetic field [7], can modify the scattering properties of an ensemble of particles. Such changes have been shown by [8] during laboratory measurements where the particles were carried and aligned by an airflow, and thus are not considered in this work.

47 Several databases of laboratory measurements provide optical properties (phase or
48 scattering functions in intensity and in polarization) of levitating particles having the same size
49 and composition, obtained by different techniques at ground or during microgravity
50 conditions [9-14]. Such experiments, for particles having small to large size parameters ($\pi D/\lambda$,
51 where D is the diameter and λ is the wavelength), assume that indeed they are randomly
52 oriented and that enough particles are considered during the measurements, from tens to
53 hundreds or thousands, to retrieve the mean optical properties. As an example, 54 different
54 orientations of the same millimetre-sized particle have been considered by [5] to retrieve the
55 mean scattering properties. Nevertheless, the authors have not quantified in detail how many
56 particles with different orientations are needed to achieve such conditions. Similarly, aerosol
57 counters used in the atmosphere to count and determine the size distribution of the solid
58 particles empirically consider their effective optical properties [16-18]. This approach works
59 well for the submicron particles that are often in large concentrations (more than tens of
60 particles cm^{-3}) but some uncertainties in size determination can arise when the concentrations
61 are too low because not enough particles are detected. Finally, experiments and numerical
62 calculations that consider individual irregularly shaped particle need also to know how many
63 different orientations are needed to establish the mean optical properties.

64 Although theoretical approaches have been proposed [15], experimental approach can
65 also be conducted to determine how many particles randomly oriented, or how many
66 different orientations of the same particle, are indeed necessary to access the mean scattering
67 properties reliably. This approach is difficult to conduct for particles with a small size
68 parameter typically in the range of 0.5-12, which corresponds to particles between 0.1 to 2.5
69 μm size in the visible domain. Such particles levitated by an air draught or during microgravity
70 conditions could aggregate, preventing to sustain a cloud of individual particles.

71 Instead of using real particles, an alternative consists in using the microwave analogy
72 [19-24] and individual particles that can be easily manipulated to control their shape and
73 orientations. The aim will be to measure the properties of a single particle at numerous
74 orientations to figure out its mean properties to mimic an ensemble of specific particles that
75 would be randomly oriented. With that concept, particles of the order of 1 μm in the visible
76 domain are of several cm when studied at a frequency of several GHz, although the size
77 parameter is conserved. Many electromagnetic scattering measurements can be conducted
78 with a single particle by changing only its orientation with respect to the incident radiation
79 beam. Such particles can be built in various materials using a 3-D printer. The permittivity of
80 a given material, or its (complex) refractive index, which is the square root of the (complex)
81 permittivity, can be chosen to reproduce the optical properties of real particles that can be
82 found in space and in Earth's atmosphere.

83
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85 **2. Conditions of microwave analogy measurements**

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87 The measurements are conducted in the anechoic chamber at the "Centre Commun
88 de Ressources en Microondes" (CCRM, Institut Fresnel, Marseille, France). Features and
89 performances are given in [25]. Briefly, the device operates for this study at 16 frequencies
90 from 3 to 18 GHz. The use of 16 frequencies from 3 to 18 GHz allows us to explore a range of
91 size parameters that vary by a factor of six for the same particle, with a refractive index that
92 can be considered almost constant for the chosen materials. The angle between the emitter
93 and receiver antennas is changed from one measurement to another, by steps of 2° , to obtain

94 the scattering function in scattering angles from -130° to 130° . The sample, mounted at the
 95 centre of the device, can be rotated after one session of measurements, to start a new session
 96 with another orientation. For the present study, measurements with 36 different orientations
 97 uniformly distributed from -180° to $+170^\circ$ were conducted by a rotation along the vertical axis
 98 in steps of 10° . The particles are placed on a polystyrene mast; then the rotation axis is vertical,
 99 *i.e.* parallel to the mast. The particles are placed first according to their greatest length
 100 (horizontally), then to their smallest length (vertically), and at about 45° (slanted) when more
 101 measurements are necessary (named inclinations in the following).

102 The device records the amplitude and the phase of the perpendicular and parallel
 103 scattered electric fields. The polarized scattering intensity I_{perp} and I_{par} with respect to the
 104 scattering plane (also so-called *S* and *P*), are the square of the amplitudes, and are used to
 105 retrieve the scattering (or phase) function for the intensity *I* and for the polarisation *P*
 106 (formulas 1 and 2).

107

$$108 \quad I = I_{\text{perp}} + I_{\text{par}} \quad (1)$$

$$109 \quad P (\%) = 100 \times (I_{\text{perp}} - I_{\text{par}}) / (I_{\text{perp}} + I_{\text{par}}) \quad (2)$$

110

111 Twelve different samples having a large variety of size, shapes, porosities and fractal
 112 dimensions have been studied, which can be representative of the main particle families that
 113 can be found in space and Earth's atmosphere. Since particles with different sizes are
 114 considered, the present study covers a size parameter range of 0.5 -12. Figure 1 presents the
 115 shape of the particles for which some physical parameters are given in table 1. The definition
 116 of the diameter is not easy in case of fluffy particles and particles with low fractal dimension,
 117 thus we provide both the gyration diameter and the equivalent diameter calculated from
 118 mean projected surface area. The size parameter is calculated from the gyration diameter for
 119 the fractal particles (AgC series except AgC181, and SootAcry) and from the equivalent
 120 diameter for the compact particles (AgC181, gravels and rough spheres).

121 For the AgC232, AgC233, AgC185, AgC186 and AgC187, the diameter of the monomer
 122 is 5.5 nm. The "SootAcryl" sample is based on a numerically generated analogue of a soot
 123 aggregate mimicking necking and overlapping phenomenon [26]. The rough spheres are
 124 derived from a perfect sphere meshed with triangles, and then perturbed modifying the
 125 distance $d(\text{OV}_i)$, with a random perturbation:

126

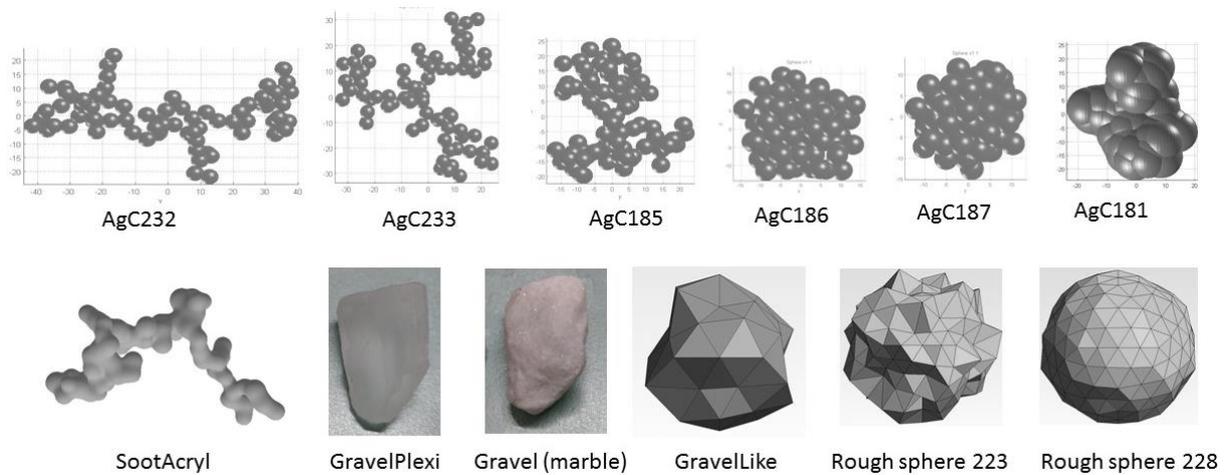
$$127 \quad d(\text{OV}_i) = R + a \cdot \text{rand}(i) \quad (3)$$

128

129 where $d(\text{OV}_i)$ is the distance from the center of the sphere *O* to the vertex *V_i* of the triangles,
 130 *rand* is a random number between -1 and 1, and *R* is the radius of the original sphere (16.25
 131 mm). For the "Rough Sphere 233" $a = 3.75$ mm, and for the "Rough Sphere 228" $a = 0.75$ mm.
 132 The roughness of the sphere and the deviation from sphericity can be estimated by the ratio
 133 D_i/D_v , where D_v corresponds to the diameter of the smaller compact sphere containing the
 134 same amount of material as the particle and D_i corresponds of the diameter of the sphere
 135 containing the particle.

136 All the particles are made by additive manufacturing using an acrylate-based resin,
 137 except the two gravels that are made of Plexiglas and marble (having similar shapes). The
 138 mean refractive index of the particles made of acrylate was measured to be $1.7 + 0.03i$ (which
 139 can be representative of some carbonaceous particles [27] but also of astrosilicate particles
 140 [28]), the refractive index of the Plexiglas is of $1.6 + 0.01i$, and the refractive index of the

141 marble is assumed to be of $2.9 + 0.2i$ for all the frequencies (which is representative of strongly
 142 optically absorbent particles).
 143



144
 145 *Figure 1: The studied samples (not on the same scale, see table 1 for the size of the samples)*
 146

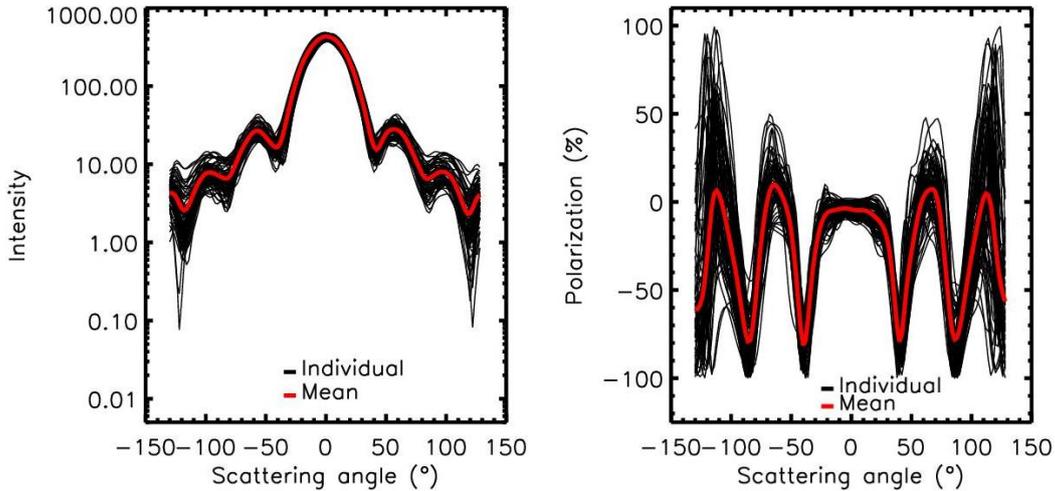
Sample	Box containing the particle (mm ³)	Gyration diameter (mm)	Equivalent diameter (mm)	Fractal dimension	Dv	Di	Di/Dv
AgC232	76 x 74 x 62	64	39	1.5	23.0	76	3.3
AgC233	61 x 39 x 67	48	37	1.7	22.9	67	2.9
AgC185	41 x 43 x 49	34	34	2.0	23.0	49	2.1
AgC186	34 x 35 x 34	23	31	2.5	23.0	34	1.5
AgC187	29 x 29 x 29	20	27	2.8	23.0	29	1.3
AgC181	46 x 28 x 50	-	40	-	35.9	50	1.4
SootAcryl	66 x 48 x 22	41	32	1.8	22.7	68.3	3.0
GravelPlexi	14 x 20 x 18	-	17	-	14.8	24.6	1.7
Gravel(marble)	14 x 20 x 18	-	17	-	17.0	25.3	1.5
GravelLike	32 x 34 x 38		32		30.6	38	1.2
Rough sphere 223	40 x 40 x 40	-	36	-	31.9	40	1.3
Rough sphere 228	34 x 34 x 34	-	34	-	32.2	34	1.05

147 *Table 1: Physical parameters of the samples. The gyration diameters and the fractal*
 148 *idimensions are calculated assuming that the spheres are not interpenetrated; the Dv*
 149 *corresponds to diameter of the smaller compact sphere containing the same amount of*
 150 *material as the particle; Di corresponds to the diameter of the sphere containing the particle*
 151

152
 153 **3. Method of measurement analysis**

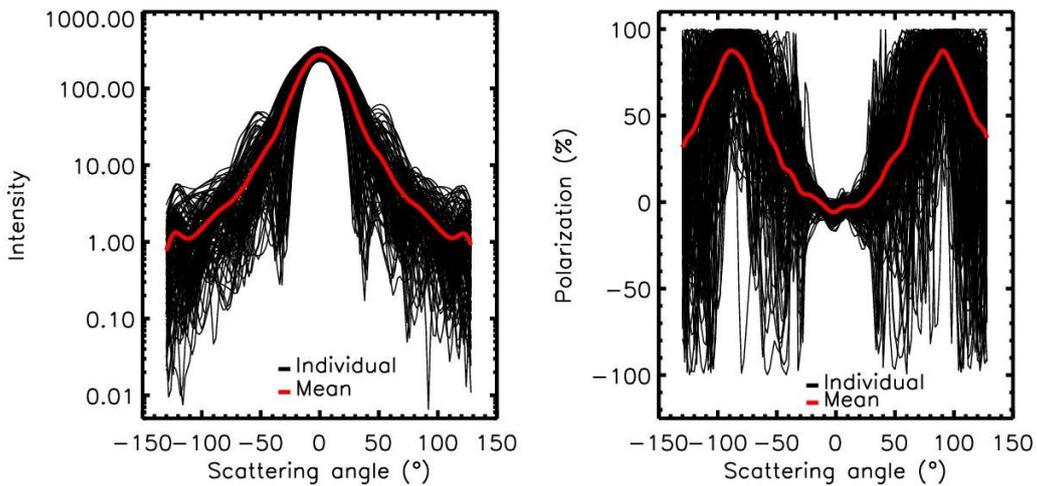
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Individual measurements of the scattering function (intensity and polarization) can exhibit strong oscillations; these oscillations are stronger as the particles are highly irregular. Figures 2 and 3 show examples of such oscillations for the rough sphere 223 and for the AgC185 at a frequency of 12 GHz, which corresponds to a size parameter of ~ 4.5 . The amplitude of the dispersion for intensity at large scattering angles is of the order of 10 for the rough sphere while the amplitude dispersion can reach a factor of 100 for the AgC185. For the polarization, the dispersion can reach at some angles the minimal and maximal possible values (-100 and 100%).



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Figure 2: Individual scattering functions for the rough sphere 223 (left: intensity; right: polarization) for the individual measurements at a given orientation and the mean of all the scattering functions



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Figure 3: Individual scattering functions for the AgC185 (left: intensity; right: polarization) for the individual measurements at a given orientation and the mean of all the scattering functions

176 To determine how many measurements are necessary to retrieve a mean scattering
177 function for intensity and polarization with a given accuracy, tens of individual measurements
178 with different orientations must be considered. The procedure is different for intensity and
179 polarization because intensity is directly retrieved from the sum of polarized measurements
180 while polarization is a relative parameter always retrieved from the ratio between the
181 difference and the sum of polarized intensity measurements and is given in a range from -
182 100% to 100%.

183 For the intensity, we calculated the mean scattering functions using a given number of
184 measurements with different orientations. At each scattering angle θ we calculate the
185 difference $Db(N, \theta)$ between the mean value of the intensity obtained for N measurements
186 and the mean value obtained for N-1 measurements, divided by the mean value of the
187 intensity for N measurements; this division act as a normalization to be able to consider
188 simultaneously the results coming for all scattering angles. Then for each value of N, we
189 calculate the standard deviation (hereafter called SDB(N)) of the normalized intensity
190 differences obtained for all scattering angles. This standard deviation, which can represent
191 the $1-\sigma$ uncertainty of the resulting mean intensity values, must decrease while the number
192 of measurements increases. This procedure can be expressed as follows:

193

$$194 \quad Db(N, \theta) = (\sum_1^N B(\theta) - \sum_1^{N-1} B(\theta)) / \sum_1^N B(\theta) \quad (4)$$

$$195 \quad SDB(N) = \sigma(Db(N, \theta)) \quad (5)$$

196

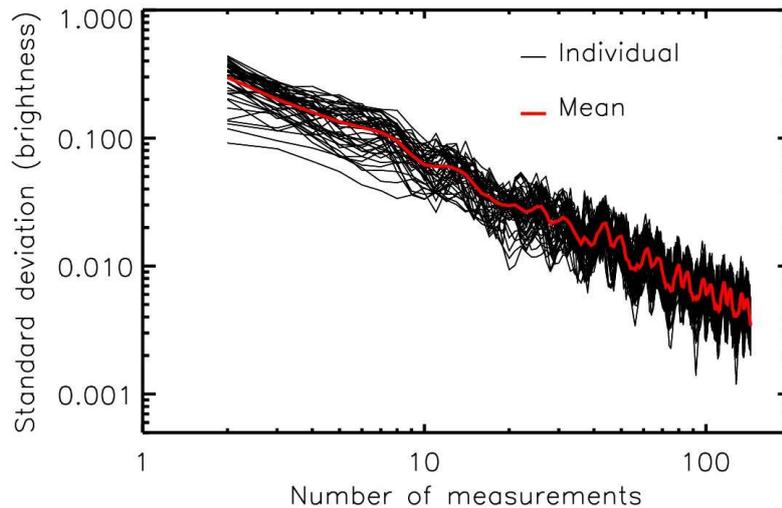
197 with N the number of measurements, B the intensity and θ the scattering angle.

198 To ensure a random distribution of the orientation when calculating the $Db(N, \theta)$
199 parameter, the calculation is conducted for different combinations on the angular orientation
200 and the inclination of the sample; then the results for SDB(N) are averaged. The choice of the
201 first orientation and the selection of the following orientations (randomly distributed or
202 considering increasing angles) do not statistically change the results for the number of
203 necessary measurements that remains within a $\pm 15\%$ uncertainty.

204 As a first step to visualise the sensitivity to the number of measurements, Figure 4
205 presents the evolution of the standard deviation with the number of measurements (the plot
206 is based on the rotation of the particles around different axes). The frequencies here
207 considered are above the Rayleigh scattering regime (size parameter greater than about 2.5
208 [29]) and different combinations of orientations and inclinations of the AgC185 sample. In log-
209 log scale, the trend is almost linear, indicating that it could be fitted by a power law that will
210 depend on the studied sample (exponent typically in the -0.8- 1.1 range).

211 Three different domains can be defined; firstly, the standard deviation decreases
212 strongly to reach an accuracy of 0.05 (*i.e.* 5%) for about 15 different orientations, secondly the
213 standard deviation slowly decreases for a number of measurements up to about 70 producing
214 a mean intensity uncertainty of about 0.01 (*i.e.* 1%), and thirdly the standard deviation slightly
215 decreases producing a mean intensity uncertainty below 0.01 (*i.e.* 1%). Increasing the number
216 of measurements will reduce again the uncertainties, but such approach does not seem
217 necessary since we can admit that a $1-\sigma$ accuracy of the order of 1% is sufficient for the
218 interpretation of the intensity measurements.

219



220
 221 *Figure 4: Evolution of the standard deviation with the number of measurements for the*
 222 *intensity of the AgC185 sample; the black lines represent the results for the different*
 223 *frequencies and the different combinations of orientations and inclinations; the red line*
 224 *represents the mean value*

225
 226
 227 For the polarization, the sums of the N orthogonal and parallel polarized scattering
 228 components are first calculated, then the polarization is calculated. The $D_p(N, \theta)$ parameter
 229 is calculated by the difference between the polarization obtained for N measurements and
 230 the polarization obtained for N-1 measurements. Finally, the SDP(N) parameter corresponds
 231 to the standard deviation of $D_p(N, \theta)$ for all scattering angles, which corresponds to the
 232 polarization uncertainty at 1- σ in absolute value (%). This procedure can be expressed as
 233 follows:

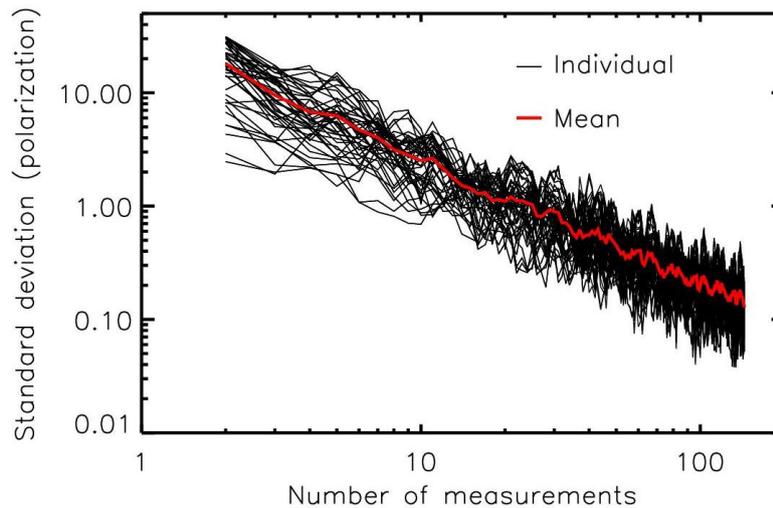
234
 235
$$P(N, \theta) = 100 \cdot (\sum_1^N I_{\text{perp}}(\theta) - \sum_1^N I_{\text{par}}(\theta)) / (\sum_1^N I_{\text{per}}(\theta) + \sum_1^N I_{\text{par}}(\theta)) \quad (6)$$

236
$$D_p(N, \theta) = P(N, \theta) - P(N - 1, \theta) \quad (7)$$

237
$$\text{SDP}(N) = \sigma(D_p(N, \theta)) \quad (8)$$

238
 239 Similar trends as those for the intensity are obtained for the polarization
 240 measurements, as shown in Figure 5 for the AgC185 sample. A 5% uncertainty is obtained for
 241 about 10 orientations, while about 50 different orientations are necessary to achieve an
 242 uncertainty of about 0.5% (1- σ); such last value is of the order of the error bars obtained for
 243 some laboratory measurements with real particles [1,13]

244



245 *Figure 5: Evolution of the standard deviation for the polarization with the number of*
 246 *measurements of the AgC185 sample; the black lines represent the results for the different*
 247 *frequencies and the different combinations of orientations and inclinations; the red line*
 248 *represents the mean value*
 249

252 4. Mean scattering functions and evolution with the size parameter

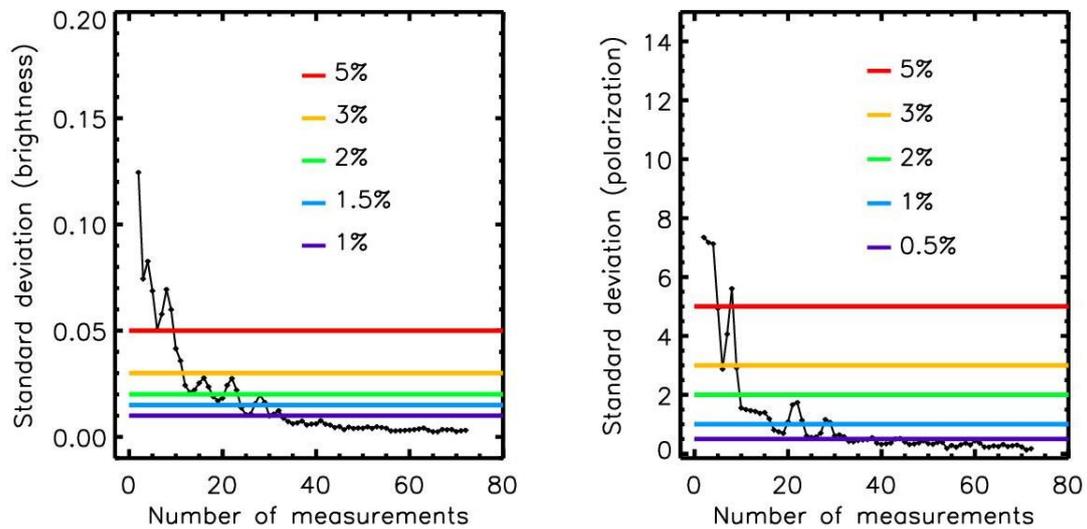
254 4.1 Measurements

255
 256 For a perfect sphere, the scattering properties can be easily retrieved using Mie
 257 scattering calculations and there is no need for several different orientations of the particle
 258 When the roughness and/or the deviation from sphericity of the particle increases, increasing
 259 the number of measurements with different orientations of the particle is needed to retrieve
 260 the mean properties.

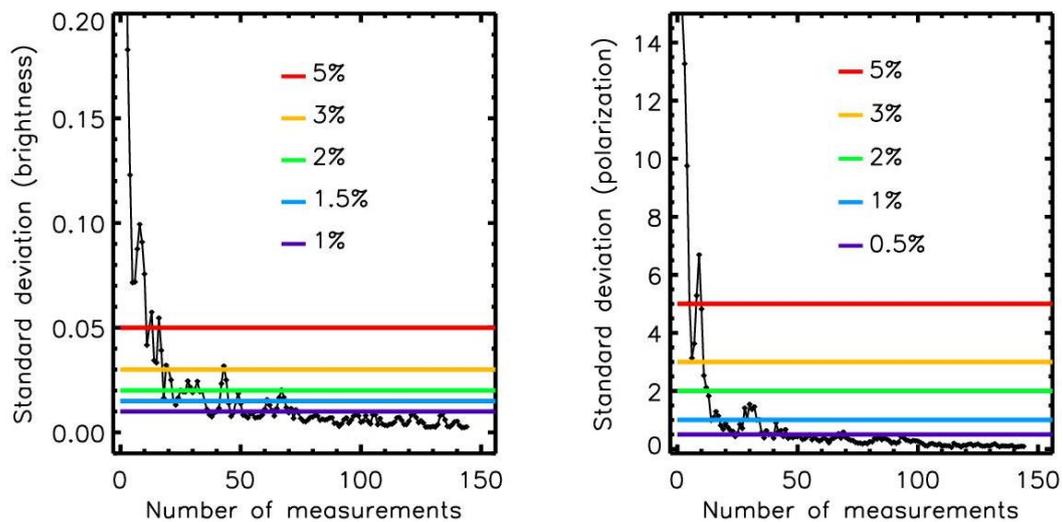
261 Previous results were calculated for all frequencies above the Rayleigh regime. It is of
 262 interest to conduct the studies at each frequency to retrieve the evolution of the standard
 263 deviation with the particles' size parameter. For each sample and for all frequencies, the
 264 number of necessary measurements where the standard deviation of the intensity drops
 265 below 5%, 3%, 2% 1.5% and 1% is searched for. Similarly, the number of necessary
 266 measurements where the standard deviation of the polarization drops below 5%, 3%, 2%, 1%
 267 and 0.5% is searched for. The lower values of 1% for intensity and 0.5 % for polarization are
 268 arbitrarily chosen but they can correspond to the typical accuracies often achieved when
 269 performing laboratory measurements with levitating particles in the visible spectral domain.
 270 All the results for the number of necessary measurements obtained for the different sample
 271 inclinations (horizontally, vertically, slanted) remains in the $\pm 15\%$ range for all the standard
 272 deviation thresholds presented above.

273 Figures 6 and 7 present examples of the evolution of the standard deviations with
 274 number of measurements for the rough sphere 223 and the AgC185, having a size parameter
 275 of 4.5. In these examples, the 1% threshold for intensity is reached for about 35 ± 5 and 100 ± 15
 276 measurements for the rough sphere 223 and the AgC185, respectively. These values decrease
 277 to 30 ± 5 and 70 ± 10 for the 1.5% threshold. The 0.5% threshold for the polarization is reached
 278 for 40 ± 5 and 70 ± 10 for the rough sphere 223 and the AgC185, respectively. These first results
 279 indicate that the number of necessary measurements to retrieve the mean optical properties

280 (hereafter called necessary measurements) is higher in the case of a non-compact particle
 281 compared to a compact one.
 282

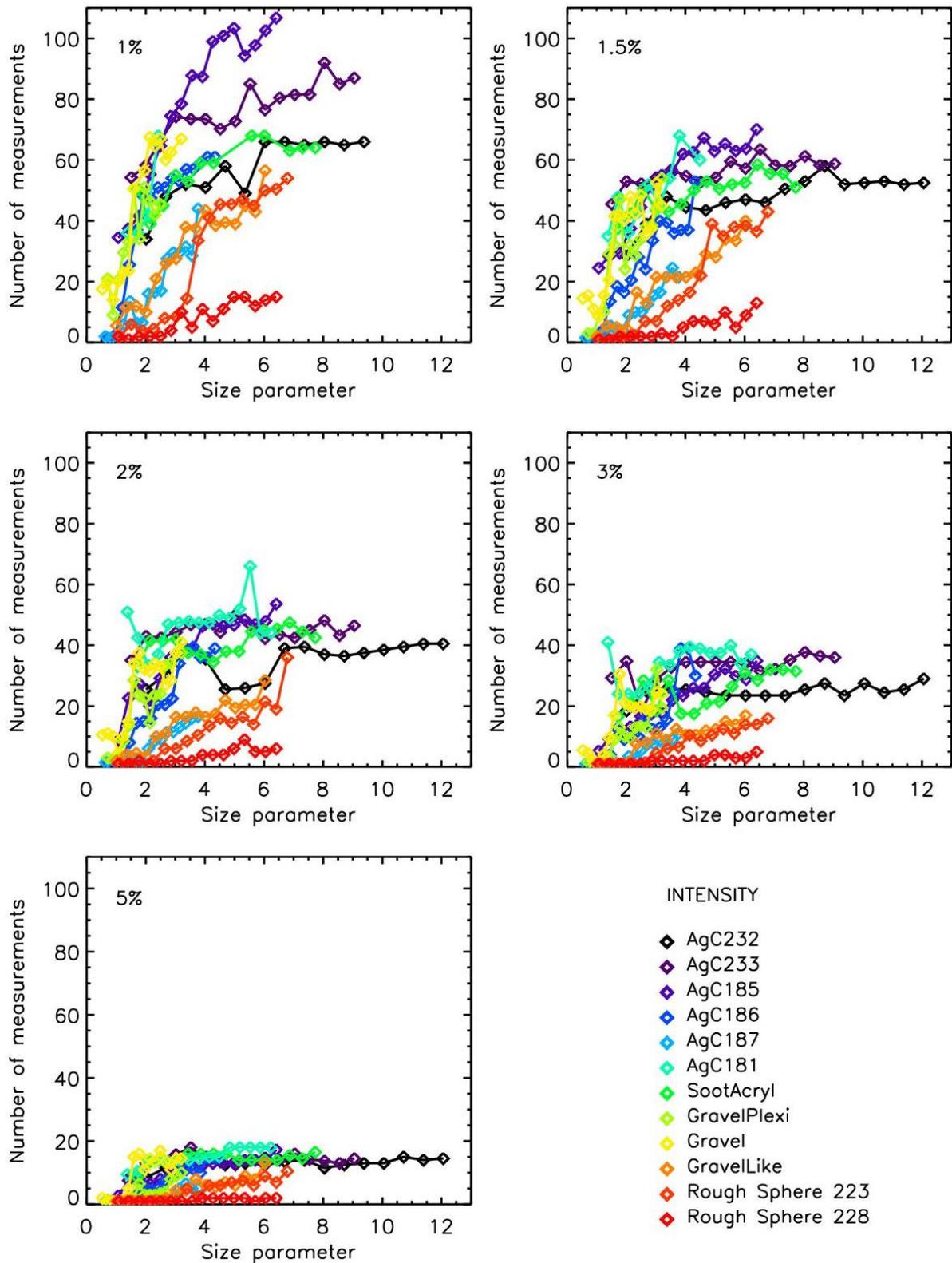


283
 284 *Figure 6: Example of the evolution of the standard deviation for the intensity and polarization*
 285 *with the number of measurements for the rough sphere 223, at a size parameter of 4.5.*
 286



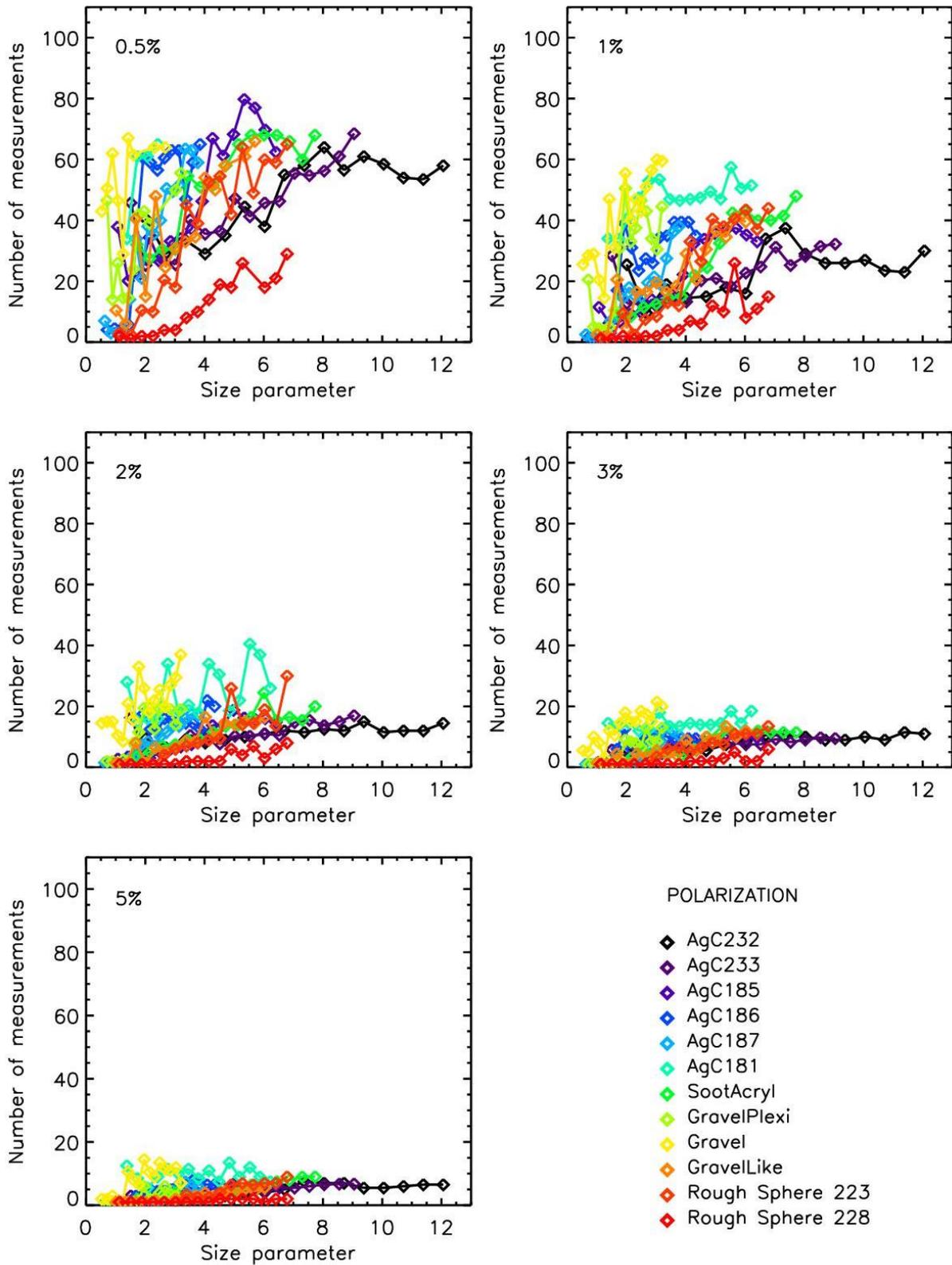
287
 288 *Figure 7: Example of evolution of the standard deviation for the intensity and polarization*
 289 *with the number of measurements for the AgC185, at a size parameter of 4.5.*
 290

291
 292 The evolution of the number of necessary measurements with size parameter for all
 293 the samples is presented in Figure 8 for the intensity and in Figure 9 for the polarization,
 294 considering the different levels of expected accuracy. Obviously, the number of necessary
 295 measurements is lower when the uncertainties is higher, and less than 20 orientations are
 296 necessary to retrieve the mean intensity and polarization phase curve with an accuracy of 5%.
 297 It also decreases with decreasing size parameter; the decrease is steeper for a size parameter
 298 below 3, corresponding to size particles below the wavelength, and is almost stable for a size
 299 parameter greater than 5. The number converges towards only one necessary measurement
 300 when the measurements are performed in the Rayleigh scattering domain.
 301



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Figure 8: Evolution of the number of necessary measurements with size parameters, to retrieve the mean intensity scattering properties, for different thresholds of uncertainties (the error bars of $\pm 15\%$ for the number of necessary measurements are omitted for clarity).



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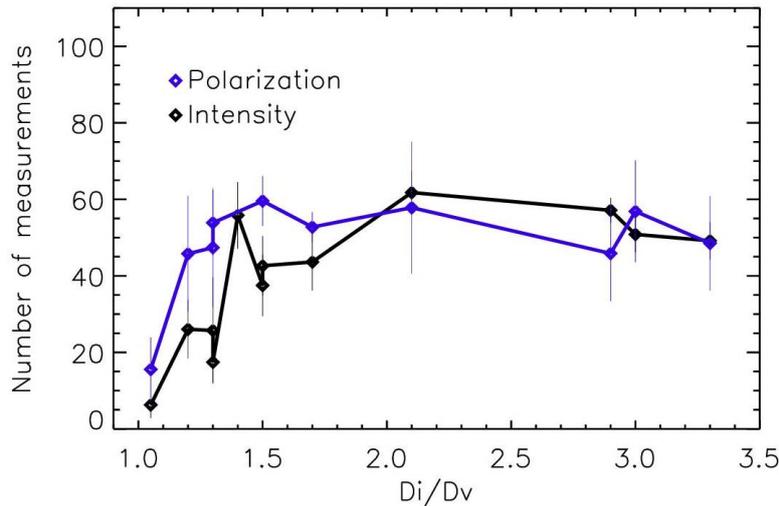
Figure 9: Evolution of the number of necessary measurements with size parameters, to retrieve the mean polarization scattering properties, for different thresholds of uncertainties (the error bars of $\pm 15\%$ for the number of necessary measurements are omitted for clarity).

315 The evolution of the necessary number of measurements to reach a given accuracy
316 strongly depends on the nature of the particles. As expected, the lower numbers (around 20
317 for intensity measurements) are obtained for the near-perfect compact sphere rough sphere
318 228, having rugosity in the $[\lambda/6, \lambda/33]$ range. Then, the value increases as the surface of the
319 particles become more irregular (rough sphere 223, GravelLike and AgC187) and the shape
320 departs from a sphere. The rough sphere 223 has a particular behaviour, starting from values
321 close to those of the rough sphere 228 for the smallest size parameters and reaching values
322 of more irregular particles for the largest size parameters. This can be understood when
323 considering that roughness depends on the frequencies in the $[\lambda, \lambda/7]$ range. The other
324 compact particles (Gravel, GravelPlexi, AgC181) and fractal particles (AgC186, SootAcryl,
325 Agc185, Agc232, AgC233) exhibits similar behaviour with the number of necessary
326 measurements in the 40-60 range (mean of 50) for intensity measurements. This
327 categorization for the particles is less obvious when considering the polarization, where only
328 the rough sphere 228 presents a behaviour that stands out from the other sample.

329 For non-compact particles composed of aggregated monomers, regarding the mean
330 scattering function convergence and the dispersion of the individual scattering function, no direct
331 dependency of the number of necessary measurements with the fractal dimension is detected. Also,
332 the results for the compact Gravel and the GravelPlexi particles are similar, which indicates
333 that for our cases the refractive index has no effect on the number of necessary
334 measurements when the imaginary part of the index indicates significant absorbing
335 properties. For compact particles, the departure from the sphericity reduces the amplitude of
336 Mie oscillations but has little effect on the number of necessary measurements. It seems that
337 the surface roughness, or the size of the irregularities with respect to the wavelength, is the
338 main parameter that drives the necessary number of measurements to retrieve the mean
339 scattering properties. When the surface irregularities or the size of the monomers is below a
340 critical value, probably smaller than about $\lambda/5$ of the wavelength, they have no effect on the
341 number of necessary measurements that have already reached a constant upper limit. The
342 number of measurements evolves also with the deviation from the sphericity (D_i/D_v
343 parameter). When considering particles with size parameter greater than 2.5, to be above the
344 transition from the Rayleigh scattering regime, the number of measurements reaches its
345 maximum value for $D_i/DV > 2$ for intensity (accuracy at 1.5%) and $D_i/DV > 1.5$ for polarization
346 (accuracy at 0.5%), as shown in Figure 10.

347 For the intensity, the 1.5% accuracy seems a good compromise between the number
348 of necessary measurements and a desired accuracy in this size parameter range. The number
349 of necessary measurements seems to reach an upper limit at about 60 ± 10 . On the other hand,
350 the fractal aggregates composed of small spherules exhibit small remaining Mie oscillations in
351 the scattering curves even if the number of measurements is large. They produce the small
352 undulations of about 1% shown in Figure 7 for more than 70 number of measurements, which
353 are difficult to totally remove (more than a hundred of different orientations could then be
354 necessary). For the polarization, an upper limit of 70 ± 10 necessary measurements is enough
355 to reach an accuracy of 0.5% in absolute value.

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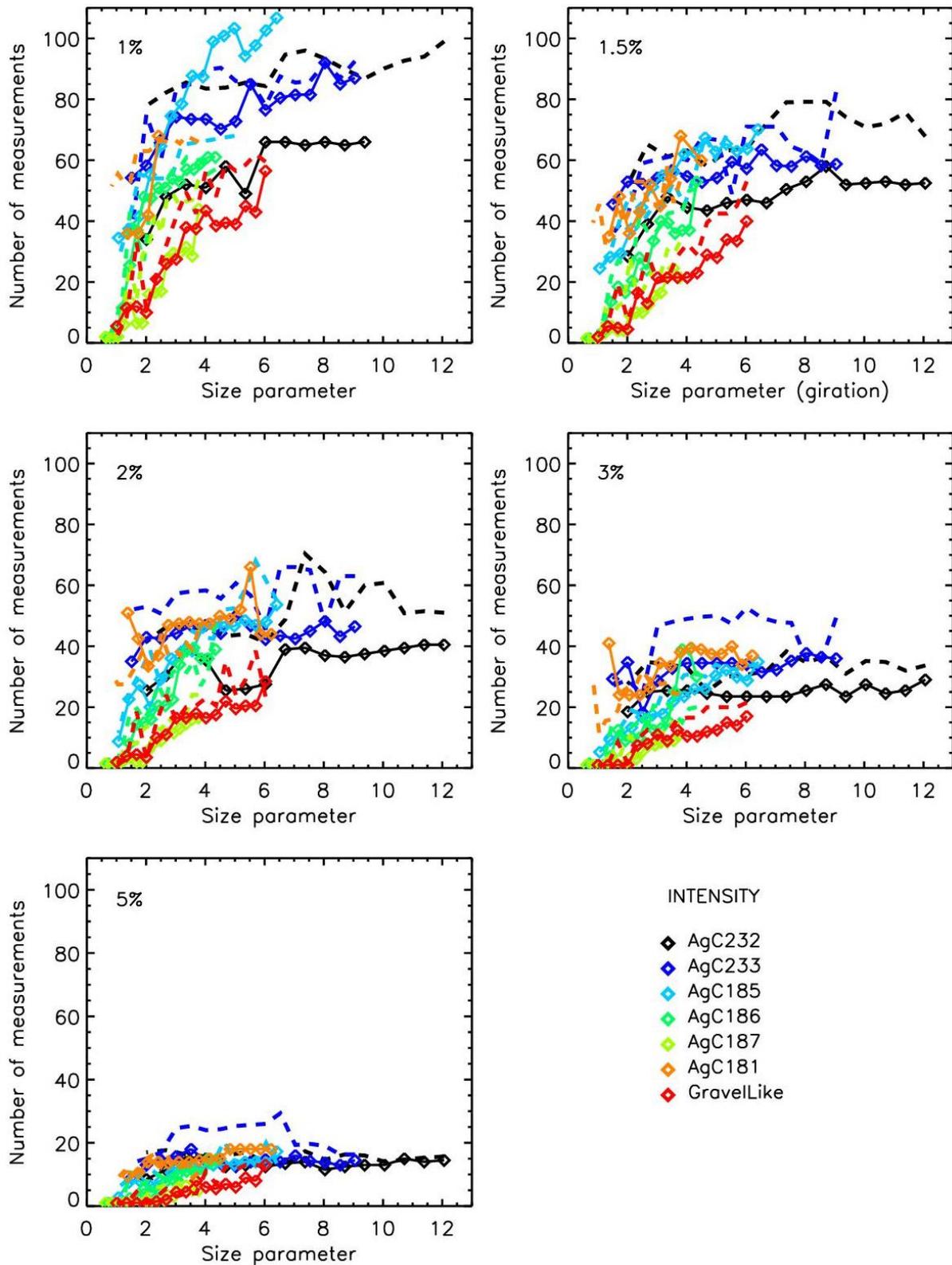
358
 359 *Figure 10: Evolution of the number of measurements as a function of deviation from*
 360 *sphericity (D_i/D_v) for particles with size parameter greater than 2.5, for intensity (accuracy at*
 361 *1.5%) and for polarization (accuracy at 0.5%)*

362
 363
 364 **4.2 Numerical calculations**

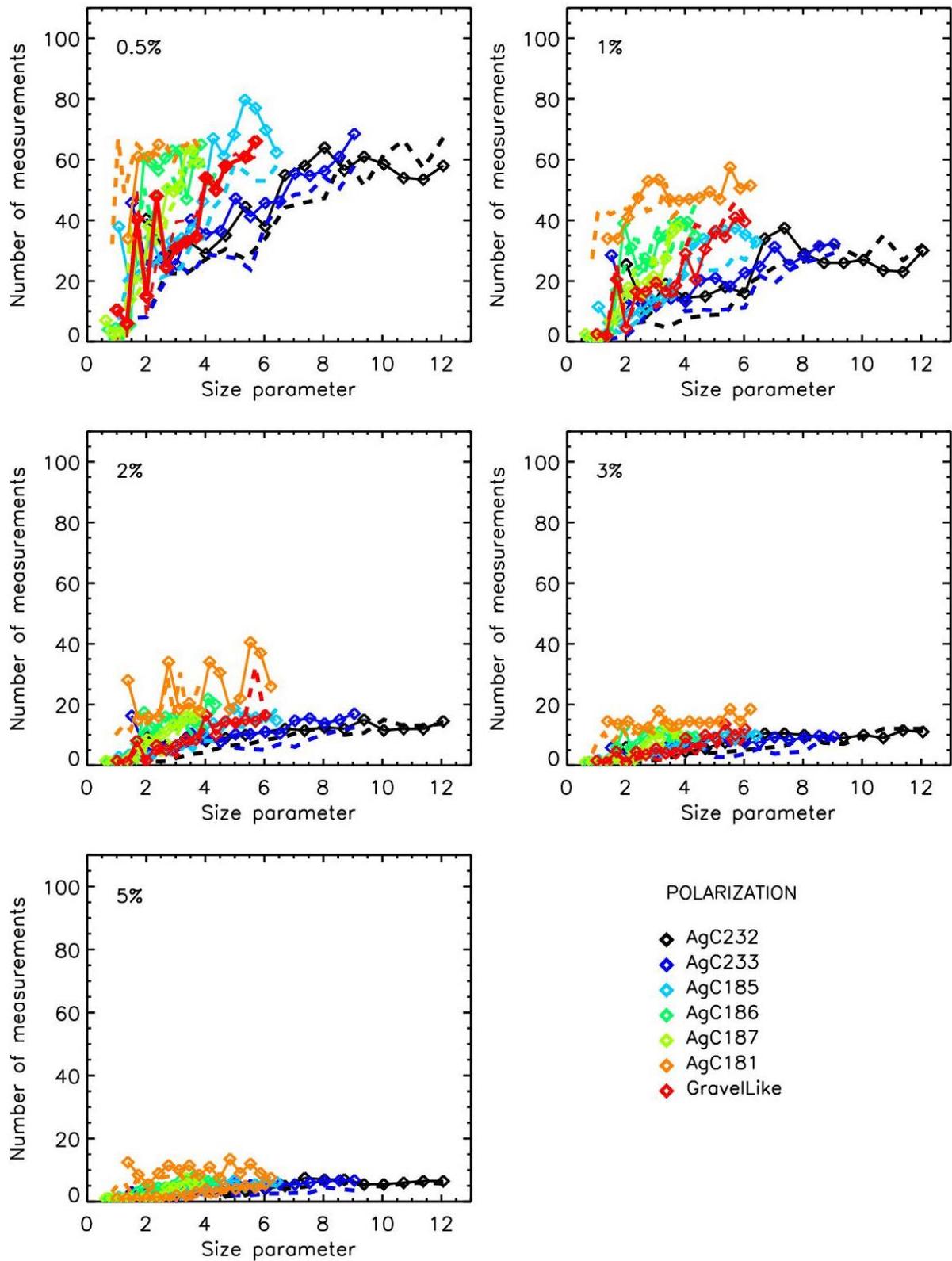
365
 366 Numerical calculations of the scattering properties of the AgC181, AgC185, AgC186,
 367 AgC187, AgC232, AgC233 and GravelLike have been conducted using the Finite Element
 368 Method presented in [30] and using the same files to describe the geometry of the objects
 369 than the one used to print them. The scattering properties are computed considering the
 370 similar orientations and inclinations of the sample as during the measurements, by steps of
 371 10° . The same method as the one used for measurements is conducted to determine the
 372 number of necessary measurements to retrieve the mean scattering properties.

373 Figure 11 shows that theoretical calculations and microwave analogy measurements
 374 have the same trends and are in correct agreement, considering the uncertainties.
 375 Nevertheless, the model for the intensity often requires a larger number of orientations to
 376 reach the same error as for the measurements. It seems that the model is more sensitive to
 377 the remaining small Mie scattering oscillations than the measurements. The noise existing
 378 naturally in the experiments can potentially help to reach more rapidly the random realization.
 379 This effect seems less visible in polarization (Figure 12), confirming that intensity and
 380 polarization are not sensitive to the same physical parameters of the particles. These results
 381 confirm that such modelling calculations can be a good approach to retrieve the mean
 382 scattering properties of fluffy and fractal particles when several tens of orientations are
 383 considered.

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385
 386 *Figure 11: Evolution of the number of necessary measurements with size parameter, to*
 387 *retrieve the intensity mean optical scattering properties, for different thresholds of*
 388 *uncertainties (the error bars of $\pm 15\%$ for the number of necessary measurements are omitted*
 389 *for clarity). Full lines: measurements; dotted lines: modelling calculations.*
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392
 393 *Figure 12: Evolution of the number of necessary measurements with size parameter, to*
 394 *retrieve the polarization mean optical scattering properties, for different thresholds of*
 395 *uncertainties (the error bars of $\pm 15\%$ for the number of necessary measurements are omitted*
 396 *for clarity). Full lines: measurements; dotted lines: modelling calculations.*
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399 The code uses a parallel direct sparse solver (Pardiso); the computation time and
400 memory requirements are strongly related to the object studied and the size of its
401 circumscribing box. Typically, for a run performed on a 28 cores SMP computer, the used
402 memory varies between 7 GB (AgC187) to 80 GB (AgC232) while the run time varies between
403 15 min (AgC187) to 11 hours (AgC232) for all frequencies. This calculation time for the
404 particles studied here (fractals with less than 75 monomers) is much lower than the tens of
405 hours necessary to retrieve experimentally the mean scattering properties from tens of
406 different orientations. Thus, this numerical approach can be used in complement with real
407 measurements to study the optical response of aggregates with various fractal dimension,
408 number of monomers, shapes, and refractive indexes. On the other hand, experimental
409 measurements could be the only solution to study particles having complex shapes (for
410 examples with different size of monomers or highly variable surface heterogeneities) and to
411 validate new numerical simulations methods or when the number of monomers or size of the
412 objects is higher.

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415 4. Conclusions

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417 The microwave analogy has allowed us to estimate how we can define the mean
418 optical scattering properties of irregularly shaped particles observed with different
419 orientations, and how they evolve with the size parameter. For all samples, the number of
420 necessary measurements is between 10 and 20 to reach a mean scattering property with an
421 uncertainty of 5% ($1-\sigma$). Then, to reach a 1.5% error in intensity and 0.5% in polarisation, the
422 number of necessary measurements strongly depends on the shape, the fluffiness, the
423 deviation from a sphere, and the surface irregularities of the sample with respect to the
424 wavelength (but not directly on their fractal dimension). For a slight rough sphere to a string-
425 like aggregate of small monomers with a fractal dimension of 1.5, the number of necessary
426 measurements increases from about 20 ± 3 to about 70 ± 10 . The higher value seems to be an
427 upper limit.

428 Such results corroborate all the previous laboratory measurements of light scattered
429 by clouds of levitating irregular randomly oriented particles where several tens of particles of
430 the same size and having a size parameter below ~ 10 are involved. They also show that
431 considering several tens of orientations of the same particle might be sufficient for considering
432 random orientation when performing modelling calculation for an aggregate composed of
433 monomers, thus reducing the calculation time. Finally, new microwave analogy
434 measurements could be conducted to better evaluate the effect of the surface roughness on
435 the mean scattering properties of compact and fluffy particles, in particular for the small size
436 parameters close to the transition to the Rayleigh scattering regime.

437

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